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## Metastable D-state spectroscopy and laser cooling of barium

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# **Metastable D-State Spectroscopy and Laser Cooling of Barium**

COVER: Red: Initial velocity distribution of thermal barium beam, Blue: Velocity distribution of barium atoms after laser cooling.

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**RIJKSUNIVERSITEIT GRONINGEN**

# **Metastable D-State Spectroscopy and Laser Cooling of Barium**

## **Proefschrift**

ter verkrijging van het doctoraat in de  
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aan de Rijksuniversiteit Groningen  
op gezag van de  
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in het openbaar te verdedigen op  
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# Chapter 1

## Introduction

In physics, the discrete symmetries Charge conjugation (C), Parity (P) and Time reversal (T) play a central role. The Standard Model (SM) [1–4] is the theory which explains the electromagnetic, weak and significant parts of strong interactions. It provides a correct description of all phenomena in particle physics observed to date<sup>1</sup>. Here gravity, which is not included in the SM, is neglected. The SM is the most successful theory we know in (particle) physics. However, it leaves a much deeper understanding open. For instance, the number of particle generations, the origin of parity violation and the fundamental fermion masses. Experimental tests of the SM are motivated to identify new physical processes that would shed light on such as yet not well understood physical facts observed in nature.

One of the urgent physics problems to be addressed is to understand the origin of the dominance of matter over antimatter in the universe, called baryogenesis. In the SM the combined charge conjugation and parity symmetry is violated and has been observed in  $K^0$  and  $B^0$  meson decay. Such CP-violation is not sufficient to explain baryogenesis [5]. This requires other sources of CP-violation. These can be explored through searches for a permanent Electric Dipole Moment (EDM) of fundamental particles. The existence of an EDM for an elementary particle would violate parity P and time reversal symmetry T. Under the assumption of an invariance of physics under the combined C, P, and T transformation (the CPT theorem [6]) a permanent EDM also violates CP.

In the Standard Model EDMs can exist. They are induced in fundamental particles through CP-violating processes as known from neutral K-meson and

---

<sup>1</sup>We leave the possible embedding of recent neutrino oscillations into the SM outside of our considerations.

B-meson systems, where they arise from higher order loops. However, they are by several orders of magnitude too small to be observed in the reach of current or planned experiments. Extensions of the Standard Model often lead to EDMs of a measurable magnitude, close to the present experimental limits. Thus, a measurement of a permanent EDM of a fundamental particle in any system (see Table 2.1) would be an unambiguous signature for physics beyond the Standard Model [7]. Because of this unique sensitivity several searches for an EDM are underway worldwide, spanning a wide spectrum of systems and experimental techniques. An experimental signature of an EDM would be the observation of a precession of the particle's spin in an imposed external electric field. Since the origin of a non Standard Model EDM is not *a priori* known, the search for this CP-violating effect must be carried out in different systems. The tightest constraints on T-violation come from atomic physics measurements. The best of these at present is an experiment which employs  $^{199}\text{Hg}$ . It yielded an upper limit on an atomic EDM of  $2.1 \times 10^{-28}$  ecm [8].

Recently, radium has been identified as a new candidate for EDM searches in neutral atoms [9]. Radium isotopes exhibit a high sensitivity to parity and time reversal violating effects due to their nuclear and atomic structure. The sensitivity to a possible EDM of the nucleons can be orders of magnitude larger than the original particle EDM. This stems from shape deformations of the nucleus. Octupole deformation leads to near degeneracy of states with opposite parity. Several isotopes of radium are known to have this property and theoretical estimates result in a factor 50-500 times enhancement for a nuclear EDM compared to the mercury atom [10, 11]. For example,  $^{225}\text{Ra}$  has a large octupole deformation in the nuclear ground state.

Radium also offers a higher sensitivity to an electron EDM due to its atomic structure [12,13]. According to the available atomic level data one can find almost degenerate levels of opposite parity states, i.e., between  $7s7p\ ^3P_1$  and  $7s6d\ ^3D_2$  states [14]. In the literature the energy difference is reported as only  $5\text{ cm}^{-1}$  and the enhancement of an electron EDM can therefore be as large as 10000 [12, 13]. This can be investigated by sensitive laser spectroscopy methods. Because of these properties radium offers a great potential for (the discovery of) a permanent electric dipole moment both of electron and nucleus.

Atomic physics has played a central role in the development of modern physics in particular in explaining the structure of atoms, in the development of quantum mechanics and the start of the most accurate field theory we know: Quantum Electro Dynamics (QED) [15]. We exploit atomic physics methods because of their potential for precision measurements. Through atomic laser spec-

troscopy, information on the nuclear charge radii, nuclear moments etc., can be obtained. Another modern atomic physics technique is laser cooling and trapping of atoms [16]. This has led to a renaissance in atomic physics. Neutral atoms can be cooled and trapped with the combination of optical and magnetic forces attaining thereby very low temperatures. Large samples of atoms can be kept substrate free and well localized in space. Almost zero first-order Doppler shift, long interaction times and high isotope selectivity can be realized. We plan to utilize atomic physics methods for EDM experiments with radioactive radium.

The rare and rather short-lived (e. g.  $^{213}\text{Ra}$ ) radioactive radium isotopes have to be produced close to the spectroscopy laboratory. The TRI $\mu$ P facility that our group is setting up at KVI, particularly provides the close proximity of isotope production and spectroscopy laboratory [17]. Using heavy-ion beams from the superconducting cyclotron AGOR we studied production of the radioactive radium isotopes. These isotopes are separated from the primary beam and other reaction products using a magnetic separator [18] that is in use since its commissioning in May 2004. The separated high-energy radium isotopes will be thermalized in a thermal ionizer stage and turned into a low-energy beam by Radio Frequency Quadrupole cooler and buncher. A low-energy beam line allows to transfer the ions to traps. They will be neutralized and trapped to carry out the high-precision experiments. This requires development of trapping methods for radium. Until recently it has not been attempted to do laser cooling and trapping on radium.

This thesis describes the first steps towards exploitation of the potential to observe symmetry violating effects in radium. In order to perform spectroscopy we have set up a laser laboratory with a variety of tunable lasers for all wavelengths of importance to these experiments. To perform EDM experiments on radium, two main scientific challenges have to be accepted:

- Determining the spectroscopic properties of radium including verification of transition wavelengths and line strengths.
- Preparing for cooling and trapping of heavy alkaline earth elements.

Because of the radioactivity of radium we have chosen to prepare the cooling and trapping with the homologue barium.

We expand the laser cooling and trapping methods to heavy alkaline earth elements barium and radium. Stable barium is well suited as a precursor for radioactive radium, because of its similar level scheme and chemical properties. We set up a laser laboratory from scratch. Semiconductor diode lasers, laser electronics were built and iodine spectroscopy was established. We investigated the

spectroscopy of metastable  $D$ -states in barium. In particular, we did the first laser spectroscopy of these states measuring the hyperfine structure and the isotope shifts. The essential input from the spectroscopy of the  $D$ -states led us to achieve for the first successful slowing of a thermal barium beam with light forces.

## Chapter 2

# Time Reversal Symmetry and Electric Dipole Moments

The fundamental discrete symmetries Charge conjugation (C), Parity (P) and Time reversal (T) are accommodated in the Standard Model [7]. The violation of parity symmetry in the weak interactions [19, 20] observed in the experimental findings can be well described in the SM but it cannot account for the physical origin of this symmetry breaking. The effects of CP or T symmetry breaking are typically small compared to the dominating electromagnetic phenomena. The observation of these effects requires state-of-the-art, high precision experiments in conducting and theoretically analyzing them. To understand these symmetries and most importantly their origin, is a driving force of experimental particle physics. Searches for a permanent electric dipole moment (EDM) of a fundamental particle belongs to this category. Such experiments play a pivotal role, because an EDM of a fundamental particle implies that the time reversal symmetry T and parity P is violated.

In the Standard Model the violation of the combined charge conjugation C and parity P first observed in the neutral  $K - \bar{K}$  meson decay is explained [23]. It can be described with a phase in the Cabibbo-Kobayashi-Maskawa matrix that accounts for quark mixing. Within the SM the observed CP-violation leads to EDMs too small to be seen in the current experiments (see Fig. 2.1). In contrast there exist other speculative theories such as Super SYmmetry (SUSY) [24], Technicolor [25, 26], Left-Right symmetric models [27] that are possible extensions to the SM. These are much favored as candidates for new physics beyond the Standard Model as the size of EDMs could be large enough to be observed at the current experimental sensitivity. An EDM could be induced in various parti-

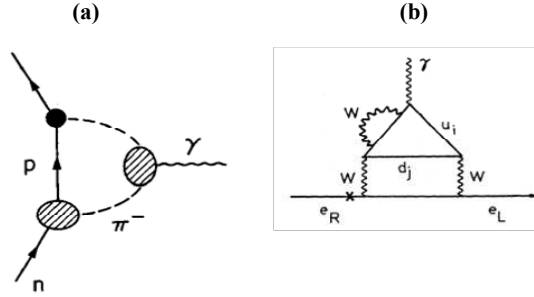


**Figure 2.1:** (a) Mechanism responsible for the electric dipole moment of the neutron in a one-loop diagram in the Standard Model. The dark blob indicates the CP-violating  $\pi NN$  interaction [figure from [21]]. (b) An example of a three-loop quark contribution to the electron-photon vertex in the Standard Model. The cross denotes a mass insertion [figure from [22]].

cles and bound systems that is sufficiently large to be observed experimentally. In general this will be the case for any model that predicts fermion compositeness and new particles (see Fig. 2.2). To explore all these possibilities we need to investigate several experimentally accessible systems to unravel the underlying SM extensions.

## 2.1 Electric Dipole Moment

A fundamental particle can have an intrinsic EDM only if T and P are violated [7]. The charge  $q$  separated from a charge  $-q$  by a distance  $r$  creates an EDM  $D \sim qr$ . The violation of fundamental discrete symmetries P and T by a particle with a permanent EDM is depicted in Fig. 2.3. The EDM would necessarily lie along its spin axis because only one vector can be defined in a quantized system. Under space inversion ( $P : r \rightarrow -r$ ) the quantity  $D$  changes direction and the particle spin direction remains unchanged. Time reversal ( $T : t \rightarrow -t$ ) changes the spin direction but leave the  $D$  direction unchanged. This results in a particle with the opposite direction of the EDM relative to the spin. Thus, if time reversal is a good symmetry, particles with spin would produce degenerate states with either  $D$  aligned parallel or anti-parallel to the spin.

### Principle of Measurement

The most frequently applied experimental method to measure the dipole moment  $d$  of a particle is to apply an electric field  $\mathbf{E}$  perpendicular to the particle spin

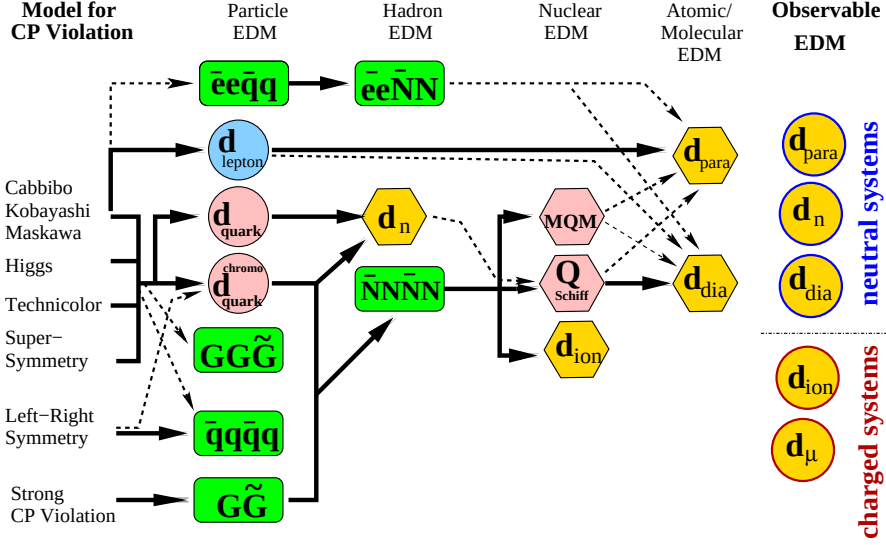


**Figure 2.2:** A variety of theoretical speculative models exist in which an EDM could be induced in fundamental particles and composite systems. Through different mechanisms or a combination of them, an EDM would be experimentally accessible[from [28]].

(ensemble average) and look for spin precession. In practice, most of the EDM experiments employ the scheme developed by Purcell et al. [29]. The particle with spin  $I$  under study is placed in external electric and magnetic fields  $\mathbf{E}$  and  $\mathbf{B}$ . The moment  $d$  interacts with electric field  $\mathbf{E}$ . This contribution can be separated from the magnetic interaction term  $-\mu \cdot \mathbf{B}$  by reversing the electric field relative to the magnetic field. The Hamiltonian for the interactions of the magnetic and electric dipole moments for such a particle may be written as

$$H = (-d\mathbf{E} + \mu\mathbf{B}) \cdot \frac{\mathbf{I}}{|\mathbf{I}|}, \quad (2.1)$$

where  $d$  and  $\mu$  are the electric and magnetic dipole moments and  $I$  is the spin. The magnetic dipole moment  $\mu$  is defined as

$$\vec{\mu} = g \frac{e\hbar}{2mc} \mathbf{I}, \quad (2.2)$$

where  $e$  is the charge of the electron,  $g$  is the gyromagnetic ratio,  $\hbar = h/2\pi$ ,  $h$  is Planck constant,  $\mathbf{I} = (\hbar/2) \cdot \boldsymbol{\sigma}$ ,  $\boldsymbol{\sigma}$  is Pauli matrix,  $m$  is the mass of a particle,  $c$  is the velocity of light, and  $\vec{I}$  is the spin of a particle. In Dirac's theory, the gyromagnetic ratio  $g$  is 2 for point-like spin 1/2 particles. Radiative corrections

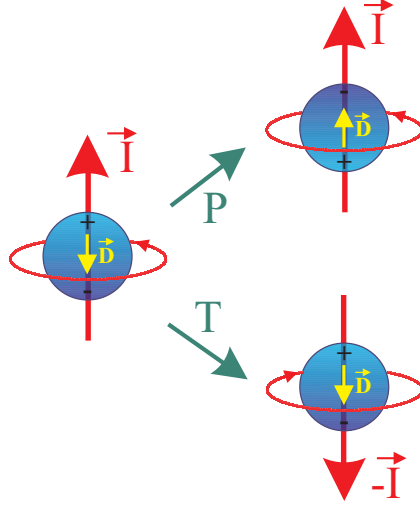


**Figure 2.3:** Particle with spin angular momentum  $I$  and dipole moment  $D$  under parity  $P$  and time reversal  $T$ . In each case, the result produces a different state with either EDM parallel or anti-parallel to  $I$ .

give rise to the anomalous magnetic moment  $a = (g - 2)/2 \simeq \alpha/2\pi \simeq 1/860$ . This deviates from the Dirac value of 2 and the deviation can be explained using quantum electro dynamics (QED). For composite objects the  $g$ -factor reflects the internal structure of these systems.

Analogously, the electric dipole moment is defined as

$$\vec{d} = \eta \frac{e\hbar}{2mc} \mathbf{I}, \quad (2.3)$$

where  $\eta$  is the quantity that is derived from the atomic physics experiments. In Dirac theory  $\eta$  is zero. In the SM  $\eta \neq 0$ , but is too small to be observed in presently possible experiments. Only in SM extensions measurable values have been predicted.

If  $t$  is replaced by  $-t$ ,  $\mathbf{E}$  does not change sign whereas  $\mathbf{B}$  and  $\mathbf{I}$  both do. Therefore, under time reversal symmetry the Hamiltonian  $H$  changes if  $d \neq 0$ . Experimentally, the difference in Larmor precession frequencies with the electric field parallel (+) and anti-parallel (−) to the magnetic field can be measured. The difference is the EDM signal given by

$$\delta\nu = \frac{2dE}{h} \frac{\mathbf{I}}{|\mathbf{I}|}, \quad (2.4)$$

$\nu$  is the spin resonance frequency.

The measurement of an EDM is a precession frequency measurement like an atomic clock. The magnetic part is well known and the small electric moment contribution has to be determined. For this atomic physics methods are well suited. An important parameter in an EDM experiment is the ultimate sensitivity of the method. The sensitivity  $\delta d$ , which is the statistical uncertainty in the measured EDM is given by

$$\delta d = \frac{\hbar}{2E\sqrt{\tau NT}}, \quad (2.5)$$

where  $E$  is the magnitude of the applied effective electric field,  $\tau$  is the spin coherence time,  $N$  is the number of atoms under study and  $T$  is the total measurement time.

The first experimental search for a permanent EDM of a fundamental particle, the neutron, was initiated almost six decades ago by Purcell and Ramsey [29]. This has led to numerous experimental measurements in various systems as well as theoretical predictions. A review on experimental and theoretical developments on EDMs has been given by Khriplovich and Lamoreaux [7] and several recent articles [22, 30, 31]. Various systems are being studied, aiming at different sensitivities to search for EDMs. Among them, the neutron and the electron take the larger share of the experimental activity.

## 2.2 EDM in Neutral Particles

### 2.2.1 The Neutron

In the initial neutron EDM experiment by Smith et al. [32] a neutron beam was used. It was guided along crossed  $\mathbf{E}$  and  $\mathbf{B}$  fields. The precession frequency of the neutron spin was measured by the method of separated oscillatory fields [33]. The main limitation in these experiments arose from the motional magnetic field, as  $\mathbf{B}$  and  $\mathbf{E}$  were not aligned perfectly along the entire length of the apparatus. With time, experimenters used ultra-cold neutrons that can be stored in magnetic bottles for hundreds of seconds. In bottle experiments motional magnetic fields average out during the course of the measurement, since the neutrons average velocity is close to zero.

The two currently active neutron EDM experiments are carried out by the Gatchina and Grenoble groups in addition to the plans at PSI and LANSCE. The Gatchina group reported a value of  $d_n = (2.6 \pm 4.0 \pm 1.6) \times 10^{-26}$  ecm for the neutron EDM with separate statistical and systematic errors [34]. They used two

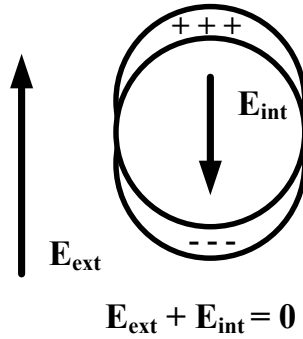


**Figure 2.4:** When an atom is placed in an external electric field the electron cloud moves so as to produce a net zero field at the nucleus. The atom is polarized in the field so that, relative to the original charge distribution, the up-field direction is more positive and the down field is more negative in charge. This induced charge distribution produces an internal field from the positive to the negative charges in a direction to cancel the external field. The cancellation is exact for a point nucleus. But in the case of atomic nucleus, which has a finite size the situation is more complex [from [31]].

adjacent neutron bottles with opposite electric fields to cancel common magnetic field drifts. In 2006, the Grenoble group obtained a result  $d_n < 3.0 \times 10^{-26} \text{ ecm}$  [35]. They used ultra cold neutrons stored in traps. Using a cohabiting atomic-mercury magnetometer spurious signals from the magnetic field fluctuations were reduced. These results give an adopted upper limit for the neutron EDM, which is  $< 3 \times 10^{-26} \text{ ecm}$ .

## 2.2.2 Atoms

### Paramagnetic Atoms

In the case of neutral atoms a quantity called nuclear Schiff moment [36] plays an important role. If we consider the neutral atom to have a point-like nucleus, then an external electric field at the nucleus is screened exactly due to rearrangement of the atomic electrons, which leads to no linear Stark effect (see Fig. 2.4). When this theorem is applied to neutral atoms the assumptions are not exactly satisfied because: the atomic electrons are relativistic, the atomic nucleus has a finite size and the electromagnetic interaction between electrons and nucleus has magnetic components. This gives rise to the Schiff moment. It consists of two parts: one is due to the P and T-violating nucleon-nucleon interaction and the other is due to core polarization via proton-neutron strong residual interaction the proton dipole moment contributes to the Schiff moment [31]. Therefore a measured atomic

EDM is a combined manifestation of these effects.

After the discovery of CP-violation in  $K$  mesons [23] it was pointed out by Sandars [37] that in heavy atoms magnetic spin-orbit interactions become quite significant. Careful calculations revealed that in heavy paramagnetic atoms the atomic EDM increases as

$$d_a \propto d_e \alpha^2 Z^3. \quad (2.6)$$

Detailed calculations show that the atomic EDM actually can be several hundred times larger than the EDM of a bare electron.

To measure the electron EDM the Berkeley group used a thermal atomic beam of thallium, a heavy paramagnetic atom and employed the technique of Ramsey separated oscillatory fields to measure the Larmor frequency [38, 39]. The motional magnetic fields were cancelled by using two oppositely directed atomic beams going through the same set of entrance and exit slits, which allows for cancellation of the first-order motional field associated with misalignment of  $\mathbf{B}$  and  $\mathbf{E}$  fields. They reported a limit on the electron EDM of  $d_e < 1.6 \times 10^{-27}$  ecm [39].

### Diamagnetic Atoms

The other type of atoms used to search for electric dipole moments associated with the nuclei are diamagnetic atoms. They have zero electronic angular momentum in the ground state and non-zero nuclear spin. Here, again the Schiff moment plays a dominant role. The Schiff moment induces an atomic EDM that is smaller than the EDM of a bare nucleus. But, it was pointed out by Khriplovich and co-workers [40] that in addition to the EDM of the valence nucleon the Schiff moment has a large contribution from the CP-odd nuclear forces that add coherently in the nucleus.

The most sensitive experiments of this type were carried out with the  $^{199}\text{Hg}$  atoms at the university of Washington, Seattle. The  $^{199}\text{Hg}$  atom is a good system in various aspects because it is heavy ( $Z = 80$ ) (see Eq. 2.6), has spin  $I = 1/2$ , has long coherence time and can be probed optically. The experimental limit of  $d(^{199}\text{Hg}) = 2.1 \times 10^{-28}$  ecm is the best limit for an EDM to date [8, 41]. Recently, the experiment has been upgraded to improve the sensitivity by a factor of 5.

### 2.2.3 Symmetry Violation in Radium

#### Electric Dipole Moment

Radium, a heavy alkaline earth radioactive element, has attracted much theoretical as well as experimental attention in the recent past. Calculations show that it

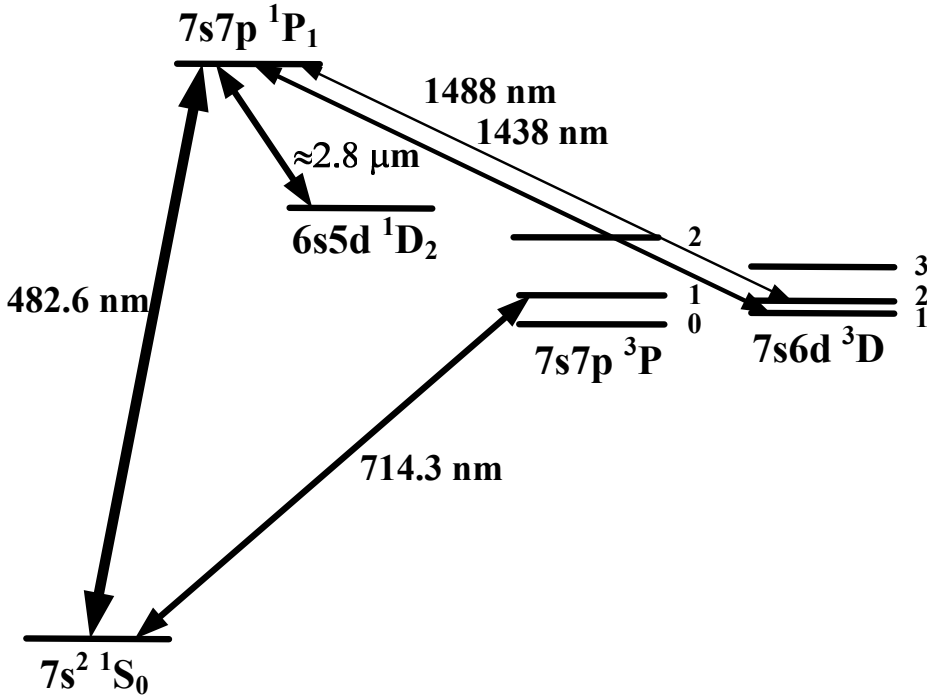


**Figure 2.5:** Level diagram of the radium atom with the wavelengths (vacuum) in nm of the transitions relevant for laser spectroscopy, laser cooling and trapping and for an EDM experiment. The wavelengths correspond to spectroscopic data given in reference [14]. The thickness of the lines indicate the strength of the transitions.

is a potential candidate for searches of a permanent EDM because of its electronic and nuclear structure. This is because the effects of time invariance and parity violation are inversely proportional to the distance between opposite parity energy levels [9, 12, 13]. In the radium atom the  $7s6d \ ^3D_2$  and the state  $7s7p \ ^3P_1$  are separated only by  $5 \text{ cm}^{-1}$  which are based on a single measurement by Rasmussen in 1934 [42, 43]. The energy level scheme (Fig. 2.5) is according to the available atomic level data [14]. The other factors that lead to enhancement in radium are the large value of the nuclear charge  $Z = 88$  (the P and T-odd effects increase with  $Z$  faster than  $Z^2$ ). Some of the radium nuclei have octupole deformation. This can be observed in the nuclear level scheme of  $^{225}\text{Ra}$  nucleus, which is shown in Fig. 2.6. The collective T, P-odd moments in octupole deformed nuclei are expected to be much greater than the same moments with spherical nuclei.

The atomic electric dipole moment appears due to the interaction between the

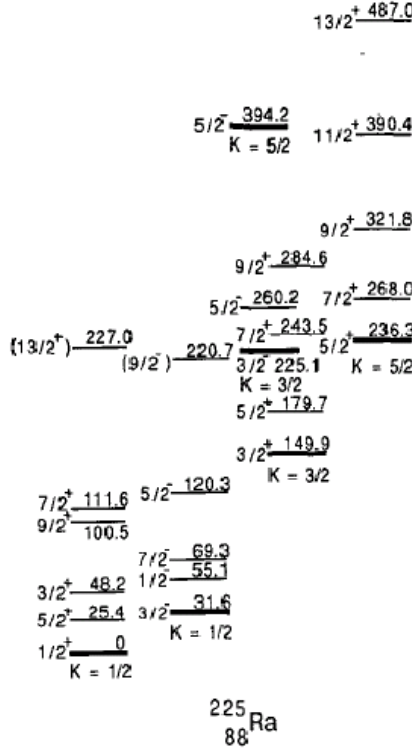


**Figure 2.6:** Nuclear level scheme of the  $^{225}\text{Ra}$  atom according to reference [44]. The opposite parity states are due to the symmetry breaking nuclear shape of octupole deformation for an oblate nucleus.

atomic electrons and the nuclear T, P-odd moments like the magnetic quadrupole moment, the Schiff moment and the electric octupole moments. These effects can also appear due to P, T-odd nuclear spin-dependent electron-nucleus interactions. The atomic EDM  $d$  is given as [9]

$$d = 2 \frac{\langle 7s6d_{3/2}, J = 2 | H_M | 7s7p_{1/2}, J = 1 \rangle \langle 7s7p_{1/2}, J = 1 | -er | 7s6d_{3/2}, J = 2 \rangle}{E_1 - E_2}, \quad (2.7)$$

where  $H_M$  is the Hamiltonian of the magnetic interaction between the nuclear magnetic quadrupole moment and the atomic electrons,  $E_1 = 13994 \text{ cm}^{-1}$  ( $7s6d \ J^P = 2^+$ ) and  $E_2 = 13999 \text{ cm}^{-1}$  ( $7s7p \ J^P = 1^-$ ) and  $-er$  is the dipole operator between  $p$  and  $d$  orbitals. The atomic EDM produced by the T, P-odd nucleon-nucleon interaction [9] is

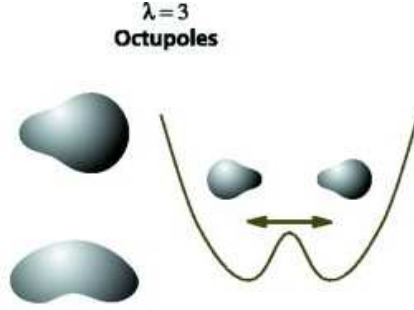


**Figure 2.7:** An example of a nucleus with an octupole deformation. The opposite parity states exist in symmetry breaking pear-shaped nucleus, which has no reflection symmetry [taken from [45]].

$$d \simeq 2 \times 10^{-20} \eta \text{ ecm} \quad (2.8)$$

where  $\eta$  is a dimensionless constant for the T, P-odd nucleon-nucleon interaction.

The atomic EDM can also be produced by the nuclear Schiff moment. The enhancement is due to the configuration mixing between the  $7s$  and  $7p_{1/2}$  states (for example, include mixing between the  $7s6d$  and  $7p7p$  configurations). The estimate of the atomic EDM due to the Schiff moment contribution comparable to the magnetic quadrupole contribution has been calculated [9] as

$$d \simeq 5 \times 10^{-20} \eta \text{ ecm}. \quad (2.9)$$

The above value is  $10^5$  times larger than the EDM of the Hg atom that was measured [8] that gives the present best limit on  $\eta$ , which is  $4.2 \times 10^{-9}$ .

The octupole deformation of some of the radium nuclei, e.g.,  $^{225}\text{Ra}$ , may lead to a factor 50 - 500 enhancement of the nuclear Schiff moment w.r.t the  $^{199}\text{Hg}$  atom [11, 46–49]. In this case, the nucleus has no longer the reflection symmetry, which can be seen in Fig. 2.7. Due to this one can observe rotational bands having states with positive and negative parities linked by strong dipole transitions [44]. The octupole deformation will have large Schiff moments because a large number of nucleons will contribute to the moments. In Fig. 2.8 we show the potential of radium with respect to the upper limits set by the experiments with various systems and constraints set on various models.

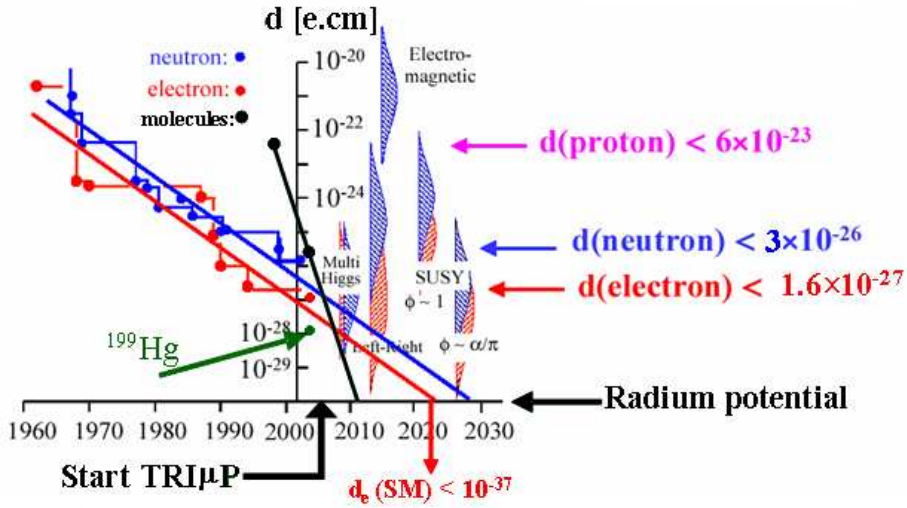


**Figure 2.8:** The progress in the experimental upper limits on EDMs in various systems with time. The values shown are the best upper limits for proton, neutron, electron and mercury experiments and their constraints on various theories beyond the Standard Model. The Standard Model limit for the electron is shown with the potential of radium in this competitive field [50].

### Parity Non-Conservation Effects

For completeness we mention further the advantages of radium in the field of parity studies, because this research may also benefit from cold and trapped atoms. The SM can be verified by the observation of neutral currents through parity non-conservation in atoms. The neutral weak interaction that does not conserve parity is mediated by the exchange of a  $Z^0$  boson between an electron and a quark inside the nucleus. In the SM, this spin-independent weak interaction is described by a Hamiltonian given by [51]

$$H_{PNC} = \frac{1}{2\sqrt{2}} G_F Q_W \gamma^5 \rho(r), \quad (2.10)$$

where  $G_F$  is the Fermi constant,  $\rho(r)$  is the nuclear density, and  $\gamma^5$  is the Dirac matrix. The weak charge of the nucleus,  $Q_W$ , is analogous to the electric charge for the electromagnetic interaction. For an atom with atomic number  $Z$  and  $N$  neutrons, the value of  $Q_W$  [52] at tree level<sup>1</sup> without radiative corrections is given

<sup>1</sup>Tree level is the lowest order of the theory

by

$$Q_W = Z(1 - 4\sin^2 \theta_W) - N, \quad (2.11)$$

where  $\theta_W$  is the Weinberg angle. This  $\gamma$ - $Z^0$  mixing angle  $Q_W$  is the atomic physics parameter to be measured and compared to the standard model. Any deviation from the tree-level value of  $Q_W$  indicates or signals a breakdown in the standard model predictions.

Atomic parity non-conservation (PNC) also occurs due to the nuclear anapole moment [51]. The anapole moment is a parity-odd, time reversal-even moment of the  $E1$  projection of the electromagnetic current. It is enhanced in heavy nuclei, increasing as  $A^{2/3}$ , where  $A$  is the mass number, and thus increasing in proportion to the nuclear surface area. The net result is that anapole contributions to the weak radiative correction will dominate. The Colorado group [53] reported PNC measurement in  $^{133}\text{Cs}$  at 0.35% level, from which a nuclear spin dependent PNC signal was extracted. This spin dependence is well above the expected signal from  $Z^0$  exchange alone but is comparable with the enhancement expected from anapole effects. For atomic parity violation a higher experimental accuracy might be possible from experiments using francium isotopes and isotopes of barium and radium ions.

Radium can be used to measure the parity non-conservation effects. The closeness of the opposite parity levels and the large nuclear charge  $Z$  give rise to the parity-violating amplitude  $E1_{PV}$  in radium about 100 times larger than a similar amplitude in cesium [9, 12]. They find that this anapole induced amplitude makes a dominant contribution to the total parity-violating amplitude in radium. This may be an advantage, since in cesium the anapole contribution has to be separated from the weak charge contribution, which is two orders of magnitude larger than the anapole contribution. The weak charge contribution in radium can be neglected because the states  $7s7p\ ^3P_1$  and  $7s6d\ ^3D_2$  have different electron angular momenta  $J = 2$  and  $J = 1$  and can not be mixed by the weak nuclear spin independent interaction.

Due to the above discussed advantages our group works towards an EDM experiment with several radium isotopes [54]. Also a group at the Argonne National Laboratory, USA has plans to use  $^{225}\text{Ra}$  for the same purpose [55]. Since the EDM of radium depends on the atomic and nuclear properties we give a review of the known experimental data and theoretical efforts in Chapter 4. Especially theorists have become interested in radium. Their results show the importance of the knowledge of the atomic wavefunctions in order to evaluate the sensitivity to an EDM and the available experimental data is not yet sufficient.

### 2.2.4 Molecules

Molecules known as polar molecules have large intrinsic EDMs. They are used to search for EDM of an electron [22]. In a simplified picture of a molecule, they have pairs of nearly degenerate states with opposite parity. Since their energy splitting is less than the thermal energy  $kT$  at room temperature, the two states act practically like a two-fold-degenerate ground state. When an external electric field  $\mathbf{E}$  is applied, the two states of opposite parities,  $|+\rangle$  and  $|-\rangle$ , mix with each other and form new energy eigenstates,  $|r\rangle \simeq (|+\rangle + |-\rangle)/\sqrt{2}$  and  $|l\rangle \simeq (|+\rangle - |-\rangle)/\sqrt{2}$  with energy eigenvalues [22]

$$E_{r,l} = \frac{1}{2}(E_+ + E_-) \pm \left[ \frac{1}{4}((E_+ - E_-)^2 + (e\langle \mathbf{r} \rangle \cdot \mathbf{E})^2) \right]^{1/2}, \quad (2.12)$$

where  $\langle \mathbf{r} \rangle$  is the transition-matrix element between  $|+\rangle$  and  $|-\rangle$ . Because  $E_{\pm}$  are almost degenerate,  $e\langle \mathbf{r} \rangle \cdot \mathbf{E}$  dominates inside the square root term in Eq. 2.12, and the energy eigenvalues are given approximated by

$$E_{r,l} = \frac{1}{2}(E_+ + E_-) \pm e\langle \mathbf{r} \rangle \cdot \mathbf{E}, \quad (2.13)$$

In this approximation the energy shift is linearly dependent on  $\mathbf{E}$ , the proportionality constant is traditionally called the “permanent EDM” of the molecule. But, this EDM is not an indication of P and T- violation. If measurements are carried out with an infinitesimally weak  $\mathbf{E}$  at zero temperature, one will find only a quadratic dependence of the energy eigenvalues on  $E$ , that is,

$$E_{r,l} = E_{\pm} \pm (e\langle \mathbf{r} \rangle \cdot \mathbf{E})^2 / (E_+ - E_-) + \dots \quad (2.14)$$

by a power series expansion in  $\mathbf{E}$ . Thus there is no linear dependence on  $\mathbf{E}$  of the energy shift. If  $T$  invariance holds, a molecule acquires only an induced EDM, which is enhanced by a small energy difference between the opposite parity states. But a permanent EDM causes a linear Stark effect even for an infinitesimally weak  $\mathbf{E}$ , which is a genuine signature of P and T-violation.

Two types of polar molecules, called paramagnetic and diamagnetic, are being used in the search for electron EDMs. In these molecules, large polarization can be achieved with modest external fields that greatly enhance an EDM. The observable energy shifts caused by an electron EDM may be 100-1000 times greater than in paramagnetic atoms. Paramagnetic molecules are obtained in two ways. One way is by using molecules that are paramagnetic in their ground states e.g., thallium fluoride (TlF) [56] and ytterbium fluoride (YbF) [57]. The other way is by populating a paramagnetic state of a diamagnetic molecule in the ground

state e.g., lead oxide (PbO) [58]. The former are thermally and chemically unstable (chemical radicals), and the experiments must be carried out in beams. This is a constraint to achieve a high production rate. The sensitivity reported for the YbF molecule measurement is  $d_e = (-0.2 \pm 3.2) \times 10^{-26}$  ecm, limited by counting statistics, with the future possible sensitivity level of  $1 \times 10^{-27}$  ecm [57].

The second approach uses diamagnetic molecules that are pumped to a paramagnetic state. The advantage is that these molecules are thermally and chemically stable. They can be stored in a cell, which gives this experiment a large advantage in sensitivity due to the large number of molecules. With a proposed ultimate sensitivity of  $d_e < 10^{-31}$  ecm for PbO molecule, the EDM limit can be improved by 4 orders of magnitude below the current limit set by the thallium experiment [58].

## 2.3 EDM in Charged Particles

One might assume that charged objects, like ions would not be suited for searches for EDMs since they are accelerated when placed in an electric field. Recently, a very novel idea was introduced for measuring an EDM of a charged particle directly. This idea originates from the muon g-2 experiment [59, 60]. For such experiments the high motional electric field is exploited, which a charged particle at relativistic speed experiences in a magnetic storage ring. In such a setup the Schiff theorem can be circumvented, because of the non-trivial geometry of the problem [31]. With an additional radial electric field in the storage region the spin precession due to the magnetic moment anomaly can be compensated, if the effective magnetic anomaly  $a_{eff}$  is small, i.e.,  $a_{eff} \ll 1$ .

### 2.3.1 Muon

The principle described above was first considered for muons. For longitudinally polarized muons injected into the storage ring an EDM would express itself as a spin rotation out of the orbital plane. The spin orientation can be found by measuring the distribution of electrons from muon decay. An EDM would appear as time dependent change of ratio of the counting rate above or below the orbit plane. The upper limit reported on muon EDM is  $< 2.8 \times 10^{-19}$  ecm [61]. For the possible muon beams at the future J-PARC facility in Japan a sensitivity of  $10^{-24}$  ecm is expected [60, 62]. In such an experiment the flux is a major limitation. For models with non-linear mass scaling of EDM's such an experiment would already be more sensitive to some certain new physics models than

| Particle          | Limit / Measurement<br>[ecm]         | Standard Model<br>Limit<br>[factor to go] | New Physics<br>Limit<br>[factor to go] |
|-------------------|--------------------------------------|-------------------------------------------|----------------------------------------|
| e                 | $<1.6 \times 10^{-27}$ (90% CL) [39] | $10^{11}$                                 | $\lesssim 1$                           |
| $\mu$             | $<2.8 \times 10^{-19}$ (95% CL) [61] | $10^8$                                    | $\lesssim 200$                         |
| p                 | $-3.7(6.3) \times 10^{-23}$ [66]     | $10^7$                                    | $\lesssim 10^5$                        |
| n                 | $<3 \times 10^{-26}$ (90% CL) [35]   | $10^4$                                    | $\lesssim 30$                          |
| $^{199}\text{Hg}$ | $<2.1 \times 10^{-28}$ (95% CL) [8]  | $\lesssim 10^5$                           | various                                |

**Table 2.1:** Limits on permanent EDMs  $d$  for electrons ( $e$ ), muons ( $\mu$ ), protons ( $p$ ), neutrons ( $n$ ) and the mercury atom ( $^{199}\text{Hg}$ ).

the present limit on the electron EDM. For certain left-right symmetric models a value of  $d_\mu$  up to  $5 \times 10^{-23}$  ecm would be possible. An experiment carried out at a more intense muon source could provide a significantly more sensitive probe to CP-violation in the second generation of particles without strangeness.

### 2.3.2 Deuteron

Another particle that is being considered actively in the recent past is the deuteron, the simplest nucleus [63–65]. Here an EDM could arise not only from a proton or a neutron EDM, but also from CP-odd nuclear forces. It was shown very recently that in certain scenarios the deuteron can be significantly more sensitive than the neutron. This is evident for the case of quark chromo-EDMs  $d_d^c$  and  $d_u^c$  of the up and down quarks yield  $d_D = -4.67 d_d^c + 5.22 d_u^c$  and  $d_n = -0.01 d_d^c + 0.49 d_u^c$ . Due to its rather small magnetic anomaly the deuteron is a particularly interesting candidate for a ring EDM experiment and a proposal with a sensitivity of  $10^{-27}$  ecm exists [60]. In this case scattering off a target will be used to observe a spin precession.

Some experimental upper limits on EDMs in various systems with different sensitivities are summarized in Table 2.1.

### 2.3.3 T-violation Searches other than EDMs

Time reversal violation (TRV) searches are also being carried out other than through the search for EDMs [67]. The approach that is complementary for searching of TRV is through certain angular correlation studies in  $\beta$ -decays [68–70]. In  $\beta$ -neutrino correlation studies the  $R$  and  $D$  coefficient (for spin polarized nuclei) [68] offers to observe new interactions in a region of potential new physics

which is similar to EDM searches. With the combined use of laser cooling and trapping technique and recoil ion momentum spectroscopy [71], the investigation of  $\beta$ -decay of the radioactive species can be carried out with high precision.

## 2.4 Experimental Methods for EDM Searches

In this section the advantages of EDM experiments with cold atoms in atom traps over traditional methods is presented. So far, atomic EDM experiments have been conducted in vapor cells [8, 39] and with atomic beams [38, 72]. Both methods have their advantages and disadvantages. In the case of vapor cell experiments high atomic density could be achieved in a smaller region and with minimum motional magnetic field effect. One can achieve long spin coherence times and does not need ultra high vacuum apparatus. The disadvantage is the spurious magnetic fields due to leakage currents. In the case of beam experiments the dominant systematic error is the motional magnetic field, less interaction time but high electric fields can be applied over the length of the apparatus.

