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Atom Trap Trace Analysis of Calcium Isotopes

Hoekstra, Steven

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Document Version

Publisher's PDF, also known as Version of record

Publication date:

2005

[Link to publication in University of Groningen/UMCG research database](#)

Citation for published version (APA):

Hoekstra, S. (2005). *Atom Trap Trace Analysis of Calcium Isotopes*. s.n.

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Chapter 6

Summary and outlook

In this thesis research has been presented on the development of a novel detection method for trace elements. The analysis of trace elements is an important tool in a wide range of research fields. Rare-isotope detection techniques have been essential to many applications, such as the well-known carbon-dating method. Over the years the promise of a rare calcium isotope, ^{41}Ca , has been recognized by many authors.

Two unique properties make ^{41}Ca suitable for a range of possible applications. The first property is the low abundance of 10^{-14} with respect to ^{40}Ca , which makes up 97 % of all calcium occurring in nature. This low abundance makes ^{41}Ca the most attractive calcium isotope to be used as a tracer in biomedical research. For example, research of calcium kinetics in human bone could greatly benefit from an efficient detection technique of ^{41}Ca . The second property is its lifetime: a ^{41}Ca atom has a half-life of 100,000 years. It is this long lifetime that makes ^{41}Ca a candidate for the development of radiocalcium dating. This technique might be applied as an independent dating of ancient sea-bottom sediments, which contains the record for the history of ice-ages on earth. However, the very low natural abundance of 10^{-14} is an enormous experimental challenge. As a consequence the few existing methods are not very practical.

We use the experimental method called Atom Trap Trace Analysis (ATTA). It is based on the interaction of laser light with the atoms. Generally, when laser light is tuned to the resonance frequency of an atom, momentum can be transferred from the laser to the atom. Using the resulting laser force the position and velocity of the atoms can be manipulated. Small differences in the electronic structure of the calcium isotopes can then be used to distinguish between the calcium isotopes. However, in a calcium vapor at room temperature these small differences are completely washed out by the Doppler effect: the resonance transition is broadened due to the velocity distribution of the atoms. Before any selection between the isotopes be attempted it is therefore essential to reduce the velocity spread of the atoms.

The use of the laser light is thus first to reduce the velocity of the atoms so that a distinction can be made between the different isotopes. Once the velocity spread is reduced sufficiently the wavelength of the laser light can be tuned to select the isotope

of interest. The force on each of the isotopes depends on the scattering rate of the laser light, and for each of the isotopes the maximum of the scattering rate is at a slightly different frequency. This selection mechanism can be used to assure that only the isotope of interest can arrive in the magneto-optical trap (MOT). This is the apparatus in which the detection of the isotopes takes place.

Trapping of the atoms in the MOT is achieved by a combination of 6 laser beams and a quadrupole magnetic field. The atoms arriving at the trap center are collected and trapped in a volume of $\sim 1 \text{ mm}^3$. The average velocity of the atoms in the trap is quickly reduced to a few m/s and the corresponding temperature of these trapped atoms is therefore only a few mK above absolute zero. During the time that an atom is in the trap (about 1/50th of a second) it scatters laser light, which can be measured by sensitive photon-detectors. The amount of light detected is thus a measure for the amount of atoms in the trap. By changing the laser frequency the different isotopes can be trapped and their abundance measured.

In the following we summarize the achievements in realizing the key experimental goals for the Alcatraz experiment, namely:

1. To evaporate, cool, select and trap calcium isotopes.
2. To reach the **sensitivity** to detect a single atom arriving in the magneto-optical trap, such that the abundance of ^{41}Ca can be determined by counting the number of atoms.
3. To reach an **isotope selectivity** of $\geq 10^{14}$ such that the interfering background signal of ^{40}Ca no longer limits the detection sensitivity of ^{41}Ca . Isotope selectivity is a measure of how much an isotope selection process favors isotope A over isotope B. Since we are interested in an isotope with a natural abundance of only 10^{-14} we need to construct a selection process that favors ^{41}Ca over ^{40}Ca by at least fourteen orders of magnitude.
4. To reach an **efficiency** in the transfer of atoms from the oven to the trap such that a measurement of ^{41}Ca atoms at a low abundance level can be done in a reasonable time. The efficiency is defined as the ratio of the number of ^{41}Ca atoms that are detected to the total number of ^{41}Ca atoms that leave the oven.

With these requirements in mind we designed and constructed the experimental setup. The experimental procedure can be summarized as follows. A calcium sample is evaporated from an oven. The resulting atomic beam is slowed down by a counter-propagating laser-beam in a Zeeman slower. The divergence of the atomic beam can be reduced by compression with optical molasses directly after the atoms leave the oven. After leaving the Zeeman slower the resulting slow atoms are deflected by laser beams in the direction of the Magneto-Optical Trap (MOT), where they are captured and detected by their fluorescence.

We have demonstrated in this thesis the evaporation, compression, slowing and deflection of a beam of calcium atoms. We have been able to load the MOT with this atom

beam, trap the atoms and detect their fluorescence. This was done with all stable calcium isotopes.

We succeeded in the detection of single calcium atoms in the magneto-optical trap. Especially challenging for these measurements is the short average trapping time of calcium atoms. Normally a calcium atom is trapped for about 20 ms due to small leak in the excited state of the trapping transition. We have been able to extend the average trapping time for the even isotopes to 275 ± 2 ms using a second laser which effectively closes this leak. For the odd isotope ^{43}Ca the trapping time could only be increased to 110 ± 2 ms because of the hyperfine structure induced by the nuclear spin of the odd calcium isotopes. Nevertheless the demonstration of a trapping time over 100 ms is an important result, since it opens up the possibility to use a separate loading and detection phase in the MOT. The use of two separate phases is expected to increase the loading rate and the detection sensitivity by at least an order of magnitude.