With the laser cooling and trapping technique, EDM measurements using an optical trap are superior to the above measurement methods. Over the cells, ultra high vacuum suppresses spurious magnetic fields due to leakage currents and allows for application of high electric fields. Smaller sample region also allows for large external electric field for the measurement of EDM. Over the atomic beams, zero average motional magnetic field  $\mathbf{v} \times \mathbf{E}$  and large density of atoms for larger EDM signal can be obtained. The advantages over both of the other methods are low in-homogeneity of external fields in the sample region and long spin relaxation lifetimes.

The basic EDM experimental setup using traps consists of a magneto-optical trap to initially cool and trap the atoms. Subsequently they can be transferred to an optical dipole trap (FORT - Far Off-Resonance Trap), where the actual EDM measurement would be performed. Also a fountain setup like for atomic clocks would be an option. Systematic effects due to orientation of static B field relative to the polarization axis of the FORT laser beams have been investigated in [73]. They find the linear Stark shift can be minimized for a configuration in which the magnetic field is along the z-axis,  $k$  along the x-axis, and the linear polarization of the light along the y-axis. Furthermore, in the case of nuclear spin  $I=1/2$  nuclei the quadratic Stark shift is eliminated. In the case of alkali metal atoms a potential problem for an EDM measurement in a trap are the cold collisions that cause spin relaxation and lead to frequency shifts [74, 75].

Fermionic alkaline earth elements, having closed electronic shells with a  $^1S_0$

ground state, such as  $^{213,225}\text{Ra}$  ( $I=1/2$ ), do not have these problems. When these atoms are spin polarized, the s-wave scattering cross-section will be zero, while higher scattering cross-sections minimize to zero at sufficiently low temperatures. With this, long spin coherence times could be achieved and collisional spin relaxation and three-body recombination rates could be reduced. With all these advantages EDM measurements using cold atoms and traps prove to be superior to the methods used currently.

## 2.5 EDMs As a Tool to Search for New Physics

Searches for EDMs can stringently test the Standard Model and set limits on the SM extensions or find physics beyond the SM. Many experiments are needed and possibly will allow to disentangle the findings in terms of the underlying possible SM extensions. New systems have been introduced recently for searches of EDMs. Among them, the radium atom seems to be a promising candidate because of various advantages over the systems studied and investigated currently in this competitive field. Radium allows to search uniquely both for nuclear and electron EDMs. New experimental techniques need to be developed to exploit this potential, which is shown in this thesis. This includes development of spectroscopic as well as cooling and trapping methods for neutral heavy alkaline earth atoms.



## Chapter 3

# TRI $\mu$ P Facility

In the periodic table of elements shown in Fig. 3.1 we find that most of the laser cooled and trapped species belong to the one valence electron (alkali) and two valence electron (alkaline earth) systems. Also noble gas atoms were trapped exploiting their metastable states. In addition, there are a few other species scattered over the periodic table that were trapped. The scientific goal of the cold atom research spans, to name a few, from precision spectroscopy to quantum fluids and from many-body physics to collision studies. Our interest is in the heavy alkaline earth elements barium and radium. Barium, which has stable isotopes, has an atomic level scheme similar to radium. It is available in metallic form that can be readily used for the experiments. Radium, a radioactive element has to be extracted from radioactive sources or produced by nuclear reactions for experiments at facilities like TRI $\mu$ P at KVI. The rare and short lived radium isotopes offer unique possibilities for investigating fundamental symmetries in physics complementary to high energy physics experiments.

World wide various experimental efforts are in progress using radioactive isotopes in high precision experiments to test the limits for the validity of the standard model. To provide short lived isotopes at KVI, Groningen, the TRI $\mu$ P (Trapped **R**adioactive **I**sotopes:  $\mu$ icro laboratories for fundamental **P**hysics) facility has been set up in the last 4 years [76]. The facility produces radioactive nuclides and provides them, after slowing down, as low energy ion beams to experimental areas. The facility is open to outside users and first experiments have been made. We will describe briefly the complete facility consisting of a production target, a magnetic separator, thermal ionizer, radio frequency quadrupole cooler and buncher, low energy beam line, and laser facility for neutral atom traps. The different components of the TRI $\mu$ P facility are schematically shown

|                       |                     |                       |                      |                      |                      |                       |                     |                       |                     |                      |                       |                     |                      |                      |                       |                       |                     |                      |                   |                      |                    |  |
|-----------------------|---------------------|-----------------------|----------------------|----------------------|----------------------|-----------------------|---------------------|-----------------------|---------------------|----------------------|-----------------------|---------------------|----------------------|----------------------|-----------------------|-----------------------|---------------------|----------------------|-------------------|----------------------|--------------------|--|
| 1<br>H<br>1.00794     |                     |                       |                      |                      |                      |                       |                     |                       |                     |                      |                       |                     |                      |                      |                       |                       | 2<br>He<br>4.00260  |                      |                   |                      |                    |  |
| 3<br>Li<br>6.941      | 4<br>Be<br>9.012182 |                       |                      |                      |                      |                       |                     |                       |                     |                      |                       |                     |                      |                      |                       | 5<br>B<br>10.811      | 6<br>C<br>12.0107   | 7<br>N<br>14.00644   | 8<br>O<br>15.9994 | 9<br>F<br>18.9984032 | 10<br>Ne<br>20.179 |  |
| 11<br>Na<br>22.989769 | 12<br>Mg<br>24.305  |                       |                      |                      |                      |                       |                     |                       |                     |                      |                       |                     |                      |                      |                       | 13<br>Al<br>26.981538 | 14<br>Si<br>28.0855 | 15<br>P<br>30.973761 | 16<br>S<br>32.066 | 17<br>Cl<br>35.4527  | 18<br>Ar<br>39.948 |  |
| 19<br>K<br>39.098     | 20<br>Ca<br>40.078  | 21<br>Sc<br>44.955910 | 22<br>Ti<br>47.867   | 23<br>V<br>50.9415   | 24<br>Cr<br>51.9961  | 25<br>Mn<br>54.938049 | 26<br>Fe<br>55.845  | 27<br>Co<br>58.933200 | 28<br>Ni<br>58.6934 | 29<br>Cu<br>63.546   | 30<br>Zn<br>65.39     | 31<br>Ga<br>69.723  | 32<br>Ge<br>72.61    | 33<br>As<br>74.92160 | 34<br>Se<br>78.96     | 35<br>Br<br>79.904    | 36<br>Kr<br>83.80   |                      |                   |                      |                    |  |
| 37<br>Rb<br>85.467    | 38<br>Sr<br>87.62   | 39<br>Y<br>88.90585   | 40<br>Zr<br>91.224   | 41<br>Nb<br>92.90638 | 42<br>Mo<br>95.94    | 43<br>Tc<br>(98)      | 44<br>Ru<br>101.07  | 45<br>Rh<br>102.90550 | 46<br>Pd<br>106.42  | 47<br>Ag<br>107.8682 | 48<br>Cd<br>112.411   | 49<br>In<br>114.818 | 50<br>Sn<br>118.710  | 51<br>Sb<br>121.760  | 52<br>Te<br>127.60    | 53<br>I<br>126.90447  | 54<br>Xe<br>131.29  |                      |                   |                      |                    |  |
| 55<br>Cs<br>132.90545 | Ba<br>137.327       |                       | 57<br>La<br>138.9055 | 72<br>Hf<br>178.49   | 73<br>Ta<br>180.9479 | 74<br>W<br>183.84     | 75<br>Re<br>186.207 | 76<br>Os<br>190.23    | 77<br>Ir<br>192.217 | 78<br>Pt<br>195.078  | 79<br>Au<br>196.96655 | 80<br>Hg<br>200.59  | 81<br>Tl<br>204.3833 | 82<br>Pb<br>207.2    | 83<br>Bi<br>208.98039 | 84<br>Po<br>(209)     | 85<br>At<br>(210)   | 86<br>Rn<br>(222)    |                   |                      |                    |  |
| 87<br>Fr<br>(223)     | Ra<br>226           |                       | 89<br>Ac<br>(227)    | 104<br>Rf<br>(261)   | 105<br>Db<br>(262)   | 106<br>Sg<br>(263)    | 107<br>Bh<br>(262)  | 108<br>Hs<br>(265)    | 109<br>Mt<br>(266)  | 110<br>(269)         | 111<br>(272)          | 112<br>(277)        |                      |                      | 114<br>(289)          | 116<br>(289)          | 118<br>(293)        |                      |                   |                      |                    |  |

Next species

|                      |                       |                     |                   |                    |                     |                    |                       |                    |                       |                    |                       |                    |                     |
|----------------------|-----------------------|---------------------|-------------------|--------------------|---------------------|--------------------|-----------------------|--------------------|-----------------------|--------------------|-----------------------|--------------------|---------------------|
| 58<br>Ce<br>140.116  | 59<br>Pr<br>140.90765 | 60<br>Nd<br>144.24  | 61<br>Pm<br>(145) | 62<br>Sm<br>150.36 | 63<br>Eu<br>151.964 | 64<br>Gd<br>157.25 | 65<br>Tb<br>158.92534 | 66<br>Dy<br>162.50 | 67<br>Ho<br>164.93032 | 68<br>Er<br>167.26 | 69<br>Tm<br>168.93421 | 70<br>Yb<br>173.04 | 71<br>Lu<br>174.967 |
| 90<br>Th<br>232.0381 | 91<br>Pa<br>231.03688 | 92<br>U<br>238.0289 | 93<br>Np<br>(237) | 94<br>Pu<br>(244)  | 95<br>Am<br>(243)   | 96<br>Cm<br>(247)  | 97<br>Bk<br>(247)     | 98<br>Cf<br>(251)  | 99<br>Es<br>(252)     | 100<br>Fm<br>(257) | 101<br>Md<br>(258)    | 102<br>No<br>(259) | 103<br>Lr<br>(262)  |

**Figure 3.1:** Periodic table of elements. Elements marked with arrows and square blocks are the neutral atoms that have been laser cooled and trapped. They have favorable level structures which allow cooling and trapping with a single laser from the ground state or a metastable excited state. The proposed heavy alkaline earth elements barium and radium are also shown. They require a number of additional lasers for repumping the atoms into states with suited transitions for cooling and trapping.

in Fig. 3.2.

Our group is interested to use the facility to perform experiments probing fundamental symmetries as described in Chapter 2. In particular we are interested in nuclear  $\beta$ -decay for T- violation studies and for CP-violation by searching for a permanent electric dipole moment with radium.

## 3.1 Isotope Production and Separation

### 3.1.1 AGOR

The superconducting cyclotron AGOR (*Accélérateur Groningen Orsay*), at KVI, accelerates both the light and heavy ions. For heavy ions the charge-to-mass ratio,  $Q/A$ , determines the achievable maximum energy. In Fig. 3.3 the available beams are shown. The maximum energy for protons is 190 MeV. Different sources are

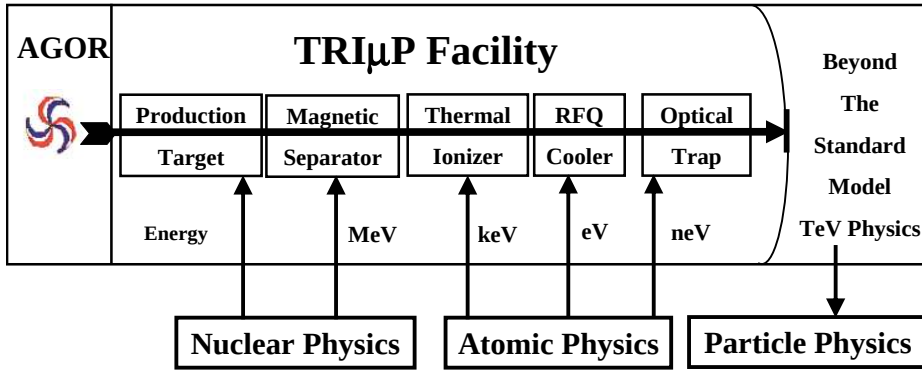


**Figure 3.2:** Conceptual view of the TRIμP facility consisting of a production target to produce radioactive isotopes by nuclear reactions with beams from the AGOR cyclotron. A magnetic separator is used for the selection of isotopes of interest. The thermal ionizer and the RFQ cooler and buncher stages are for slowing the high energy isotopes. Atom traps are for storing and for conducting high precision experiments with the goal to search for physics beyond the Standard Model [17].

available to produce a wide range of primary beams. To accelerate heavy ions like lead (Pb) the vacuum needed to be improved to be better than  $10^{-7}$  mbar in the accelerator. Overall the AGOR cyclotron is upgraded to provide beams for the TRIμP facility. The goal is 1 kW of beam for any accelerable ion. So far 100 W has been reached, which makes AGOR, a state-of-the-art cyclotron with high beam current for heavy ions.

### Production Target

A hydrogen gas target cooled to liquid nitrogen temperature is in use that is compatible with the typical beams and inverse reaction dynamics for which the separator is designed. The use of a gas target has important benefits over solid targets. The polyethylene targets used during the commissioning phase of the magnetic separator showed rapidly diminishing hydrogen content at a few tens of nA beam current. The target was built by a collaborating group at North Carolina University, USA [77].

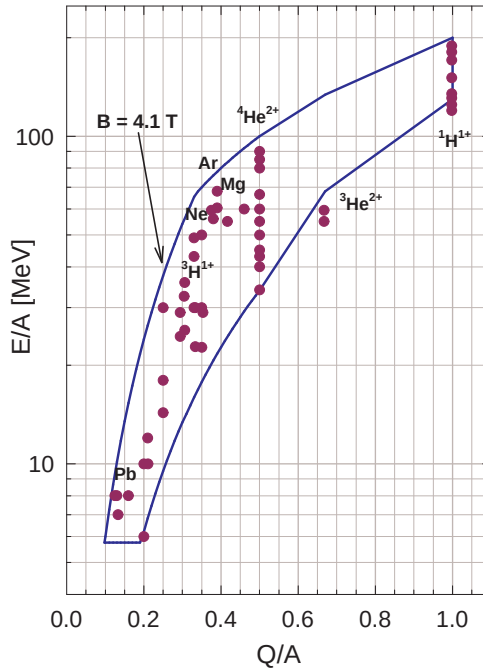


**Figure 3.3:** Operating diagram and available beams from the AGOR superconducting cyclotron at KVI. Charge-to-mass ratio ( $Q/A$ ) determines the maximum achievable energy for the ion beams. The boundary line indicates the design limit of the cyclotron for the ions that can be accelerated.

### 3.1.2 The Magnetic Separator

The different types of nuclear reactions that will be used required the design of a dual-mode combined fragment and recoil magnetic separator [17]. The separator was designed at KVI. The layout of the magnetic separator is shown in Fig. 3.4. It consists of two pairs of dipole magnets for the particle momentum selection, four pairs of quadrupoles for focussing and components such as beams stops, beam profile detection and slits. Two target positions,  $T1$  and  $T2$ , are foreseen for the different kinematics of the various production methods. It was built in 2003 and commissioned in May 2004. The magnetic separator can be operated in two modes. One is the fragmentation mode and the other is the gas-filled mode. In the fragment-separator mode, target  $T1$  is used. In the gas-filled mode of operation, for fusion-evaporation reactions gas filling of the second part is used to efficiently

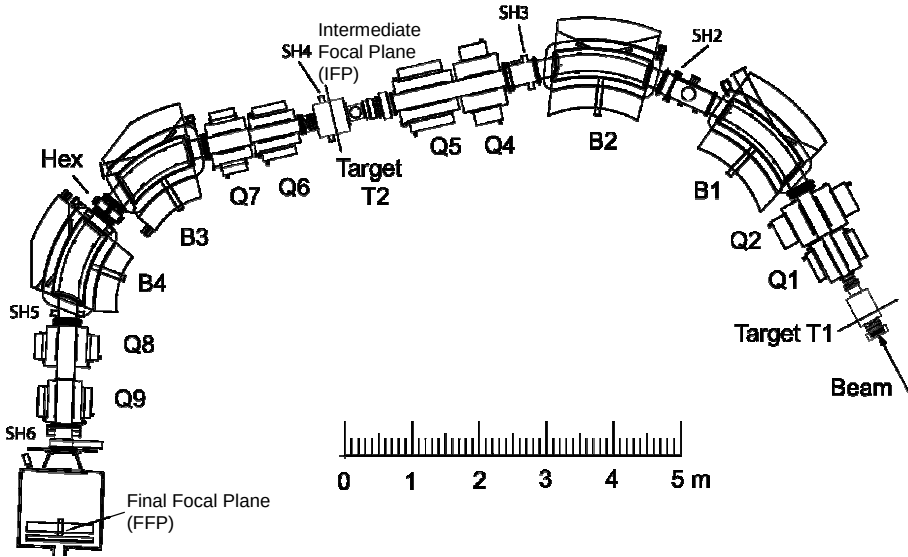


**Figure 3.4:** Layout of the TRIμP dual-mode magnetic separator. The separator can be used in fragment-separator (target position T1) and in gas-filled (target position T2) mode. B1-B4 are the dipole magnets for particle momentum selection and Q1-Q9 are the quadrupole magnets for focussing. SH1-6 are the slit systems and a hexapole Hex.

collect heavy ion products which are spread out over many atomic charge states. For this the target station T2 is used.

The TRIμP separator is made available for various physics experiments by the user community in collaboration with our group. They include production of  $^{19}\text{Ne}$  to measure the half-life for the  $^{19}\text{Ne}$  to  $^{19}\text{F}$  decay in collaboration with the North Carolina State University group [78]. An experiment to study particle unstable states in  $^{12}\text{C}$  populated in the  $\beta$ -decays of  $^{12}\text{N}$  and  $^{12}\text{B}$  was carried out. This experiment is led by the Aarhus university group [79]. The details of the magnetic separator such as design, construction and commissioning can be found in the thesis of Emil Traykov [80].

### Fragment-separator mode

The fragmentation mode was tested with the  $^1\text{H}(^{21}\text{Ne}, ^{21}\text{Na})\text{n}$  reaction in inverse kinematics. Initially, the basic ion-optical parameters are tested using the 43 MeV/nucleon  $^{21}\text{Ne}^{7+}$  beam ( $B\rho = 2.8 \text{ Tm}$ ). After successful commissioning

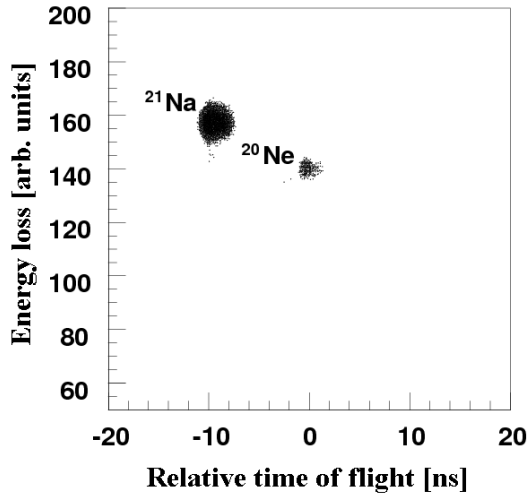


**Figure 3.5:** The time-of-flight (TOF) spectrum (relative to  $^{20}\text{Ne}$  shown at the focal plane of the magnetic separator consists of only  $^{21}\text{Na}$  and a small contamination of stable  $^{20}\text{Ne}$ , which could be reduced to below 0.5%. Here  $^{21,20}\text{Na}$  isotopes have the same magnetic rigidity ( $B\rho$ ) but different velocities. Hence  $^{21}\text{Na}$  arrives at the detector earlier than  $^{20}\text{Ne}$ .

and verification of ion optics and design parameters, the first radioactive  $^{21}\text{Na}$  isotope was produced using the (p, n) reaction in inverse kinematics with this  $^{21}\text{Ne}$  beam. A silicon detector of 20 mm diameter and thickness of 150  $\mu\text{m}$  placed in the focal plane of the magnetic separator was used to measure the yield of  $^{21}\text{Na}$ . The spectrum shown in Fig. 3.5 is the time-of-flight spectrum of particles arriving at the detector. The typical  $^{21}\text{Na}$  production rates during the commissioning were  $8 \times 10^4$  particles per second. The  $^{21}\text{Na}$  isotope beam was used for first physics experiments to measure the  $\beta$ -decay branching ratio in  $^{21}\text{Na}$  to the excited state of  $^{21}\text{Ne}$  at 350.5 keV [81]. The population of this state is of significance for  $\beta - \nu$  correlation experiments of  $^{21}\text{Na}$  planned by our group.

### Gas-filled Separator Mode

The separator has been tested in gas-filled mode. In this mode the first part of the separator acts as a beam line and the second part as a recoil separator. The vacuum (first part) and gas-filled sections (second part) are separated by a thin Havar foil of 2.5  $\mu\text{m}$ . This mode is required for the production of radioactive radium isotopes for the EDM experiment. For example,  $^{213}\text{Ra}$  can be produced by the fusion-evaporation reaction in inverse kinematics. In these reactions the products

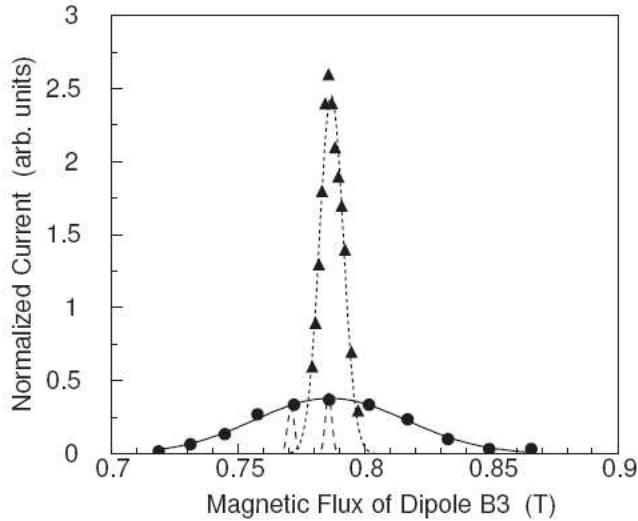


**Figure 3.6:** Experimental charge-state distribution of a  $^{206}\text{Pb}$  beam in the focal plane of the TRI $\mu$ P separator setup. The full circles represent the charge state distribution of  $^{206}\text{Pb}$  beam without gas filling of the second part of the separator after passing the Havar foil and the carbon target. The full triangles represent the all charge states of  $^{206}\text{Pb}$  beam concentrated in a single peak with the gas filling of the second part of the separator.

are not fully stripped. They leave the target with charge states that are widely distributed. The gas in the second part of the separator causes reaction products to loose and gain electrons rapidly. As a result they move along a trajectory with an effective average charge. It also provides additional differential stopping to separate primary beam and the recoil products further.

To test the separator in gas-filled mode a  $^{206}\text{Pb}^{29+}$  beam of 8.4 MeV/nucleon is passed through a Havar window of  $2.5\ \mu\text{m}$  thickness before impinging on a  $4.2\ \text{mg}/\text{cm}^2$  thick  $^{12}\text{C}$  target.  $^{206}\text{Pb}$  instead of the more abundant  $^{208}\text{Pb}$  was used because the  $^{206}\text{Pb}(^{12}\text{C}, 5n)^{213}\text{Ra}$  has the maximal cross-section. By passing through the Havar foil the lead beam is stripped to a distribution of charge states centered near  $60^+$ . The inverse kinematics allows  $^{213}\text{Ra}$  recoils to get out of the carbon target. The results of the testing of the separator in gas-filled mode are shown in Fig. 3.6. The charge distribution of the lead beam after passing the Havar foil and the carbon target without gas filling is given by the full circles. A total of eleven charge states were observed with a maximum intensity at  $q = 60$ . A Gaussian distribution fit with a full width at half maximum (FWHM) of 9%

$\delta p/p$  is shown as a line through the points. A more detailed measurement over two charge states indicated by the two dashed Gaussian curves, show a FWHM of 0.37%  $\delta p/p$  for individual charge states. The triangles represent a scan after filling the second part of the separator after the Havar foil with 2.5 mbar Argon gas. The eleven separated charge states are now concentrated in a single peak corresponding to an average charge state due to the statistical process of charge-state changes in the gas. It can be observed from the fitted dotted curve that the resulting distribution is well reproduced by a Gaussian distribution with a FWHM of 1.4%  $\delta p/p$ .

## 3.2 Low Energy Beam Preparation

In order to perform the precision measurements, radioactive ions produced need to be cooled and trapped in atom traps. The cooling procedure needs to be fast and efficient in view of the short lifetime of the isotopes. This is done with the thermal ionizer and the radio frequency quadrupole cooler and buncher.

### 3.2.1 Thermal Ionizer

The thermal ionizer shown in Fig. 3.7 is used as a first stage of cooling to stop and re-ionize fast radioactive nuclides produced and separated with the TRI $\mu$ P separator. The main principle of the thermal ionizer is to stop the high-energy ions in a solid. It is ideal for alkali and alkaline earth elements that have low ionization potential [82]. The thermal ionizer consists of a stack of 10 tungsten foils each 1  $\mu$ m thick and separated by 1 mm to stop the reaction products. The catcher is placed in a tungsten cavity of diameter 30 mm and length 25 mm. The cylinder wall with a thickness of 0.1 mm is heated by electron bombardment and radiation from a tungsten filament surrounding it.

The reaction products pass a few thin tungsten foils, serving as heat shields and energy degraders. After stopping, the reaction products will diffuse out of the foil at these high temperatures. The time scale depends on the diffusion constant, which is highly sensitive to the temperature [82]. Collisions with the surface of the foil and the cavity body ionize these atoms with a high probability. The reaction products can now be extracted as ions with a suitable electric field. The difference between the ionization potential of the reaction product and the foil material determines the ionization probability and the re-neutralization after each collision with the surface. The thermal ionizer was tested successfully in December 2005 using the reaction  $^1\text{H}(^{20}\text{Ne}, ^{20}\text{Na})\text{n}$  at 22.7 MeV/nucleon [83]. Up to

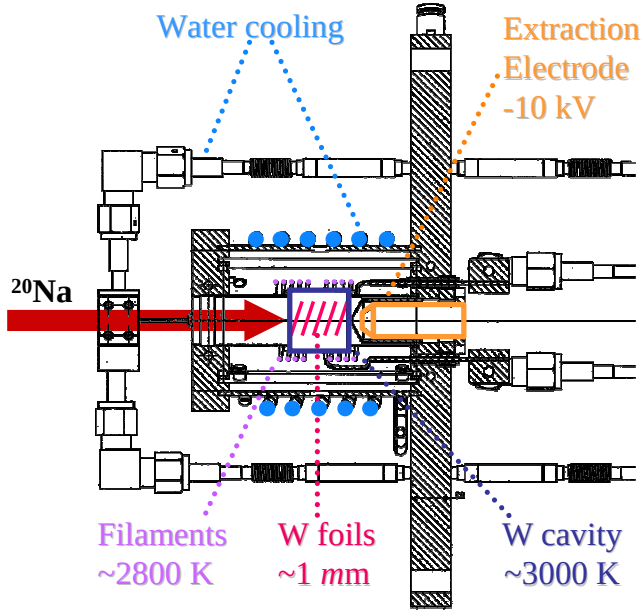


**Figure 3.7:** Side view of the thermal ionizer. The  $^{20}\text{Na}$  beam from the magnetic separator enters from left and stops inside a stack of tungsten (W) foils. Atoms diffuse out of the tungsten foils at high temperature. Collisions with the surface of the foils and the tungsten cavity body ionize these atoms, which can be extracted with an electric field.

now at least 2.5% of the primary beam of  $^{20}\text{Na}$  ions was extracted at a cavity temperature of at most  $2100^\circ\text{C}$ . Improvements of the thermal ionizer are under way. First experiments with  $^{21}\text{Na}$  isotope are planned.

### 3.2.2 Radio Frequency Cooler and Buncher

A segmented radio frequency quadrupole cooler and buncher is the second stage of the cooling procedure in the TRI $\mu$ P facility, which is shown in Fig. 3.8. Input ions coming from the thermal ionizer have an energy spread of several eV. The RFQ cooler and buncher consists of two identical segmented RFQ units of 330 mm length, separated by a small aperture for the purpose of differential pumping. The quadrupole rods are 10 mm apart and segmented to apply an axial field gradient. The segments are connected by a dc resistor chain which allow for

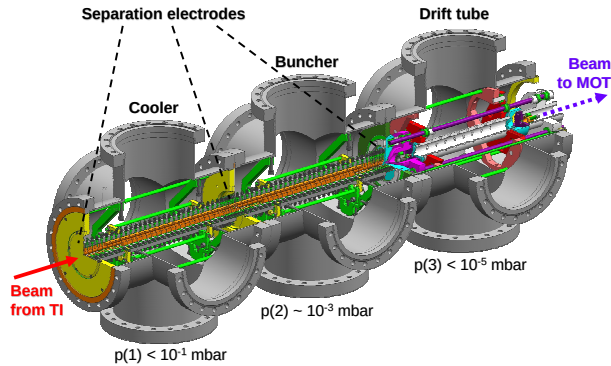


**Figure 3.8:** 3D-drawing of the segmented RFQ cooler, the RFQ buncher and the pulsed drift tube placed in standard NW160 vacuum chambers. Isotope beam from the thermal ionizer is further slowed down in energy with the RFQ1 having buffer gas. RFQ2 is to trap the ions and extract them in pulsed mode with a drift tube. The singly charged ions will be transported to the neutral atoms traps (MOT).

a dc potential along the axis, while the radio frequency is capacitively coupled to the segmented electrodes [84, 85].

The RFQ system was commissioned using  $^{23}\text{Na}^+$  ions with energies of 10-30 eV. In the first RFQ ions are slowed and transversely cooled by collisions with helium buffer gas at a pressure of about  $3 \times 10^{-2}$  mbar. A small drag potential of about 0.5 V/cm moves the ions along the axis. The second RFQ operates at a pressure ten times lower than the first one. The fine segmentation along the axis provides the possibility of confinement in all three dimensions. This is used to accumulate ions and then extract them in a pulse. The axial potential is shaped to allow for the trapping of ions near to the exit. The potential depth is on the order of 5 V. The ions can be ejected into a pulsed drift tube accelerator by switching the last electrode by several 10 V. Preliminary measurements indicate that more than 30% of the ions entering the device are transferred into the drift tube. The drift tube is pulsed and the ion pulse is detected by a micro channel plate (MCP) in the low energy beam line. Measuring the transmission of the ions and optimizing the settings are the main issues of the commissioning. Simulations, commissioning measurements of the radio frequency cooler and buncher and other technical details are described in the thesis of Emil Traykov [80].

The ion pulses from the drift tube are transported in an electrostatic low energy beam line, consisting of electrostatic elements for steering and focussing of

the beam to the experiments.

### 3.2.3 Atom Traps

Neutralization and laser cooling and trapping is the last step to collect the radioactive isotopes in magneto optical traps, where the proposed experiments will be conducted. For neutralization of the low energy and singly charged ion beam coming out of the low energy beam line a 25 micron thick yttrium foil is used. It is mounted inside the accumulation Magneto Optical Trap (MOT) opposite the incoming ion beam located at end of the low energy beam line. The ions are converted to neutral atoms by a hot (900 °C) yttrium foil and trapped in the MOT.

We accomplished the accumulation MOT for sodium atoms using an oven with stable sodium [86]. It is located inside a Pyrex glass cell where care was taken to have a small size to optimize the capture efficiency.

A magneto optical trap for  $\beta - \nu$  correlation measurements with  $^{21}\text{Na}$  is being set up. In order to allow for investigation of systematic errors, which are important for a precision measurement, a large magneto optical trap chamber has been designed and fabricated [87]. Preliminary tests like magnetic field mapping and testing the ultra high vacuum conditions are completed.

## 3.3 Laser Facility at TRI $\mu$ P

High precision experiments will be carried out using modern atomic physics techniques like laser cooling and trapping of atoms. In the case of alkali atoms like sodium laser cooling and trapping techniques are well established and KVI has expertise. But, for our other experimental work we need to develop the laser cooling and trapping techniques. They have not been attempted for laser cooling or trapping till date to the best of our knowledge. One of the tasks within the TRI $\mu$ P project as a facility is the establishment of the infrastructure for the atomic laser spectroscopy. Prior to that there was no laser spectroscopy laboratory available at KVI. We started with the development of grating stabilized diode lasers, built our experimental setups and developed spectroscopic techniques. The laser developments were needed to provide light at different wavelengths. For the lasers, control electronics like temperature controllers and high voltage amplifiers were developed. In addition to the above signal control electronics for the frequency stabilization and modulation of the lasers and the optical detectors for sensitive low-level light detection were designed and developed.

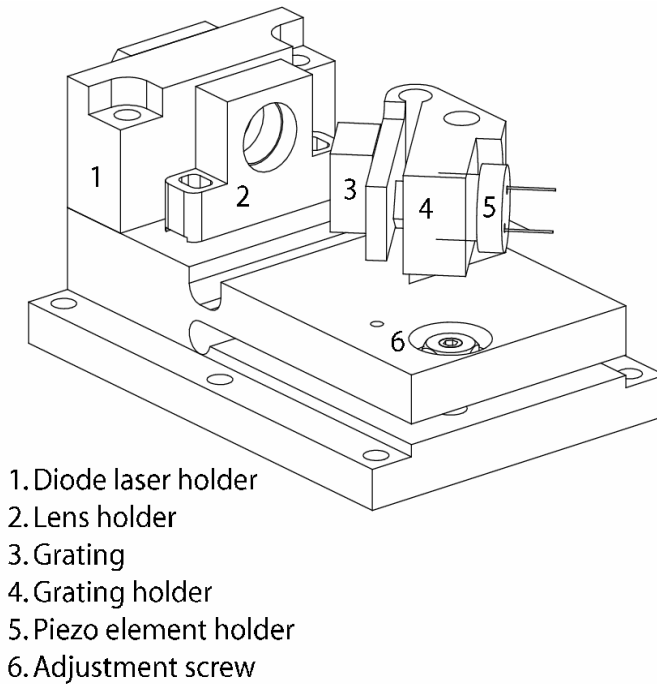


**Figure 3.9:** A schematic drawing of the mechanical set up of the Extended Cavity Diode Laser (ECDL) in Littrow configuration.

### 3.3.1 Diode Lasers

Semiconductor diode lasers with single and stable output frequency are in use for high-resolution spectroscopy and laser cooling and trapping of atoms because of their wavelength selection and compactness [88, 89]. A review on the properties of semiconductor diode lasers and their usage in atomic physics experiments can be found elsewhere [90]. We use grating stabilized diode laser systems, which were built in our laboratory for barium spectroscopy. We describe the layout in this section.

Laser diodes are extremely sensitive to optical feedback. Making use of this sensitivity, light is coupled back in to the laser diode from a grating as wavelength selective element. The frequency tuning of the diode laser can be achieved by three parameters: the temperature of the laser diode, the injection current of the diode and the tilting angle of the grating. The tuning range for each of the parameters is typically a few GHz. The combination of grating and current tuning allows mode hop free tuning ranges of several 10 GHz.

1. Temperature influences both the optical path length of the laser cavity and the gain curve of the laser diode used. The frequency of the laser diode decreases with increasing temperature. For a laser diode the typical temperature tuning range is 0.06 nm/K ( $\cong$  30 MHz/mK). It can be used for scanning, however, due to the temperature controlling requirements this is only useful for slow scanning of the laser in spectroscopy experiments.
2. Tuning of the frequency of diode laser can be achieved by tilting the angle of the grating. The fine tuning of the tilting angle of the grating can be accomplished with a piezo electric transducer (PZT) element. For our experiments we use the diode lasers in the Littrow configuration (Fig. 3.9). In this configuration, the grating is aligned such that the first order diffraction light returns directly to the laser diode. The amount of light which is fed back is selected by the grating. It ranges from 5-50% depending on the laser diode. The output is the zeroth order reflected beam that can be 50% to 80% of the original output power of the laser diode. The angle at which the incident beam strikes the grating sets the wavelength of the output beam, given by

$$\lambda = 2d\sin\theta \quad (3.1)$$

where  $\lambda$  is the wavelength of light,  $d$  is the distance between the grooves of the grating and  $\theta$  is the angle of incidence with the grating normal. The wavelength tunes with the angle  $\theta$ . This angle is varied by changing the voltage on the PZT. As the voltage increases,  $\theta$  decreases, and the wavelength decreases. The frequency tuning range of the diode laser with PZT is 300 MHz/V. The modulation frequency is limited to  $\approx$ 1.5 kHz due to mechanical resonances of the grating support.

3. Changes of the injection current affect both the diode temperature and carrier density that changes the index of refraction of the active area, which in turn affects the wavelength. Thus the wavelength of the laser can be tuned with the injection current. Increase in the injection current decreases the frequency of the laser diode. Typical tuning range of current is 4 GHz/mA. The modulation frequency of a few hundred MHz can be attained with injection current.

For barium we use diode lasers at 791.3 nm to drive the  $6s^2 \ ^1S_0 - 6s6p \ ^3P_1$  transition and to populate the metastable triplet  $D$  states (see Section 4.5). To

obtain the long term frequency stability we used iodine ( $I_2$ ) spectroscopy to lock these lasers to a transition in molecular iodine described in the next section 3.3.2.

### 3.3.2 Iodine Spectroscopy

Iodine is a well known secondary frequency standard for frequency stabilization of lasers [91]. Iodine, a heavy diatomic molecule, has a large number of bound ro-vibrational states for each electronic configuration. A thorough description of the diatomic molecular spectra and the nomenclature used in such systems can be found elsewhere [92]. Transitions from the ground state  $X^1\Sigma_{0g}^+$  to the four low-lying excited states  $A'^3\Pi_{2u}$ ,  $A^3\Pi_{1u}$ ,  $^3\Pi_{2u}$ , and  $B^3\Pi_{2u}$  lie close to the near infrared/visible region of the spectrum (see [93]). The A-X spectrum lies in the near infrared region (from 890 nm to beyond 1.3  $\mu$ m) and, being relatively weak, is obscured by the stronger B-X spectrum at short wavelengths [94,95]. The B-X spectrum extends from 500 nm to around 900 nm and has been the subject of our spectroscopic measurements. The transition is allowed by the  $0^+ \leftrightarrow 0^+$  selection rule and provides a good example of the break down of the  $\Delta S = 0$  selection rule in molecules of atoms with high element number. The transition is quite intense giving rise to the violet color of iodine vapor.

In heavy molecules with highly charged nuclei the spin-orbit interaction may be so strong that  $L$  and  $S$  are not uncoupled by the electrostatic field of the nuclei. The  $L$  and  $S$  couple to give a resultant  $J_a$  which is then coupled to the internuclear axis with a component  $\Omega\hbar$ . The electronic angular momentum  $\Omega$  and the angular momentum  $N$  of nuclear rotation then form the resultant angular momentum  $J$ . This is known as Hund's case (c) and is the best representation of the coupling in iodine. The selection rules that apply in this regime are summarized below.

$$\Delta\Omega = 0, \pm 1; \Delta S = 0; \Delta J = 0, \pm 1;$$

$\Delta J=0$  is forbidden for  $\Omega = 0 \rightarrow \Omega = 0$ ;  $0^+ \leftrightarrow 0^+$ ,  $0^- \leftrightarrow 0^-$ ,  $0^+ \leftrightarrow 0^-$ . The selection rule for  $J$  gives rise to the familiar  $Q$  ( $\Delta J=0$ ),  $P$  ( $\Delta J=-1$ ) and  $R$  ( $\Delta J=+1$ ) branches. However, if both states have  $\Omega = 0$  then the  $Q$  branch no longer occurs.

Lasers stabilized to iodine transitions are widely used as reliable frequency standards [96–100]. High resolution saturated absorption spectroscopy of molecular  $I_2$  hyperfine transitions deliver a natural frequency grid in the 500 nm-900 nm range. The absence of an intrinsic electric dipole moment, leading to extremely small perturbation by external electromagnetic fields as well as the strong and relatively narrow natural linewidth of the hyperfine structure transitions make it

an ideal secondary frequency standard. There are more than 60000 iodine lines, which means that there is about one line for every 5 GHz.

### Experimental setup

The diode laser used for the iodine spectroscopy is in an extended cavity configuration. The laser diode is a 50 mW (GaAlAs-HL7851G) single-mode laser at 785 nm. The highly divergent light from it is collimated with an aspheric lens (Thorlabs C230TM-B). Around 30% of the output power is fed back by means of a 1200 lines/mm Al-coated reflection diffraction grating (Edmund Optics) placed in Littrow configuration, which is held on a piezo electric transducer that changes the angle of the grating to tune the wavelength of the laser. The laser diode current is controlled by a Thorlabs LDC 201 laser diode driver. The temperature of the laser diode is maintained by a 33 W Peltier element sandwiched between the diode mounting block and the anodized aluminium base plate holding the entire setup. The temperature of the laser diode is measured with an analogue AD590 temperature sensor and it is controlled to drift  $<1$  mK in 1 second with an in house built PID controller. A stable single-mode output beam of power 13 mW at 791.3 nm is achieved by tuning the grating angle with the piezo element, adjusting the injection current and the temperature respectively. The typical operating current of the diode at 13 mW is 122 mA.

The collimated output beam from the diode laser is elliptical in nature. It is made roughly circular  $\sim 1.2$  mm diameter by an anamorphic prism pair. The circular beam passes through a Faraday isolator (LINOS Model: DL1-1; S. No 449) to minimize the unwanted optical feedback from the optics downstream, which would cause instability of the laser. With the help of a wedge around 8% of the beam is split to the wavelengths meter and to the Fabry-Perot (FP) cavity. The former (*High finesse Model: w5/6 UV, 250-1100 nm*) measures the wavelength to an accuracy of  $\pm 1$  GHz. The latter is made of two mirrors of radius of curvature -500 mm that were glued to a cylindrical quartz spacer of length 35 mm with one of the mirrors sitting on a piezo element. It has a Free Spectral Range (FSR) of 3 GHz and a finesse of 750 to observe if the laser is in a multiple frequency mode. Mode matching to the FP-cavity is attained with a lens of focal length 250 mm (f6 in Fig. 3.10) placed at a distance of 300 mm. The transmitted light from the cavity is focussed onto a fast Si-photodiode (Thorlabs FDS010) with a lens of f5 = 50 mm (Fig. 3.10).

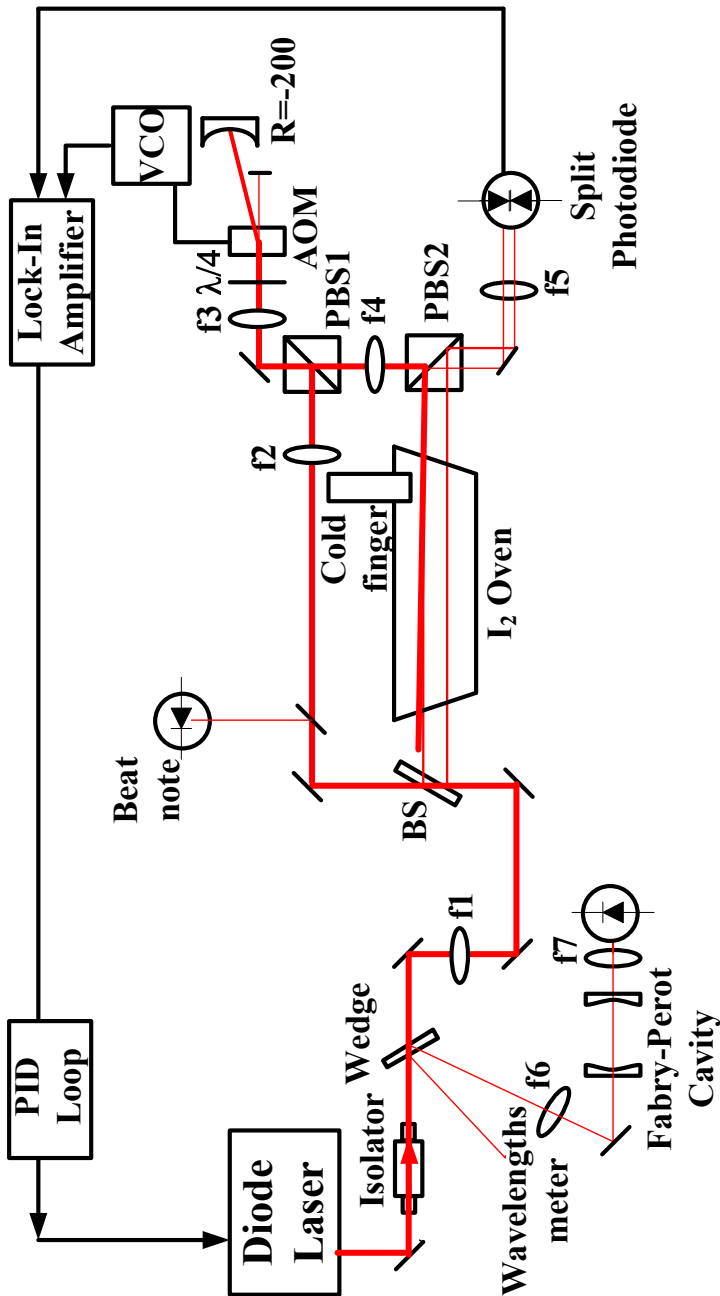


**Figure 3.10:** Set up for the frequency modulation saturated absorption spectroscopy of iodine. Linear  $\perp$  linear polarization is used for the probe and pump beams. The pump beam is frequency modulated with an acousto optic modulator driven by voltage controlled oscillator. The signals are detected with a split photodiode.

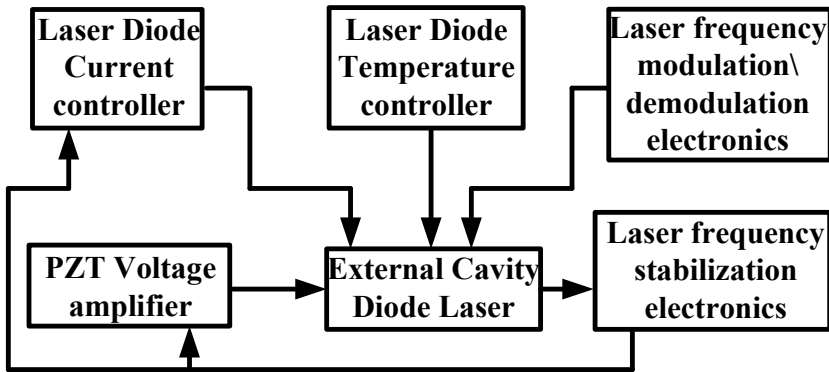


**Figure 3.11:** Block diagram of the control electronics of the diode laser for frequency modulation saturated absorption spectroscopy.

### The Iodine Oven

To stabilize the diode laser frequency at barium atomic transition of 791.3 nm the nearest ro-vibrational transition in  $I_2$  is the P(52)(0-15) transition at 791.3 nm. It has a total of 15 lines. The line marked ‘a1’ in Fig. 3.12 is the nearest single component to the barium transition and is blue detuned from it. This transition in  $I_2$  could be reached with higher ro-vibrational levels. To populate a high vibrational state, higher temperature is needed. But at higher temperatures the  $I_2$  pressure in the cell will be enormous, and study of Doppler-free saturation absorption spectroscopy will not be possible due to collisional broadening of lines. This is solved with a cold finger attached to the  $I_2$  cell by the supply of  $I_2$  molecules to maintain a low pressure in the cell. Thus, the temperature of the cold finger determines the vapor pressure of the cell, while the wall temperature of the cell determines the fraction of  $I_2$  vapor in the vibrational level.

In order to satisfy the above requirement an oven was designed and fabricated capable of reaching temperatures up to 600°C, while a copper tube cooled with a Peltier element acts as a cold finger. The cold finger of the  $I_2$  cell is maintained at a temperature of 30°C. The temperature of the oven is measured at the center of the cell using a Thermocoax type K-NiCr thermocouple (41 $\mu$ V/1°C).

### Optical Layout and Spectroscopy

The experimental arrangement for the frequency-modulation saturated absorption spectroscopy [101,102] is shown in Fig. 3.10. We use linear  $\perp$  linear polarization for the probe and pump beams.

The light from the diode laser after the Faraday isolator is focused with a  $f_1 = 750$  mm lens. A thick glass blank is positioned in its path to generate two weak probe beams of power less than about 0.5 mW and beam radius of about 0.4 mm is passed through  $I_2$  vapor cell (length 540 mm) placed inside the oven. The transmitted strong beam after a lens of focal length  $f_2 = 400$  mm is directed through a polarizing beam splitter (PBS1) for double passing through an acousto optic modulator (AOM). The double passing is advantageous to maintain directional stability of the laser beam that can otherwise lead to systematic shifts of the peak positions. Further, there are two advantages of modulating the frequency of the laser beam with an AOM instead of the grating angle or modulation of the injection current of the diode. One is the linearity of the scan, since the voltage controlled oscillator (VCO) that determines the AOM frequency has a linear transfer function. Secondly, the scan axis has absolute frequency calibration once the voltage to frequency transfer function of the VCO is known. Also, modulation of the injection current of the laser diode produces amplitude modulation on the signal thus affecting the signal to noise ratio. The block diagram of the diode laser control electronics and frequency stabilization electronics is also shown (Fig. 3.11).

The vertically polarized strong beam is focused using a  $f_3 = 200$  mm lens, into a 80 MHz AOM (IntraAction MT-IR-80). The separation between the zero order and first order beams is 15 mrad. The zero order beam is blocked with an iris diaphragm. A  $\lambda/4$  plate is used before the AOM to change the polarization of the retro-reflected beam from the focussing mirror of  $R = -200$  mm and it could also be used to vary the power of the pump beam by ‘misalignment’. The focussing mirror is placed on a micrometer translation stage to obtain maximum double passing diffraction efficiency by fine adjusting the position of the mirror. The double passing diffraction efficiency thus obtained is about 67%.

Now, the horizontally polarized double passed beam is focussed with a lens  $f_4 = 500$  mm into the  $I_2$  cell giving a 0.4 mm diameter beam in the overlapping region. This frequency shifted double passed beam is the strong ‘pump’ beam for the spectroscopy experiment. The polarizing beam splitter cube (PBS2) is used to superpose the pump and probe beams. The PBS2 is aligned, such that the horizontally polarized pump beam is transmitted directly to overlap with one of the probe beams. The vertically polarized probe beams passing opposite to it, however, are reflected and focussed with  $f_5 = 50$  mm lens on to a split-photodiode.

The typical power of the pump beam was 6 mW after double passing the AOM, and the beam radius  $w$  ( $=1/e$  amplitude radius) in the cell is about 0.4 mm. The frequency modulation signal for the saturation spectroscopy is obtained from

a VCO operated at the center frequency of the 80 MHz AOM by applying a voltage of 0.42 V with a function generator (HP 3314A). The output RF power of the VCO is +11 dBm. It is attenuated down to -2 dBm with a 13 dB attenuator. The output of the attenuator is given to a RF power amplifier (Mini-circuits, Model-ZHL-1-2W), to drive the AOM. The modulation frequency and modulation depth are controlled by the function generator. The typical modulation frequency is 90.5 kHz and the modulation depth is 4 MHz. The demodulated first derivative signal of the Lorentzian absorption signal is obtained with a 1-100 kHz lock-in amplifier (Scitech). The spectrum of the P(52)(0-15) transition measured by a low-noise split photodiode (UDT sensors model: SPOT-2D). The electronics circuit diagram of the split-photodiode amplifier is given in the Appendix B.1.

We get a good agreement of the iodine spectrum of the P(52)(0-15) transition compared with the spectrum generated with a Doppler width of  $\pm 0.0003 \text{ cm}^{-1}$  by commercial software based on the results of [103]. The generated spectrum is shown in Fig. 3.12. From the resolution of the multiplets we can estimate the linewidth of the transitions to be less than 10 MHz. The iodine spectrum is used to stabilize the laser frequency. The shape of the signal is ideal for a locking application.

The laser frequency is tuned to the line marked ‘a1’ in Fig. 3.12. The derivative like signal after the lock-in amplifier is given to a servo loop that has two paths. One is the signal to be fed back to the piezo of the diode laser and the other is to the injection current of the laser diode. The piezo path consists of a proportional gain stage and an integrator, which is used to stabilize the frequency to the zero crossing of the derivative signal. The current path consists of proportional gain amplifier to reduce the low frequency noise. The laser remained locked for several hours without problems.

The linewidth of the  $\text{I}_2$  laser was measured with the amplitude of the error signal before and after frequency locking the laser that is calibrated in frequency. It is found that the linewidth of the diode laser after frequency stabilization is less than 1 MHz. The method used to measure the linewidth of the laser does not provide a good estimate, but it served our purpose. One can measure the linewidth and stability of the laser by optical heterodyne measurement. By locking two different lasers to the adjacent single components of the P(52)(0-15) transition, i.e., locking the lasers to ‘a1’ and ‘a10’ components and measure the beat frequency with a fast photodiode. This gives better accuracy for the frequency measurement.

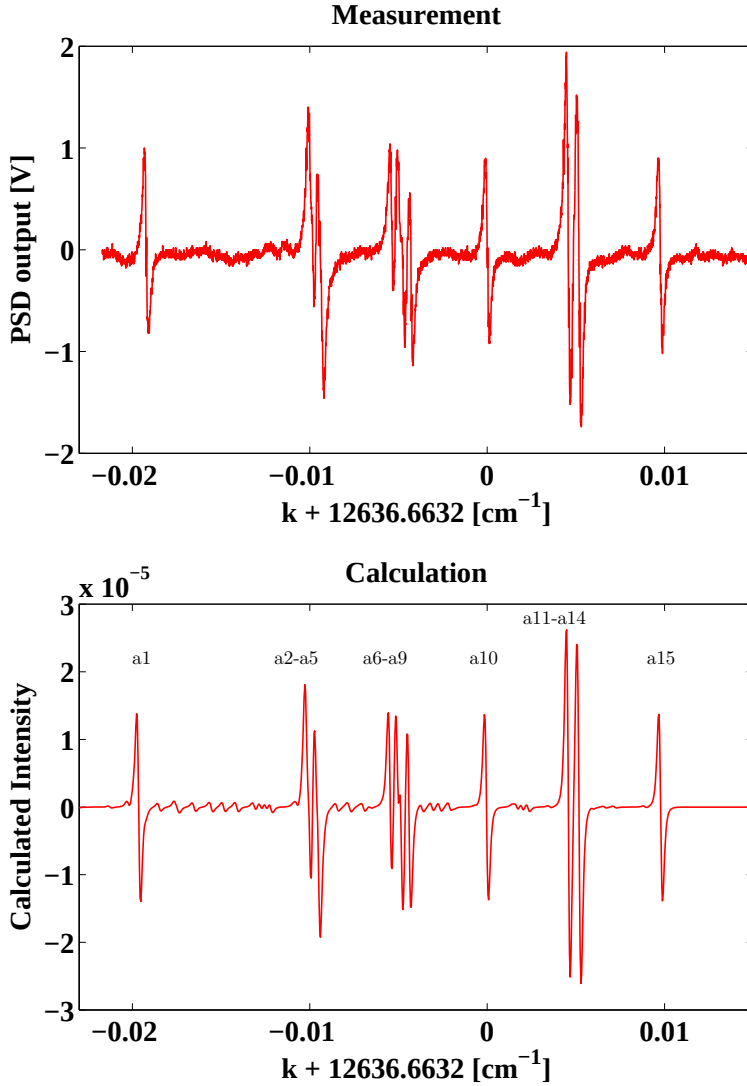


**Figure 3.12:** (a) The measured hyperfine spectrum of  $P(52)(0-15)$  transition by frequency modulation saturated absorption spectroscopy of iodine. The iodine oven temperature is  $560(10)^\circ\text{C}$ . The sensitivity of the lock-in amplifier used is  $300 \mu\text{V}$  that corresponds to 10 V. (b) The first derivative of the Lorentzian hyperfine spectrum of  $P(52)(0-15)$  transition in iodine [103]. The parameters used are temperature  $T = 560^\circ\text{C}$  and hyperfine linewidth  $= 0.0003\text{cm}^{-1}$  corresponding to a resolution of 9 MHz for each component.

### 3.4 TRI $\mu$ P - Studying Fundamental Interactions and Symmetries

In the new TRI $\mu$ P facility we can produce rare isotopes like  $^{213}\text{Ra}$  and  $^{21}\text{Na}$ . Both are ideal candidates for fundamental tests of the Standard Model. The separator as a facility has been used for first physics experiments and all the ingredients have been tested. The production target and the magnetic separator are in use by the scientists and user community. In the near future we hope to improve the thermal ionizer. The radio frequency quadrupole cooler and buncher and low energy beam line was constructed and commissioned. We set up a laser facility with the development of diode lasers, electronics for signal control and detection and established iodine spectroscopy as a frequency reference. In conclusion, the combination of all the different stages to a facility will make the research for fundamental symmetries with radium and sodium possible at KVI.



## Chapter 4

# Radium and Barium: Atomic Properties

Laser spectroscopy is an ideal tool for precise determination of atomic system properties. It not only provides information on the electron cloud, but also e.g., the nuclear spin, charge distribution, the magnetic dipole and electric quadrupole moment of the nuclei. In particular, isotope shifts and hyperfine structure depend on nuclear and atomic properties. Their knowledge improves the understanding of the structure of the atom and atomic nucleus. As an example, laser spectroscopy is a precise method for determining mean charge radii of nuclei of radioactive atoms [104]. Other properties, like an EDM, which might have their origin in nuclear forces, can be amplified due to the atomic structure and become visible by precise laser spectroscopy. This is exploited in experiments with different atoms in many searches for an EDM [8].

In order to prepare for an EDM experiment, the atomic and nuclear properties of the atom of interest have to be determined. Thus extensive studies through laser spectroscopy are indispensable. Since radium has been identified as a good candidate for an experimental search for an EDM we develop laser spectroscopy to determine the missing atomic and nuclear properties. In the literature very scarce information is available on the radium atom. Only a few energy levels, transition rates and life times of states of interest for an EDM experiment have been determined so far. The available data as well as the information, which has to be determined is discussed below. Part of the reason is that radium itself is only available in small quantities since most isotopes are short-lived and the radioactivity poses a problem. In the future short-lived isotopes of radium, like  $^{213}\text{Ra}$  with  $I \neq 0$ , will be available at TRI $\mu$ P facility [18].

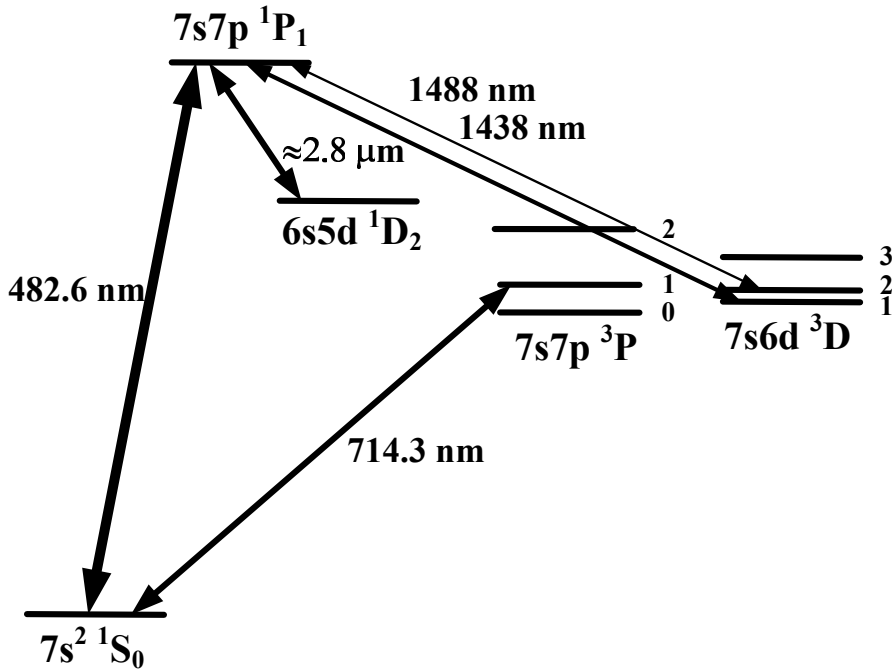


**Figure 4.1:** Level diagram of the radium atom with the wavelengths (vacuum) in nm of the transitions relevant for laser spectroscopy, laser cooling and trapping and for an EDM experiment. Wavelengths are according to [14]. The thickness of the line indicates the strength of the transition.

In section 2.2.3 of Chapter 2 the advantages of radium for searches of electric dipole moments was discussed. Here we give an overview on the available experimental data on radium atom as well as the spectroscopy work that needs to be performed as a precursor for an EDM experiment.

## 4.1 Radium Atom

The energy level scheme of low lying states of atomic radium is shown in Fig. 4.1. Radium ( $^{226}\text{Ra}$ ) was discovered in 1898 by M. Curie. Other short lived isotopes were recognized later. The first spectroscopic measurements of radium dates back to 1934. Rasmussen [42, 43] measured the energy levels of radium ion and atom from a discharge lamp using a grating monochromator. The interpretation of these measurements was corrected by Russel [107]. Absorption spectroscopic

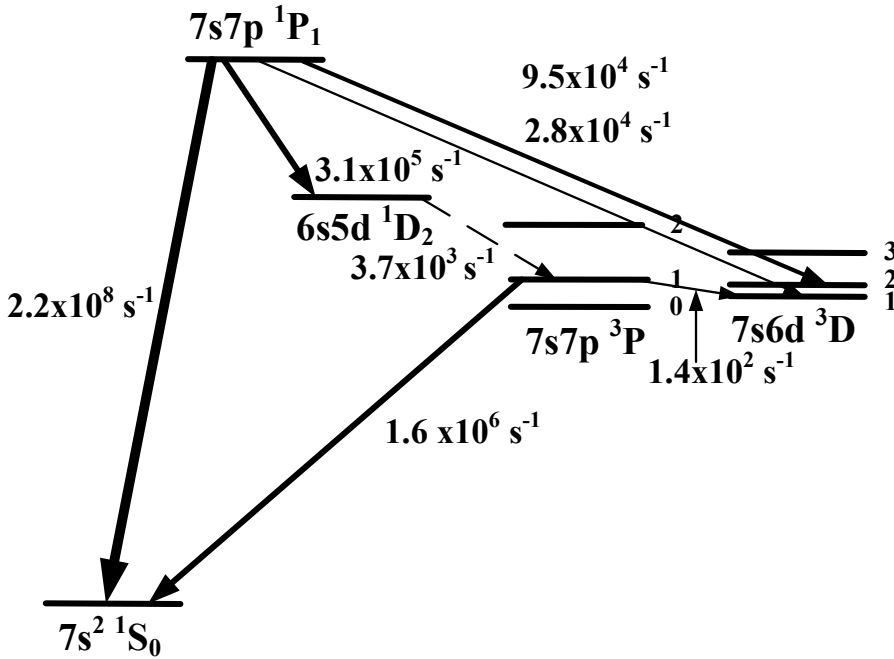


**Figure 4.2:** Level diagram of the radium atom with the transition rates calculated by K. Pachucki [105]. The measured value for the  $^1S_0 - ^3P_1$  transition by Scielzo et al. [106] is  $2.37(12) \times 10^6 \ s^{-1}$ . The thickness of the lines indicates the strength of the transitions.

measurements of the  $^1P_1$  Rydberg series in radium are reported in [108, 109].

The first direct laser spectroscopic measurements on radium were reported in 1983 by Ahmad et al. [104, 110]. They made a systematic investigation of isotope shifts and hyperfine structures of radium atoms and radium ions of the  $7s^2 \ ^1S_0 - 7s7p \ ^1P_1$  transition at 482.6 nm. The isotopes were produced at the ISOLDE facility at CERN. The method of on-line collinear fast beam laser spectroscopy was used to obtain the nuclear spins and moments. Later, Wendt et al. [111] continued the measurements on the  $7s^2 \ ^1S_0 - 7s7p \ ^3P_1$  transition at 714.3 nm and confirmed the previously evaluated moments. The nuclear magnetic moments of  $^{225}\text{Ra}$  and  $^{213}\text{Ra}$  were determined to be  $-0.7338(15)\mu_N$  and  $0.6133(18)\mu_N$  [112], respectively. The nuclear magnetic moment of  $^{213}\text{Ra}$ , according to theory, is  $0.607(12)\mu_N$  [113]. This is in good agreement with experiment. Because of the interest in radium as a good candidate for an EDM measurement the group of R. Holt recently started spectroscopy of radium. They focus on the  $^1S_0 - ^3P_1$  intercombination line at 714.3 nm. They determined an absolute frequency of

| Radium Excitation energies [ $\text{cm}^{-1}$ ] |           |                         |                         |            |
|-------------------------------------------------|-----------|-------------------------|-------------------------|------------|
| State                                           | Ref. [14] | Ref. [114] <sup>a</sup> | Ref. [114] <sup>b</sup> | Ref. [115] |
| $7s7p\ ^1P_1$                                   | 20716     | 21156                   | 21148                   | 20450      |
| $\ ^3P_1$                                       | 13999     | 14072                   | 14096                   | 14027      |
| $\ ^3P_2$                                       | 16689     | 16855                   | 16855                   | 16711      |
| $7s6d\ ^1D_2$                                   | 17081     | 17806                   | 17737                   | 17333      |
| $\ ^3D_2$                                       | 13994     | 13974                   | 13907                   | 13980      |
| $\ ^3D_1$                                       | 13716     | 13672                   | 13609                   | 13727      |

**Table 4.1:** Excitation energies of low states in radium. Reference [14] are the experimental values. Reference [114]: ‘a’ is Dirac-Coulomb (DC) and ‘b’ is Dirac-Coulomb-Breit (DCB) excitation energies calculated based on relativistic coupled cluster (RCC) method. Reference [115] is based on configuration interaction method and many-body perturbation theory.

that transition of  $13999.269(0.001)\text{ cm}^{-1}$  by referencing it to the  $13999.2459\text{ cm}^{-1}$  line in molecular iodine and they found the lifetime of the  $\ ^3P_1$  state to be  $422(20)\text{ ns}$  [106]. In particular they are using this transition for laser cooling and trapping of radium.

There are several theoretical many body calculations of this multi-electron system. Recently there has been an increased effort by the theoretical community for a better estimation of the transition energies and lifetimes of the states. Various methods are used to calculate the transition energies and lifetimes of the states in radium [105, 114–117]. Generally the calculated excitation energies reported are higher than those from the experiment. The deviation is in between  $200 - 400\text{ cm}^{-1}$  for the low excitation energies and increases for the highest levels. The lifetime of the  $7s6d\ ^3D_2$  level in radium was calculated [12, 115, 118] to be 3.3 to 15 seconds. However, there is no experimental data to test these calculations. New measurements will help to constrain the atomic wave functions for theoretical calculations and will improve the overall quality of the theoretical predictions.

A prerequisite for a radium EDM experiment are the following measurements: The first and foremost is the knowledge of the energy levels of radium to a GHz accuracy. This is because the energy level diagram shown in Fig. 4.1 is based on the 1934 data [43], which has a limited resolution. Optical spectroscopy has to be carried out on the singlet and triplet  $P$  ( $7s7p\ ^1P_1$ ,  $7s7p\ ^3P_1$ ) and  $D$ -states ( $7s6d\ ^1D_2$ ,  $7s6d\ ^3D_{1,2}$ ) respectively. Especially for an EDM measurement the positions of the  $\ ^3D_{1,2}$  levels with respect to the triplet  $P$ -states play an important role. This is because the actual position of the  $7s6d\ ^3D_2$  energy level with respect

| Isotope          | $^{138}\text{Ba}$ | $^{137}\text{Ba}$ | $^{136}\text{Ba}$ | $^{135}\text{Ba}$ | $^{134}\text{Ba}$ | $^{132}\text{Ba}$ | $^{130}\text{Ba}$ |
|------------------|-------------------|-------------------|-------------------|-------------------|-------------------|-------------------|-------------------|
| Nuclear spin (I) | 0                 | 3/2               | 0                 | 3/2               | 0                 | 0                 | 0                 |
| Abundance %      | 71.7              | 11.3              | 7.8               | 6.6               | 2.4               | 0.1               | 0.1               |

**Table 4.2:** Natural isotope distribution of barium, spin and abundance [121].

to the  $7s7p\ ^3P_1$  level is crucial for the enhancement of the P and T-odd effects considered in references [12, 13]. This enhancement is due to the very small energy difference of  $5\text{ cm}^{-1}$  between the opposite parity states  $7s7p\ ^3P_1$  and  $7s6d\ ^3D_2$ . A significant change in the position of the  $^3D_2$  state would severely affect the enhancement.

The other requirement for experiment on parity and time reversal invariance violation in radium is the measurement of the lifetimes of the  $^1D_2$ ,  $^3D_1$  and  $^3D_2$  states, which reveal information on the electronic wave function at the location of the nucleus. In addition, lifetimes are also important for laser cooling and trapping of radium using  $^1S_0$ - $^1P_1$  transition and they provide information on the decay rates.

## 4.2 Barium Atom

Barium has a similar atomic structure as radium and we use it as a precursor to develop the laser spectroscopic techniques which we can apply to the radium atom. Our spectroscopic work on barium allowed us to establish the first laser cooling of barium.

Here we describe the spectroscopy of barium. The focus is on the laser spectroscopy of the  $6s5d\ ^1D_2$  -  $6s6p\ ^1P_1$ ,  $6s5d\ ^3D_2$  -  $6s6p\ ^1P_1$  and the  $6s5d\ ^3D_1$  -  $6s6p\ ^1P_1$  transitions (see Fig. 4.3). We report the first laser spectroscopy studies of these transitions. In particular, we measure the repumping rates and hyperfine structure of these transitions. In addition, we were able to determine for the first time the isotope shift of the metastable  $6s5d\ ^3D_1$  state. Natural barium with several isotopes was used. This gives access to the isotope dependence of nuclear charge radii.

Barium is an alkaline earth element (group II). Its atomic number is 56 and the electron configuration is  $[\text{Xe}]\ 6s^2$  i.e., it has two s valence electrons. The ground state  $LS$  configuration is  $6s^2\ ^1S_0$ . The natural abundance of barium isotopes with their nuclear spin is given in Table 4.2. The energy levels relevant for spectroscopy and laser cooling are shown in Fig. 4.3. The transition rates shown in Fig. 4.4 are taken from [106, 119, 120].

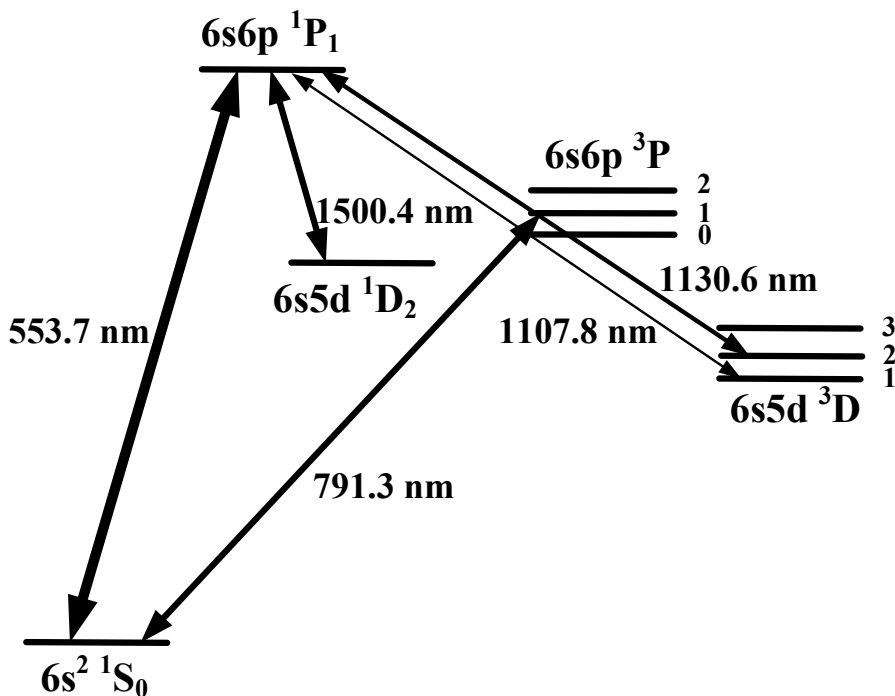


**Figure 4.3:** Level diagram of the barium atom with all the wavelengths (vacuum) in nm for the transitions used in laser spectroscopy of the metastable  $D$ -states and for laser cooling experiment. Wavelengths are according to [119]. The thickness of the lines indicate the strength of the transitions.

The first excited state of barium is the singlet  $6s6p\ ^1P_1$  state. We have measured the lifetime of the  $^1P_1$  state using the Hanle effect to be  $8.0(5)$  ns in good agreement with the value of  $8.2(2)$  ns [122] reported in the literature. This results in a natural line width of  $19(1)$  MHz. The transition rate of  $6s^2\ ^1S_0 - 6s6p\ ^1P_1$  transition is  $1.19(1) \times 10^8\ \text{s}^{-1}$  [119]. This transition is the only option for laser cooling of barium. Since it exhibits large leak to metastable  $D$ -states, this is not a closed two level system and we have to use lasers to drive the  $D - P$  transitions.

The atomic transitions from the excited  $^1P_1$  state, relevant for laser cooling other than the  $^1S_0 - ^1P_1$  transition are the  $^1D_2 - ^1P_1$  transition at 1500.4 nm, the  $^3D_2 - ^1P_1$  transition at 1130.6 nm and the  $^3D_1 - ^1P_1$  transition at 1107.8 nm respectively. The wavelengths and transition rates were determined by Fourier transform spectroscopy using a hollow-cathode discharge [119]. The measured transition probabilities of these transitions are  $2.5(2) \times 10^5\ \text{s}^{-1}$ ,  $1.1(2) \times 10^5\ \text{s}^{-1}$

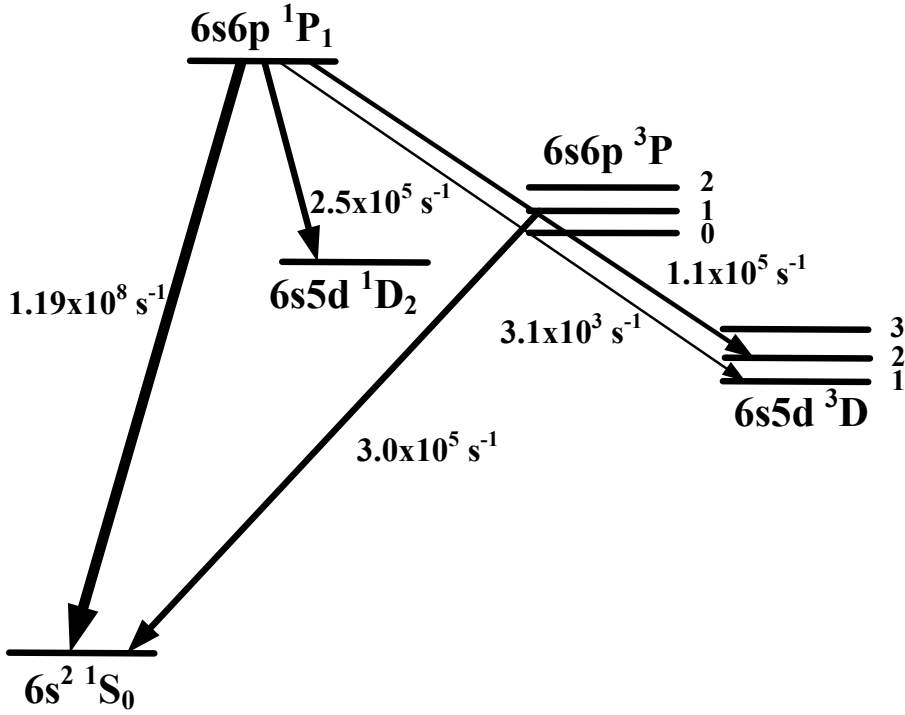


**Figure 4.4:** Level diagram of barium atom with transition rates for the transitions used in optical spectroscopy of the metastable  $D$ -states and in the laser cooling of barium experiment. The experimental transition rates are according to the references [106, 119, 120].

and  $3.1(5) \times 10^3 \text{ s}^{-1}$  [119] respectively. To the best of our knowledge there are no direct laser spectroscopic measurements of these transitions reported in the literature.

While using the strong  $1S_0 - 1P_1$  transition for laser cooling atoms leak into the  $1D_2$ ,  $3D_2$  and  $3D_1$  states. The branching ratio ( $B_r$ ) into the metastable  $D$ -states when we use the numbers from Table 4.3.

$$B_r = A_{leak}^{-1} = \frac{A_{1P_1, 1S_0}}{A_{1P_1, 1D_2} + A_{1P_1, 3D_2} + A_{1P_1, 3D_1}} \approx 330(30) \quad (4.1)$$

On average an atom in the cooling cycle scatters  $B_r$  photons before it decays into one of the metastable  $D$ -states. This is a constraint for laser cooling.

For simplicity we discuss here the slowing of the atom with a laser beam

| Upper Level              | Lower Level            | Wavelength [nm] | $A_{ik}$ [ $10^8 \text{ s}^{-1}$ ] |
|--------------------------|------------------------|-----------------|------------------------------------|
| $^1P_1$                  | $^1S_0$                | 553.7           | $1.19(1)^a$                        |
| <i>8.2(2) ns</i> [122]   | $^1D_2$ (0.25 s) [123] | 1500.4          | $0.0025(2)^a$                      |
| <i>8.0(5) ns</i>         | $^3D_2$ (60 s) [123]   | 1130.6          | $0.0011(2)^a$                      |
| Our measurement          | $^3D_1$                | 1107.8          | $0.000031(5)^a$                    |
| $^3P_1$                  | $^1S_0$                | 791.3           | $0.0030(3)$                        |
| <i>1345(14) ns</i> [106] | $^3D_2$                | 2723.0          | $0.0012(1)^b$                      |
|                          | $^3D_1$                | 2923.3          | $0.0032(3)^b$                      |

**Table 4.3:** The energy levels, lifetimes in *italic*, wavelengths (vacuum) and transition rates of the barium atom relevant for our experiments. Experimental transition rates: ‘a’ [119], ‘b’ [120]. Lifetimes from [106,122] are the experimental values and from [123] are the calculated values.

against the velocity direction of the atoms and we assume that the momentum transfer in the emission cycle averages to  $\Delta p = 0$ . This becomes clear when we look at the velocity change of an atom after scattering  $n$  photons

$$v_r = \frac{\Delta \vec{p}_a}{m_{Ba}} = \frac{n \hbar \vec{k}_\gamma}{m_{Ba}}, \quad (4.2)$$

where  $\vec{p}_a$  is the momentum of the atom,  $n$  is the number of scattered photons and  $\vec{k}_\gamma$  is the photon wave vector and  $\hbar = h/2\pi$ ,  $h$  is Planck constant. For barium the velocity changes by  $v_r = 0.52 \text{ cm/s}$  due to a single photon kick. Scattering of  $B_r$  photons means a fraction of  $1/e$  of the atoms are left for cycling while the velocity change is only  $1.7 \text{ m/s}$ , hardly enough to speak of laser cooling. To achieve further slowing, the atoms have to be pumped back into the cooling cycle. Hence, laser spectroscopy of the  $D - P$  transitions, which we are reporting here is a prerequisite to laser cooling.

In the future, the experimental methods which we employed for spectroscopy of the  $D$ -states in barium can also be applied to radium. There are no experimental data available on the  $D - P$  transitions in radium. Such data will not only help for laser cooling but are also indispensable input to improve theoretical many-body calculations, which is needed for an evaluation of a radium EDM experiment.

## 4.3 Experimental Setup

For the experiment we need an atomic beam source for barium, appropriate lasers, a detection and a data acquisition system . We explain the details of our setup below.

### 4.3.1 Barium Oven

The design of the atomic beam oven used for barium experiments is similar to the oven used for the Alcatraz experiment [124]. It is adapted from the design of an oven used by the Time and Frequency group of the Physikalisch-Technische Bundesanstalt, Braunschweig, Germany.

The central part of the oven is a stainless steel tube. The base (back end) of the steel tube has a small cavity, which can be filled with the element of interest. To measure the temperature of the oven a thermocouple is attached to the heated cavity. The front end of the cavity is an exit channel of 1 mm diameter and 10 mm length . The stainless steel tube is surrounded by a cylindrical tube made of  $\text{Al}_2\text{O}_3$ . The  $\text{Al}_2\text{O}_3$  ceramic holds a tantalum heating wire, which is thread back and forth through thin holes in the  $\text{Al}_2\text{O}_3$  periphery to provide uniform heating. The entire set up of the oven chamber is mounted on a standard CF35 vacuum flange, which has feedthroughs for the heater current and for a thermocouple to measure the temperature of the oven. The temperature of the oven is regulated with a PID controller. It can be heated to  $1000^\circ\text{C}$ .

We use two ovens for the barium experiments. One oven is for the spectroscopy of the  $^1\text{S}_0 - ^1\text{P}_1$  transition serving as a frequency reference for the laser cooling experiments. It contains natural barium (with all isotopes). The typical operating temperature of the ovens is around 800 K.

For laser cooling experiments the second oven is filled with isotopically enriched  $^{138}\text{Ba}$ . The enriched barium has  $^{138}\text{Ba}$  - 99.7 % in purity supplied by [125] in the form of barium carbonate ( $\text{BaCO}_3$ ). To produce a barium beam, finely powdered zirconium as a reduction agent is mixed with the barium carbonate and heated in the oven. The oven was operated for 12 hours at the maximum temperature to transfer the  $\text{BaCO}_3$  to  $\text{BaO}$  and  $\text{CO}_2$  gas.

### 4.3.2 Vacuum System

The vacuum vessel for barium spectroscopy and cooling experiments consists of a six-way cross and a trap chamber. It is shown in Fig. 4.13. The entire system is maintained at a typical pressure of  $1 \times 10^{-9}$  mbar by a 10 l/s (Gamma

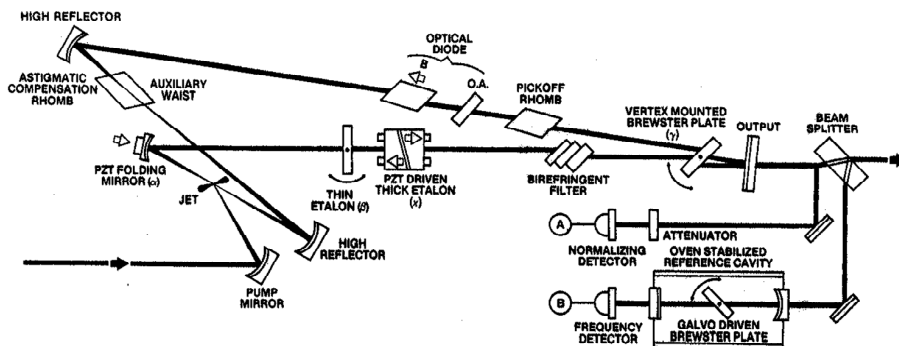


**Figure 4.5:** Optical layout of a Coherent CRR-699 Ring Dye Laser [taken from the Coherent manual].

Vacuum) ion pump. The inside walls of the trap chamber are coated with SPI conductive carbon paint [126] to minimize the scattered light. Baffling, extension ports and AR coated optical windows are also used to reduce the scattered light seen that causes a background count rate in the detector (a photomultiplier tube or a photodiode).

### 4.3.3 Laser Systems

#### Dye Laser

A Coherent CR-699-21 ring dye laser (see Fig. 4.5) at 553.7 nm was used for spectroscopy of the  $^1S_0 - ^1P_1$  transition. The CR-699 is a unidirectional, travelling wave, ring dye laser. It provides a single frequency, tunable, stabilized output of several hundred milliwatts of power, capable of scan operation across 30 GHz range. Unidirectional, travelling wave operation of the dye laser is achieved by an optical diode employing the Faraday effect. The diode construction is such that light passing through in one direction undergoes a small polarization rotation and subsequently, reflection losses at the Brewster surfaces in the laser. Light travelling in the opposite direction undergoes no net rotation and hence, less loss per round trip.

The single frequency operation is achieved by the use of three optical elements: A three-plate birefringent filter, a thin etalon and a piezoelectric driven thick etalon.

1. The three-plate birefringent filter has a passive bandwidth of about 380 GHz and its use reduces the laser line width to about 2 GHz.

2. The thin etalon has a free spectral range of 200 GHz, which is sufficient to enforce single longitudinal mode operation.
3. A piezoelectric driven thick etalon with 10 GHz free spectral range allows to select a single mode.

The CR-699 employs active stabilization, which is servo-locked to a temperature stabilized reference cavity. An error signal from the reference cavity is used to adjust the dye laser cavity length by driving a high-frequency piezoelectric mounted folding mirror and a scanning Brewster plate. To scan the laser in the lock mode, the length of the reference cavity is varied, which is accomplished using a galvo driven Brewster plate in the reference cavity. The dye laser line width as quoted by the manufacturer is 1 MHz.

A Coherent Verdi-V10 (10W) diode pumped solid-state DPSS, single frequency, continuous wave laser at 532 nm optically pumps the Pyrromethene567 dye which is solved in ethylene glycol. The concentration of the dye solution used for optimum performance at 553.7 nm is 1 gram of dye in 1.5 liter of ethylene glycol. We achieved 350 mW single frequency laser output at 553.7 nm with 5 W pump power and a lasing threshold of 3.2 W pump power. At higher dye concentrations the optimum performance is shifted to longer wavelength. The normal operating pressure of the dye circulator is 6 bar. The typical life time of the dye is 200 Wh.

### Fiber laser system

The advent of erbium (Er), ytterbium (Yb) or (Er/Yb) doped fiber amplifiers with the development of UV writing of gratings in photosensitized fibers has paved the path for distributed feedback (DFB) fiber lasers.

In the infrared we use three commercial custom built fiber laser systems (KOHHERAS) for barium spectroscopy of the  $^1D_2 - ^1P_1$  transition (1500.4 nm), the  $^3D_2 - ^1P_1$  transition (1130.6 nm) and the  $^3D_1 - ^1P_1$  transition (1107.8 nm) and for laser cooling of barium.

The principle of the DFB fiber lasers is shown in Fig. 4.6. The three basic parts of a DFB fiber laser are a semiconductor pump laser, a fiber Bragg grating (FBG) and an active, doped fiber. The ytterbium (1107.8 nm and 1130.6 nm) ytterbium/erbium (1500.4 nm) doped fiber is the gain medium of the fiber laser. The semiconductor diode laser acts as a pump laser at 980 nm for the wavelength output in the range of 1020-1610 nm. The fiber Bragg grating defines a cavity as it is written onto the doped fiber by UV-illumination of the fiber through a phase mask. The wavelength is preselected to within a fraction of a nm. The wavelength



**Figure 4.6:** Principle and basic components of a DFB fiber laser [taken from KOHERAS brochure]. It consists of a fiber Bragg grating written in an actively doped fiber pumped at 980 nm or 1480 nm with a semiconductor laser.

tuning is provided by changing the temperature and is limited to about 0.4 nm. An additional advantage is the  $TEM_{00}$  output mode from the fiber.