The isotope selectivity and the efficiency of the various parts of the experiment have been investigated by means of computer simulations. We found it to be essential for these computer simulations to take into account the random nature of the absorption and emission processes, and therefore we developed a Monte Carlo simulation program.

First we will discuss the isotope selectivity, then the efficiency. We have reached an isotope selectivity such that the contribution of ^{40}Ca to the background signal, when measuring any of the other calcium isotopes, is less than the fluorescence of a single trapped atom, and thereby is not the limiting factor of the sensitivity. We have estimated the number of atoms that leave the oven at $5 \cdot 10^{13}$ atoms per second. Combining these numbers we can thus say that the total present experimental isotope selectivity is $> 5 \cdot 10^{13}$. Compared to existing experimental approaches the essential improvement that enabled us to reach this isotope selectivity was the deflection stage. We have developed a method to measure the isotope selectivity of the deflection stage and found a selectivity of at least $5 \cdot 10^4$.

The measured isotope selectivity of the different parts of the experiment is compared with estimates from the simulation in table 6.1. We conclude from the simulations that with the present laser power that is available the estimated isotope selectivity can be sufficient for the detection of ^{41}Ca at the natural abundance level. If more laser power would be available the isotope selectivity can be increased further by adding laser compression stages directly before the entrance to the Zeeman slower and at the deflection point. The total power requirement for these two compression stages is roughly 30 mW. We have done a measurement of the isotope selectivity of a 2D compression stage which gave good qualitative and quantitative agreement with the simulations.

The total efficiency was experimentally determined from the loading rate of the MOT. We have been able to measure a loading rate of the MOT of $1.1 \cdot 10^8$ ^{40}Ca atoms per second. For the number of atoms leaving the oven per second we have to rely on an estimation of $5 \cdot 10^{13}$ atoms per second. Combining these numbers we find a total efficiency of $2.2 \cdot 10^{-6}$. The partial efficiency of the different components of the experiment could experimentally not be measured. In table 6.2 the total efficiency of the present experiment is compared with the estimate from the simulation. Added here is an estimated value of the efficiency that becomes possible if extra laser power would be available to

Part of the experiment	Experimentally demonstrated		Estimate from simulation	
	present	possible	present	possible
Compression@oven	/	$2 \cdot 10^4$	/	$2 \cdot 10^4$
Zeemanslower	-	-	50	50
Deflection	$> 5 \cdot 10^4$	$> 5 \cdot 10^4$	10^5	10^5
Compression@deflection	/	-	/	10
Trapping	-	-	$2.5 \cdot 10^7$	$2.5 \cdot 10^7$
Total Isotope selectivity:	$> 5 \cdot 10^{13}$	-	$1.2 \cdot 10^{14}$	$2.5 \cdot 10^{19}$

Table 6.1: A comparison of the experimental results on the isotope selectivity of the experiment with estimates from the simulations. Also indicated is the efficiency that becomes possible by adding two compression stages to the experiment. ‘/’ indicates that this component was not implemented, ‘-’ indicates that this value could not be measured separately

Part of the experiment	Experimentally demonstrated		Estimate from simulation	
	present	possible	present	possible
Total Efficiency:	$2 \cdot 10^{-6}$		$1 \cdot 10^{-6}$	$3 \cdot 10^{-2}$

Table 6.2: A comparison of the experimental results on the efficiency of the experiment with estimates from the simulations. Also given is the efficiency that becomes possible by adding more laser power for the compression of the atomic beam and a separate loading and detection phase

add the two above-mentioned compression stages to the experiment. Also the expected improvement of the efficiency due to the use of a separate loading and detection phase is taken into account.

The importance of the addition of laser power to be able to implement the compression stages is clear. Therefore we have performed test measurements on the efficiency of a compression stage. We found good qualitative and quantitative agreement between the experiments and simulations, strengthening us in the opinion that it is indeed possible to greatly improve the efficiency by adding a second laser system with an intensity of at least 35 mW.

Coming back to the four key experimental goals of the Alcatraz experiment we can conclude that we have achieved the first three goals, thereby significantly improving the performance and understanding of the ATTA method. Concerning the fourth goal, the efficiency we have achieved in the experiment is sufficient to start working on samples enriched in ^{41}Ca . The sensitivity we have reached with ATTA is presently similar to that of resonance ionization mass spectrometry (RIMS). These two methods can become very important for biomedical applications of ^{41}Ca , where enriched samples are used. The ease of use and small size offered by ATTA are important issues for use in a clinical environment.

A measurement of the natural abundance of ^{41}Ca would still take too long: but the experiments and simulations show that by making more laser power available to the experiment the efficiency can be increased. At the expected efficiency of $3 \cdot 10^{-2}$ we could count 36 atoms per hour from sample containing ^{41}Ca at a 10^{-14} -level. Such a

performance would make ATTA the most sensitive ^{41}Ca detection method available.

Besides the improvement of the efficiency one of the steps that has to be taken to get there is to calibrate the experiment. A series of calibrated samples in the range of 10^{-6} to 10^{-13} exists [116] which can be used to calibrate the experimental setup. The calcium in these samples is in the form of CaCO_3 . A modified oven design, allowing for higher temperatures of ~ 1000 K which are necessary to dissociate these CaCO_3 molecules, is needed to produce a beam of calcium atoms. Such a type of oven has already been shown to work. Special care should be taken in the design of this oven to make sure that the divergence of the atomic beam is kept as small as possible, since we know that this is the largest loss factor in the experiment. Handling of CaCO_3 is also essential for potential future biomedical applications such as Osteoporosis monitoring or pharmaceutical studies on medications, to compensate a surplus loss of bone calcium.