Additionally the fiber laser can be tuned by a PZT. The tuning range is about 0.3 nm. The fiber laser wavelength can be modulated by modulating either the period of the grating, or the refractive index of the fiber. Both of these parameters are affected by the strain of the fiber. The simplest way of wavelength modulation of a fiber laser is with a PZT that affects the strain on the fiber.

Non-linearity in tuning with PZT is an issue for fiber lasers. When the power of the laser changes the frequency of the fiber laser tends to drift due to change in temperature of the fiber. To overcome this, power of the fiber lasers are set to the required value and once in thermal equilibrium measurements are taken. The operating temperature and PZT voltages of the three fiber laser systems are given in Table 4.4.

#### 4.3.4 Fluorescence Detection System

The detection (imaging) system (see Fig. 4.7) used to detect fluorescence for spectroscopic studies and for laser cooling of barium is a photomultiplier tube (PMT) in photon counting mode. It is a Hamamatsu PMT (R7205-01) with low dark current and high sensitivity at 550 nm. The fluorescence from the radiative decay of the  $^1P_1$  state atoms is imaged with a telescopic lens system. It consists

| Distributed Feedback (DFB) Fiber lasers               |                       |                     |                       |
|-------------------------------------------------------|-----------------------|---------------------|-----------------------|
| Specification                                         | 1500.4 nm             | 1130.6 nm           | 1107.8 nm             |
| Max. Output power                                     | 15 mW                 | 90 mW               | 150 mW                |
| Linewidth                                             | <48 kHz               | <50 kHz             | <100 kHz              |
| Thermal wavelength tuning coefficient                 | 1 2.5 pm/K            | 10 pm/K             | 9 pm/K                |
| Thermal tuning range (25°-55°)                        | 368 pm                | 300 pm              | 260 pm                |
| Piezo wavelength tuning coefficient (0 - 90 V)        | 3.7 pm/V<br>493 MHz/V | 3 pm/V<br>704 MHz/V | 2.6 pm/V<br>635 MHz/V |
| Piezo wavelength tuning coefficient (Our Measurement) | 292(10) MHz/V         | 410(4) MHz/V        | 420(4) MHz/V          |
| Temperature °C                                        | 44.7                  | 38.2                | 41.7                  |
| PZT Voltage (V)                                       | 4.6                   | 3.3                 | 3.1                   |

**Table 4.4:** Optical specifications of the three fiber lasers provided by the manufacturer KOHERAS. Our measured values of the piezo wavelength tuning coefficient are also shown. Last row is the operating temperature of the fiber lasers for the wavelengths and PZT voltages of the  $^{138}\text{Ba}$  resonances. In section 4.5.2 and 4.6.2 frequency calibration methods for the fiber lasers is described.

of a lens of focal length  $f = 30$  mm. The fluorescence is imaged on to a pin hole. The pinhole has a diameter of 1.0(1) mm. From the magnification of the imaging system ( $34/250 = 1/7$ ) we estimate the fluorescence from a region with diameter of 7 mm is imaged onto the PMT. After the pinhole a 25.4 mm lens collimates the light again. An interference filter of bandwidth 10 nm with a transmission of  $\epsilon_{trans} = 45\%$  centered at 550 nm is placed in-front of the PMT.

The output signal from the PMT is fed into a dual bipolar amplifier (LRS 333) to amplify the pulses. A discriminator with a low threshold converts the pulses to logic NIM signals which can be counted by pulse counters (Racal 1999) with variable integration time.

The detection efficiency  $\epsilon$  depends on solid angle  $\Omega$ , the quantum efficiency of the PMT,  $\epsilon_{PMT}$ , and the transmission efficiency of the interference filter,  $\epsilon_{trans}$ , i.e.,

$$\epsilon = \Omega \cdot \epsilon_{PMT} \cdot \epsilon_{trans}, \quad (4.3)$$

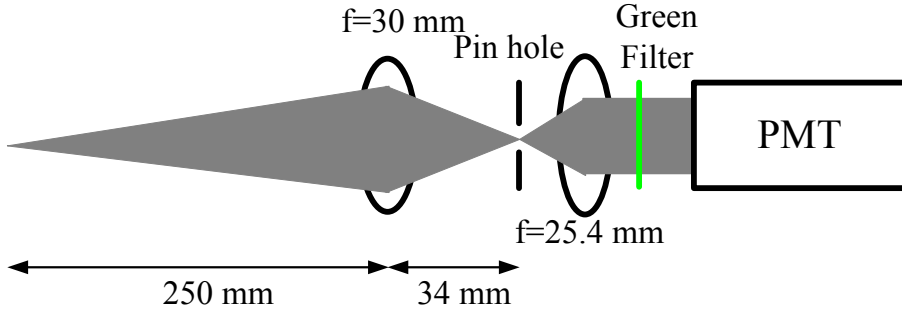


**Figure 4.7:** Fluorescence detection system used for the experiments. Fluorescence is imaged with a 30 mm lens on to a pin hole and the collimated fluorescence is spatially filtered and detected with a PMT.

with  $\varepsilon_{PMT} = 0.07(1)$ ,  $\varepsilon_{trans} = 0.45(2)$  and a solid angle for the light collection of  $\Omega = 5(1) \cdot 10^{-4}$  we get

$$\varepsilon = 1.6(4) \times 10^{-5}. \quad (4.4)$$

We can estimate the atomic flux with the detection efficiency  $\varepsilon$ , the number of photons detected and the laser beam size. With a laser beam diameter  $d$  of 1 mm the PMT count rate was 20 kHz. The fluorescence is detected from atoms passing through the laser and the imaging system selects a region  $l$ , 7 mm along the laser. The estimated atomic flux  $F$  is

$$F = \frac{\text{Countrate}}{\varepsilon \cdot d \cdot l}, \quad (4.5)$$

which was about  $10^{10}\text{ cm}^{-2}\text{s}^{-1}$  for an oven temperature of  $550^\circ\text{C}$  at a distance of 60 cm from the oven. In addition, to detect the 553.7 nm fluorescence we use a home built low-noise, high-sensitive and high-gain photodiode amplifier.

#### 4.3.5 The Data Acquisition System

In this section, the data acquisition system used to collect and store the data for barium experiments is described. The block diagram of the data acquisition system is shown in Fig. 4.8. The data acquisition system consists of the following parts: frequency counters, a SR245 interface module, a USB-to-GPIB controller module and a computer.

The SR245 ADC module is used to set the voltages for the piezo elements of the dye laser and fiber lasers to scan their frequencies. It has 8 analog I/O chan-

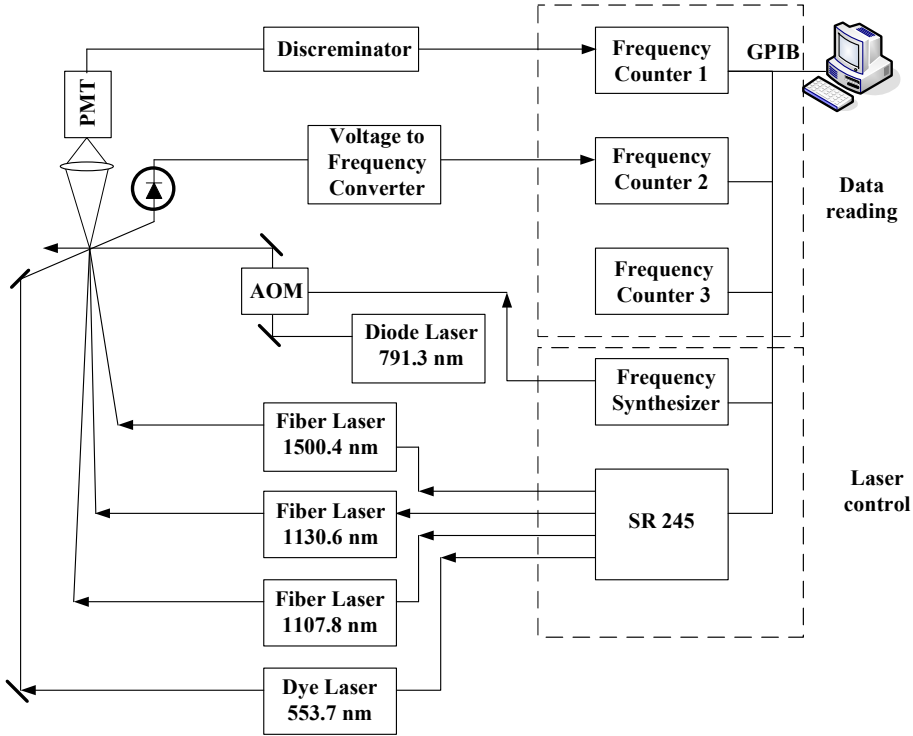


**Figure 4.8:** Data Acquisition system for the optical spectroscopy and laser cooling of barium experiments. Computer controlled GPIB is used to set the voltages to the lasers, read the data from the frequency counters and to scan the frequency of synthesizers.

nels with 13-bit resolution over the  $\pm 10.24$  VDC full scale range, with 0.05% accuracy.

A USB-to-GPIB controller (ICS Electronics model:488-USB) with a USB interface to the PC is used. It is operated with a Windows XP PC that supports this driver. Programs written in C++ language are used to set the laser frequencies, scan the laser frequencies, scan acousto optic modulators with frequency synthesizers, monitor the frequency lock status of the diode lasers and to count photons.

## 4.4 Spectroscopy of the $^1S_0$ - $^1P_1$ Transition

The  $6s^2 \ ^1S_0 - 6s6p \ ^1P_1$  transition at 553.7 nm is the only transition available for laser cooling in barium. In our experiments it is extensively used for the

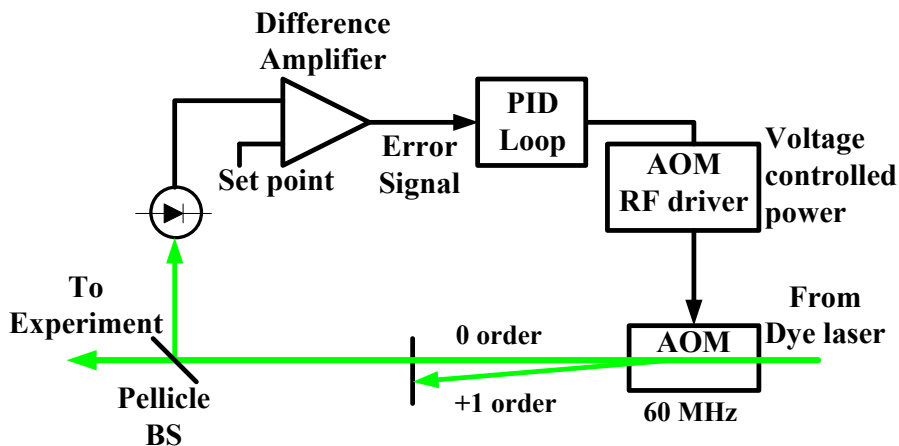


**Figure 4.9:** Layout of the optics and electronics employed for the external power stabilization of the dye laser. The power detected on the photodiode remains constant by controlling the diffraction efficiency of the AOM.

sensitive probing of other transitions. Here we give the basic properties of the spectroscopy. In particular we use this transition for frequency stabilization of the dye laser and to measure the lifetime of the  $^1P_1$  state and later for the spectroscopy of the  $D$ -states and laser cooling experiments. Precision spectroscopy of this transition can be found in the literature [121, 127–132].

#### 4.4.1 Stabilization of the Dye Laser

The Coherent dye laser nozzle and dye circulator has limitations. The system is unstable if operated at pressures higher than 5 bar. The laser output power has fast dropouts on the scale of  $10\ \mu\text{s}$  due to the air bubbles in the dye. This causes problems for frequency stabilization as well as power stabilization. To overcome this the Coherent dye laser nozzle was replaced with a nozzle RDSN 02 from Radiant Dyes. In addition we use a high pressure dye circulator RD1000 CW with an air cushion RDAC 20 from Radiant Dyes. This helped to achieve a more stable operation without fast dropouts of the dye laser. Still we have a slow drift of the laser frequency and power fluctuations at the level of 5%.

To overcome these problems we implemented external stabilization of power and frequency. Power fluctuations can occur due to change in pressure of the dye jet, mechanical disturbances, thermal fluctuations of the laser dye and aging of the laser dye.

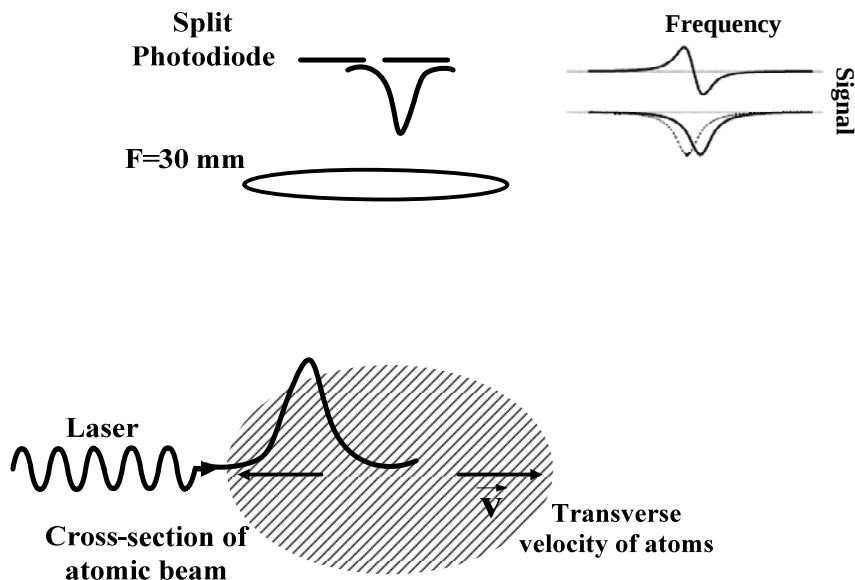


**Figure 4.10:** Principle of the Doppler shift method. Making use of the atomic beam divergence the position dependent fluorescence is detected with a split photodiode. Two identical Lorentzian line shapes offset by half their linewidth and the difference between the two Lorentzian line shapes is shown.

The schematic layout of the dye laser power stabilization is shown in Fig. 4.9. The power is regulated by the diffraction efficiency of an acousto optic modulator (AOM) in the light path. The light for the experiment is taken from the zeroth order beam of the AOM. The amount of light is monitored by a photodiode after splitting off a few percent with a beam splitter. From the detected light level we subtract a set point value in a difference amplifier. The resultant signal is an error signal which indicates that the power deviates from the set point. The error signal is used to control the diffraction efficiency of the AOM by controlling the RF power. With the servo loop closed the power stability of the system is improved and the fluctuations are less than 1% of the output power of the dye laser. The fluctuations of the power of the dye laser with power stabilization OFF and ON is shown in Fig. 4.12.

The dye laser is frequency stabilized to the  $^1S_0$  -  $^1P_1$  transition in barium employing a Doppler shift method. The method makes use of the divergence of an

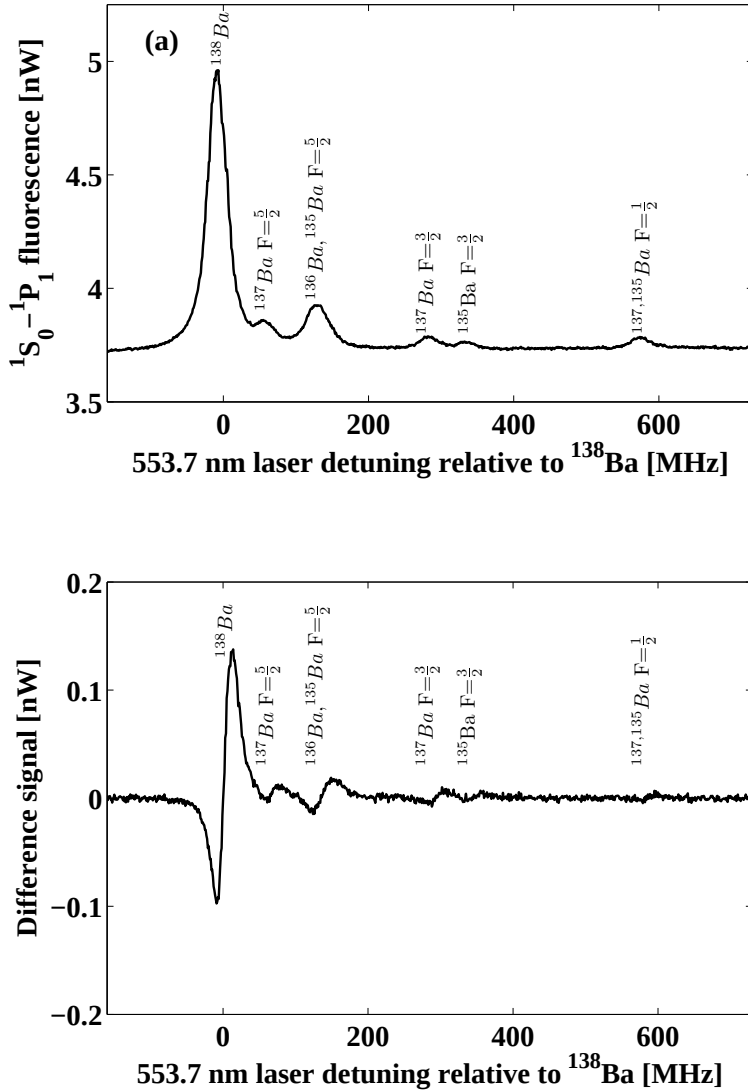


**Figure 4.11:**  $^1S_0 - ^1P_1$  fluorescence from natural barium. The fluorescence is detected with an amplified split photodiode with a sensitivity of 0.2 V/nW. **(a)** Fluorescence spectrum of the atomic beam when intersected by 553.7 nm laser beam at an angle of  $90^\circ$  from one segment of the split photodiode. The  $^{136}\text{Ba}$  peak also contains a  $^{135}\text{Ba } F = 5/2$  component. **(b)** Dispersion-like (error) signal measured from the difference between the two photodiode sections. It is obtained using the Doppler shift method shown in Fig. 4.10. The  $^{138}\text{Ba}$  isotope is used for stabilization because it has the largest signal to noise.

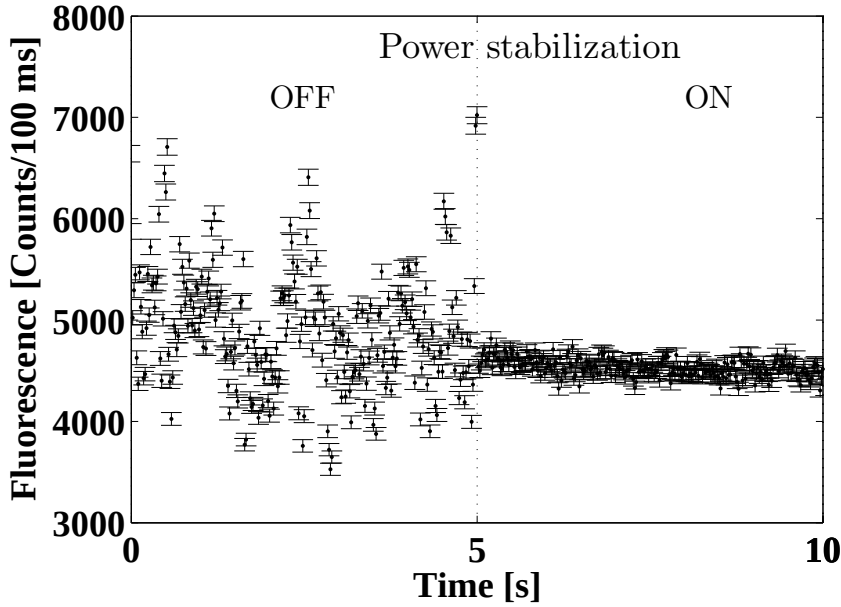


**Figure 4.12:** Fluorescence of the  $^1S_0$  -  $^1P_1$  transition detected by a photomultiplier tube when the dye laser is frequency locked to the  $^{138}\text{Ba}$  resonance with external power stabilization OFF and with external power stabilization ON.

atomic beam. The principle is shown in Fig. 4.10. The barium beam from the oven intersects with the laser beam at a right angle. The fluorescence light is imaged with a 30 mm lens on to a split photodiode with two zones, separated by 0.13 mm distance. The signal from one segment is shown in Fig. 4.11(a). The two signals from the two halves of the split-photodiode are subtracted in a difference amplifier. The difference signal is shown in Fig. 4.11(b). It exhibits a zero-crossing with a high slope, which is used for the dye laser frequency stabilization. It is amplified and fed back to the external scan input of the laser after appropriate integration. The long term drift of the dye laser frequency is minimized by locking to the  $^1S_0$  -  $^1P_1$  transition and a long term frequency stability within 1 MHz is achieved (Fig. 4.12).

#### 4.4.2 $^1P_1$ state Lifetime

Measurement of lifetime  $\tau$  of a state gives information about the absolute transition probability of a transition. It is defined as

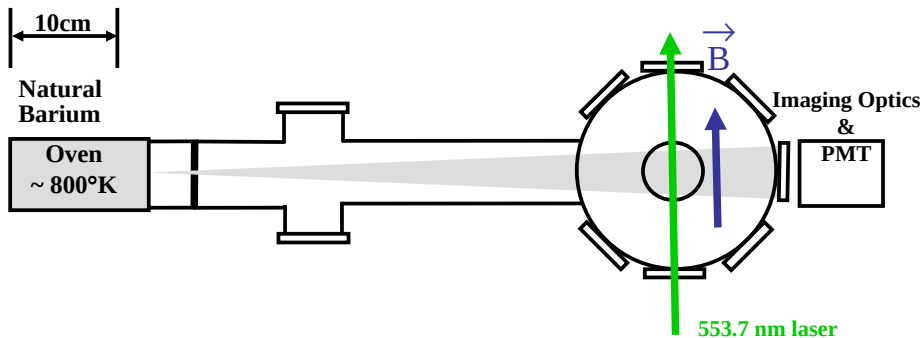


**Figure 4.13:** Setup to measure the  $^1P_1$  state lifetime in barium. The polarization of the 553.7 nm laser and magnetic field direction are perpendicular to each other. The fluorescence is detected with a PMT perpendicular to the direction of both magnetic field and polarization of the laser.

$$\tau = \frac{1}{\sum_i A_{ik}} = \frac{\lambda^3}{\langle i|r|k \rangle^2}, \quad (4.6)$$

where  $i$  is the lower state and  $k$  is the higher state. This yields transition dipole matrix elements. Thus, lifetime measurements are important tests as well as input for the computed wave functions and models of the electron distribution in complex atoms like heavy alkaline earth elements.

We used the Hanle effect [133,134] to measure the lifetime of the  $^1P_1$  state by measuring the intensity distribution of the 553.7 nm fluorescence in an external homogeneous magnetic field. The polarization of the 553.7 nm laser light is perpendicular to the magnetic field, which is applied perpendicular to the atomic barium beam. We detected the fluorescence at different magnetic fields. Shown in Fig. 4.14 are the measurements for the  $^{138}\text{Ba}$  and  $^{136}\text{Ba}$  isotopes respectively. Atoms in a homogeneous magnetic field  $B = \{0,0,B_z\}$  are excited by the polarized electromagnetic wave

$$E = E_y \cos(\omega t - kx), \quad (4.7)$$

the frequency  $\omega$  is on resonance with the  $^1S_0 - ^1P_1$  transition. The field-dependent term falls to half its maximum value at the magnetic fields  $B_{\pm}$  given by

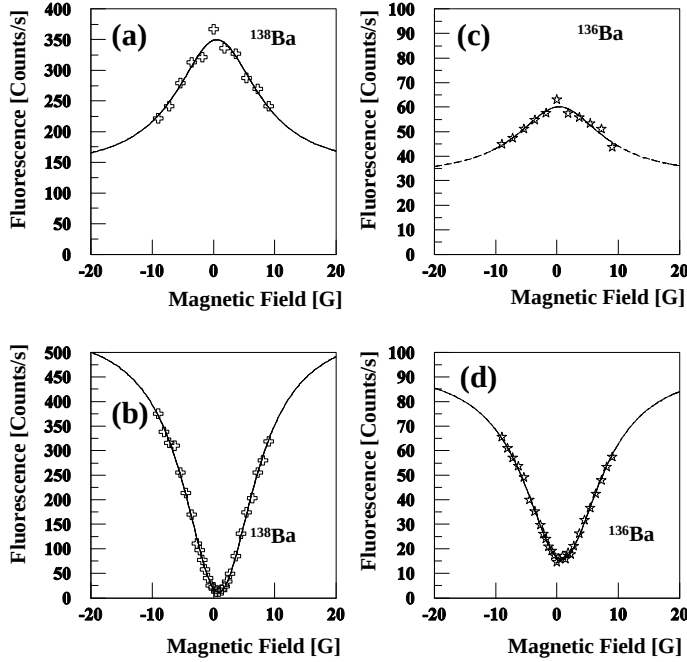


**Figure 4.14:** Lifetime measurement of the  $^1P_1$  state in barium using Hanle effect. **a, c** corresponds to the polarization of the laser perpendicular to the direction of the PMT for  $^{138,136}\text{Ba}$  isotopes. **b, d** corresponds to the polarization of the laser in the the direction of the PMT for  $^{138,136}\text{Ba}$  isotopes. For  $^{136}\text{Ba}$  isotope at zero magnetic field the fluorescence does not go to zero because of the residual fluorescence from the  $F = 5/2$  state of the  $^{135}\text{Ba}$  isotope that overlaps with  $^{136}\text{Ba}$  in the  $^1S_0 - ^1P_1$  transition.

$$\Gamma = 2\omega_L = 2\frac{g_J\mu_B B_{\pm}}{\hbar}, \quad (4.8)$$

where  $g_J$  is Lande factor,  $\hbar = h/2\pi$ ,  $h$  is Planck constant,  $\mu_B$  is Bohr magneton. The full width at the half maximum of the signal ( $\Delta B_{1/2}$ ),  $\Gamma = 1/2\pi\tau$ , is related to the radiative lifetime through the relation

$$\tau = \frac{\hbar}{2\pi g_J \mu_B \Delta B_{1/2}}. \quad (4.9)$$

The Lande factor  $g_J = 1$  for the  $^1P_1$  state,  $\mu_B/\hbar = 1.4 \text{ MHz/G}$  and from the width of the measured depolarization curve, the excited state lifetime is determined. The

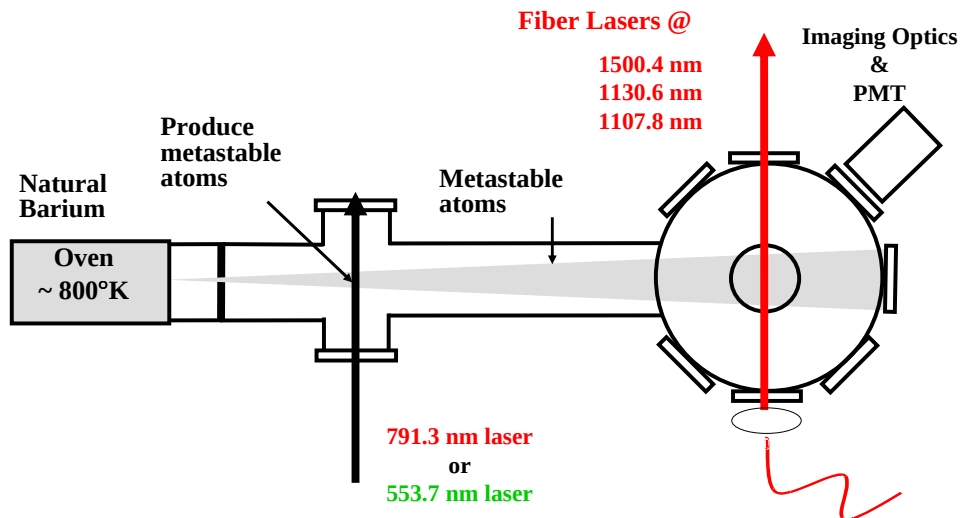


**Figure 4.15:** Set up for production and detection of barium atoms in the metastable D-states. Region 1 is to populate the metastable states with 553.7 nm or 791.3 nm laser. Region 2 to probe the metastable atoms driven by the infrared lasers by detecting the 553.7 nm fluorescence from the  $^1P_1$  state with a PMT.

results are fitted with a Lorentzian function. From the magnetic field FWHM of the curve was extracted. In Fig. 4.14 the minimum is not exactly at zero magnetic field because of the residual magnetic field due to the magnetization of the the experimental chamber and the earth's magnetic field. We did not shield any external fields. Using the Eq. 4.9, the lifetime of the  $^1P_1$  state was obtained to be 8.0(5) ns, which is in good agreement with the reported value of 8.2(2) ns [122].

## 4.5 Laser Spectroscopy of Metastable D-states

To conduct laser spectroscopy and to find the transitions we realized an experimental setup that satisfies two requirements. First the production of metastable atoms and second the probing of the population of the metastable states by laser spectroscopy. The layout of the experimental setup is shown in Fig. 4.15.

We provide two separate regions for the interaction with laser beams. In the first section we pump the atoms optically into the metastable states. This is done either by the  $^1S_0 - ^1P_1$  transition using the dye laser or the  $^1S_0 - ^3P_1$  transition using a diode laser (Fig. 4.15). In the second interaction region we probe the metastable

atoms by driving the atom to the  $^1P_1$  excited state with the corresponding laser. The spatial separation of metastable production and probing is advantageous for the suppression of scattered light onto the detector. The transitions are always detected by the fluorescence at 553.7 nm from the  $^1P_1 - ^1S_0$  transition since the visible light is easily detected with a photomultiplier tube. Here we explain the principle and show different methods of frequency calibration of the scanning of the fiber laser with the PZT voltage.

#### 4.5.1 Production and Detection of Metastable Atoms

Conventionally, metastable barium states are populated in a plasma discharge inside the atomic-beam oven [135] or by electron bombardment on the atomic beam [136]. Typically a few percent of the atoms can be transferred to the metastable states. Unlike the above techniques, we use optical pumping and exploit the branching ratio of the  $^1P_1$  and the  $^3P_1$  states to populate the metastable  $D$ -states. The two feasible optical methods are by the  $^1S_0 - ^1P_1$  transition and the  $^1S_0 - ^3P_1$  transition.

When the barium atoms are excited by a laser at 553.7 nm from the  $^1S_0$  state to the  $^1P_1$  state they can also decay to the  $^1D_2$ ,  $^3D_2$  and  $^3D_1$  metastable states. The ratio of population is 1 : 1/2.3 : 1/80.

The  $^3D_2$  and  $^3D_1$  states can also be populated by driving the  $^1S_0 - ^3P_1$  transition at 791.3 nm. Around 60% of the barium atoms decay to the  $^3D_2$  and  $^3D_1$  states and 40% decay back to the ground state. Cycling the  $^1S_0 - ^3P_1$  transition effectively populates the triplet  $D$ -states with a ratio of 65% in the  $^3D_2$  and 35% in the  $^3D_1$  state. Here we employ a diode laser, which is locked to the ‘a1’ line of the P(52)(0-15) hyperfine transition in molecular  $I_2$  (Chapter 3.3.2).

#### Detection of the D-P Transitions

For observing the  $D - P$  transitions we use a sensitive indirect method. The fluorescence at 553.7 nm of the  $^1P_1$  state decay to the  $^1S_0$  ground state is detected rather than the fluorescence in the infrared. The advantages of this method are:

1. The laser wavelength of the driving transition is at a different wavelength than the wavelength of the detected photons. It can be separated easily by an interference filter.
2. PMT's in the visible region have high gain and good quantum efficiency ( $\simeq 10\%$ ) and hence provide good signal to noise ratio for single photon counting. The infrared fluorescence from transitions to  $^1D_2$  and  $^3D_{1,2}$  states

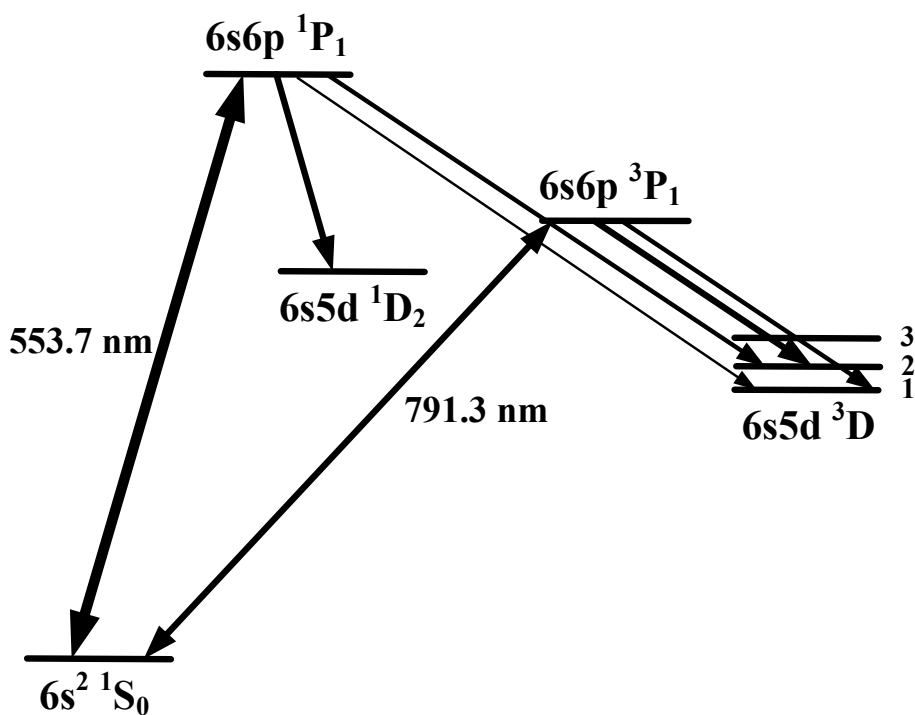


**Figure 4.16:** Two possible options to populate the triplet D-states using the  $^1S_0 - ^1P_1$  transition at 553.7 nm and the  $^1S_0 - ^1P_1$  intercombination line at 791.3 nm.

is weaker than the fluorescence to the ground state. Detection of fluorescence at 1500.4 nm, 1130.6 nm and 1107.8 nm is difficult with a photodiode and PMT's in the infrared have very low quantum efficiency and have to be cooled to minimize the dark counts.

3. The possibility to use the  $^1S_0 - ^1P_1$  transition to probe the ground state population efficiently. Since we can cycle the  $^1S_0 - ^1P_1$  transition we scatter many photons per atom.
4. We can extract quantitative information of repumping of atoms to the  $^1S_0$  state for laser cooling experiments.

Based on these considerations we realized the experimental setup for state selective sensitive laser spectroscopy of metastable barium with high signal to noise ratio. The latter is of particular importance in view of planned spectroscopy of radium.

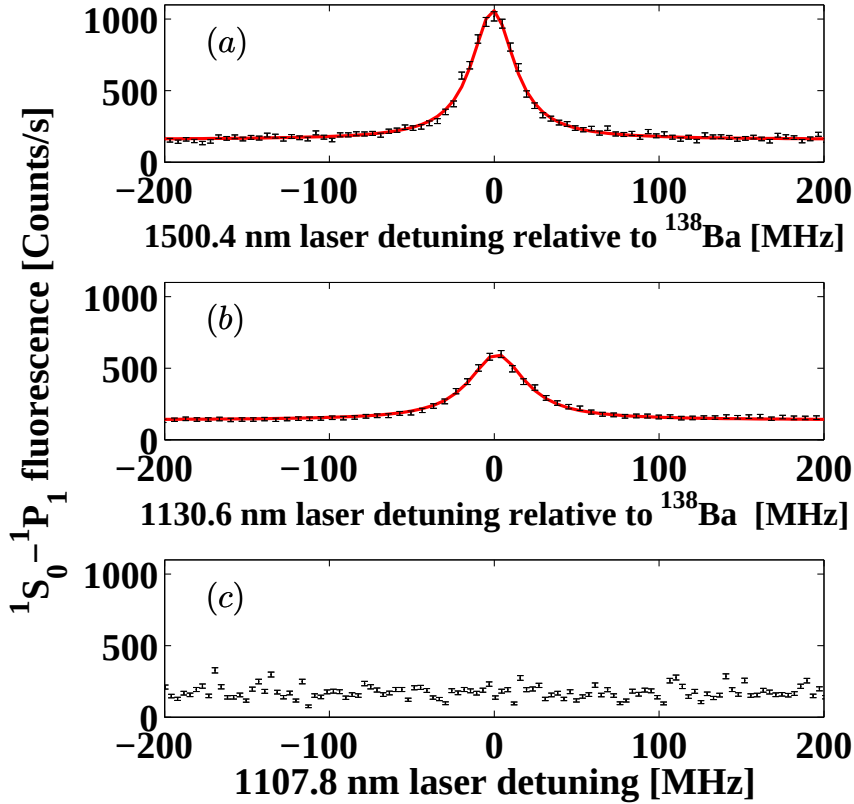


**Figure 4.17:** (a)  $^1D_2 - ^1P_1$  transition detected by 553.7 nm fluorescence with a PMT. The line through the data points is a fit of a Lorentzian function with a linewidth of 31(2) MHz, which is greater than 19 MHz. (b)  $^3D_2 - ^1P_1$  transition observed by detecting fluorescence at 553.7 nm with a PMT. The line through the data points is a fit of Lorentzian line shape with a line width of 41(2) MHz. In both cases the broadening is due to the angle made by the infrared laser beams with respect to the atomic beam. (c) The  $^3D_1 - ^1P_1$  transition detected by 553.7 nm fluorescence. In all the three cases metastable D-states are populated by driving the  $^1S_0 - ^1P_1$  transition. Since the population of the  $^3D_1$  state is 80 times less than the  $^1D_2$  state we cannot detect it. For all the plots each data point has an integration time of 1 second.

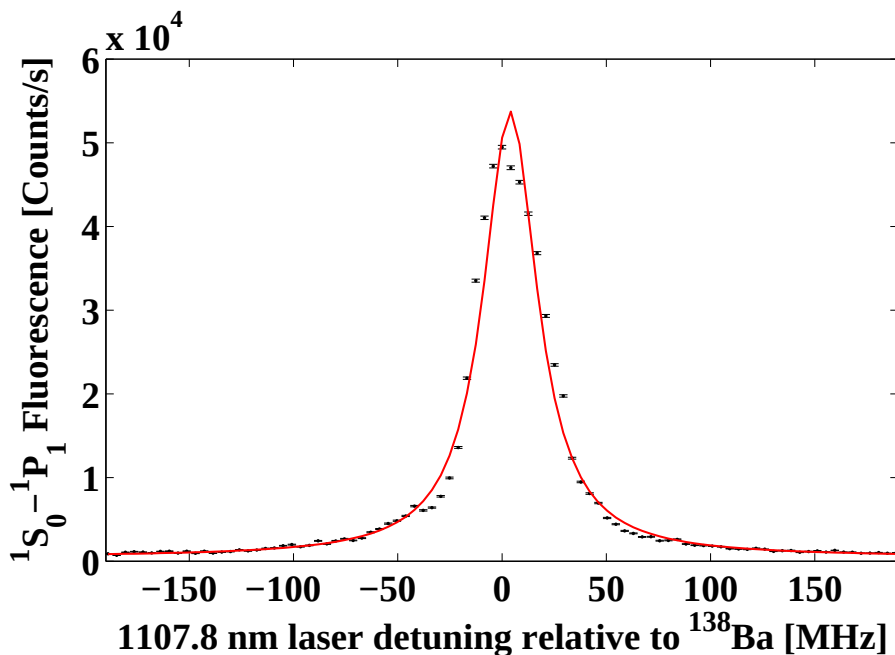


**Figure 4.18:** Detection of  $^3D_1 - ^1P_1$  transition when  $^3D_1$  state is populated through  $^3P_1$  level by 791.3 nm diode laser. Each data point is taken with an integration time of 1 second. The line through the data points is fit by a Lorentzian function. The linewidth is 32(2) MHz.

#### 4.5.2 Even Isotopes

The  $^{138}\text{Ba}$  atoms in the  $^1S_0$  ground state are pumped to the  $^1D_2$  state with the 553.7 nm laser in resonance with the  $^1S_0 - ^1P_1$  transition. After setting the temperature of the fiber laser appropriately we scan the laser by the PZT. The 1500.4 nm resonance detected through fluorescence at 553.7 nm is shown in Fig. 4.17(a). The linewidth of a Lorentzian function fitted to the data is 31(2) MHz. This is larger than the natural linewidth of 19 MHz due to the excited state lifetime. The broadening is due to the alignment of the 1500.4 nm laser beam with respect to the atomic beam.

The steps described for the  $^1D_2 - ^1P_1$  transition are repeated to tune the fiber laser to resonance for the  $^3D_2 - ^1P_1$  transition at 1130.6 nm. The resonance is shown in Fig. 4.17(b). The ratio of the peak amplitudes of the  $^1D_2$  and  $^3D_2$  states provide the information on the ratio of population in these states, which is 2.0(3). This is in agreement with the value 2.3(1) reported by Bizzari et al. [119].

Further, comparing the count rate for the  $^1D_2$  and  $^3D_1$  states we can estimate the ratio of the population in these transitions. Since the count rate is on the order of 800 Hz for the  $^1D_2 - ^1P_1$  transition we would expect a count rate of  $\approx 10$  Hz for the  $^3D_1 - ^1P_1$  transition. With a background rate due to scattered light of the order of 200 Hz we have a  $S/N$  of only 0.7. Thus a better experimental configuration was necessary. For this we use a diode laser at 791.3 nm to drive the  $^1S_0 - ^3P_1$  transition. Then we populate about 35% of all the atoms in the  $^3D_1$  state rather than about 1%.

The experimental setup consists of two diode lasers that are frequency offset locked. One of the diode lasers is frequency stabilized to the ‘a1’ line of the P(52)(0-15) transition at 791.3 nm in molecular iodine, which is the frequency reference for the  $^1S_0 - ^3P_1$  transition in barium. The spectroscopy of iodine was described in section 3.3.2. The frequency offset locking technique of two diode lasers is described in the Appendix A. The diode laser used to populate triplet  $D$ -states is frequency offset locked to the diode laser stabilized to iodine. It is tuned to the  $^{138}\text{Ba}$  resonance of the  $^1S_0 - ^3P_1$  transition. Between 3-5 mW power from the diode laser is available during the measurements. The fiber laser is scanned with the PZT over the resonance. A typical spectrum is shown in Fig. 4.18.

During the experiments we observed that the passive frequency stability of the fiber lasers is good and we observe a drift of less than 13(2) MHz/h. In order to achieve a higher stability the lasers could be locked to temperature stabilized Fabry-Perot cavities.

Frequency detuning of the repump lasers plays an important role in laser cooling experiments. This is discussed in Chapter 5. The frequency scanning with the PZT voltage of the fiber lasers is calibrated using two methods: In the first method, we used a resonant type electro-optic modulator (EOM) for the 1500.4 nm and 1130.6 nm lasers. The EOM is used for the generation of frequency side bands and its working principle and construction is described in the Appendix C. The lithium tantalate crystal is driven at a frequency of 33.90 MHz. Typical RF power levels used to achieve a modulation index of the order of 1 is 5 dBm.

We detect the side bands by spectroscopy of the metastable states. The side bands observed are shown in Fig. 4.19. From a fit we determine the side band spacing to 84(2) mV (1130.6 nm) and 116(3) mV (1500.4 nm) for a modulation frequency of 33.9 MHz. We obtain from this measurement a scanning of 404(10) MHz/V and 292(10) MHz/V respectively.

The second method used to calibrate the fiber laser is the Zeeman splitting of energy levels in a uniform external magnetic field. Here we populated the

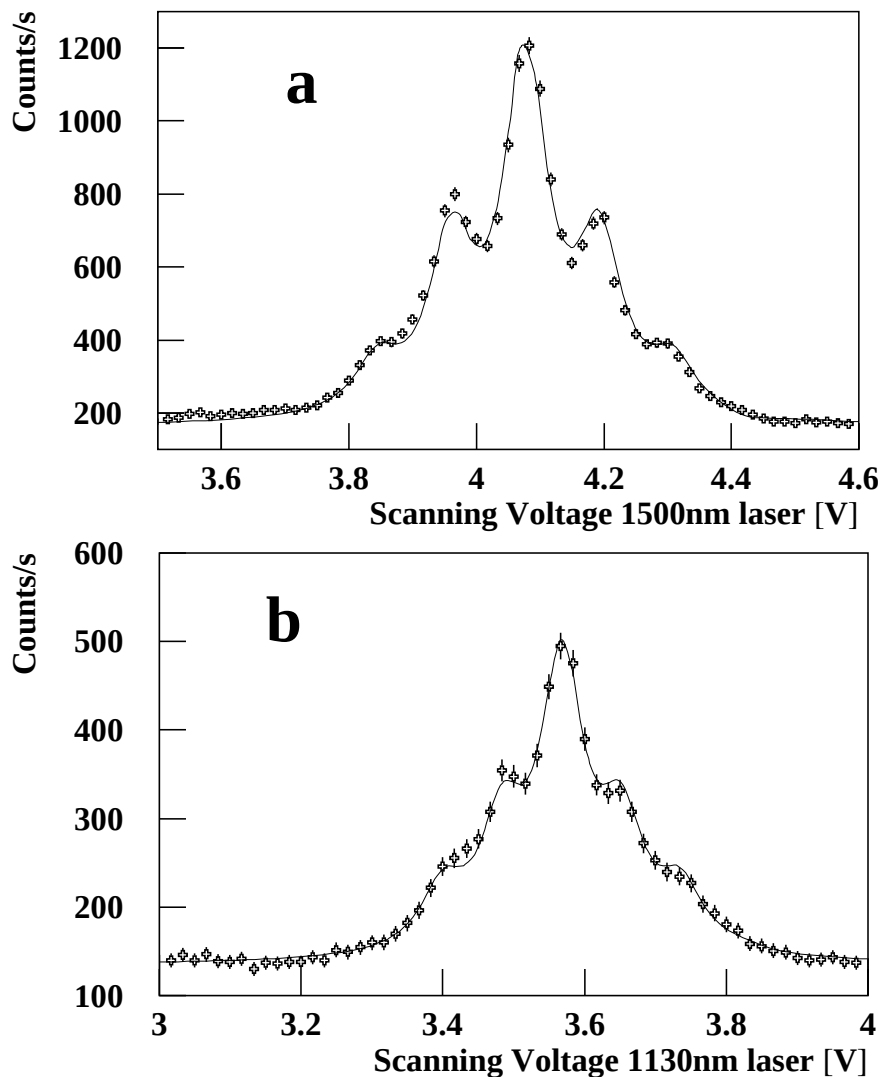


**Figure 4.19:** The spectral width of the (a) 1500.4 nm and (b) 1130.6 nm lasers are broadened by an EOM with modulation frequency of 33.9 MHz. The sidebands are clearly visible in the spectrum and are used to calibrate the tuning of the fiber lasers. The line is a fit taking three sidebands into account.

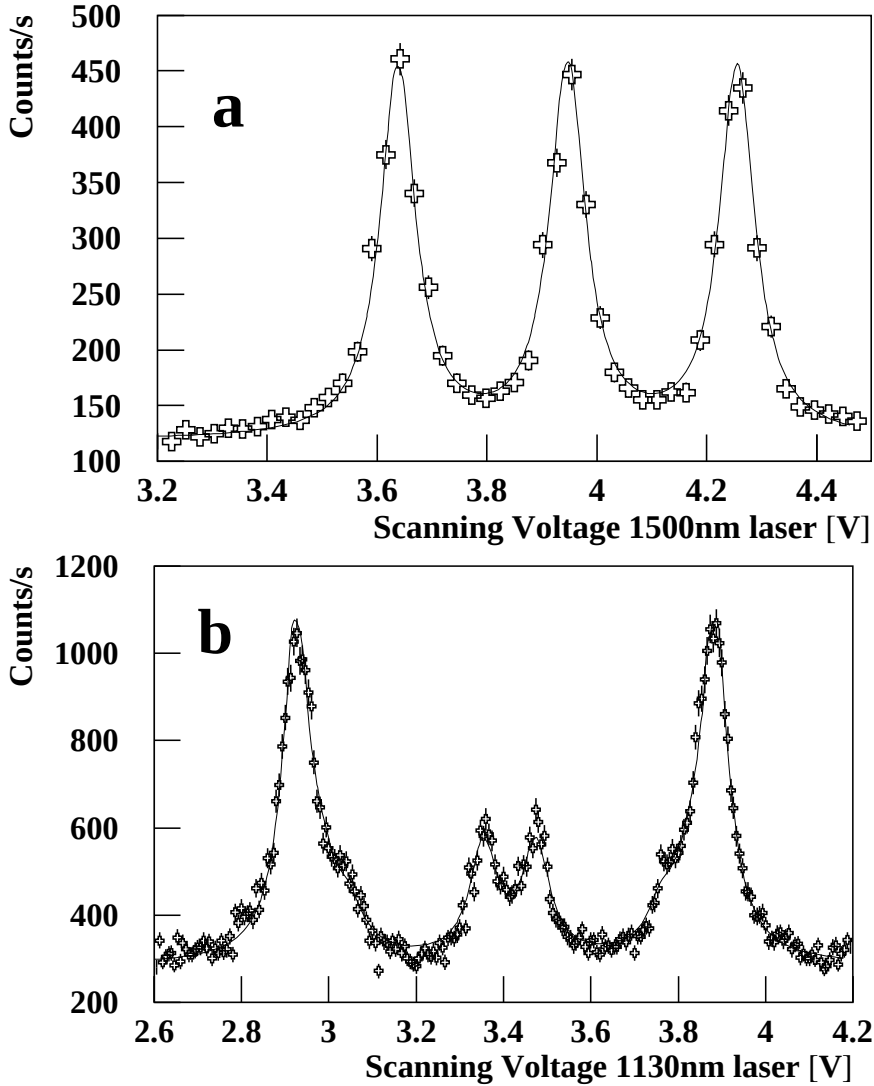


**Figure 4.20:** Spectrum of the D-P transitions in an external magnetic field of 65(3) Gauss by detecting the 553.7 nm fluorescence of the  $^1P_1$  state. (a) With the detuning of the 1500.4 nm laser. (b) With the detuning of the 1130.6 nm laser. Each data point is taken with an integration time of 1 second. The fit through the data is a Lorentzian function.

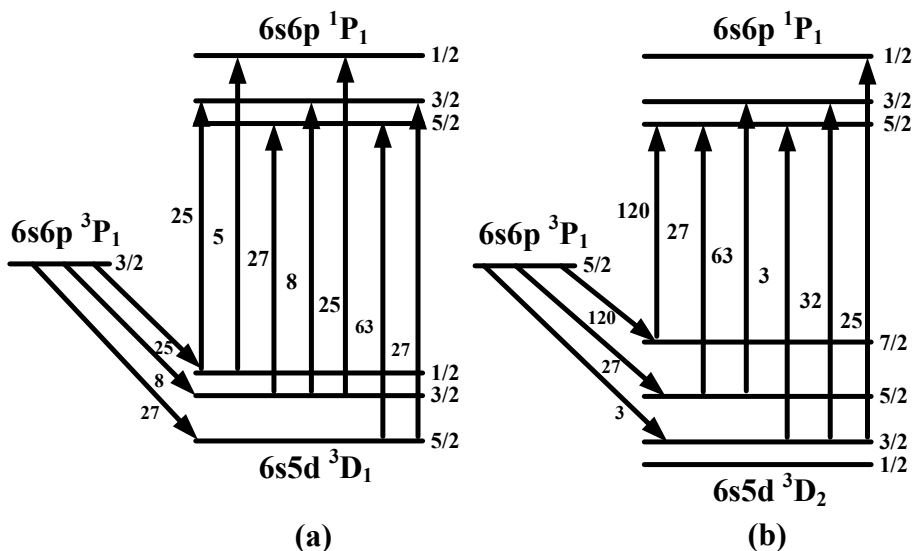


**Figure 4.21:** (a) Possible transitions from the hyperfine states of the  $^{137,135}\text{Ba}$  isotopes for the  $^3P_1 - ^3D_1$  and  $^3D_1 - ^1P_1$  transitions. The calculated strengths for each component of all the transitions are given. (b) Possible transitions from the hyperfine states of the  $^{137,135}\text{Ba}$  isotopes for the  $^3P_1 - ^3D_2$  and  $^3D_2 - ^1P_1$  transitions. The calculated strengths for each component of all the transitions are shown. Strengths are multiplied with a common factor and are given in the smallest possible integer numbers.

$^1D_2$  level with the 553.7 nm laser. An external magnetic field of 65(3) Gauss is applied perpendicular to the  $^{138}\text{Ba}$  atomic beam. The laser is perpendicular to the atomic beam to reduce the Doppler effect. The resulting spectrum when scanning the laser consists out of three peaks (1500.4 nm) based on the selection rules  $\Delta J = 0, \pm 1$  (Fig. 4.20). The voltage difference between the three peaks determined from a fit is 0.31(1) V (1500.4 nm) for an applied magnetic field of 65(3) Gauss. The splitting between the peaks is  $\mu \cdot \mathbf{B}$ . With this we get a scanning of 294(20) MHz/V for this laser. In Table 4.4 we give all the calibrations for the fiber lasers. Their values differ significantly from the scanning sensitivity given by the manufacturer.

## 4.6 Hyperfine Structure of Triplet D-States

### 4.6.1 Expected Hyperfine Spectrum

The interaction between the electron and the moments of the nucleus results in hyperfine structure for an atom. For isotopes with nuclear spin  $I$ , the hyperfine splitting can be written as

$$\nu = \frac{1}{2}C \cdot A + B \cdot C1, \quad (4.10)$$

where

$$C = F(F + 1) - I(I + 1) - J(J + 1),$$

$$C1 = \frac{3C(C + 1) - I(I + 1)J(J + 1)}{8I(2I - 1)J(2J - 1)},$$

where  $A$  is the magnetic dipole interaction constant,  $B$  is the electric-quadrupole coupling constant,  $J$  is the angular momentum of the electron and  $F$  is the total angular momentum. It is important to know how the isotopes with nuclear spin ( $I \neq 0$ ) influence the transition strength within the hyperfine states of these isotopes. This plays an important role in laser cooling and trapping because of the optical pumping effects that arise while using polarized light.

For barium isotopes with nuclear spin  $I = 3/2$  one can find the hyperfine splitting for the triplet  $D$ -states as well as for the singlet and triplet  $P$  state in literature. The hyperfine splitting frequencies of the  $^3D_1 - ^1P_1$  transition known from references [121, 135] are given Table 4.5 and for the  $^3D_2 - ^1P_1$  transition in Table 4.6. We can use the position of these lines for additional calibration of the fiber lasers. The hyperfine structure splitting is sensitive to the overlap of the electronic wave function with the nucleus. A measurement of the hyperfine structure of the  $P$  and  $D$ -states in radium is needed (for laser cooling as well). This will be important for evaluations of possible EDM.

### Transition Strengths: Calculation

The calculation of transition strengths for the triplet  $^3D_1$  and  $^3D_2$  states to the singlet  $^1P_1$  state of barium is presented.

Transition strengths are governed by selection rules and define the probability of the transition occurring. The square of the dipole transition matrix element  $\mu_{eg}$  gives the transition strength. The dipole transition matrix element is given by

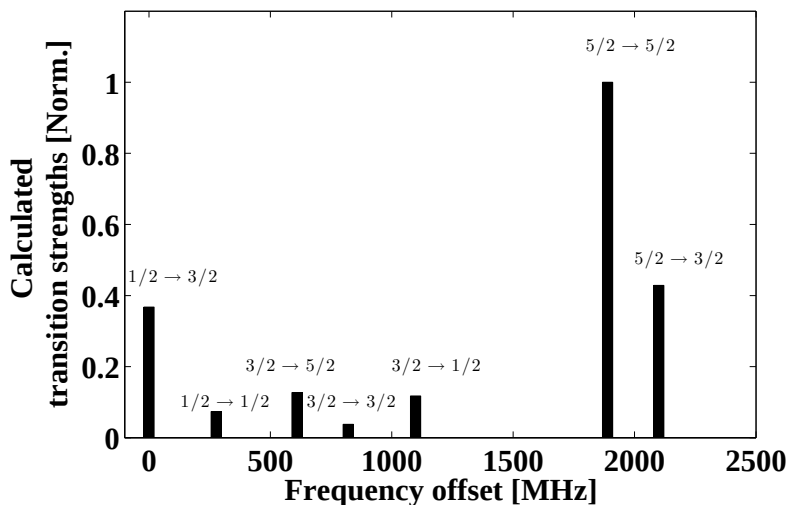


**Figure 4.22:** Calculated transition strengths of the  $^{137}\text{Ba}$  isotope for the  $^3D_1 - ^1P_1$  transition. In the spectrum the transition strengths are normalized to the  $5/2 \rightarrow 5/2$  transition. The frequencies are relative to the  $1/2 \rightarrow 3/2$  transition.

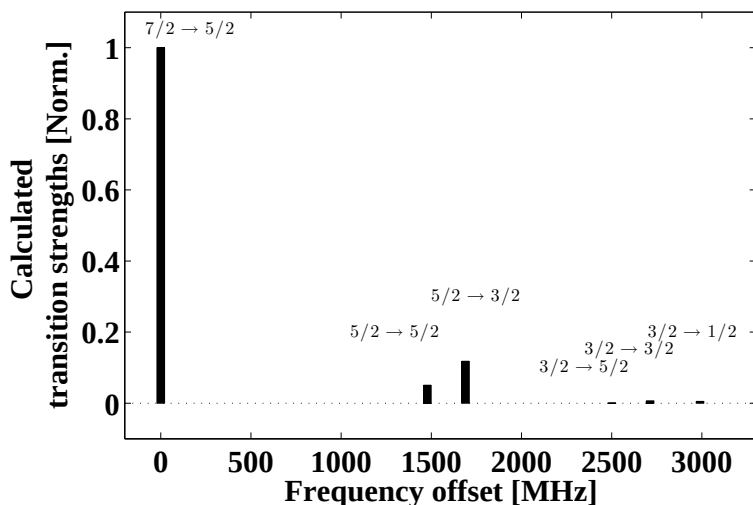


**Figure 4.23:** Calculated transition strengths of the  $^{137}\text{Ba}$  isotope for the  $^3D_2 - ^1P_1$  transition. In the spectrum the strengths are normalized to the  $7/2 \rightarrow 5/2$  transition. The frequencies are relative to the  $7/2 \rightarrow 5/2$  transition.

$$\mu_{eg} = (-1)^{1+L'+S+J+J'+I-M'} \sqrt{(2J+1)(2J'+1)(2F+1)(2F'+1)} \times \\ \left\{ \begin{array}{ccc} L' & J' & S \\ J & L & 1 \end{array} \right\} \left\{ \begin{array}{ccc} J' & F' & I \\ F & J & 1 \end{array} \right\} \left( \begin{array}{ccc} F & 1 & F' \\ M_F & q & M'_F \end{array} \right), \quad (4.11)$$

where  $I$  is the nuclear spin,  $S$  is the spin quantum number and the terms with in  $\{ \}$  and  $( )$  are Wigner 6j and 3j symbols respectively. The quantum numbers without an accent are ground state terms and with an accent are excited states.  $q$  indicates the polarization of the light with  $q = 0$  for linearly polarized light and  $q = \pm 1$  for right and left circularly polarized light. The transition matrix consists of a radial and an angular component. The radial part is the same for all the transitions between two terms in the ground state and the excited state, and is therefore only a numerical factor that determines the absolute value of the coupling.

In the calculation of transition strengths, all the hyperfine transitions of the  $^{137,135}\text{Ba}$  isotopes for the  $(L, S, J) {}^3P_1$  state to  $(L', S', J') {}^3D_{1,2}$  states and  $(L, S, J) {}^3D_{1,2}$  states to the  $(L', S', J') {}^1P_1$  state contain a common factor

$$A1 = (2J+1)(2J'+1) \left\{ \begin{array}{ccc} L' & J' & S \\ J & L & 1 \end{array} \right\}^2, \quad (4.12)$$

The transition strengths are equally probable for the different polarization of the light i.e.,  $q = M'_F - M_F = 0, \pm 1$  for the allowed transitions. It is a common factor, which is equal to 1 in equation 4.11 for the purpose of our calculation

$$\sum_{M_F \rightarrow M'_F} \left( \begin{array}{ccc} F & 1 & F' \\ M_F & q & -M'_F \end{array} \right)^2 = 1, \quad (4.13)$$

Hence for the calculation of the transition strengths equation 4.11 reduces to the following

$$A1 \times (2F+1)(2F'+1) \left\{ \begin{array}{ccc} J' & F' & I \\ F & J & 1 \end{array} \right\}^2. \quad (4.14)$$

The calculated transition strengths for the  ${}^3P_1 - {}^3D_1$  transition and for the  ${}^3D_1 - {}^1P_1$  transition for different initial  $F$  values and final  $F'$  values for  $J = 1$  to  $J' = 1$  are shown in Fig. 4.21(a). Similarly, the calculated transition strengths for the  ${}^3P_1 - {}^3D_2$  transition and for the  ${}^3D_2 - {}^1P_1$  transition for different initial  $F$  values and final  $F'$  values for  ${}^3P_1(J=1) - {}^3D_2(J=2)$  and  ${}^3D_2(J=2)$  to  ${}^1P_1(J'=1)$  are shown in Fig. 4.21(b).

| $^3D_1 - ^1P_1$ transition ( $^{137}\text{Ba}$ ) |                 | Relative transition strengths |             |             |
|--------------------------------------------------|-----------------|-------------------------------|-------------|-------------|
| $F \rightarrow F'$                               | Frequency [MHz] | Calculation                   | Measurement | Ratio [M/C] |
| $1/2 \rightarrow 3/2$                            | 1264.29(25)     | 0.37                          | 1.15(2)     | 3.10(5)     |
| $1/2 \rightarrow 1/2$                            | 987.34(42)      | 0.0735                        | 0.243(8)    | 3.3(1)      |
| $3/2 \rightarrow 5/2$                            | 654.37(21)      | 0.13                          | 0.137(6)    | 1.08(5)     |
| $3/2 \rightarrow 3/2$                            | 443.23(25)      | 0.038                         | 0.066(4)    | 1.74(11)    |
| $3/2 \rightarrow 1/2$                            | 166.28(42)      | 0.12                          | 0.154(6)    | 1.28(5)     |
| $5/2 \rightarrow 5/2$                            | -624.6(21)      | 1.00                          | 1.00(2)     | 1.00(2)     |
| $5/2 \rightarrow 3/2$                            | -835.74(25)     | 0.43                          | 0.46(1)     | 1.07(2)     |
| $^3D_1 - ^1P_1$ transition ( $^{135}\text{Ba}$ ) |                 | Relative transition strengths |             |             |
| $F \rightarrow F'$                               | Frequency [MHz] | Calculation                   | Measurement | Ratio [M/C] |
| $1/2 \rightarrow 3/2$                            | 1113.32(26)     | 0.37                          | 1.13(3)     | 3.05(8)     |
| $1/2 \rightarrow 1/2$                            | 889.56(44)      | 0.0735                        | 0.27(1)     | 3.7(1)      |
| $3/2 \rightarrow 5/2$                            | 592.26(22)      | 0.13                          | 0.15(1)     | 1.15(8)     |
| $3/2 \rightarrow 3/2$                            | 389.37(26)      | 0.038                         | 0.067(5)    | 1.76(13)    |
| $3/2 \rightarrow 1/2$                            | 165.61(44)      | 0.12                          | 0.147(8)    | 1.23(7)     |
| $5/2 \rightarrow 5/2$                            | -556.09(22)     | 1.00                          | 1.00(3)     | 1.00(3)     |
| $5/2 \rightarrow 3/2$                            | -758.98(26)     | 0.43                          | 0.48(2)     | 1.11(5)     |

**Table 4.5:** The first column gives all the possible transitions of the  $^3D_1 - ^1P_1$  based on the selection rules for a dipole transition. The second column gives the frequencies of the transitions known from the hyperfine splitting measurements of the  $^3D_1$  and  $^1P_1$  states from centroid to centroid [121, 135]. The third column gives the calculated transition strengths that are normalized to the strongest transition. The fourth column consists of the transition strengths from our measurement normalized to the strongest transition. The fifth column gives the ratio of strengths from the measurement and calculation.

The transition strengths of the seven transitions shown in Fig. 4.22 are the product of the transition strengths of the  $^3P_1 - ^3D_1$  transition and the  $^3D_1 - ^1P_1$  transition respectively when the  $F = 3/2$  hyperfine state of the  $^3P_1$  level is populated. Similarly the transition strengths of the 6 transitions shown in Fig. 4.23 are the product of the transition strengths of the  $^3P_1 - ^3D_2$  transition and the  $^3D_2 - ^1P_1$  transition respectively when the  $F = 5/2$  hyperfine state of the  $^3P_1$  level is populated.

#### 4.6.2 Results from the Triplet D-states

To populate the hyperfine levels in the triplet  $D$ -states we locked the diode laser to the  $^1S_0, F = 3/2 - ^3P_1, F = 3/2$  transition. The selection rules allow to populate

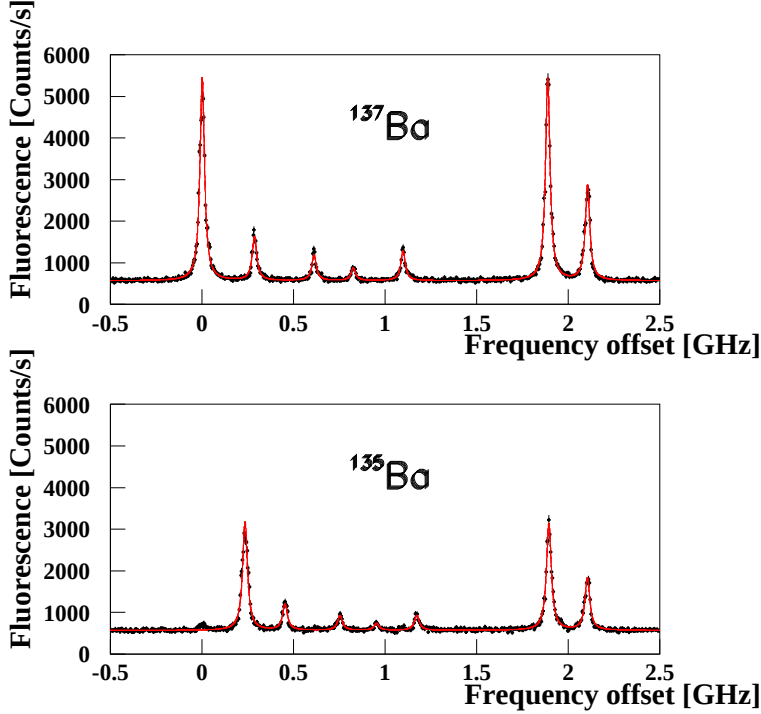


**Figure 4.24:** Measured hyperfine spectra of  $^{137,135}\text{Ba}$  isotopes for the  $^3D_1 - ^1P_1$  transition when frequency of the 1107.8 nm laser is scanned 3 GHz. Each point has an integration time of 1 second. The line through the data points is a fit of a Lorentzian function with a halfwidth of 20(1) MHz. The frequency scale is relative to the  $1/2 \rightarrow 3/2$  transition (1st peak of the  $^{137}\text{Ba}$  isotope).

the  $F = 1/2, 3/2$  and  $5/2$  states of the  $^3D_1$  level for  $^{137}\text{Ba}$  and  $^{135}\text{Ba}$ , which is shown in Fig. 4.21(a).

The hyperfine splitting of  $^{137}\text{Ba}$  and  $^{135}\text{Ba}$  isotopes for the  $^3D_1 - ^1P_1$  transition are measured simultaneously (see Section 4.7). The 1107.8 nm laser frequency is scanned spanning the entire hyperfine spectrum of 3 GHz. The measured hyperfine spectrum of the  $^3D_1 - ^1P_1$  transition for  $^{137}\text{Ba}$  and  $^{135}\text{Ba}$  isotopes is shown in Fig. 4.24. All the seven transitions are fitted with a sum of Lorentzian functions of the form

$$F_\nu = C + \sum_{i=1}^7 \frac{A_i}{1 + \left(\frac{\nu - \nu_i}{\Gamma/2}\right)^2} \quad (4.15)$$

where  $C$  represents the background counts,  $i$  is the sum of all the hyperfine com-

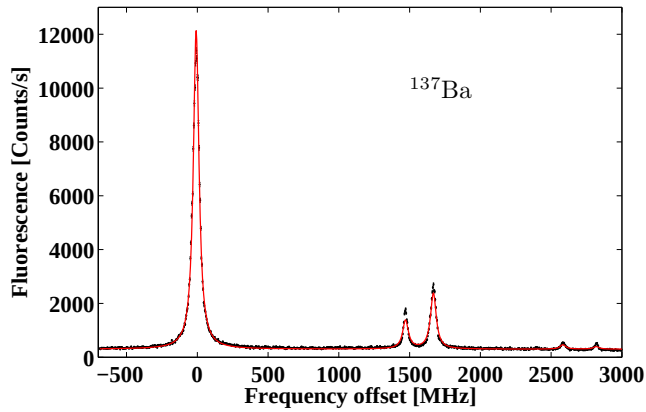


**Figure 4.25:** Measured hyperfine spectrum of the  $^{137}\text{Ba}$  isotope for the  $^3\text{D}_2 - ^1\text{P}_1$  transition when frequency of the 1130.6 nm laser is scanned 4 GHz. Each point has an integration time of 1 second. The line through the data points is a fit of a Lorentzian line shape with a line width of 20(1) MHz. The frequency scale is relative to the  $7/2 \rightarrow 5/2$  transition (first peak of the  $^{137}\text{Ba}$  isotope). Here the diode laser is locked to  $^{137}\text{Ba}$   $F = 5/2$  state of the  $^3\text{P}_1$  level. In the spectrum the frequencies are relative to the frequency of  $7/2 \rightarrow 5/2$  transition.

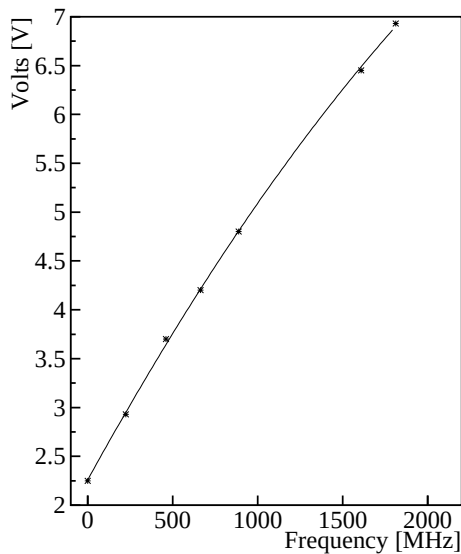


**Figure 4.26:** Calibration of the 1107.8 nm laser. Plotted are the voltages at the peak amplitude of each transition with the frequencies from the hyperfine splitting measurements. The fit is a second order polynomial function.

| $^3D_2 - ^1P_1$ transition ( $^{137}\text{Ba}$ ) |                 | Relative transition strengths |             |             |
|--------------------------------------------------|-----------------|-------------------------------|-------------|-------------|
| $F \rightarrow F'$                               | Frequency [MHz] | Calculation                   | Measurement | Ratio [M/C] |
| $7/2 \rightarrow 5/2$                            | 1405.99(21)     | 1.0                           | 1.00(1)     | 1.00(1)     |
| $5/2 \rightarrow 5/2$                            | -72.42(21)      | 0.051                         | 0.088(3)    | 1.72(6)     |
| $5/2 \rightarrow 3/2$                            | -283.56(25)     | 0.118                         | 0.175(4)    | 1.48(3)     |
| $3/2 \rightarrow 5/2$                            | -1096.05(21)    | 0.0006                        | -           | -           |
| $3/2 \rightarrow 3/2$                            | -1307.19(25)    | 0.007                         | 0.014(1)    | 2.00(14)    |
| $3/2 \rightarrow 1/2$                            | -1584.14(42)    | 0.005                         | 0.0080(8)   | 1.60(16)    |

**Table 4.6:** The first column is the six possible transitions of the  $^3D_2 - ^1P_1$  transition based on the selection rules of dipole transition. The second column is the frequency of the transitions from centroid to centroid from the hyperfine splitting measurements of the  $^3D_2$  and  $^1P_1$  states [121, 135]. The third column is the calculated transition strengths that are normalized to the  $7/2 \rightarrow 5/2$  transition. The fourth column consists of the transition strengths normalized to the  $7/2 \rightarrow 5/2$  transition from our measurement. The fifth column is the ratio of the strengths from the measurement and calculation.

ponents of the respective isotope,  $A$  is the amplitude of each component,  $\Gamma$  is the halfwidth maximum and  $\nu_i$  is the center of the hyperfine component.

The frequencies of the measured spectrum are plotted relative to the  $1/2 \rightarrow 3/2$  transition. The transition strengths from the calculation, the measurement and their ratios are given in Table 4.5.

A similar measurement was done for the  $^{137}\text{Ba}$  isotope for the  $^3D_2 - ^1P_1$  transition at 1130.6 nm. Here the  $^3D_2$  sub levels  $F = 3/2, 5/2$  and  $7/2$  (Fig. 4.21(b)) were populated by driving the  $^1S_0 F = 3/2 - ^3P_1 F = 5/2$  transition with the diode laser.

The 1130.6 nm laser frequency is scanned by 4 GHz, spanning the entire hyperfine spectrum of the  $^{137}\text{Ba}$  isotope for the  $^3D_2 - ^1P_1$  transition. The measured hyperfine spectrum is shown in Fig. 4.25. The analysis is done in the same way as for the  $^3D_1 - ^1P_1$  transition.

The measurements are compared to the expected values derived in section 4.6.1 and added to Table 4.6. We observe a discrepancy of relative transition strengths, which can be accounted for by the polarization of the infrared lasers with respect to the direction in which we collect the fluorescence.

### Calibration of Infrared Lasers

We scan the fiber laser (frequency) with a PZT in terms of voltage. We used the hyperfine structure measurement for the calibration of the scanning of fiber

lasers with the piezo element. In the manual we can find this conversion factor (see Table 4.4).

We determine the PZT voltage at the center of each peak from our measurement. The hyperfine splitting frequencies of the  $^3D_1$ ,  $^3D_2$  states for the  $^{137,135}\text{Ba}$  isotopes are known from the Atomic Beam Magnetic Resonance (ABMR) experiment [135, 136]. The hyperfine splitting of the  $^{137,135}\text{Ba}$  isotopes for the  $^1P_1$  state are also known [121]. From these, frequencies of the different hyperfine transitions of the  $^3D_1 - ^1P_1$  transition are calculated.

The measured PZT voltages at the peak of each resonance are plotted versus the frequencies of the transitions (Fig. 4.26). There is a small deviation from a linear behavior. The quadratic term in the fit shown is the non-linearity in the stretching of the fiber by the PZT. These are reproducible over the voltage range used in the experiments.

The value obtained from the fit for the 1107.8 nm laser is 420(4) MHz/V. The measured conversion factor deviates from the value 635 MHz/V provided by the manufacturer. It is 1.5 times higher than our measurement. The error in our measurement is limited by the resolution of the device used to set the PZT voltage. Similarly, the 1130.6 nm fiber laser was calibrated with the  $^3D_2 - ^1P_1$  transition and the value is 410(4) MHz/V.

## 4.7 Isotope Shifts

The isotope shift of an atomic transition frequency between two different isotopes of an atom with mass numbers  $A_1$  and  $A_2$  is decomposed as a sum of normal mass shift ( $\delta v_{NMS}$ ), specific mass shift ( $\delta v_{SMS}$ ) and field shift ( $\delta v_{FS}$ ) [121]

$$\delta v = \delta v_{NMS} + \delta v_{SMS} + \delta v_{FS} \quad (4.16)$$

The normal mass shift arises due to the reduced mass correction to the energy levels in an atom and is calculated by

$$\delta v_{NMS} = v \frac{m_e}{m_p} \frac{A_1 - A_2}{A_1 A_2} \quad (4.17)$$

where  $m_e$  is the electron mass,  $m_p$  is the proton mass and  $v$  is the transition frequency. The specific mass shift is caused by the momentum correlations among orbital electrons. This is a constant for all the isotopes and insensitive to the nuclear component. It is derived from the theoretical estimates.

The field shift arises due to the change in the spatial distribution of the nuclear charge between the different isotopes, which is given as

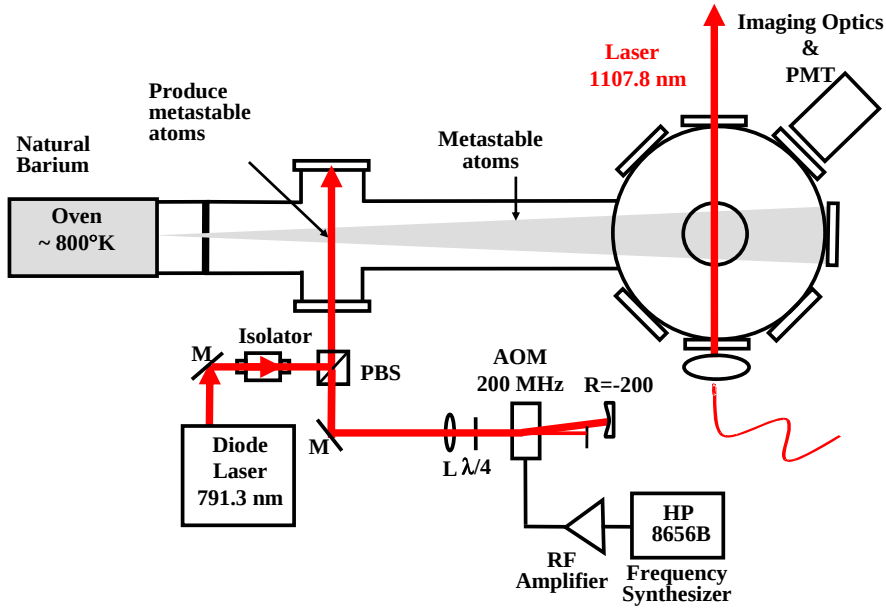


**Figure 4.27:** In region 1 using 791.3 nm laser with an AOM in double pass configuration different isotopes are populated to the  $^3D_1$  state. In region 2 we measured the isotope shift and hyperfine structure with the 1107.8 nm laser by spectroscopy of the  $^3D_1-^1P_1$  transition for different isotope pairs.

$$\delta v_{FS} = F \delta \langle r^2 \rangle \quad (4.18)$$

where  $F$  is an electronic factor and  $\langle r^2 \rangle$  is the expectation value of the square of charge distribution of the nucleus. This provides information on the overlap of wavefunctions with the nucleus, which for us is of interest for an EDM experiment with the radium atom.

Barium isotopes with nuclear spin  $I = 0$  have no hyperfine structure in their ground state and excited state. Due to this, the isotope shift between different isotopes can be derived directly from the measurement. Because of the long term drifts of the fiber lasers with temperature, we have to measure the transition for different isotopes at the same time. To achieve this, 791.3 nm laser light from the diode laser is passed through an AOM at a center frequency of 200 MHz in double pass configuration (see Fig. 4.27). We toggle the frequency to the AOM with a frequency synthesizer by switching fast between the different frequencies for the isotopes. Typical switching times are limited by the HP 8656B synthesizer to about 50 ms. This short switching time allows us to reduce the uncertainty of the

| Isotope pair | Isotope shift [MHz] |
|--------------|---------------------|
| 138-137      | -183.4(1.0)         |
| 138-136      | -109.2(1.0)         |
| 138-135      | -219.9(1.0)         |
| 138-134      | -122.3(2.5)         |
| 138-130      | -174.7(1.2)         |

**Table 4.7:** Isotope shifts of the  $^1S_0 - ^3P_1$  transition at 791.3 nm in barium [137].

| F   | $^{135}\text{Ba}$ [MHz] | $^{137}\text{Ba}$ [MHz] |
|-----|-------------------------|-------------------------|
| 5/2 | 1535.70(3)              | 1715.48(6)              |
| 3/2 | -1001.23(3)             | -1108.98(3)             |
| 1/2 | -2604.62(6)             | -2928.49(6)             |

**Table 4.8:** Hyperfine splitting frequencies of the  $^3P_1$  state from atomic beam double resonance measurements from the centroid [138].

measurement due to slow drifts of the fiber laser frequencies. The experimental resolution of this transition in our experiment is below 2 MHz. The spacing allows us to select the isotope by the frequency of the diode laser. This method applies to all isotopes because the isotope shift is large compared to the linewidth of the transition.

We make use of the known isotope shift and hyperfine structure in the  $^3P_1$  level. The isotope shift of barium isotopes for the  $^3P_1$  level measured by Grundevik et al. [137] are given in Table 4.7. The hyperfine structure in the  $^3P_1$  excited state in barium isotopes with nuclear spin  $I = 3/2$  were measured by zu Putlitz [138] with the optical double-resonance technique, using a hollow cathode

| F state | Isotope pair | $\Delta\nu$ [MHz] |
|---------|--------------|-------------------|
|         | 138-136      | -109.2(1.0)       |
|         | 136-134      | -13.1(2.7)        |
| 5/2     | 137-135      | 143.3(1.4)        |
| 3/2     | 137-135      | -144.3(1.4)       |
| 1/2     | 137-135      | -360.4(1.4)       |

**Table 4.9:** Absolute difference frequencies of the corresponding sub-levels of the  $^3P_1$  state for different isotope pairs. The difference frequencies allow for isotope selective population of the metastable D-states. The uncertainties are dominated by the isotope shift measurements of the  $^3P_1$  state.

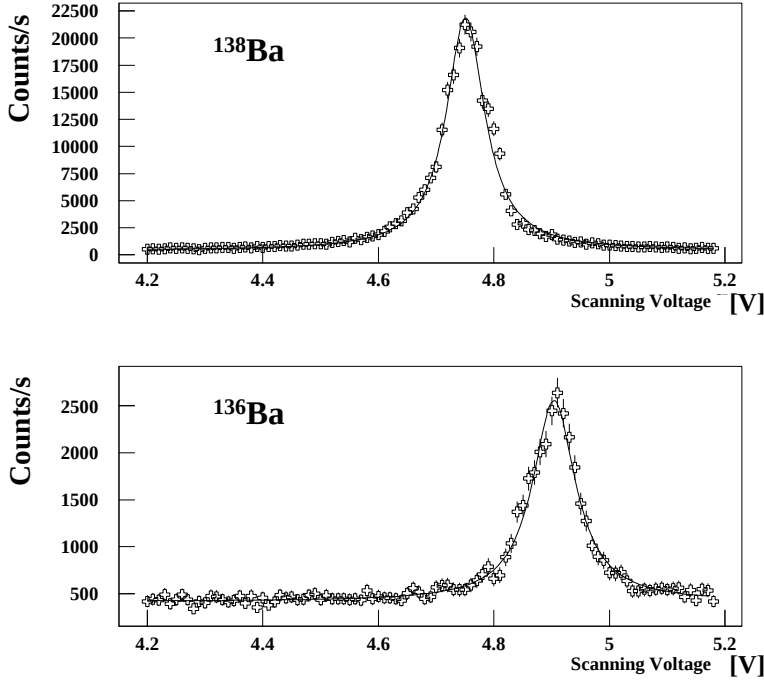


**Figure 4.28:** Spectra of the  $^{138,136}\text{Ba}$  isotopes. They are taken with 100 msec integration time. The line through the measured data is Lorentzian fit function. The isotope shift between  $^{136}\text{Ba}$  and  $^{134}\text{Ba}$  is -71(5) MHz.

lamp as a light source and the  $A$  and  $B$  parameters were extracted.

Using optical pumping with lasers allows us to provide a state selective and isotope selective metastable beam. The different frequencies for the population transition, the  $^1S_0 - ^3P_1$  transition is well known from previous experimental results on isotope shifts [137] and hyperfine structure [138]. The relevant data is compiled in Table 4.7 and Table 4.8. In Table 4.9 we give the frequency differences for the same transition in the different isotopes we used.

When the diode laser is toggled between the  $^{138}\text{Ba}$  and  $^{136}\text{Ba}$  isotopes we can determine the isotope shift of the  $^3D_1 - ^1P_1$  transition. The spectra obtained from one such a scan are shown in Fig. 4.28. The two resonance spectra are fitted with Lorentzian functions. The linewidth is 0.081(4) V ( $^{138}\text{Ba}$ ) and 0.090(6) V ( $^{136}\text{Ba}$ ) and the splitting between the peaks is 0.15(2) V. Without isotope selective

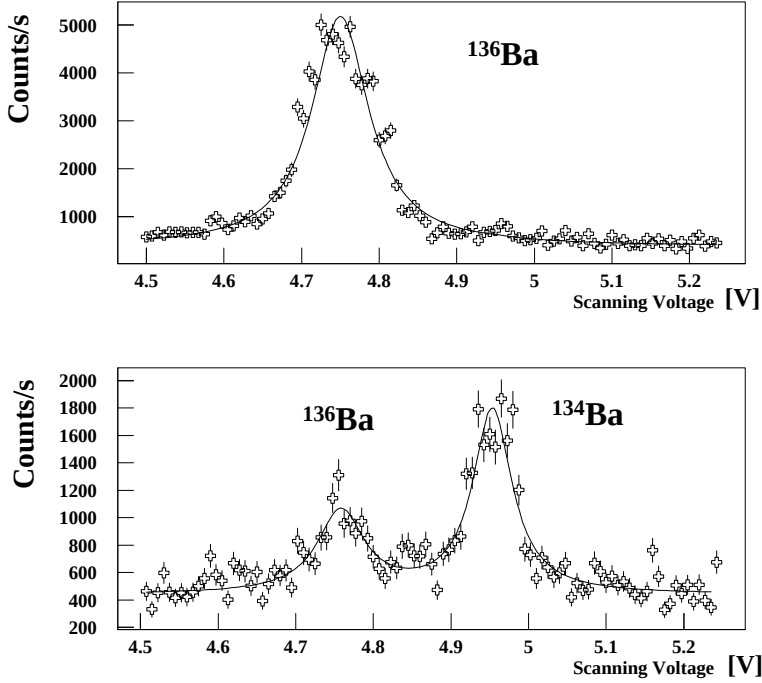


**Figure 4.29:** Isotope shift of  $^{136}\text{Ba}$  and  $^{134}\text{Ba}$ . They are taken with 100 ms second integration time. The line through the measured data is a Lorentzian fit function. In the  $^{134}\text{Ba}$  also  $^{136}\text{Ba}$  isotope is seen. The isotope shift between  $^{136}\text{Ba}$  and  $^{134}\text{Ba}$  is - 93(6) MHz.

population of the metastable states the two peaks would overlap and the determination of the line center would require much higher statistics. In addition, we find from the peak intensities the abundance ratio  $^{138}\text{Ba} : ^{136}\text{Ba}$  to 10.0(4) : 1, which is in good agreement with the natural abundance ratio of 10.9 : 1 found in literature [121]. Using the calibration of fiber lasers we get an isotope shift of -71(5) MHz and a linewidth of 34(1) MHz and 38(2) MHz, respectively.

In a similar manner we measured the isotope shift between  $^{136}\text{Ba}$  and  $^{134}\text{Ba}$  isotopes in the  $^3D_1$  state. The isotope shift between  $^{136}\text{Ba}$  and  $^{134}\text{Ba}$  of the  $^1S_0 - ^3P_1$  transition is just 13.1(2.7) MHz [137]. The narrow intercombination line with natural linewidth of 120 kHz permits still the isotope selective population of the triplet  $D$ -states. Although our experimental resolution of the transition is on the

| Isotope | Width [V] | Center [V] | Amplitude [Counts/s] |
|---------|-----------|------------|----------------------|
| 138     | 0.081(4)  | 4.75(1)    | 21500(400)           |
| 136     | 0.090(6)  | 4.90(2)    | 2150(80)             |
| 136     | 0.091(3)  | 4.75(1)    | 4070(60)             |
| 134     | 0.062(4)  | 4.95(1)    | 1340(70)             |

**Table 4.10:** Fit parameters of the different isotopes for the  $6s5d\ ^3D_1 - 6s6p\ ^1P_1$  transition in barium.

| Odd isotopes: $^3D_1 - ^1P_1$ transition |                     |                                   |                     |
|------------------------------------------|---------------------|-----------------------------------|---------------------|
| Transition                               | $\Delta\nu_1$ [MHz] | $\Delta\nu_2 - \Delta\nu_3$ [MHz] | Isotope shift [MHz] |
| 1/2 $\rightarrow$ 3/2                    | -229.3(3.3)         | 151.0(0.4)                        | -78.4(3.3)          |
| 1/2 $\rightarrow$ 1/2                    | -167.3(2.5)         | 97.8(0.6)                         | -69.5(2.6)          |
| 3/2 $\rightarrow$ 5/2                    | -143.2(2.6)         | 62.1(0.3)                         | -81.1(2.6)          |
| 3/2 $\rightarrow$ 3/2                    | -128.3(4.2)         | 53.8(0.4)                         | -74.4(4.2)          |
| 3/2 $\rightarrow$ 1/2                    | -76.2(2.2)          | 0.7(0.6)                          | -75.5(2.3)          |
| 5/2 $\rightarrow$ 5/2                    | -6.1(0.6)           | -68.5(0.3)                        | -74.6(0.7)          |
| 5/2 $\rightarrow$ 3/2                    | 1.2(1.0)            | -76.7(0.4)                        | -75.5(1.1)          |

**Table 4.11:** Measured isotope shifts between  $^{137}\text{Ba}$  and  $^{135}\text{Ba}$  for the  $6s5d\ ^3D_1 - 6s6p\ ^1P_1$  transition.

order of 2 MHz due to the Doppler broadening in the divergent atomic beam, we still have a good selectivity of the isotopes.

We determined the isotope shift of the  $^3D_1 - ^1P_1$  transition with the same method. We use the  $^1S_0 - ^3P_1\ F = 3/2$  transition for the isotope selective population. The frequency difference is 143.3(1.4) MHz. We take one data point on the one isotope then switch the 791.3 nm laser before we change the laser frequency of the 1107.8 nm laser. The obtained spectra are shown in Fig. 4.29. The linewidth is in good agreement with the  $^{138}\text{Ba}$  and  $^{136}\text{Ba}$  isotope measurements. From a fit to the data we determine the isotope shift to 0.2(1) V. This converts to a shift of -93(6) MHz for this isotope pair. The ratio in abundances is 3.0(1) : 1 in good agreement with the expectation for natural barium. The appearance of both isotopes in the lower spectrum can be explained by the not perfect isotope selectivity in the population process due to the small isotope shift.

We derived the isotope shift between the  $^{137}\text{Ba}$  and  $^{135}\text{Ba}$  isotopes for the  $^3D_1 - ^1P_1$  transition from the measured hyperfine spectrum. For this we used hyperfine splitting frequencies from the centroid for the  $^{137,135}\text{Ba}$  isotopes of the  $^3D_1$  and  $^1P_1$  states respectively. The difference frequencies ( $\Delta\nu_1$ ), (see Table 4.11) of

the seven transitions from the measurement are calculated. Substituting the measured values and difference frequencies of the hyperfine splitting for both states with the relation below, the isotope shift between  $^{137}\text{Ba}$  and  $^{135}\text{Ba}$  isotopes is derived.

$$IS = \Delta\nu_1 + [\Delta\nu_{2_{137}} - \Delta\nu_{3_{135}}] \quad (4.19)$$

where

$$\Delta\nu_{2_{137}} = [hfs(^3D_1) - hfs(^1P_1)]_{137}$$

$$\Delta\nu_{3_{135}} = [hfs(^3D_1) - hfs(^1P_1)]_{135}$$

We get seven values from the different components. The most sensitive ones are the components where the transitions nearly coincide in frequency ( $5/2 \rightarrow 5/2$  and  $5/2 \rightarrow 3/2$ ). These data points are less sensitive to the frequency calibration of the fiber laser. The isotope shift between  $^{137}\text{Ba}$  and  $^{135}\text{Ba}$  isotopes is  $-75.3(5)$  MHz.

The isotope shifts in the  $^3D_1$  state between the even barium isotope pairs and odd isotope pair derived from our measurement are summarized in Table 4.12. In the literature such data are analyzed using a King plot [139]. In most cases the King plot will be a straight line. The gradient of the line gives the ratio of field factors for the two transitions and the intercept is related to the difference in specific mass shifts. In Fig. 4.30 we show a King plot for the isotope shift data of the  $^1S_0$ - $^3P_1$  transition as abscissa and our measured isotope shift data of  $^3D_1$ - $^1P_1$  transition as ordinate. The values shown for each isotope pairs are the modified shifts ( $\delta_M$ ) for the both transitions, which is given as

$$\delta_M = \nu_{Meas} - \nu_{NMS} \quad (4.20)$$

where  $\nu_{Meas}$  is the measured isotope shift and  $\nu_{NMS}$  is the calculated normal mass shift for each isotope pair. However, there are instances of deviation from a straight line as we observe in our case. This is seen also in Samarium [140]. The deviations may be due to the effects of electron-electron correlation in two electron systems [141]. The nuclear charge radii part may not be prominent. There are no theoretical predictions to compare our results. A theoretical analysis could shed light on this issue.

## 4.8 Metastable D-state Spectroscopy

In this chapter, various techniques employed for the spectroscopy of the metastable  $D$ -states in barium were established. They are required for barium laser cooling experiments.

| Isotope shifts: $^3D_1$ - $^1P_1$ transition |         |         |            |
|----------------------------------------------|---------|---------|------------|
| Isotope pair                                 | 138-136 | 136-134 | 137-135    |
| Isotope shift [MHz]                          | -71(5)  | -93(6)  | -75.3(0.5) |

**Table 4.12:** Measured isotope shifts for the  $6s5d\ ^3D_1$  -  $6s6p\ ^1P_1$  transition in barium.

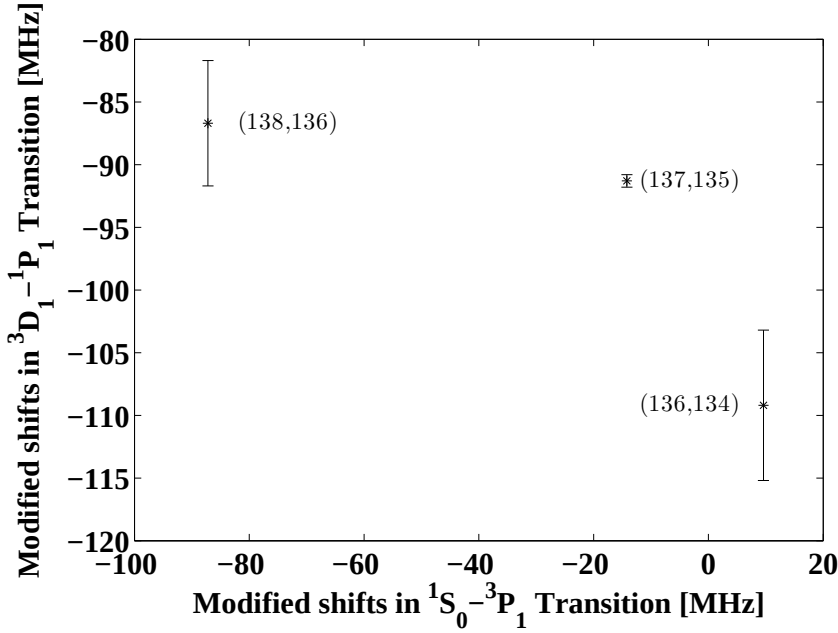


**Figure 4.30:** A King plot for the modified measured isotope shifts of the  $^3D_1$ - $^1P_1$  transition versus the modified shifts of the  $^1S_0$ - $^3P_1$  transition taken from reference [137].

We use the hyperfine structure measurements of the  $^3D_1$  -  $^1P_1$  and  $^3D_2$  -  $^1P_1$  transitions for the  $^{137}\text{Ba}$  and  $^{135}\text{Ba}$  isotopes for the calibration of fiber lasers. We compared the measured transition strengths and we attributed the observed deviations to the angular distribution of the fluorescence. We report the first measurement of the isotope shifts between  $^{138,136}\text{Ba}$  and  $^{136,134}\text{Ba}$  and odd  $^{137,135}\text{Ba}$  isotopes in the  $^3D_1$  level respectively. The techniques developed for the spectroscopy of the metastable  $D$ -states should be applicable to radium.



## Chapter 5

# Laser Cooling of Heavy Alkaline Earth Atoms

Modern atomic physics techniques like laser cooling and trapping of neutral atoms has become a vital tool for a variety of experiments. Of them, to mention are Bose-Einstein condensation, high precision measurements, optical frequency standards and studies on fundamental symmetries. The advantage of neutral atom trapping is that atoms can be confined in a small volume in high densities, at low kinetic energies providing long observation and storage times with excellent isotope selectivity. Furthermore, optical pumping allows to prepare the trapped sample in well defined atomic states. Laser cooling and trapping has so far been demonstrated with alkali elements lithium (Li) [142], sodium (Na) [143], potassium (K) [144, 145], rubidium (Rb) [146, 147], cesium (Cs) [148, 149] and francium (Fr) [150], light alkaline earth elements magnesium (Mg) [151], calcium (Ca) [152] and strontium (Sr) [152], except beryllium (Be), noble gases helium (He) [153], neon (Ne) [154], argon (Ar) [155], krypton (Kr) [155, 156] and xenon (Xe) [157], except radon (Rn), transition elements chromium (Cr) [158, 159] and silver (Ag) [160], ytterbium (Yb) [161] and erbium (Er) [162]. Recently, radium has been added to the list [55]. The weak intercombination line  $^1S_0$ - $^3P_1$  is sufficient for trapping, but the trapping efficiency is small.

The light alkaline earth elements have a strong  $^1S_0$ - $^1P_1$  transition, which can be approximated by a two-level system. Magnesium has a closed  $3s^2$   $^1S_0$  -  $3s3p$   $^1P_1$  transition with all the metastable  $D$ -states higher than this transition making it a two level system [151]. Strontium and calcium atoms have low lying singlet  $D$ -states connected to the first excited  $^1P_1$  state with negligible leakage rate. For calcium [163] and strontium [164] longer storage times in a MOT have

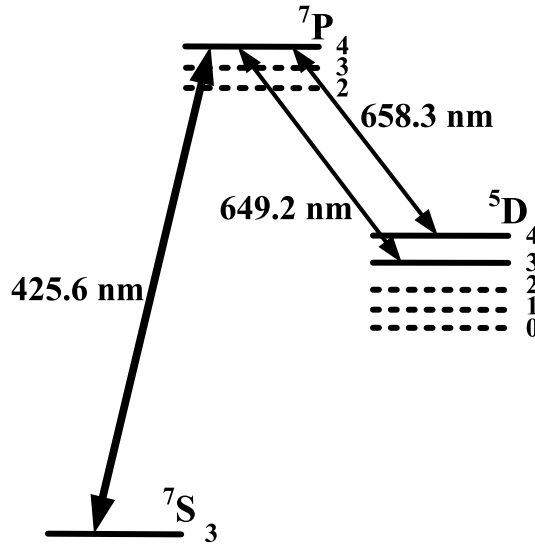


**Figure 5.1:** Partial term scheme of chromium used for laser cooling and trapping experiment [159].

been demonstrated when an appropriate repumping laser is added to the scheme. In calcium, atoms from the singlet  $D$ -state are repumped via the higher  $4s5p\ ^1P_1$  state rather than to the  $4s4p\ ^1P_1$  state used in the cooling transition.

In contrast to the above, the atomic level structure is different in heavy alkaline earth atoms barium and radium. They have the metastable  $D$ -states lying lower than the excited  $^1P_1$  state. In barium the strong  $6s^2\ ^1S_0 - 6s6p\ ^1P_1$  transition is the only option for cooling, while radium offers the possibility to use the weaker intercombination line  $^1S_0 - ^3P_1$  for cooling in addition to the  $7s^2\ ^1S_0 - 7s7p\ ^1P_1$  transition [55].

In barium, the leakage rate to the  $D$ -states is so large that laser cooling without repumping lasers is not possible. In addition, repumping has to go through the  $6s6p\ ^1P_1$  state, because excitation to higher levels would allow branching into many more states. Since the cooling transition and the repumping transition have a common excited state, we expect coherent Raman resonances. These will be discussed in detail in Section 5.1, together with their implication for laser cooling.

To illustrate the problem of repumping we will discuss briefly laser cooling of chromium. The level structure with its  $\Lambda$ -systems is shown in Fig. 5.1. Laser cooling and trapping has been demonstrated with and without repumping from the two metastable  $D$ -states and the cooling transitions use a common excited state [159]. Of all trapped atomic systems it has the largest similarity to laser

cooling of barium. We give a simple estimate to emphasize the issues for the case of barium.

In this estimate we compare the average velocity change of an atom without repumping. The average velocity change is given by

$$\Delta v = A_{leak}^{-1} \times v_r \quad (5.1)$$

where  $A_{leak}^{-1} = 2500$  is the leak rate to the  $D$ -states in chromium and the recoil velocity  $v_r = \hbar k/m = 1.8$  cm/s. Thus a velocity change of  $\Delta v = 45$  m/s can be achieved, while still a fraction of  $1/e$  of the atoms remain in the cooling cycle. Below 45 m/s there is a fraction  $1 \times 10^{-5}$  of the atoms in thermal distribution from their atomic source. It was reported that less than 100 atoms could be trapped without repumping lasers. Adding the repumping of atoms using the additional diode lasers from the  $D$ -states helped to increase the number of atoms in the MOT by a factor of 45 and to achieve longer storage times. The repump lasers are focussed on the trapped sample and high intensities are easily achieved. The repumping transitions are in the visible region that are conveniently accessible with diode lasers at 658.3 nm and 649.2 nm respectively.

Now we estimate what fraction of barium atoms we could stop without repump lasers. The branching ratio to the three  $D$ -states is given in Eq. 4.1. On average only  $A_{leak}^{-1} = 330(30)$  photons are scattered before the atom is in one of the  $D$ -states. Also the recoil velocity  $v_r = 0.52$  cm/s is smaller than for chromium. Using the relation 5.1, we arrive at a velocity change of only  $\Delta v = 1.7$  m/s. At these low velocities there are no atoms in an atomic beam, because of collisions in the nozzle. This effect is known as the Zacharias effect [33].

To bring a barium atom from a velocity of 45 m/s to rest the atom has to scatter about  $N_{45m/s} = 8650$  photons. This is  $N_{45m/s}/A_{leak}^{-1} = 26$  times more than  $A_{leak}$ . A fraction of only

$$P_{cc} = \exp\left(\frac{-N_{45m/s}}{A_{leak}^{-1}}\right) = 10^{-11} \quad (5.2)$$

remain in the cooling cycle. This makes trapping without repumping not feasible. To achieve a fraction of  $1/e$  of the atoms remaining in the cooling cycle the atoms have to be optically pumped out of the metastable states about 26 times.

The time the atoms spend in the metastable  $D$ -states depends on the optical pumping rate, which is related to the Rabi frequency (Eq. 5.41). In addition, this drives two photon Raman transitions from the ground state to the metastable  $D$ -states. The consequence is that depending on the detuning of the repump lasers

and cooling laser the atoms can get transferred efficiently into the metastable states. On average this increases the population compared to a situation in which the optical pumping can be done via another level. In atoms with nuclear spin other states of the hyperfine manifold can be used for repumping.

Though Raman transitions are present for barium, they are not a strong constraint for laser cooling. At high enough intensities for the repump lasers we get an equal probability to find the atom in one of the states connected by the laser transitions. Depending on the number  $N$  of states involved, for barium, the ground state and three metastable  $D$ -states, we will have a fraction of  $1/N$  of the atoms in each state.

We will describe the theoretical treatment and the experimental observation of coherent Raman resonances. Using these results we discuss the setup for slowing a barium atomic beam by light forces. We show the first successful slowing of barium and we give an analysis of slowing results using our knowledge of Raman resonances in a dynamic calculation.

## 5.1 $\Lambda$ - System

### 5.1.1 Theoretical Treatment of Coherent Raman Resonances in a $\Lambda$ -System

Atoms having a common excited state with multiple ground states exhibit the phenomenon of coherent Raman resonances. As an example, consider a three-level system, having two ground states connected to a common excited state, a  $\Lambda$ -system. In the  $\Lambda$ -configuration the atoms interacting with near-resonant bichromatic light fields show additional resonances (Fig. 5.2).

When the detuning  $\Delta$  from the excited state of both lasers is the same, atoms are transferred into a superposition of state  $|1\rangle$  and  $|3\rangle$  being the two groundstates which is expressed by an additional two photon Rabi frequency  $\Omega_{13}$ . Hence, resonant excitation and fluorescence are suppressed. To the observer this process appears as a *dark resonance* with a narrow line width, which is in principle determined by the excited state properties. Experimentally, in 1976, dark resonances were first observed in sodium [165]. A review on developments and status of the coherent dark state spectroscopy experiments can be found in for example [166, 167].

For the theoretical discussion we consider the  $\Lambda$ -system with the  $^1S_0$ ,  $^1P_1$  and one of the metastable states. In particular we use the  $^1D_2$  state as an example. This treatment can easily be extended to a four level system.

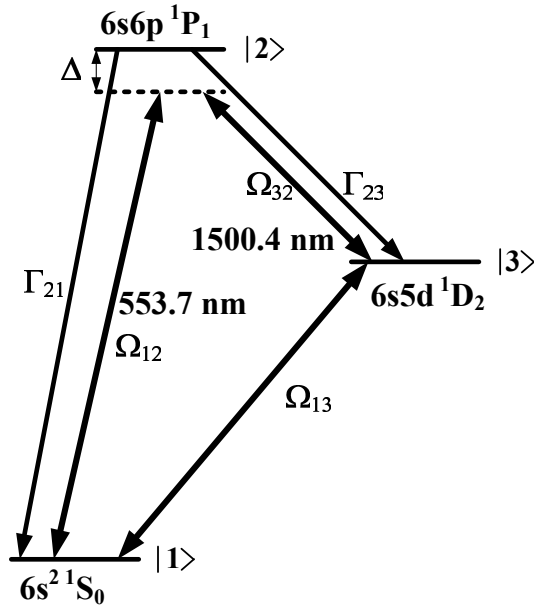


**Figure 5.2:** Simplified barium level scheme showing only one metastable D state with the atomic eigenstates  $|1\rangle$ ,  $|2\rangle$  and  $|3\rangle$  and the coherent light fields with Rabi frequencies  $\Omega_{12,32}$ , decay rates  $\Gamma_{21,23}$  and detuning  $\Delta$ .

### Interaction of a $\Lambda$ -system with Coherent Light Fields

The energy level scheme of the barium atom used for the calculation is shown in Fig. 5.2. The  $1P_1$  excited state decays to the  $1S_0$  ground state and weakly to the  $1D_2$  metastable state. With the bichromatic excitation, the scattered light of the barium atom contains spectral components at  $553.7 \text{ nm}$  and negligible  $1500.4 \text{ nm}$  light.

A theoretical model describing the atom-laser interaction for a three-level  $\Lambda$ -system, interacting with two coherent light fields is described. The formalism adapted here is similar to the calculations for the barium ion [168, 169]. Here the atom is treated quantum mechanically whereas the coherent laser fields are treated as classical electromagnetic waves  $\vec{E}_g \sin(\omega_g t)$  and  $\vec{E}_r \sin(\omega_r t)$  as they are of high photon numbers. They have linewidths  $\Gamma_g$  and  $\Gamma_r$  respectively. The spontaneous emission of photons is treated as a decay from  $|2\rangle$  to  $|1\rangle$  and  $|3\rangle$  with the rates  $\Gamma_{21}$  and  $\Gamma_{23}$ . The quantities  $\Omega_{12}$  and  $\Omega_{32}$  are Rabi frequencies and denote the strength of the coupling between the atom and the electric field.

The Hamiltonian of the atom, using the states 1, 2 and 3 as the basis, is

defined by

$$\hat{H}_{atom} |a\rangle = \hbar\omega_a |a\rangle \quad \text{with } a = 1, 2, 3 \quad (5.3)$$

with  $\omega_a$  being the atomic Bohr frequencies. The atom-laser interaction is restricted to the dipole interaction. Then the interaction Hamiltonian can be written as:

$$\begin{aligned} \hat{H}_{int} = -\{ & \vec{D}_{12} \cdot \vec{E}_g i [|2\rangle\langle 1| e^{-i\omega_g t} - |1\rangle\langle 2| e^{i\omega_g t}] + \\ & \vec{D}_{32} \cdot \vec{E}_r i [|2\rangle\langle 3| e^{-i\omega_r t} - |3\rangle\langle 2| e^{i\omega_r t}] \} \end{aligned} \quad (5.4)$$

where  $\vec{D}_{12}$  and  $\vec{D}_{32}$  are the dipole matrix elements for the transition from  $|1\rangle$  to  $|2\rangle$  and  $|3\rangle$  to  $|2\rangle$ , respectively. Here the terms which rotate at twice the optical frequency and the non-resonant terms e.g.,  $\vec{D}_{12} \cdot \vec{E}_r$  are neglected (rotating wave approximation) [170].

The Hamiltonian can also be written in matrix formalism with

$$\begin{pmatrix} c_1 \\ c_2 \\ c_3 \end{pmatrix} \equiv c_1 |1\rangle + c_2 |2\rangle + c_3 |3\rangle \quad (5.5)$$

$$H_{atom} = \hbar \begin{pmatrix} \omega_{12} & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & \omega_{32} \end{pmatrix} \quad (5.6)$$

with  $\omega_{12} = \omega_1 - \omega_2$  and  $\omega_{32} = \omega_3 - \omega_2$ . The energy zero has been set to  $|2\rangle$ . The interaction Hamiltonian written as

$$H_{int} = \hbar \begin{pmatrix} 0 & \frac{\Omega_{12}}{2} e^{+i\omega_g t} & 0 \\ \frac{\Omega_{12}}{2} e^{-i\omega_g t} & 0 & \frac{\Omega_{32}}{2} e^{-i\omega_r t} \\ 0 & \frac{\Omega_{32}}{2} e^{+i\omega_r t} & 0 \end{pmatrix} \quad (5.7)$$

where

$$\hbar\Omega_{12} := \vec{D}_{12} \cdot \vec{E}_g \quad (5.8)$$

$$\hbar\Omega_{32} := \vec{D}_{32} \cdot \vec{E}_r \quad (5.9)$$

The complete Hamiltonian can be written as

$$H = H_{int} + H_{atom} = \hbar \begin{pmatrix} \omega_{12} & \frac{\Omega_{12}}{2} e^{+i\omega_g t} & 0 \\ \frac{\Omega_{12}}{2} e^{-i\omega_g t} & 0 & \frac{\Omega_{32}}{2} e^{-i\omega_r t} \\ 0 & \frac{\Omega_{32}}{2} e^{+i\omega_r t} & \omega_{32} \end{pmatrix} \quad (5.10)$$

The Hamiltonian does not describe the spontaneous decay of the  $|2\rangle$  level into the  $|1\rangle$  and  $|3\rangle$  levels. To understand our measurements it is not necessary to use a full quantum-mechanical treatment of the electro magnetic fields. It is sufficient to treat the spontaneous decay as a rate  $\Gamma_{21}$  ( $\Gamma_{23}$ ) with which the state  $|2\rangle$  decays into  $|1\rangle$  ( $|3\rangle$ ). The resulting state of the atom is no longer a pure state and one can use the density matrix formalism.

### Density Matrix Formalism

The density operator  $\hat{\rho}$ , written in the basis of atomic eigenstates  $|a\rangle$ , is

$$\hat{\rho} = \sum_{a,b=1,2,3} \rho_{ab} |a\rangle\langle b| \quad (5.11)$$

The elements  $\rho_{11} = \langle 1|\rho|1\rangle$ ,  $\rho_{22}$  and  $\rho_{33}$  are the expectation values for finding the atom in the states  $|1\rangle$ ,  $|2\rangle$  and  $|3\rangle$  respectively, such that

$$\text{Trace}(\hat{\rho}) = \rho_{11} + \rho_{22} + \rho_{33} = 1 \quad (5.12)$$

The time evolution of the density operator is governed by the Liouville equation

$$\frac{d\hat{\rho}}{dt} = -\frac{i}{\hbar}[H, \hat{\rho}] + \mathcal{L}_{damp}(\hat{\rho}) \quad (5.13)$$

where the first part corresponds to the Schrödinger equation and  $\mathcal{L}_{damp}$  describes the spontaneous decay of the upper state. The operator  $\mathcal{L}_{damp}$  has the general form

$$\mathcal{L}_{damp}(\hat{\rho}) = -\frac{1}{2} \sum_m [\hat{C}_m^\dagger \hat{C}_m \hat{\rho} + \hat{\rho} \hat{C}_m^\dagger \hat{C}_m - 2\hat{C}_m \hat{\rho} \hat{C}_m^\dagger] \quad (5.14)$$

The operators  $\hat{C}_m \hat{C}_m^\dagger$  describe the different dissipative processes. The first two terms  $\hat{C}_m^\dagger \hat{C}_m \hat{\rho}$  and  $\hat{\rho} \hat{C}_m^\dagger \hat{C}_m$  describe the decay of the excited levels. The term  $2\hat{C}_m \hat{\rho} \hat{C}_m^\dagger$ , which is called feeding term, describes the decay into the lower states. Here this is the decay from  $|2\rangle \rightarrow |1\rangle$  and  $|2\rangle \rightarrow |3\rangle$ . Also, the decoherence through the finite bandwidth  $\Gamma_g$  and  $\Gamma_r$  of the driving laser fields can be included in  $\mathcal{L}_{damp}$ .

In this case of three level atom we have the decay from the  $P$  level to the  $S$  level, described by

$$\hat{C}_{21} = \sqrt{\Gamma_{21}} |1\rangle\langle 2| \quad (5.15)$$

and from the  $P$  to the  $D$  level

$$\hat{C}_{23} = \sqrt{\Gamma_{23}} |3\rangle\langle 2| \quad (5.16)$$

The finite linewidth of the lasers can be introduced by the operators

$$\hat{C}_g = \sqrt{2\Gamma_g} |1\rangle\langle 1| \quad (5.17)$$

$$\hat{C}_r = \sqrt{2\Gamma_r} |3\rangle\langle 3| \quad (5.18)$$

By writing

$$\frac{d\hat{\rho}}{dt} = \mathcal{L}\hat{\rho}(t) \quad (5.19)$$

with

$$\mathcal{L}\hat{\rho} = -\frac{i}{\hbar}[H, \hat{\rho}] + \mathcal{L}_{damp}(\hat{\rho}) \quad (5.20)$$

the density operator  $\rho$  at the time  $t$  can now be expressed in terms of  $\hat{\rho}(0)$  by

$$\hat{\rho}(t) = e^{\mathcal{L}t} \hat{\rho}(0) \quad (5.21)$$

Finally, we transform the system into the rotating frame of the laser light fields with

$$U = \begin{pmatrix} e^{-i\omega_g t} & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & e^{-i\omega_r t} \end{pmatrix} \quad (5.22)$$

and get the transformed density and Hamiltonian operators

$$\hat{\rho}' = U\hat{\rho}U^\dagger \quad (5.23)$$

$$H' = UHU^\dagger - i\hbar U \frac{dU^\dagger}{dt} \quad (5.24)$$

The resulting Hamiltonian  $H'$  is

$$H' = \hbar \begin{pmatrix} \Delta_g & \frac{\Omega_{12}}{2} & 0 \\ \frac{\Omega_{12}}{2} & 0 & \frac{\Omega_{32}}{2} \\ 0 & \frac{\Omega_{32}}{2} & \Delta_r \end{pmatrix} \quad (5.25)$$

with the detunings

$$\Delta_g = \omega_g - \omega_{12} \quad (5.26)$$

$$\Delta_r = \omega_r - \omega_{32} \quad (5.27)$$

The damping terms remain unchanged.

### Optical Bloch Equations

The Liouville equation 5.13 can be transformed into a system of linear equations by setting

$$\vec{\rho} := (\rho_{11}, \rho_{12}, \dots, \rho_{23}, \rho_{33}) \quad (5.28)$$

$$\frac{d\vec{\rho}_i}{dt} = \sum_j M_{ij} \vec{\rho}_j \quad (5.29)$$

where  $\rho_{ab} = \langle a | \vec{\rho} | b \rangle$  and we have introduced the  $N^2 \times N^2$  matrix  $M$ . These equations are called optical Bloch equations. The time evolution  $\vec{\rho}(t)$  with a given  $\vec{\rho}(0)$  is

$$\vec{\rho}(t) = \exp(Mt) \vec{\rho}(0). \quad (5.30)$$

The matrix  $M$  is uniquely determined by  $\mathcal{L}$  in Eq. 5.13, i.e., it contains all information relevant for the evolution of the system, i.e., the laser intensities, detunings and decay constants. All the terms have been introduced in such a way that  $\rho$  remains normalized, i.e.,

$$\sum_i \rho_{ii}(t) = 1 \quad (5.31)$$

In order to obtain the steady state solution ( $\vec{\rho}(\infty) = \text{constant} \leftrightarrow \vec{\rho}'=0$ ) is given by

$$\sum_j M_{ij} \vec{\rho}_j = 0 \quad (5.32)$$

To solve Eq. 5.32 with the correct normalization, one of the equations must be replaced by the normalization condition Eq. 5.31. The time evolution of  $\vec{p}(t)$  and the steady state solution  $\vec{p}(\infty)$  are calculated numerically with a computer program written in matlab [169, 171].

The numerical approach allows us to calculate the Raman spectrum. In order to compare the numerical result to the experiment we have to include the finite time the atom spends in the laser beam. Since in the experiments the laser beams are perpendicular to the atomic beam in order to reduce the Doppler broadening, the time is given by the laser beam diameter and the velocity of the atoms. We use a Maxwell-Boltzmann velocity distribution for the atoms.

Depending on the detuning of the  $^1D_2 - ^1P_1$  transition laser (1500.4 nm) with respect to the  $^1S_0 - ^1P_1$  transition (553.7 nm) frequency the atoms are driven coherently between the  $S$  and the  $D$ -states without passing through the  $P$  state. If both detunings are equal the effective Rabi frequency can be approximated to

$$\Omega_{13} = \frac{\Omega_{12}\Omega_{32}}{2\Delta} \quad (5.33)$$

under the approximation that the detuning  $\Delta$  is much larger than the linewidth  $\Gamma$  of the transition. The dark resonance spectrum calculated with the parameters used in the experiments is shown in Fig. 5.5. Because we operate in the limit of  $\Delta$  on the order of linewidth of the transitions we have to solve the full optical Bloch equations.

### 5.1.2 Coherent Raman Resonances: Experiment

The two photon Raman transition allowed us to determine the Rabi frequency of the repumping transitions. We will discuss the  $^1D_2 - ^1P_1$  transition in detail. The experimental setup used to observe the dark resonances in a  $\Lambda$ -system is shown in Fig. 5.3. It consists of the 553.7 nm laser and the 1500.4 nm laser.

| Transition      | Rabi frequency in rad/s<br>@ I = 1mW/cm <sup>2</sup> |
|-----------------|------------------------------------------------------|
| $^1S_0 - ^1P_1$ | $22 \cdot 10^6$                                      |
| $^1D_2 - ^1P_1$ | $4.7 \cdot 10^6$                                     |
| $^3D_2 - ^1P_1$ | $2.0 \cdot 10^6$                                     |
| $^3D_1 - ^1P_1$ | $0.32 \cdot 10^6$                                    |

**Table 5.1:** Rabi frequencies for different transitions in barium. The numbers are calculated based on the wavelength and the decay rates to the different states.

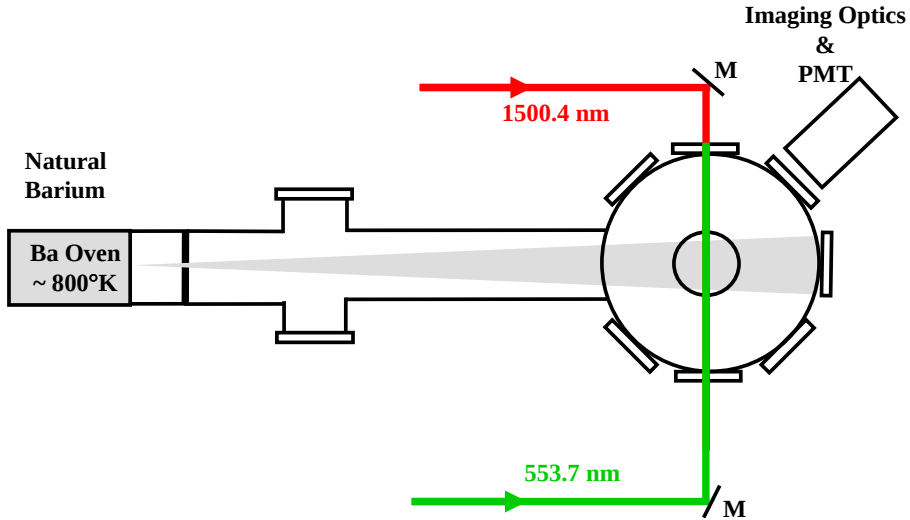


**Figure 5.3:** Setup to observe the coherent Raman resonances. A natural barium atomic beam is used. 553.7 nm laser light and 1500.4 nm laser light are overlapped and passed perpendicular to the barium atomic beam to reduce the Doppler broadening. The fluorescence from the  $6s6p\ ^1P_1$  state is detected with a PMT.

The two laser beams are passed perpendicular to the barium atomic beam and are counter propagating. They are collimated and are overlapped along the path of interaction with the atomic beam. The beam radius of the 1500.4 nm laser was 1.0(0.2) mm with Gaussian beam profile and the 553.7 nm laser beam was shaped by a rectangular aperture of size  $3 \times 3\text{ mm}^2$ . The fluorescence signal from the  $^1P_1 - ^1S_0$  transition is detected by a photomultiplier. This probes the population in the excited state or  $\rho_{22}$  of the density matrix. We compare the experimental spectra with numerical calculations taking the geometry of the setup into account. In particular, the laser beam at 553.7 nm was about a factor of two larger in diameter than the infrared laser and we have to take into account the time the atoms spend in the laser beams depending on their velocity.

The 1500.4 nm laser was red detuned by  $\Delta = -13.5(5)$  MHz with respect to the  $^1D_2 - ^1P_1$  transition of the  $^{138}\text{Ba}$  resonance and the 553.7 nm laser frequency was scanned across the resonance. The output power of the 1500.4 nm laser used was 10(1) mW corresponding to an intensity of  $320(120)\text{ mW/cm}^2$ . This corresponds to a Rabi frequency of  $\Omega_{32} = 90(20) \cdot 10^6\text{ rad/s}$ . For the 553.7 nm laser, a power

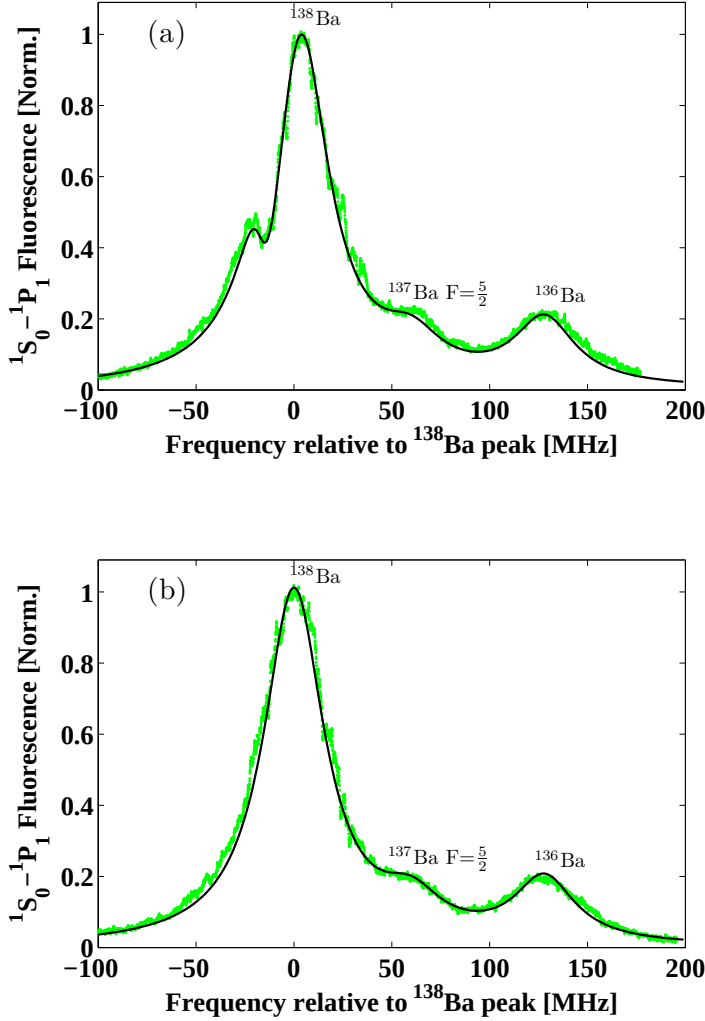


**Figure 5.4:** Fluorescence spectrum of the  $^1P_1$  state detected with a beam of natural barium. The upper figure (a) has the 1500.4 nm laser frequency detuned by -13.5 MHz with respect to the  $^{138}\text{Ba}$  resonance. The solid line is the result of the optical Bloch equations with parameters  $\Omega_{12}=120 \cdot 10^6$  rad/s for the 553.7 nm laser and  $\Omega_{32}=100 \cdot 10^6$  rad/s. The lower figure (b) is in the absence of the repump laser. The line is the result of the optical Bloch equation calculation with the parameters  $\Omega_{12}=120 \cdot 10^6$  rad/s. For both calculations the laser linewidth for the 553.7 nm laser  $\Gamma_g=1$  MHz and  $\Gamma_r=50$  kHz for the 1500.4 nm laser. The spectra are normalized to the peak amplitude. The only parameter, which has been adjusted is the ratio of the coherent Raman transition fraction to the ordinary fluorescence spectrum. The latter is about 45 % of the  $^{138}\text{Ba}$  fluorescence.

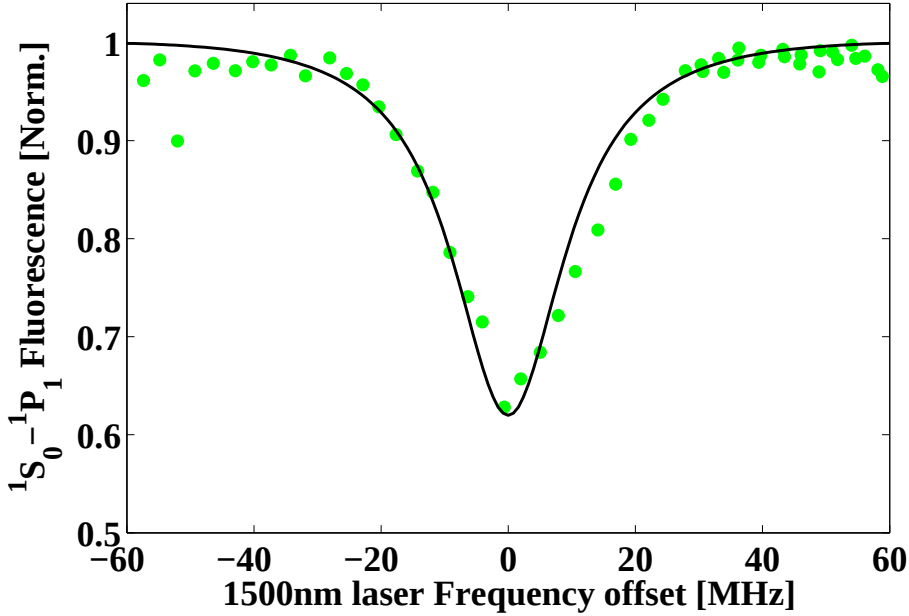


**Figure 5.5:** The measured dark resonance spectrum of the  $^1P_1$  state. Here the 553.7 nm laser is frequency locked to  $^{138}\text{Ba}$  resonance peak and the 1500.4 nm laser frequency is scanned. The solid line is the solution of the optical Bloch equations with the parameters  $\Omega_{12}=100 \cdot 10^6 \text{ rad/s}$ ,  $\Delta_g=0 \text{ MHz}$  and  $\Gamma_g=1 \text{ MHz}$  for the 553.7 nm laser;  $\Omega_{32}=3.0 \cdot 10^6 \text{ rad/s}$ , and linewidth  $\Gamma_r=5 \text{ kHz}$  for the 1500.4 nm laser. About 50% of the signal stems from atoms which only cross the 553.7 nm laser beam. The only parameter, which has been adjusted is the ratio of the coherent Raman transition fraction to the ordinary fluorescence spectrum. The latter is about 45 % of the  $^{138}\text{Ba}$  fluorescence, in agreement with Fig. 5.4. The fluorescence is normalized to the peak values.

of 2.2(1) mW was used corresponding to an intensity of 24(1) mW/cm<sup>2</sup> or a Rabi frequency of  $\Omega_{12} = 120(3) \cdot 10^6 \text{ rad/s}$  respectively. The laser linewidth for the two lasers is much less than the Rabi frequencies and the decay rate of the  $^1P_1$  state.

In Fig. 5.4(a) we show the observed spectrum which contains different barium isotopes. The dip on the left side of the  $^{138}\text{Ba}$  resonance is the two photon Raman transition. The fluorescence is reduced because of the direct coupling of the  $^1S_0$  ground state and the  $^1D_2$  state. The depth of the observed dip contains information on the Rabi frequency  $\Omega_{32}$ . Further, the size of the dip depends on the overlapping of the two laser beams and their alignment with respect to the atomic beam. Shown in Fig. 5.4(b) is the spectrum in the absence of the 1500.4 nm

laser. The solid line in both figures is the numerical solution of the optical Bloch equations. The parameters are in good agreement with the calculated Rabi frequencies.

Dark resonances can also be observed in another experimental configuration. For this, the 553.7 nm laser was frequency locked to the peak of the  $^{138}\text{Ba}$  isotope resonance using the Doppler shift method described in Section 4.10 and the 1500.4 nm laser frequency was scanned over the resonance. The spectrum is shown in Fig. 5.5. As long as the 1500.4 nm laser detuning is different from the detuning at 553.7 nm we observe the unperturbed scattering from the  $^1S_0$ - $^1P_1$  transition. When the 1500.4 nm laser and the 553.7 nm laser have the same detuning  $\Delta$  the scattering rate decreases, because of coherent population transfer to the metastable  $^1D_2$  state. The width of the observed dip is 20(1) MHz, in good agreement with the calculation. This indicates that Doppler broadening due to improper overlapping was small. We again get a good agreement with the result from optical Bloch equation with a Rabi frequency  $\Omega_{32}$  of  $120 \cdot 10^6$  rad/s, and a fraction of 50% of the atoms passing only through the 553.7 nm laser beam.

The determined Rabi frequency is of the order of  $10^8$  rad/s. Thus we can expect that we achieve Rabi frequencies larger than  $10^7$  rad/s with a beam diameter of 5mm with the available laser power of 10 mW from the fiber laser. For the repumping of the  $^3D_2$  state we expect similar Rabi frequencies at the same power (Table 5.1).

## 5.2 Laser Cooling of Barium

In this section we discuss aspects of laser cooling relevant for our system. A detailed information on laser cooling in general can be found elsewhere [16]. A simple approach to describe the laser cooling principle is to consider a closed two level atomic system. One parameter that plays an important role is the scattering rate  $\gamma_p$ . It is defined as the ratio of population of the excited state  $P_e$  to the life time  $\tau$  of the excited state. It is written as

$$\gamma_p = \frac{P_e}{\tau} = \frac{\gamma}{2} \frac{S}{1+S} \frac{1}{1 + (\frac{2\delta}{\gamma})^2} \quad (5.34)$$

where  $\gamma$  is the decay rate and  $\gamma' = \gamma \cdot \sqrt{1+S}$  is the power broadened linewidth of the atomic transition.  $\delta = \omega_{laser} - \omega_0$  is the detuning of the laser frequency  $\omega_{laser}$  with respect to the  $\omega_0$  of the atomic transition. Power broadening becomes important when the intensity of the laser is greater than the saturation intensity.  $S$  is the saturation parameter of the atomic transition that is defined as

$$S = \frac{I}{I_s} = 2 \frac{\Omega^2}{\Gamma^2} \quad (5.35)$$

where  $\Omega$  is the Rabi frequency and  $\Gamma$  is the linewidth of the transition and  $I_s$  is the saturation intensity defined as

$$I_s = \frac{\pi \hbar c}{3 \lambda^3 \tau} \quad (5.36)$$

where  $\hbar$  is the Planck constant,  $c$  is the velocity of light and  $\lambda$  is the wavelength of the atomic transition.

### 5.2.1 Requirements for Slowing Barium

To demonstrate the slowing of an atomic beam of barium we have chosen the cooling beam to be counter propagating to the atomic beam. The repumpers are also counter propagated to the atomic beam. This limits the range of cooling force to a part of the velocity spectrum from an effusive beam depending on the linewidth of the laser. The central velocity which is effected depends on the laser detuning.

Here we give a rough estimate of the time the atoms spend in the metastable states and compare them with our experimental values reported in Section 5.2.3. For stopping barium atoms of velocity  $v$ , assuming a constant acceleration  $a$ , over a distance  $d$  a time  $\delta t$

$$\delta t = \frac{2d}{v}, \quad (5.37)$$

is needed. The constant acceleration can be achieved by chirping the laser frequency or by broadening of the spectral width of the lasers.

During the slowing process the atom needs to scatter  $N_p = v/v_r$  photons, where  $v_r$  is the single photon recoil velocity. The minimum scattering rate  $\gamma_m$  to achieve a sufficient deceleration

$$\gamma_m = \frac{N_p}{\delta t}, \quad (5.38)$$

which has to be smaller than  $\gamma_p$  given in Eq. 5.34. We estimate the fraction of time the atoms have to be in the cooling cycle in order to achieve this scattering rate. Here we assume that all the lasers are on resonance all the time. This could be achieved by chirping the lasers. Assuming that we work at saturation intensity for the cooling transition we find in steady state a fraction of

$$f = \frac{\gamma_m}{\gamma_p} = \frac{4\gamma_m}{\gamma}, \quad (5.39)$$

of the atoms in the ground state. This is the result from a rate equation, which does not describe the coherent Raman transitions, which we will discuss in detail below. In addition, driving the cooling transition at saturation intensity results in an excited state population of  $f/4$ . The atom will be in the metastable states for the rest of the time.

With these assumptions we calculate the required repumping rate  $r$ , written as

$$r = \frac{N_p}{A_{leak}^{-1} \delta t (1 - 5f/4)} \quad (5.40)$$

For a weak excitation the repumping rate  $r_r$  is determined by the Rabi frequency  $\Omega_{32}$  at the repump wavelength and the decay rate  $\Gamma_{21}$  of the cooling transition,

$$r_r = \frac{\Omega_{32}^2}{\Gamma_{21}}. \quad (5.41)$$

As a numerical example, we assume a starting velocity of  $v = 120$  m/s for the atoms. The distance over which atoms are interacting with the lasers in our apparatus is  $d = 0.6$  m. This gives a slowing time  $\delta t = 10$  ms (Eq. 5.37). To stop the atoms the number of scattered photons  $N_p$  is then 23000 and the minimum scattering rate  $\gamma_m$  equates to  $2.3 \cdot 10^6$ /s (Eq. 5.38). Assuming that the cooling beam at 553.7 nm is at saturation intensity, we arrive at a fraction of at least  $5f/4 = 0.1$  (Eq. 5.39) in which the atoms have to be in the cooling cycle. Thus the repumping rate has to be larger than 8000/s (Eq. 5.40). The minimal required Rabi frequency  $\Omega_{32} \geq 10^6$  rad/s corresponds to an intensity of about 0.1 mW/cm<sup>2</sup> (1500.4 nm). Higher intensities would allow for larger cooling forces. This low intensity would be sufficient if the cooling transition and the repumping transition would not use a common excited state.

For investigating the effect of the coherent Raman transition on the repumping rate we have solved the optical Bloch equations for the three different level configurations (Fig. 5.6). The cooling force is proportional to the number of scattered photons, which is

$$N_{scatter} = \frac{\rho_{22}}{\tau} \quad (5.42)$$

where  $\rho_{22}$  is the probability to be in the excited state and  $\tau$  is the lifetime of the excited state.

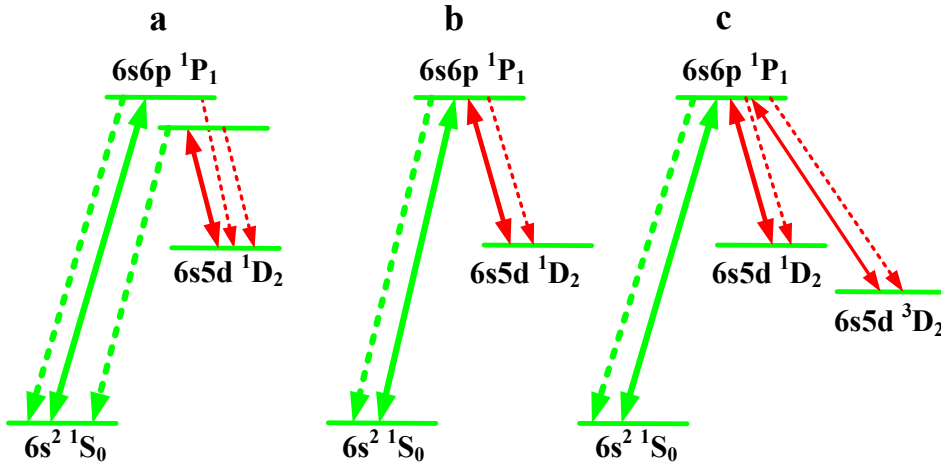


**Figure 5.6:** Three different level configurations for repumping. (a) Repumping through different excited state; (b) Three level  $\Lambda$ -system; (c) Four level system as used in the experiments.

We compare the requirements for the different situations. A lower repump laser intensity can be used when different excited states are involved. The result for the population of the excited state is shown in Fig. 5.7. The calculation was done for different detunings at  $\Omega_{\text{repump}} = 3.5 \cdot 10^6$  rad/s and on resonance at saturation intensity of the cooling transition. We see that the population  $\rho_{22}$  in the excited state is about 0.13. The calculation for the three level  $\Lambda$ -system was done for different linewidths of the repump laser. A larger linewidth effectively washes out the two-photon resonance, but the area under the curve gets smaller, indicating lower efficiency in recycling to the cooling transition. The population of the ground state is 0.04 for the two photon resonance. Integrating over the full spectrum the repumping is about a factor of two less efficient than in case (a) and the Rabi frequency should be larger by this factor to achieve the same effective cooling.

In a more realistic case of the four-level system with ( $\Omega_{23} = 3.5 \cdot 10^6$  rad/s and  $\Omega_{24} = 2.5 \cdot 10^6$  rad/s), the population of the excited state appears to be even less. These Rabi frequencies correspond to  $0.75 \text{ mW/cm}^2$  and  $1.6 \text{ mW/cm}^2$  respectively. It is at maximum 0.03, which in the above numerical example was determined as the minimal required value for the population. In order to understand laser cooling with strong repumping requirements a full simulation of the slowing process might be helpful.

Since the detuning of the lasers from the resonance is Doppler sensitive

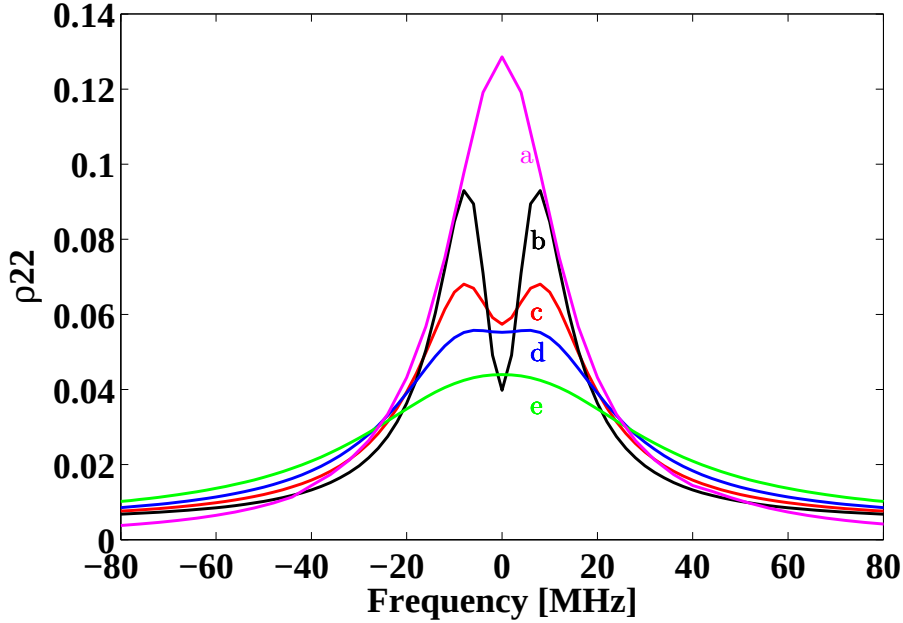


**Figure 5.7:**  $^1P_1$  state ( $\rho_{22}$ ) population at different linewidths of the repump lasers: (a) non-coherent population probability of  $^1P_1$  state, (b) 1 MHz, (c) 5 MHz, (d) 10 MHz and (e) 20 MHz.

the lasers are not always in resonance. While the velocity changes, they move through the resonance. As a result, we expect that we need higher intensities in the experimental realization than from this simple estimate.

### 5.2.2 Different Slowing Methods

Neutral thermal atomic beams have been cooled by various methods. Of them to mention are Zeeman slower method [172], frequency chirping of lasers [173], low-velocity intense source method [174], white light cooling technique [175, 176] and two step deceleration of an atomic beam [177]. Here we discuss briefly the advantages and disadvantages of these methods for barium atoms.

#### Zeeman Slowing Technique

During laser cooling, an atom is decelerated as it receives the momentum of photons from a laser beam counter propagating to its direction of motion. While

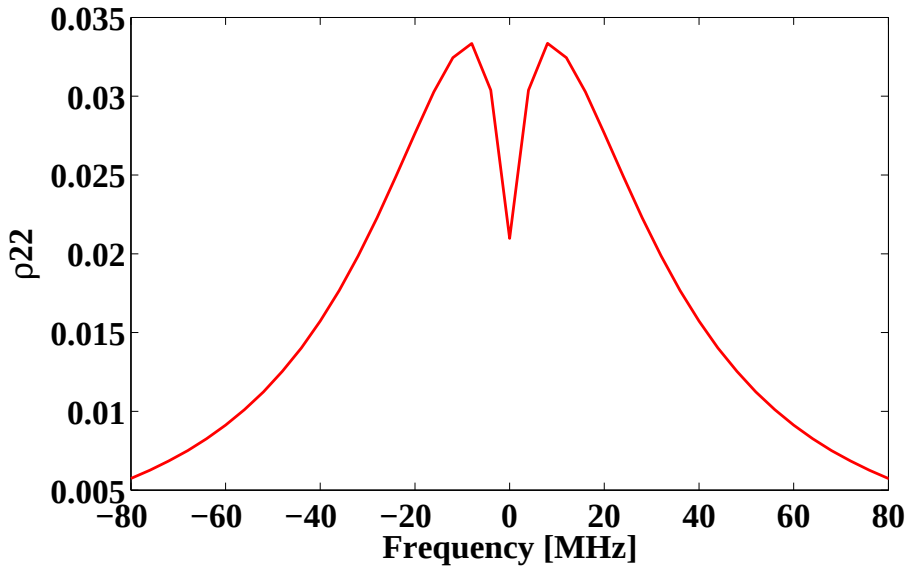


**Figure 5.8:**  $^1P_1$  state ( $\rho_{22}$ ) population for the four level system with  $\Omega_{23}=3.5 \cdot 10^6$  rad/s and  $\Omega_{23}=2.5 \cdot 10^6$  rad/s.

slowing an atom with a suitable wavelength laser light each absorbed photon gives a momentum 'kick' of  $\Delta p = \hbar k$  to a slowing atom. Subsequently, to bring it to rest tens of thousands of photons have to be scattered as quickly as possible. With the method of decelerating atoms by Zeeman tuned slowing, atoms pass through a spatially varying magnetic field that is designed to keep the atomic transition resonant with the laser by compensating the decelerated atom's changing Doppler shift with an equal but opposite Zeeman shift [172].

Decelerating barium atoms using Zeeman slower method may not be a feasible option because of the need of repump lasers. The barium  $^{138}\text{Ba}$  ( $I = 0$ ) atoms in an external magnetic field experience Zeeman splitting. The  $^1P_1$  singlet state has 3  $m_J$  states. The  $^1D_2$  state has 5  $m_J$  states and the  $^3D_{1,2}$  states have 3 and 5  $m_J$  states respectively. For perfect circular polarization the excited state decays into 3 magnetic substates of  $^1D_2$  and  $^3D_2$  and 2 states in  $^3D_1$ . The  $g_J$  factors for these transitions are given in Table 5.2.

In practice the polarization is not perfect and the other magnetic substates are populated. The energy level splitting in an external magnetic field is

$$\Delta f = \frac{m_J g_J \mu_B}{h} B \quad (5.43)$$

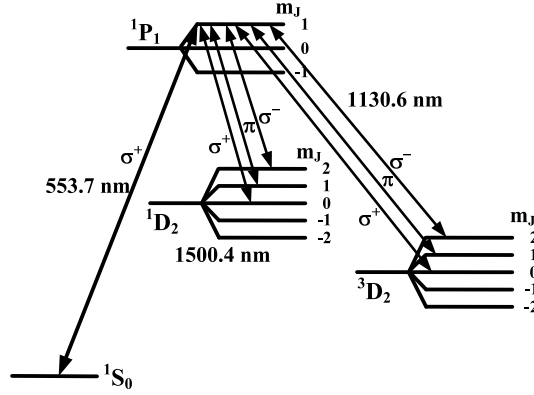


**Figure 5.9:** Splitting of the  $^1P_1$ ,  $^1D_2$  and  $^3D_2$  levels of barium atoms with nuclear spin zero in an external magnetic field. The possible  $m_J$  states that will be populated when barium is driven by right circularly polarized are shown.

where  $m_J$  is the magnetic quantum number,  $g_J$  is the Lande  $g$ -factor of the state,  $\mu_B$ , Bohr magneton,  $h$  is the Planck constant and  $B$  is the external magnetic field.

The magnetic field  $B_{max}$  for a Zeeman slower aiming for an initial velocity of  $v_{max}$  of the atom is

$$B_{max} = \frac{v_{max}}{\lambda g_J m_J \mu} \quad (5.44)$$

where  $\lambda = 553.7$  nm is the wavelength of the  $^1S_0$ - $^1P_1$  transition. For a barium atom of velocity  $v_{max} = 350$  m/s we arrive at a magnetic field of 450 G and a frequency splitting of 630 MHz. To cover the entire Zeeman splitting range for all the substates of the metastable  $D$ -levels, the repump lasers need to cover a frequency range of

$$\Delta f_{rep} = 2(g_J^P m_{Jmax}^P - g_J^D m_{Jmax}^D) \mu B. \quad (5.45)$$

Thus  $\Delta f_{rep}$  is on the order of twice the Zeeman splitting of the  $^1P_1$  state. In a detailed analysis one has to take the selection rules for the dipole transitions in a

| g <sub>J</sub> values of levels in barium |         |         |         |         |         |
|-------------------------------------------|---------|---------|---------|---------|---------|
| Level                                     | $^1S_0$ | $^1P_1$ | $^1D_2$ | $^3D_2$ | $^3D_1$ |
| g <sub>J</sub>                            | 0       | 1       | 1       | 7/6     | 1/2     |

**Table 5.2:** The  $g_J$  values of the levels involved in laser cooling of barium.

magnetic field into account.

However, the Doppler shift is inversely proportional to wavelength  $\lambda$  of the transition. Thus the Doppler shift cannot be compensated for all the transitions at the same time and a broad spectrum of the repump lasers is required. The situation is complicated in the case of isotopes with nuclear spin ( $I \neq 0$ ) because of the large hyperfine structure.

### Frequency Broadened Lasers

Another approach to achieve laser cooling of atoms is using a broad frequency spectrum of the laser light for the cooling transition, also called white light cooling [175]. In this method, there is always some portion of the light spectrum that is in resonance to compensate change in the velocity of the atom. For barium this may be an option. The broad frequency spectrum of the repump lasers is necessary to keep the Doppler shifted atoms in resonance.

We estimate the required intensity to 2-4 mW/cm<sup>2</sup> in 19 MHz bandwidth to achieve a Rabi frequency of  $5 \cdot 10^6$  rad/s. To cover a range up to the most probable velocity, which is 350 m/s, one needs a bandwidth of around 230 MHz for the 1500.4 nm or 12 times the power, and 310 MHz for the 1130.6 nm or 16 times the power required in a 19 MHz bandwidth.

The frequency broadening of the transition can be achieved by using an EOM with frequency side bands spacing of about the linewidth of the transition. To achieve the same bandwidth by power broadening of the transition one needs much more power, since Rabi frequencies of 1 GHz would be required. This corresponds to intensities for example  $\cong 50$  W/cm<sup>2</sup> (1500.4 nm), respectively  $\cong 250$  W/cm<sup>2</sup> (1130 nm). These intensities pose significant limitations in an experiment.

### 5.2.3 Experimental Results of Laser Cooling of Barium

The estimates for the required intensities show that the best way to achieve slowing of a barium atomic beam is with overlapping cooling and repumping lasers against the atomic beam. The experimental setup is shown in Fig. 5.10. We used enriched barium (<sup>138</sup>Ba) because fluorescence from slow <sup>138</sup>Ba atoms overlaps with the fluorescence of other isotopes at higher velocity which are more abundant. In addition, <sup>138</sup>Ba has no hyperfine structure. The 553.7 nm laser provides light for the cooling beam counter propagating to the atomic beam and a probe beam to measure the velocity distribution at 45° relative to the atomic beam.

The frequencies needed for the cooling and probe beams are generated using acousto optic modulators (Fig. 5.10). The acousto optic modulator AOM1 is used for the output power stabilization of the 553.7 nm laser (Section 4.4.1). The acousto optic modulator denoted as AOM2 is used as a variable beam splitter to provide power for cooling and probe beams. With the RF power to AOM2 we regulate the power into the zero order and first order beams. The zero order beam is passed through AOM3. We use '+1' order that is frequency shifted by ( $f_3$ ) relative to the output of the dye laser. It is utilized as cooling beam with 30 mW power available for the experiment. A fraction of the laser light (1 - 2 mW) is used in a second atomic beam apparatus to provide an absolute frequency reference to the barium transition.

The '-1' order beam, which is frequency shifted by AOM2 ( $f_2 = 60$  MHz), is passed through AOM4 ( $f_4$ ) to produce the probe beam. We use AOM4 in double pass configuration for the '+1' order to allow for scanning the frequency without changing the position of the laser beam. The beam crosses the atomic beam at 45° angle as shown in Fig. 5.10.

The difference frequencies  $\delta f$  between the cooling beam and probe beam are given by the AOM frequencies  $f_2$ ,  $f_3$  and  $f_4$  by

$$\delta f = 2 \cdot f_4 - f_3 - f_2 \quad (5.46)$$

The two beams probe different velocity classes of the atomic beam. Because of the different angles of the beams relative to the atomic beam they scan at a different rate across the Doppler profile (Fig.5.11). The detuning for the cooling beam is  $\delta_c = v/\lambda$ , while for the probe beam we have  $\delta_p = v \cdot \cos\alpha/\lambda$ , where  $\alpha = 45^\circ$ . The beams are both on resonance at a characteristic velocity  $v_0$  depending on the difference frequency  $\Delta f$

$$v_0 = \frac{\Delta f \cdot \lambda}{(1 - \cos\alpha)} \quad (5.47)$$

where  $v_0$  is the velocity class of the atoms both cooling and probe laser beams interact with at  $\Delta f$ ,  $\lambda$  is the wavelength of the  $^1S_0 - ^1P_1$  transition and  $\alpha = 45^\circ$  angle relative to the atomic beam.

To scan across the velocity profile of the atomic beam we scan the dye laser frequency. This changes the detuning of the cooling beam as well as the probe beam at the same time. This method is advantageous over using the double pass AOM3 for tuning the probe beam, because we can scan over a larger frequency range, while keeping alone the probe beam power constant.

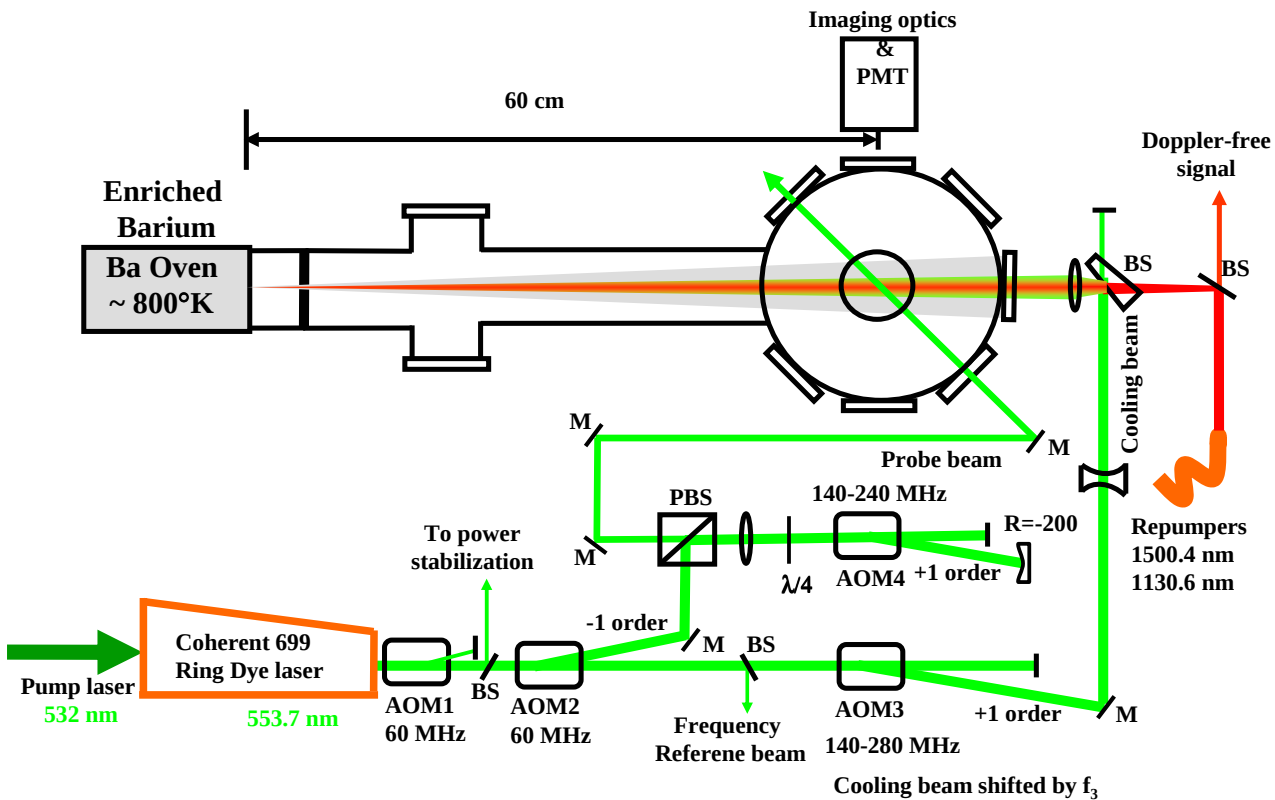


Figure 5.10: Setup for laser cooling of barium atoms.

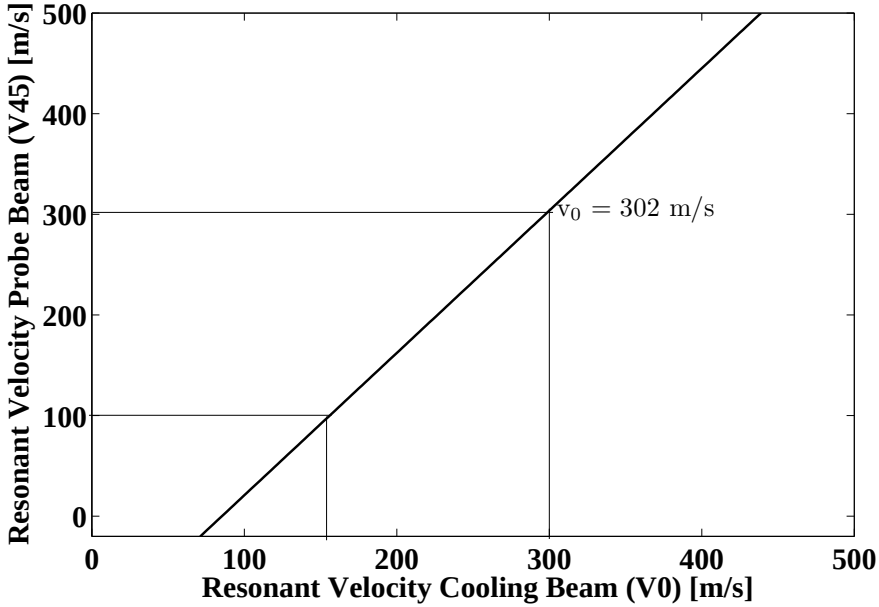


**Figure 5.11:** Velocities class of the atoms probed by the cooling beam and the probe beam. At a specific detuning both beams probe the same velocity class of atoms. This would be  $v_0 = 302$  m/s for a  $\delta f = 160$  MHz (Eqn. 5.46 AOM frequencies  $f_3 = 200$  MHz and  $f_4 = 210$  MHz).

The two repump lasers for the  $^1D_2 - ^1P_1$  (1500.4 nm) and the  $^3D_2 - ^1P_1$  (1130.6 nm) transitions are overlapped with the cooling beam. The main power of the two repump lasers is utilized for laser cooling. The laser beams at 1500.4 nm and 1130.6 nm are combined on a dichroic beam splitter. 1-2 mW power of the laser light from the repump lasers is used as a reference to find the Doppler free resonance of the respective repump lasers. The beams cross the atomic beam perpendicularly in the trap chamber and the fluorescence is observed by the same detection system (not shown in the Fig. 5.10).

The two repump laser beams are combined with the 553.7 nm cooling beam on a second beam splitter. At this point we are left with 44% of the output power for the 1500.4 nm laser and 30% for the 1130nm laser.

The combined beams are counter propagated to the atomic beam to achieve long interaction times. All the three beams are focussed to match the divergence of the atomic beam in order to achieve higher intensities. The atoms can interact with the laser beams over a length of 60 cm. The initial beam size is about 3 mm

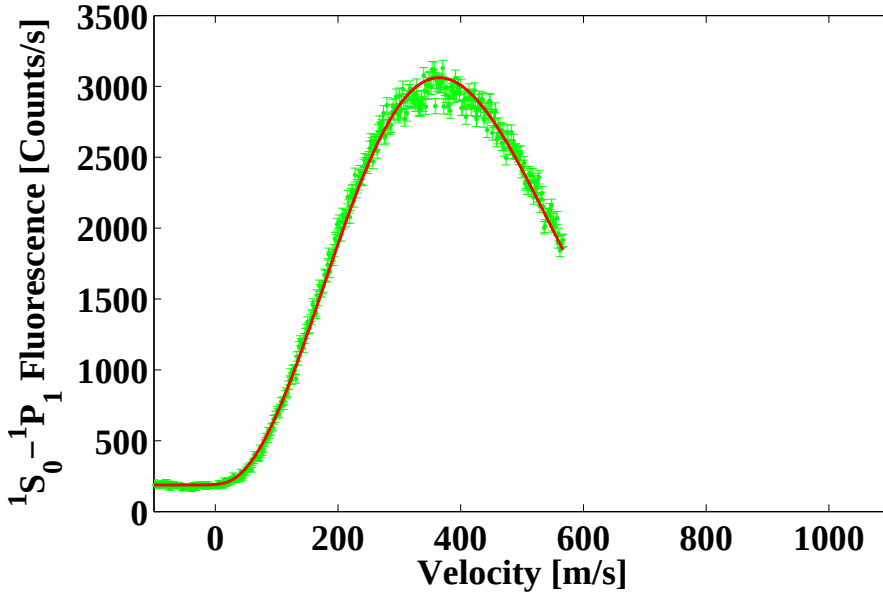


**Figure 5.12:** The velocity distribution of barium atoms from an effusive oven measured with probe beam at  $45^\circ$  to the atomic beam. From the fit we extract a mean velocity of  $250(20)$  m/s, which corresponds to a temperature of  $800(40)^\circ$ .

radius for the infrared and about twice the size for the 553.7 nm.

The probe beam at  $45^\circ$  to the atomic beam is aligned to intersect the cooling beam in the middle of the trap chamber. During the measurement the probe beam power is typically between 0.8 to 1.2 mW. The fluorescence from the atoms is observed with the detection system described in Section 4.3.4. The different power changes the overall fluorescence signal but does not effect its shape.

## Results

A typical longitudinal velocity distribution of the barium atoms from the atomic oven is measured with the probe beam at an angle of  $45^\circ$ . In Fig. 5.13 we show two examples where the velocity distribution is denoted with **(a)**. This velocity distribution is used as a reference spectrum when we affect the distribution by the cooling lasers. The spectrum agrees with the velocity distribution from an effusive oven (Fig. 5.12). The velocity spectrum is fitted assuming a Maxwell Boltzmann velocity distribution of atoms from an effusive oven and taking into consideration the laser beam size and the linewidth of the transition. This is

because the barium atom scatters maximum  $A_{leak}^{-1}$  photons in the time it stays in the interaction region before it ends up in one of the metastable  $D$ -states.

The fit to the spectrum agreed with the zero velocity calibration from a Doppler-free spectrum of barium isotopes measured in a second atomic beam setup. The two measurements are in good agreement with in the experimental error of about 10 m/s, which is due to the alignment of the laser beams with respect to the atomic beam.

The fluorescence spectrum measured when both the probe beam and cooling beam interacting with the  $^{138}\text{Ba}$  atomic beam is denoted as **(b)** in both figures. For velocities below  $v_0$ , where cooling and probe beams address the same velocity class we get a strong depletion, because of the optical pumping to the  $D$ -states. For the upper plot  $v_0 \simeq 230$  m/s and for the lower plot 320 m/s. The fluorescence in the absence of the repump lasers has two origins. First, the cooling beam does not cover the full atomic beam divergence and second a small reflection from one of the windows gives a probe beam at  $135^\circ$  relative to the atomic beam. The latter increases the fluorescence for negative velocities, meaning atoms are moving away from the beam.

In order to achieve cooling we overlapped the two repump laser beams with the atomic beam and the cooling beam at a specific detuning relative to the Doppler free frequency. At the right value for the detuning of the repump lasers we see a clear increase of atomic flux at lower velocities, a sign for laser cooling of barium atoms. The two figures denoted with **(c)** in Fig. 5.13 show optimization for two different velocity classes of the atoms and at different detuning of the repump lasers.

To get a quantitative estimate on the cooling we plot the enhancement of flux versus the difference of velocity excited by cooling and probe beam. In Fig. 5.14 **(I)** the enhancement of flux is over a wide range of velocity which extend down to zero velocity. From this figure we can estimate the velocity change of the atoms to  $\Delta v = 40$  m/s for atoms around 200 m/s. Atoms starting at lower velocity have been slowed further since they spend a longer time in the cooling laser beam. To slow an atom by 40 m/s it has to scatter  $N_p = \Delta v/v_r = 7500$  photons. This can only be possible if the atom is pumped back from metastable  $D$ -states about  $n_r = N_p/A_{leak}^{-1} = 22$  times. The time the atom had spend in the cooling beam over the distance of 0.6 m is  $t = d/v = 3$  ms. Thus the average time in a  $D$ -state is around  $140 \mu\text{s}$ . The laser intensities used in the experiment are about  $18 \text{ mW/cm}^2$  and  $64 \text{ mW/cm}^2$  corresponding to Rabi frequencies of  $19 \cdot 10^6 \text{ rad/s}$  and  $16 \cdot 10^6 \text{ rad/s}$  respectively. The Rabi frequencies are larger than our estimate above by a factor of 5. The main reason for this difference is probably the overlap

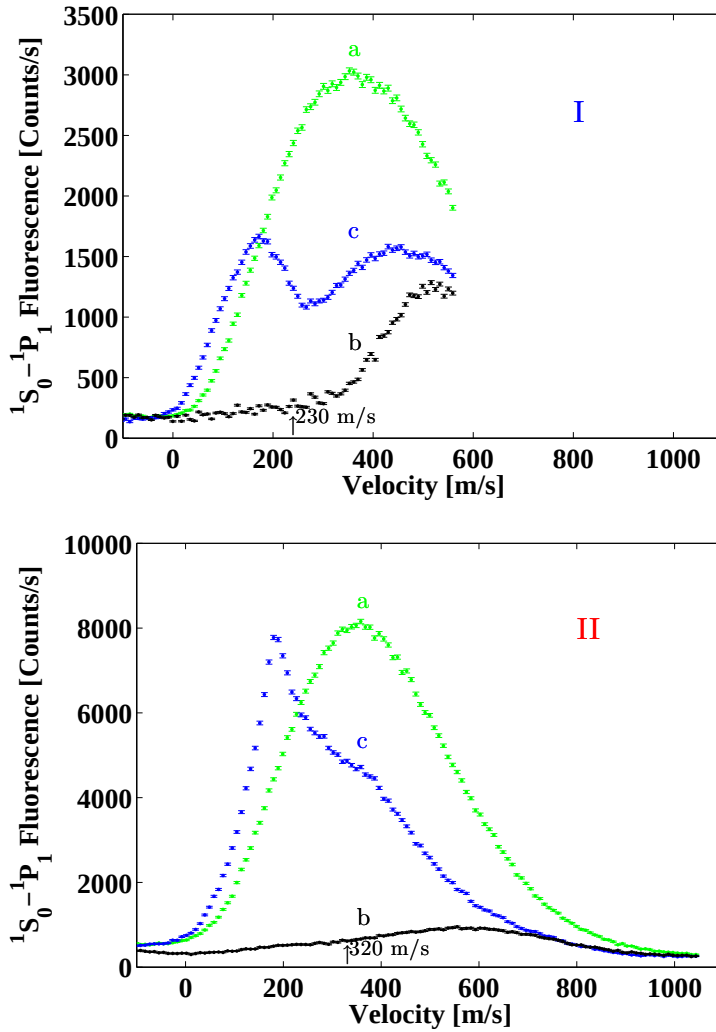


**Figure 5.13:** Examples of laser cooling results for two different detuning of the lasers. **I:**  $v_0=230\text{ m/s}$  and **II:**  $v_0=320\text{ m/s}$ . **(a)** Longitudinal velocity distribution of the atoms measured with 553.7 nm probe beam at  $45^\circ$  to the atomic barium beam. **(b)** Velocity distribution of the atoms measured with the cooling beam counter propagating the atomic beam and 553.7 nm probe beam at  $45^\circ$  to the atomic beam. The fluorescence of the atoms with velocities less than  $v_0$  is almost equal to the background counts because the atoms decayed to the metastable D-states. Only fluorescence from the faster atoms is detected. **(c)** Velocity distribution of the atoms measured with the 553.7 nm cooling beam and repump laser beams counter propagating to the atomic beam and 553.7 nm probe beam at  $45^\circ$  to the atomic beam.

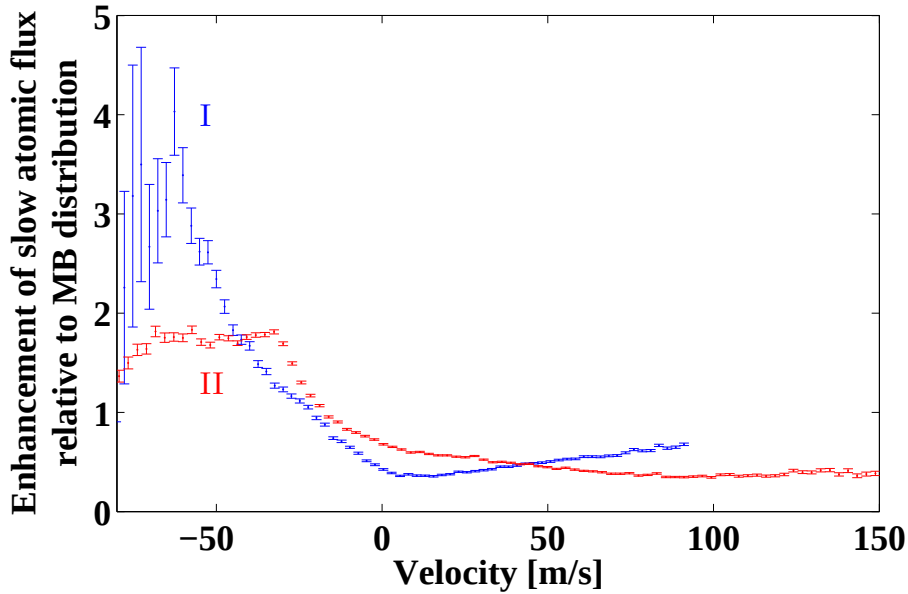


**Figure 5.14:** Increase in the flux of slow atoms after laser cooling. We plot the enhancement of flux over the velocity distribution from the oven versus the difference of velocity with which the cooling beam is on resonance and the probed velocity which is a measure of the slowing of the atoms. The increase is the ratio of spectra (c) to (a) in Fig. 5.13. the enhancement of flux is over a wide velocity range in **I** where as it is flat after a certain velocity range in **II**.

of all the repump laser beams with the atomic beam and the cooling laser beam in the interaction region.

Apart from the geometrical overlap of laser beams and the atomic beam the cooling result depends on several parameters. These are, in particular, the repump lasers power, detuning and the frequency spectrum of the repump lasers. The effects due to a change in these parameters are reflected in the velocity spectrum after the slowing. Thus our observation is always the effect of the changed parameter averaged over the whole velocity distribution of the atomic beam. To extract the effect on a particular velocity class, one would need a full numerical solution of the slowing process. Without that, we can only give a qualitative picture.

We investigated the influence of repump lasers intensity on the enhancement of lower velocity atom flux. The observed spectra are shown in Fig. 5.15. They show the result for 3 different intensities of the repump lasers. A spectrum with-

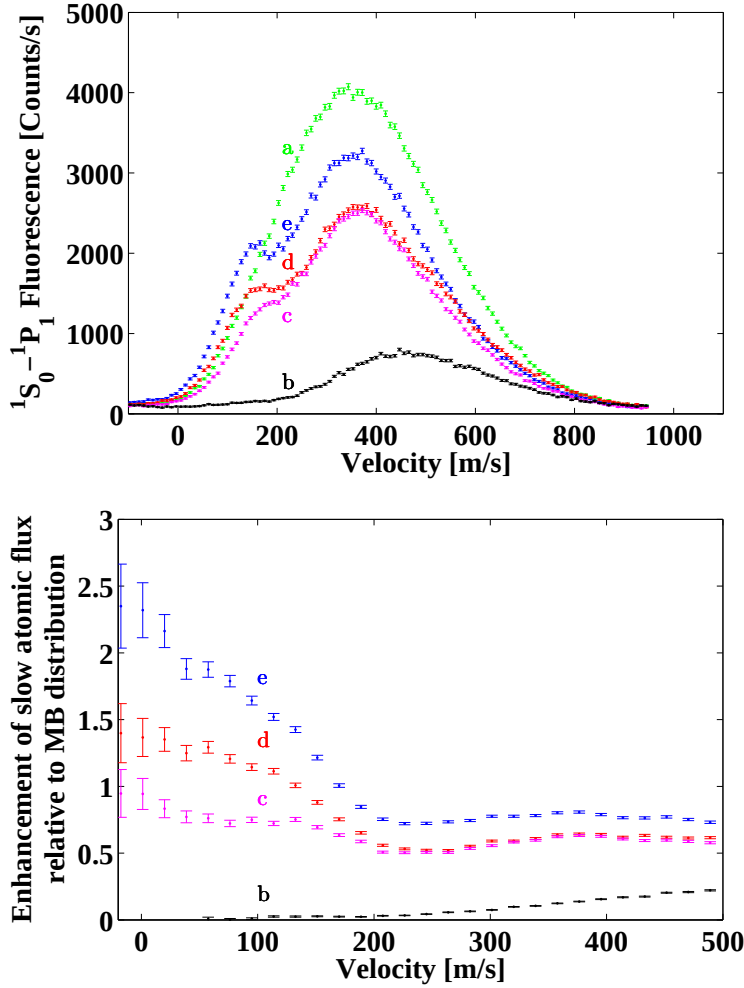


**Figure 5.15:** Velocity distribution measured with cooling laser and at different output power of the repump lasers against the atomic beam and the probe laser at  $45^\circ$  to the atomic beam. **(b)** with no repump lasers. **(c)** Enhancement of atom flux at repump lasers intensities of 20(4) mW/cm<sup>2</sup> (1130.6 nm) and 5(1) mW/cm<sup>2</sup> (1500.4 nm), **(d)** 40(8) mW/cm<sup>2</sup> and 10(2) mW/cm<sup>2</sup>, **(e)** 60(12) mW/cm<sup>2</sup> and 15(3) mW/cm<sup>2</sup> respectively. The bottom plot shows the enhancement at low velocities at different powers. Further increase in the repump lasers power did not increase the slow barium atom flux.

out repumping is added for reference. The slow atom flux in the velocity distribution of atoms increases with higher power of the repump lasers. A further increase of the intensities of the repump lasers did not increase the atoms at lower velocities. This indicated that due to the changing of the Doppler shift, the atoms moved out of resonance and could not be slowed further. The intensities of the lasers were not large enough to cause power broadening and thus allow a larger velocity range to be slowed.

The measurements were repeated for different detuning frequency of the repump lasers (Fig. 5.16). We observe that changing the detuning of the repump lasers by about half the linewidth of the transition influences the repumping and the slowing of atoms. This strong dependance is expected from the calculations of the multilevel system (Fig. 5.7). In particular, the dark Raman resonance can cause that the atoms cannot be slowed further than the velocity, at which it appears, because effective repumping has ceased. One can assume that at higher intensities for the repump lasers the effect of the detuning is less. With our experimental setup we could not explore higher intensities due to the limitation in the power of the fiber lasers.

As a third parameter, we investigated the influence of the spectral width for the repump lasers. For these measurements we introduces an EOM in the pass of the infrared lasers. Depending on the RF-power applied to the resonant EOM the spectral width is broadened (as an example see Fig. 4.19). In Fig. 5.17 several spectra are shown. In the caption the effective spectral width of the repump laser is given. The intensity of the lasers is held constant. The largest enhancement at low velocities is achieved with 60 MHz (1130 nm) and 35 MHz (1500 nm). Increasing the spectral width further lead to a decrease in slow atoms down to the level of no broadening due to the EOM. Here, the power is distributed over a larger frequency range and the effective repumping rates are reduced. Still, at velocities below 50 m/s, there is a small advantage over the case without frequency broadening. The longer time of flight of the atoms can compensate for the smaller repump rates.

The results of the measurements with variations in the parameters fit well with our expectations. In order to get a better picture on the various contributions to the cooling a single velocity barium beam would be advantageous. Such a beam could be produced by a velocity selector with chopping wheels.

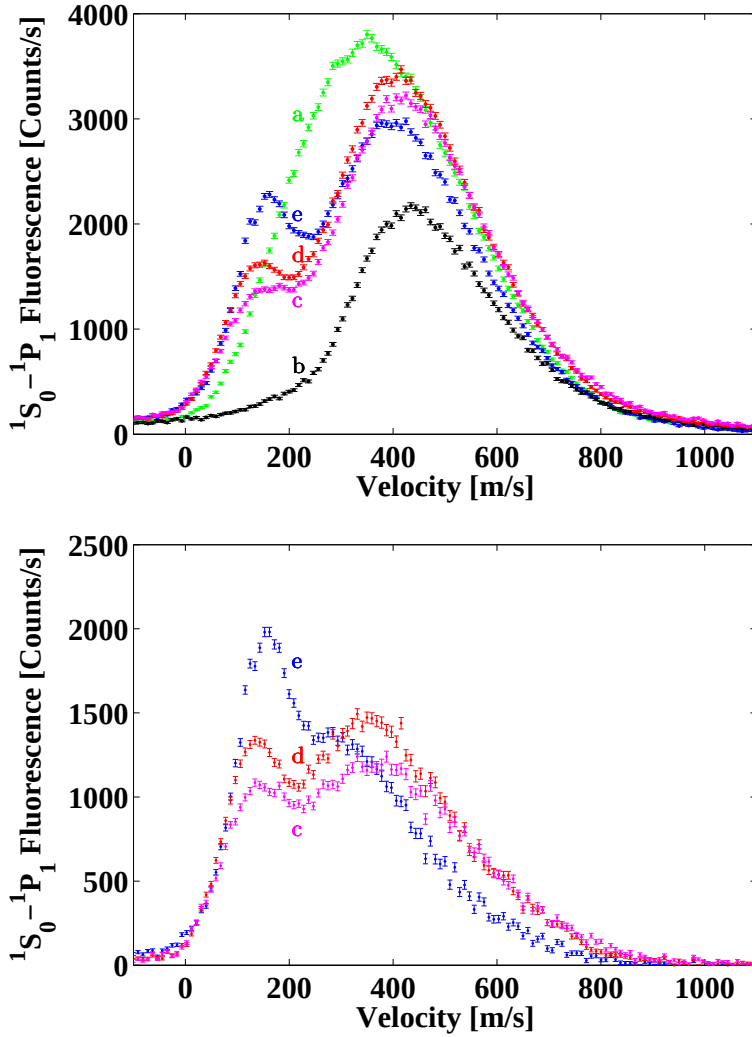


**Figure 5.16:** Velocity distribution of the barium atoms measured with cooling laser, for 3 different detuning of the repump lasers. (a) initial velocity distribution, (b) depletion of velocity spectrum due to optical pumping to dark states, and (c-e) with different detunings of the repumper lasers. In (d) the detuning is -20(1) MHz (1130 nm) and -24 MHz (1500 nm) relative to (c) and in (e) the detuning is -8(1) MHz (1130 nm) and -15 MHz (1500 nm) relative to (c). Thus a change of less than the linewidth of the transition has a large consequence for the slowing process. The effect over no repump lasers in fluorescence is shown in the lower figure.

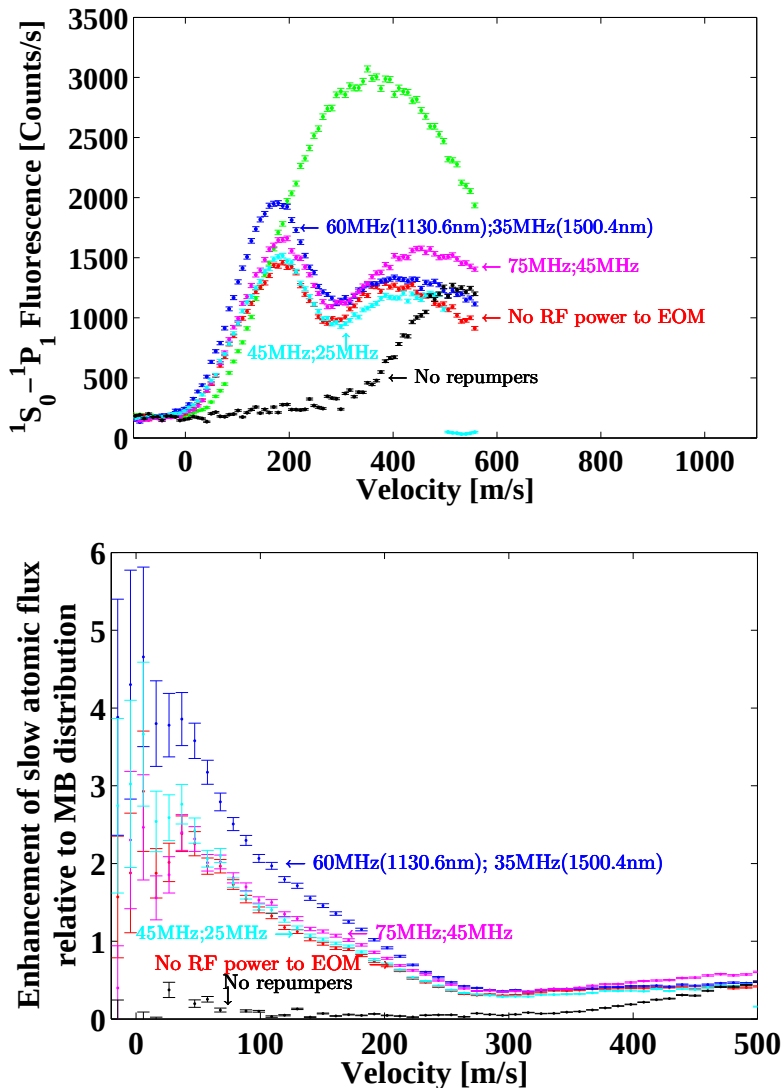


**Figure 5.17:** Velocity distribution of atoms measured with cooling laser and frequency broadened repump lasers by an EOM counter propagating the atomic beam and probe laser at  $45^\circ$  to the atomic beam. The repump lasers are detuned by -60 MHz. Enhancement of slow barium atom flux without RF power to infrared lasers EOM, at different RF power to EOM : -2 dBm corresponding to a frequency broadening of 45 MHz (1130.6 nm) and 25 MHz (1500.4 nm), 0 dBm corresponding to 60 MHz and 35 MHz and and +2 dBm: 75 MHz and 45 MHz. The enhancement increases with increasing RF power to the EOM but at +2 dBm it decreases because the frequency broadening is large for the repump lasers so that the repump lasers are in resonance with high velocity atoms.

## 5.3 Cooling of Atoms with Complex Level Structure

In this chapter, we have investigated the possibilities for laser cooling of atoms with a complex level structure. In particular, we discussed the heavy alkaline earth element barium. We deal with two features.

- A strong leak in the singlet-singlet  $^1S_0 - ^1P_1$  cooling transition at 553.7 nm to the metastable  $D$ -states. The leakage is stronger than in any other atom which has been laser cooled so far. Without repumping the velocity can only be changed by less than 2 m/s before the atoms are pumped into one of the metastable states.
- The repumping from the metastable  $D$ -states has to go via the excited state of the cooling transition. During repumping we see the effects of the coherent Raman transitions in this level system.

We showed that these features do not cause a major limitation for laser cooling of such atoms.

We have discussed the behavior of a  $\Lambda$ -system barium interacting with two coherent light fields. The properties of the two-photon transition that exhibit coherent Raman resonances are observed and compared to the results from a numerical solution of the optical Bloch equations. The extracted Rabi frequencies are large enough to allow for slowing a barium atomic beam. We demonstrate the first successful slowing of a thermal atomic beam by about 60 m/s. At velocities below 30 m/s we see an enhancement of the flux of atoms of nearly a factor of 5 over the thermal distribution from the atomic beam. In addition, we showed that the detuning of the repump lasers play an important role in achieving efficient slowing. The slow atom flux, which we have achieved was limited by the amount of laser power available for the repump lasers.

It should be possible to capture the slowed atoms from the beam in a magneto optical trap. The results are directly applicable to laser cooling of radium.



## Chapter 6

# Summary and Outlook

This thesis is written in the framework of the research of the TRI $\mu$ P group at KVI. Described are the first steps toward the search for a permanent electric dipole moment (EDM) of the radium atom, which violates simultaneously parity (P) and time reversal symmetry (T). Any observation of an EDM would be an unambiguous signature for physics beyond the Standard Model of the electroweak interactions.

We have presented the Standard Model prediction of EDMs for fundamental particles which are well below the limits set by the current experimental efforts with various systems. The Radium atom has drawn in recent years considerable attention by both the theoretical and the experimental physics community for studying discrete symmetries like parity, time reversal and charge conjugation. The high sensitivity of radium arises from the large polarizability due to the near degeneracy of the  $7s7p\ ^3P_1$  and  $7s6d\ ^3D_2$  levels, which are of opposite parity. An energy difference of only  $5\text{ cm}^{-1}$  between these levels is indicated from the available spectroscopic data. To confirm this, improved spectroscopic studies are required as an input for theoretical studies and for the design of the EDM experiment. In particular, the term energies and the lifetimes of the singlet and triplet  $P$  and  $D$ -states are needed to identify a suitable laser cooling scheme.

Short lived radium isotopes with nuclear spin  $I \neq 0$ , are not readily available for the experiments. They have to be extracted from radioactive sources or produced by nuclear reactions at accelerator facilities like the TRI $\mu$ P facility at KVI.

We have presented the various stages for the production, separation and slowing of the short lived radioactive isotopes of our interest at TRI $\mu$ P. Since only small quantities of short lived radioactive atoms are available and because of

long observation times sensitive experiments will be based on laser cooled and trapped atoms.

To develop spectroscopic techniques and laser cooling schemes we used stable isotopes of barium, because of its similarity in its energy level diagram to radium. For spectroscopy, metastable singlet and triplet  $D$ -states were populated by optical methods rather than using the conventional method of atomic discharges. The singlet  $D$  state is populated by driving the  $6s^2\ ^1S_0 - 6s6p\ ^1P_1$  transition. To populate the triplet  $6s5d\ ^3D_{1,2}$  states efficiently we exploited the branching ratio of the  $6s6p\ ^3P_1$  level by optical pumping with the narrow intercombination line. To excite this  $6s^2\ ^1S_0 - 6s6p\ ^3P_1$  transition at 791.3 nm we built diode lasers. We have achieved long term frequency stability of these lasers by referencing the frequency to a nearby transition in molecular iodine  $I_2$ , an often used frequency standard. With this setup we have performed several measurements.

We have reported the first direct optical spectroscopy of the  $6s5d\ ^1D_2 - 6s6p\ ^1P_1$  (1500.4 nm), the  $6s5d\ ^3D_2 - 6s6p\ ^1P_1$  (1130.6 nm) and the  $6s5d\ ^3D_1 - 6s6p\ ^1P_1$  (1107.8 nm) transitions respectively.

We optically resolved the hyperfine structure of the  $6s5d\ ^3D_1$  and  $6s5d\ ^3D_2$  levels for the  $^{137,135}\text{Ba}$  isotopes. The intensities of the measured hyperfine spectra are compared with the transition strengths calculated using Clebsch-Gordan coefficients. The hyperfine structure splitting is compared to previous microwave measurements for the  $^3D_1 - ^1P_1$  and  $^3D_2 - ^1P_1$  transitions, which are of higher precision. These are used for the calibration of the frequency scanning of the fiber lasers at 1107.8 nm and 1130.6 nm lasers. In addition, we used frequency side bands generated with an EOM and Zeeman splitting in a magnetic field to calibrate the fiber laser frequencies scanning.

In an atomic beam of natural barium we find the majority of the atoms distributed over the isotopes  $^{138-134}\text{Ba}$ . This allowed us to perform the first measurement of isotope shifts of the  $6s5d\ ^3D_1 - 6s6p\ ^1P_1$  transition. The observed shift between  $^{138}\text{Ba}$  and  $^{136}\text{Ba}$ ,  $^{136}\text{Ba}$  and  $^{134}\text{Ba}$  isotopes and between the  $^{137}\text{Ba}$  and  $^{135}\text{Ba}$  isotopes are given in Table 6.1. The analysis with a King plot shows that electron-electron correlations may play a larger role for the values of the isotope shift. A better theoretical understanding would be desirable because of possible consequences for an EDM measurement utilizing atoms in one of the metastable  $D$ -states.

We report the experimental observation of coherent Raman resonances in barium for the first time. To understand these resonances quantitatively we solved the optical Bloch equations and compared the results with the experimental observation. We extracted the relevant Rabi frequencies for the  $\Lambda$ -system and estimated

| Isotope pair        | 138-136 | 136-134 | 137-135    |
|---------------------|---------|---------|------------|
| Isotope shift (MHz) | -71(5)  | -93(6)  | -75.3(0.5) |

**Table 6.1:** Measured isotope shifts of the  $6s5d\ ^3D_1 - 6ssp\ ^1P_1$  transition in barium.

the transition rates, relevant for the laser cooling experiment.

We have achieved the first slowing of a barium atomic beam using enriched  $^{138}\text{Ba}$ . These results have a far reaching influence for atoms, in which a closed cooling cycle can only be found with several laser fields at different wavelengths, adding to the complexity of the experimental setup. The results from this study can easily be extended to other atoms with similar level structure, in particular radium.

The only option for laser cooling of barium is offered by the  $6s^2\ ^1S_0 - 6s6p\ ^1P_1$  transition. Unlike the other atoms that have been laser cooled and trapped, the barium atoms are easily pumped into metastable  $D$ -states. Without repumping no significant change in the velocity of the atoms can be achieved. They have to be pumped back through the  $6s6p\ ^1P_1$  state used in the cooling transition. This makes barium a multiple  $\Lambda$ -system. The repumping of atoms through a common excited state results in coherent Raman resonances.

In our experimental setup for slowing of barium atoms we employed a co-propagating cooling laser and infrared repump lasers travelling against the atomic beam. We have measured the velocity distribution from our atomic beam source with a 553.7 nm probe laser at  $45^\circ$  to the atomic beam at a distance of 0.6 m from the oven. We see a clear sign of cooling with cooling laser and repump lasers at appropriate frequencies. The shift in the velocity of the atoms can be larger than 50 m/s. At velocities below 30 m/s we see an enhancement of the thermal beam of flux of atoms nearly a factor of 5.

The next step is to capture the slowed atomic beam into a magneto-optical trap. In the future, we need to expand cooling to isotopes with hyperfine structure. In this case, the repump lasers should cover the frequency spectrum of the hyperfine structure splitting. This can be achieved by electro-optical modulators.

In the future, the slowing of atoms have to be optimized. Frequency chirping of the lasers look like a promising option as well as white light cooling. This can improve the accessible range of velocities from an atomic beam and it shows advantages over Zeeman slowing techniques, especially for isotopes with hyperfine structure.

In addition, the geometry of the experiment can be optimized. A critical parameter is the overlap of the infrared repump lasers with the atomic beam.

Furthermore, the infrared repump lasers should be frequency stabilized. The first steps were taken to lock them to Fabry-Perot cavities.

A theoretical treatment of the laser cooling for barium should be done by solving the equation of motion and the optical Bloch equation simultaneously for the whole cooling process. This can be based on our treatment of the multilevel system. The results of the calculation will go beyond our simple estimates and allow to design the laser system for radium laser cooling.

We have demonstrated the spectroscopy of metastable  $D$ -states in barium and have used the results to set up the first slowing of a thermal barium beam. All these results will be used for the setup of a radium spectroscopy and cooling experiment at KVI, where we are developing the online production of radium as well as a radium atomic beam. Within the next few years we expect to report similar results on spectroscopy and trapping with radium.

## Appendix A

# Frequency Offset Locking of Lasers

The frequency offset locking of lasers [178–180] also called frequency difference stabilization is described. The schematic layout of the electronics employed for frequency offset locking of the two diode lasers is shown in Fig. A.1.

Around 1 mW of laser light is taken from the two diode lasers. They are overlapped and coupled into a single mode fiber that is connected to a fast, high bandwidth silicon (Si) photodiode (Thorlabs SUV-FC 2.2 GHz), which measures the beat note. The output signal of the photodiode at a beat frequency of  $\nu_1$  MHz is amplified by a  $2 \times 10$  dB two stage radio frequency amplifier. After amplifying the signal, part of the signal is split off for monitoring the beat signal. The output of the amplifier is mixed down (Mini-Circuits ZMX-2000) with the output of a local oscillator in our case a frequency synthesizer (HP 8656B) operating at a frequency  $\nu_2$  MHz. The radio frequency signal is further amplified with a radio frequency amplifier. Subsequent, output of the amplifier is split into two equal parts, one part passes through a band pass filter at 16.6 MHz. It acquires a frequency dependent phase shift which we can easily detect by recombining the signal with the original signal on a phase detector. As a result we get an dispersive like signal that can be used to frequency lock the laser.

The error signal is used to lock the offset diode laser using a PID servo loop [181]. One servo loop path with an proportional amplifier and integrator controls the piezo of diode laser adjusting the length of the external cavity and a second servo loop with proportional gain controls the laser diode injection current for suppression of the low frequency noise. The beat note could be scanned over several hundred MHz keeping both the lasers locked.

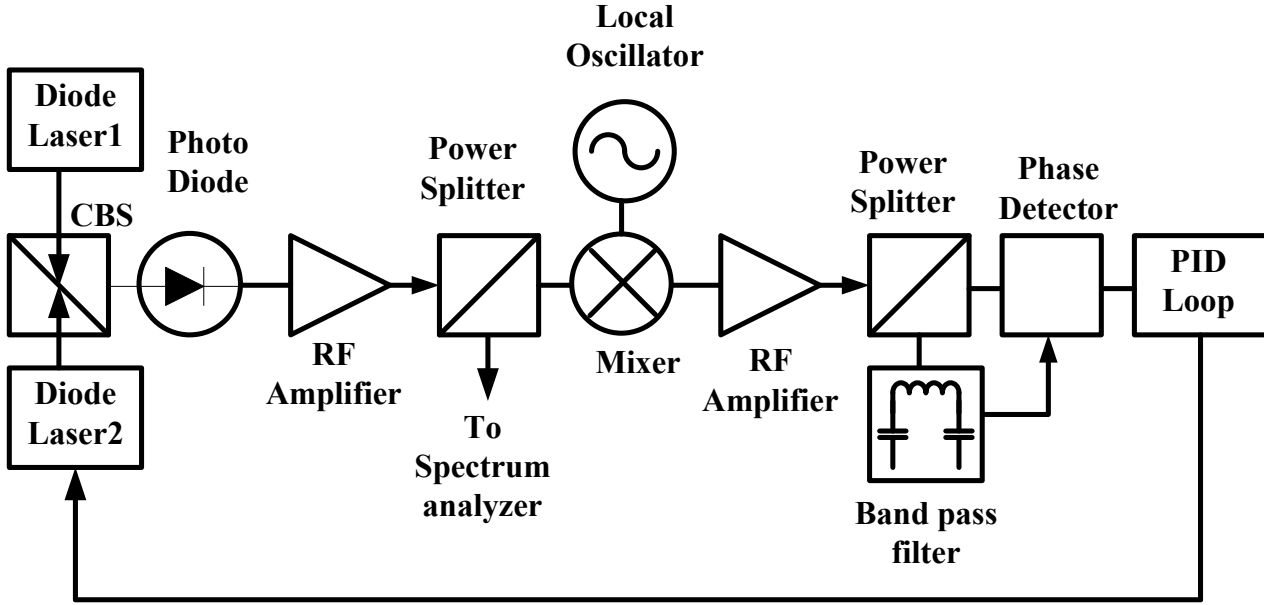


**Figure A.1:** Electronic scheme for the frequency offset locking of the diode lasers.

## Appendix B

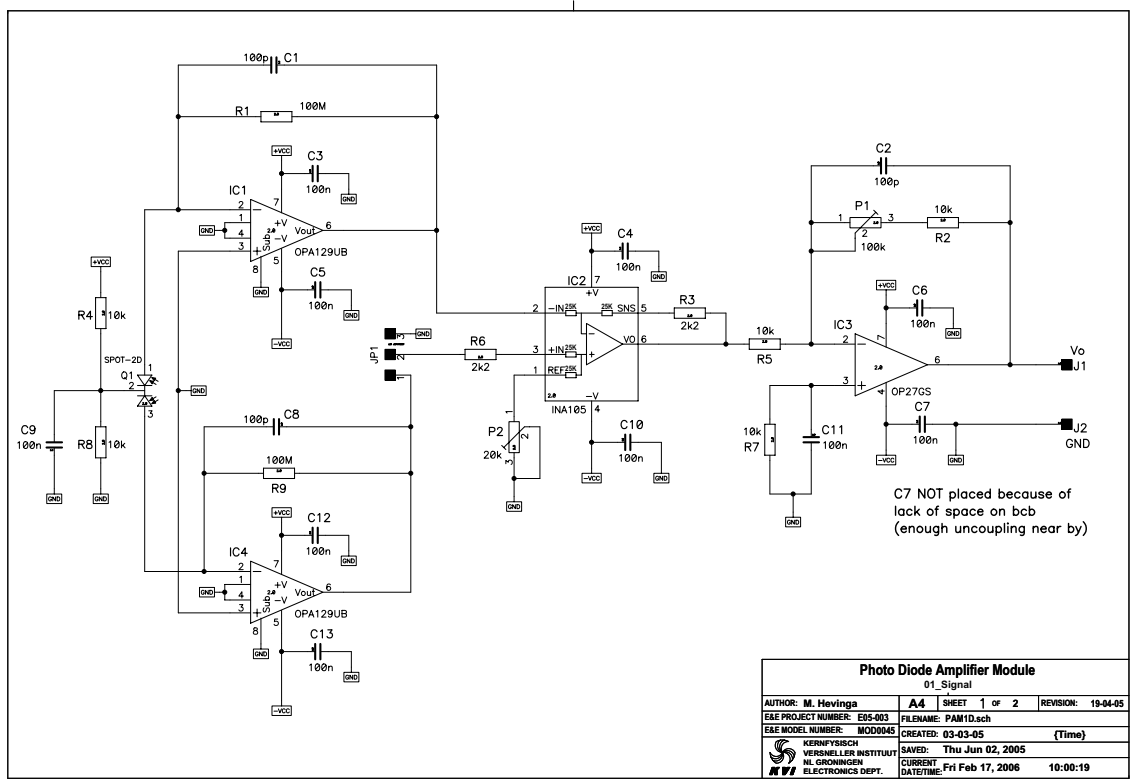
# Photodiode Amplifier

The photodiode transimpedance amplifier was designed to satisfy the following conditions.

- To detect very small light signals.
- Highest sensitivity and high gain.
- High bandwidth advisable at lower gain.
- Compact and cost-effective.

The designed photodiode amplifier is a multifunctional device which can be operated in both photovoltaic and photo conductive mode. It is designed such that a split-photodiode can also be used for measurements where difference signals are needed. We use this for frequency stabilizing the dye laser. Here it operates as difference amplifier with a variable gain stage of gain 1-10 and an option for adjusting offsets of the signals. The sensitivity of the photodiode amplifier is 1 V/100pW. It can be used for low-level light detection like fluorescence measurements. The advantages over PMT is that it is less sensitive to higher light levels and it has a high quantum efficiency (QE) in the wavelength range 400 nm-1100 nm.

**Figure B.1:** The schematic electronics layout of the split photodiode amplifier used for iodine spectroscopy and dye laser frequency stabilization.



## Appendix C

# Electro Optic Modulator

The electro-optic modulator *EOM* was built for generation of white light for the 553.7 nm laser light of  $^1S_0$ - $^1P_1$  transition,  $^1D_2$ - $^1P_1$  transition (1500.4 nm) and  $^3D_2$ - $^1P_1$  transition (1130.6 nm) respectively. The device utilizes a lithium tantalate  $\text{LiTaO}_3$  crystal in a resonant driving circuit.

### C.0.1 Electro-Optic Effect

A detailed explanation of the electro-optic effect can be found in most non-linear optics text books [182]. The propagation of electromagnetic waves in anisotropic crystals is best described by the index ellipsoid

$$\frac{x^2}{n_x^2} + \frac{y^2}{n_y^2} + \frac{z^2}{n_z^2} = 1 \quad (\text{C.1})$$

where  $x$ ,  $y$  and  $z$  are chosen to be in the same direction as the principal dielectric axes of the crystal (the axes along which the  $D$  and  $E$  vectors in a beam of light are parallel). For a uniaxial crystal the index of ellipsoid takes the form:

$$\frac{x^2}{n_o^2} + \frac{y^2}{n_o^2} + \frac{z^2}{n_e^2} = 1 \quad (\text{C.2})$$

The application of an external electric field to a medium which does not possess inversion symmetry may induce a change in the ordinary and extraordinary refractive indices - the *electro-optic effect*. The magnitude and direction of the induced birefringence depends on the magnitude and direction of the applied field relative to the dielectric axes of the crystal. The effect of the electric field may be described by a modified index of ellipsoid.

$$\left(\frac{1}{n^2}\right)_1 x^2 + \left(\frac{1}{n^2}\right)_2 y^2 + \left(\frac{1}{n^2}\right)_3^2 + 2\left(\frac{1}{n^2}\right)_4 yz + 2\left(\frac{1}{n^2}\right)_5 xz + 2\left(\frac{1}{n^2}\right)_6 xy = 1 \quad (\text{C.3})$$

In zero applied field Eq. C.3 reduces to Eq. C.2. The changes in the coefficients  $(\frac{1}{n^2})_i$  caused by the applied field can be determined by the elements  $r_{ij}$  of a 6x3 tensor specific to the material, called the ‘electro-optic tensor’, such that:

$$\Delta\left(\frac{1}{n^2}\right)_i = \sum_{j=1}^3 r_{ij} E_j \quad i = 1, \dots, 6 \quad (\text{C.4})$$

### C.0.2 The Lithium Tantalate Crystal

The characteristics, specifications and initial tests of the LiTaO<sub>3</sub> crystal(s) used in the present measurements are presented in the reference [183]. In this section, some of the important features are described. The crystal is 25 mm long, 0.75 mm wide and 1 mm high. It is placed between two electrodes so that a transverse electric field could be applied along the optic axis i.e., the z-direction (Fig. C.1). Under such conditions the crystal remains uniaxial, but with a modified index ellipsoid.

$$\left(\frac{1}{n_o^2} + r_{13}E\right)(x^2 + y^2) + \left(\frac{1}{n_e^2} + r_{33}E\right)z^2 = 1 \quad (\text{C.5})$$

### C.0.3 Frequency Modulation

Frequency modulation of the light transmitted by the lithium tantalate crystal may be achieved by the application of a sinusoidally varying electric field. The effect of an applied voltage of the form  $V = V_0 \sin \omega_m t$  is to change the output phase of the light by:

$$\delta \sin \omega_m t = \frac{\pi L}{\lambda d} n_e^3 r_{33} V_0 \sin \omega_m t \quad (\text{C.6})$$

If the electric field of the incident light is described by  $E_{IN} = A \cos \omega_c t$ , then the field of the transmitted light may be written as:

$$E_{OUT} = A \cos(\omega_c t + \delta \sin \omega_m t) \quad (\text{C.7})$$

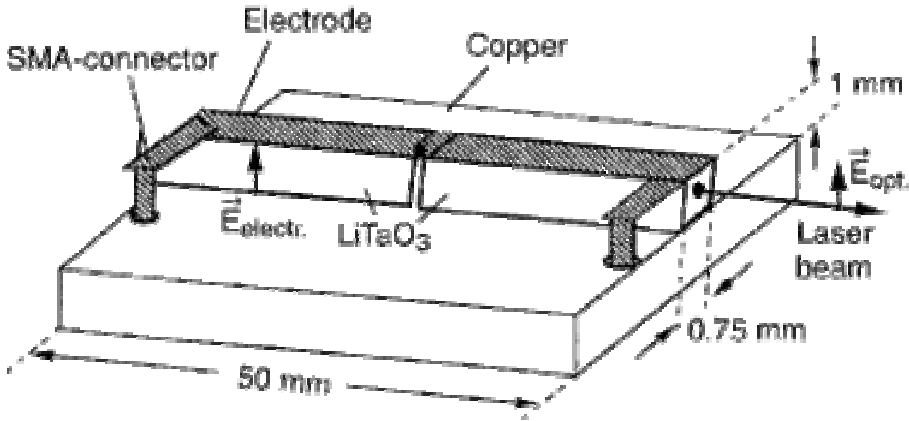


**Figure C.1:** The EOM in travelling-wave configuration. LiTaO<sub>3</sub> crystal is used to produce frequency side bands [figure taken from reference [184]].

The light is said to be phase modulated with a modulation index  $\delta$ . Eq. C.7 can be rewritten as:

$$E_{OUT} = A[J_0 \cos \omega_c t + J_1 \cos(\omega_c + \omega_m)t - J_1(\delta) \cos(\omega_c - \omega_m)t + J_2(\delta) \cos(\omega_c + 2\omega_m)t + J_2(\delta) \cos(\omega_c - 2\omega_m)t + \dots] \quad (C.8)$$

Frequency sidebands, separated by the modulation frequency  $\omega_m$ , are therefore generated. The two first order sidebands have the opposite phase. The distribution of energy between the carrier and the sidebands is determined by the modulation index  $\delta$ . The ratio of the amplitude of the first sideband to that of the carrier is given by:

$$\frac{E_1}{E_0} = \frac{J_1(\delta)}{J_0(\delta)} \quad (C.9)$$

#### C.0.4 The Resonant Circuit

The simplest way to achieve the high modulation voltage required at the relatively high frequencies ( $\omega_m/2\pi = 33.90$  MHz) desirable in the laser cooling experiments depends on the line width of the transition. This is accomplished by incorporating the electro-optic crystal into a resonant circuit with relatively high  $Q$  factor. The circuit used in this experiment is illustrated in Fig. C.3.  $C_x$  represents the parallel plate capacitance of the lithium tantalate crystal between the two electrodes.  $L$  represents an inductor with a high  $Q$ -factor. The resistance

of the inductor winding is represented by  $R$ . Finally,  $C$  represents an optional, additional capacitance placed in parallel with the crystal to change the resonant frequency of the circuit.

The resonant frequency of the circuit in Fig. C.3 is  $\omega_0 = 1/\sqrt{LC_x}$ . At this frequency the impedance of the circuit is real and equal to the resistance  $R$ . The voltage across the resistance is then equal to the voltage across the whole circuit and a current of magnitude  $V/R$  flows. The voltage across the inductance  $L$  and the capacitance  $C_x$  is zero. The magnitude of the voltage across the capacitance alone is

$$\frac{V_c}{V} = \frac{1}{\omega_0 C_x R} = Q \quad (\text{C.10})$$

Therefore, at the resonant frequency, the voltage across the lithium tantalate crystal is enhanced by a factor equal to the  $Q$ -factor of the resonant circuit.

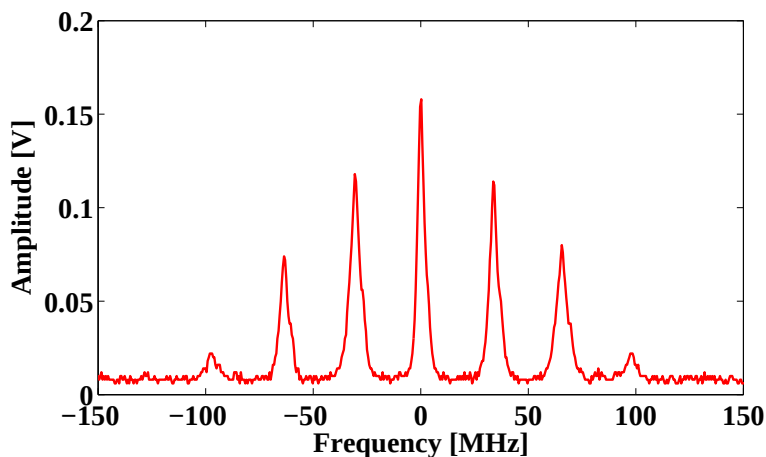


**Figure C.2:** The sidebands observed after an EOM at a modulation frequency of 33.9 MHz from a HeNe laser.

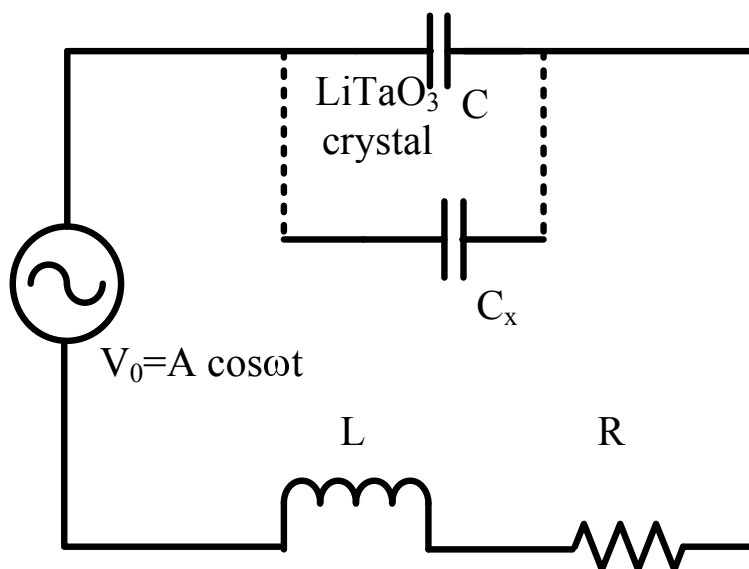


**Figure C.3:** Resonance circuit used for the  $\text{LiTaO}_3$  crystal to have resonance frequency at 33.9 MHz.



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# Samenvatting

Symmetrieën zijn een waardevol concept gebleken in de beschrijving van de natuur. De discrete symmetrieën pariteit (P), ladingsconjugatie (C) en tijdsomkeer (T) spelen een belangrijke rol in de beschrijving van interacties op sub-atomaire schaal. Behoud van pariteit betekent dat gebeurtenissen in een systeem en hetzelfde systeem, maar gespiegeld, op precies dezelfde manier verlopen als in het spiegelbeeld van elkaar. Pariteits-schending of -breking betekent dat de afloop van gebeurtenissen in een spiegelbeeldige wereld niet meer voorspeld kan worden door in een spiegel te kijken. Ladingsconjugatie houdt hetzelfde in als geldt voor pariteit maar nu vergelijkt men een wereld bestaande uit materie en antimaterie. Ten slotte verstaat men onder invariantie bij tijdsomkeer, dat de natuurwetten niet afhankelijk zijn van de richting waarin de tijd verloopt.

Het kwam als een grote verrassing in 1957 dat in het radioactieve verval van  $^{60}\text{Co}$  zowel pariteit (P) als ladingsconjugatie (C) volledige geschonden bleken te zijn. Dit is een gevolg van de zwakke wisselwerking, die dit radioactieve verval mogelijk maakt. De gecombineerde symmetrie (CP) leek wel behouden te zijn. In 1967 bleek dit echter toch niet het geval te zijn, zoals in het verval van neutrale kaonen (elementaire deeltjes) kon worden vastgesteld. Er zijn tot nu toe geen experimentele aanwijzingen dat CPT, de combinatie van C, P en T, geschonden wordt, wat impliceert dat de CP en T symmetrieën equivalent zijn.

De schending van CP symmetrie speelt een essentiële rol in de verklaring waarom uit de oerknal wel materie is overgebleven en geen anti-materie. De mate van CP schending, zoals onder andere gevonden in het kaon verval, is echter niet voldoende om de materie/antimaterie asymmetrie in ons heelal te verklaren, en er moeten dus nieuwe, nu nog onbekende, vormen van CP schending zijn. De zoektocht naar meetbare permanente elektrische dipoolmomenten (EDM) van fundamentele deeltjes is een veelbelovende manier om deze nieuwe fysica te vinden. De reden is dat een EDM zowel de pariteits- als de tijdsomkeer-symmetrie schendt, en zelfs met de meeste nieuwe en nauwkeurige technieken alleen meetbaar is als er nieuwe vormen van CP schending zijn.

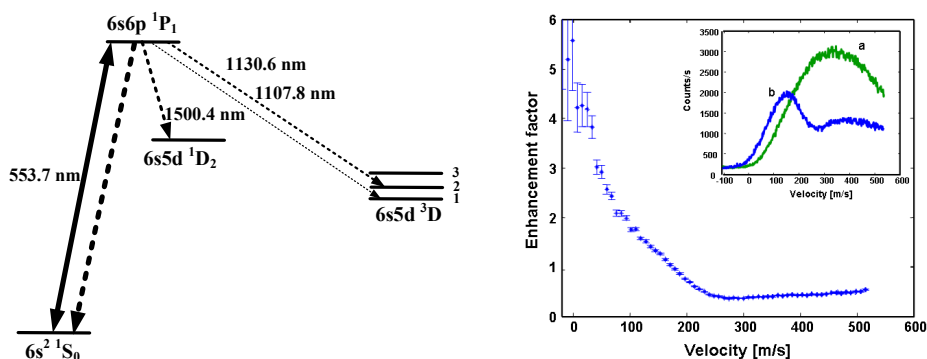


**Figure C.4:** Links: niveau schema van het barium atoom. De (vacuüm) golflengten, zoals bepaald door middel van laser spectroscopie aan de metastabiele D-toestanden, zijn aangegeven in nanometers. De overige overgangen gebruikt voor het laserkoelingsexperiment zijn eveneens gegeven. Rechts: relatieve toename van de flux van langzame atomen. Inzet (a): snelheidsverdeling van de thermische barium bundel. (b) de snelheidsverdeling na laserkoeling.

Het radium atoom (Ra), een radioactief aardalkali element, blijkt een veelbelovende kandidaat te zijn. Als er nieuwe vormen van T of CP schending zijn, zullen die in het Ra atoom (kern en elektronen) en in de Ra kern versterkt worden waardoor de gevoeligheid voor deze schendingen vele malen groter wordt. De TRI $\mu$ P-faciliteit (Trapped Radioactive Isotopes:  $\mu$ icrolaboratories for fundamental Physics) wordt op dit moment in gereedheid gebracht om deze (en andere) radioactieve isotopen te produceren, en te gebruiken in precisie experimenten. De faciliteit bestaat uit een productiestation waar met behulp van deeltjes uit de versneller AGOR van het KVI radioactieve kernen kunnen worden gemaakt en een aantal trappen om de gewenste isotopen te isoleren, af te remmen, en te vangen in een atoomval.

In het EDM experiment zal gebruik gemaakt worden van een aantal concepten uit de atoomfysica, zoals het koelen en invangen van atomen met laserlicht. Dit levert atomen die nagenoeg stilstaan. Ter voorbereiding op het EDM experiment met radium atomen moeten de laser koel- en vangtechnieken voor zware aardalkali elementen aangepast en ontwikkeld worden. In eerste instantie wordt dit gedaan door stabiel barium te gebruiken, dat een atomaire structuur heeft vergelijkbaar met die van radium. Laserkoeling van barium is het onderwerp van dit proefschrift.

Laserkoeling en vangst vereisen een gesloten atomaire overgangscyclus, waarbij voortdurend een atoom met laserlicht wordt aangeslagen om vervolgens weer

naar dezelfde toestand te vervallen waaruit het aangeslagen werd. De atomen die tot nu toe gevangen konden worden in een dergelijke lichtval hebben allen een nagenoeg gesloten cyclus, met verwaarloosbare lekken naar andere toestanden. Voor barium atomen zal echter gemiddeld 1 op de 330 cycli een verval plaatsvinden naar een metastabiele toestand (D-toestanden). Het atoom keert dus niet meer terug naar het uitgangspunt, waardoor het niet verder gekoeld kan worden. Hierdoor zal de snelheid van de barium atomen met niet meer dan 2 m/s verminderd worden. Dit lek vormt dus een ernstige complicatie. Voor verdere koeling zullen de atomen uit de D-toestanden teruggebracht moeten worden in de koel-cyclus met daarvoor geschikte "terugpomplasers".

Een eerste vereiste hierbij is de precieze bepaling van de overgangen naar de metastabiele D-toestanden in barium door laser spectroscopie. De golflengten voor de overgangen zijn bepaald op respectievelijk 1500.4 nm, 1130.6 nm en 1107.8 nm. Deze meting is gebruikt om de isotoopverschuiving van deze toestanden te bepalen (zie figuur C.4).

Wij zijn er als eerste in geslaagd een atomaire bariumbundel te vertragen. De gemeten snelheidsverdeling van een thermische bundel is te zien in fig. C.4. De toename van langzame atomen is het resultaat van laserkoeling. De snelheid van deze atomen is met meer dan 50 m/s afgenomen. Voor snelheden beneden 30 m/s is de toename in bundel flux nagenoeg een factor 5. De flux van atomen met een snelheid beneden 10 m/s moet nog verder verbeterd worden. De volgende stap is het invangen van de vertraagde bundel in een magneto-optische val.

Het werk beschreven in dit proefschrift is de eerste stap op weg naar een nieuw type EDM experiment, waarin radium gebruikt zal worden vanwege zijn intrinsieke gevoeligheid. Hierbij is een volledig nieuw laserlaboratorium opgezet en ingericht. Het eerste belangrijke resultaat is een werkend koelingsschema voor zware aardalkali atomen gedemonstreerd met barium. De spectroscopische technieken die hiervoor ontwikkeld zijn zullen ook gebruikt worden om meer over de atomaire structuur van radium te weten te komen.



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