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Published in:
The Journal of Chemical Physics

DOI:
[10.1063/1.456151](https://doi.org/10.1063/1.456151)

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Document Version
Publisher's PDF, also known as Version of record

Publication date:
1989

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

Citation for published version (APA):

Meijer, H. A. J., Zeegers, T., Pelgrim, T. J. C., Heideman, H. G. M., & Morgenstern, R. (1989). The use of weak magnetic fields in collisions of polarized atoms. *The Journal of Chemical Physics*, 90(2), 729-737.
<https://doi.org/10.1063/1.456151>

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The use of weak magnetic fields in collisions of polarized atoms

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(Received 14 July 1988; accepted 28 September 1988)

We have experimentally and theoretically investigated the behavior of excited Na atoms under the combined influence of resonant laser light and a weak magnetic field. The use of weak magnetic fields gives another possibility to vary the polarization of the atoms, additional to variation of the polarization of the exciting laser. We calculated in detail the influence of weak magnetic fields on the density matrix describing the excited atoms, using generalized rate equations. We checked the results of our model experimentally, at the $\text{Na}(3^2S_{1/2}, F_l = 2) \rightarrow (3^2P_{3/2}, F_u = 3)$ transition, excited with circularly polarized light. We determined the polarization of the excited atoms, by measuring the fluorescence light at various angles, as a function of both magnetic field strength and laser irradiance. Model and experiment agreed very well. We applied our model to the associative ionization of the excited Na atoms. We measured the ion signal as a function of magnetic field. Analyzing the results, we could determine a specific associative ionization cross section, which is not accessible in our experimental geometry without the use of magnetic fields. Thereby we showed that the application of magnetic fields in polarization dependence studies can provide us with additional information about the process.

I. INTRODUCTION

In collision experiments with excited atoms it is possible to prepare the collision partners very accurately regarding their polarization. Especially when the atoms are excited by laser light this can be easily achieved by changing the polarization of the laser. Normally one tries to avoid possible disturbing effects during the excitation process, e.g., caused by magnetic fields, so that one can rely on the relatively simple theoretical calculations of the polarization of the excited atoms. However, one can also deliberately use (weak) magnetic fields in a controlled way. By doing so one can vary the polarization of the excited atom in another way, and might be able to learn more about the mechanism of the particular collision.

We present a model which describes the excitation process of a degenerate two-level system in the presence of a magnetic field. This model is applied to the $F_l = 2 \rightarrow F_u = 3$ transition of the Na D_2 line ($3^2S_{1/2} \rightarrow 3^2P_{3/2}$), excited with left-hand circularly polarized (σ^+) light. To allow a relatively simple description, the magnetic field is taken to be perpendicular to the laser propagation direction. Furthermore the magnetic field is assumed weak (≤ 2 G), so that Zeeman splitting can be neglected.

To our knowledge, up to now only strong magnetic fields (up to 100 kG) have been used in collision experiments.¹ Then, individual Zeeman sublevels of the excited state can be excited separately. Advantage of this method is the rather straightforward way in which the population of the excited state can be described and the polarization can be varied (although some problems with the hyperfine splitting might complicate the situation). Disadvantages are, however, the loss of signal because only one sublevel of the

ground state can be excited, and the fact that the atoms will be lost for excitation after a few cycles, because they decay to the wrong sublevel of the ground state. Finally, even the shift in energy of the excited state, although relatively small (1 meV typically at 100 kG), might influence the collision process. Especially the associative ionization process, in which there is only 51 meV energy difference between the $3p-3p$ infinite distance energy and the well depth of the Na_2^+ potential,^{2,3} may be sensitive to this energy shift. In our approach we avoid this difficulties.

II. AIM

An example of what can be achieved with weak magnetic fields can best be illustrated in a classical way. In our experiment, the laser intersects the atomic beam (and thus the axis in the direction of the initial collision velocity) at (nearly) right angles. Without the use of a magnetic field, it is impossible in this arrangement to prepare an oriented excited state (this is a state in which the magnetic substates M and $-M$ are differently populated) with respect to the collision velocity axis. Excitation with linearly polarized (π) light cannot orient an excited state, excitation with circularly polarized (σ^+) light does orient the excited state completely, but only with respect to the laser propagation direction. As can be seen from Fig. 1 excitation with σ^+ light leads to a perfect symmetric projection of the angular momentum $\mathbf{J}$ to the collision velocity axis. If we only apply a magnetic field $\mathbf{B}$, and no exciting field, the $\mathbf{J}$ vector will precess about $\mathbf{B}$ (Fig. 2). If we apply both, the $\mathbf{J}$ vector will on one hand again try to precess about $\mathbf{B}$, but on the other hand, will try to precess about the laser propagation axis. So one would expect that as a result, $\mathbf{J}$ will precess about some axis, at an angle φ with the laser propagation axis (Fig. 3). In that case, the projection on the collision velocity axis will not be

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FIG. 1. Classical picture of the precession of the angular momentum about the laser propagation direction. This J is the angular momentum of an atom excited by left-hand circularly polarized light. With respect to the laser propagation axis the projection of J is constant: M_J . With respect to the relative collision velocity direction the projection varies in a symmetric way between the classical boundaries M^- and M^+ .

symmetric anymore, M^- will be favored over M^+ . The angle φ , and thus also the population ratio M^+/M^- depends in this picture on the relative strength of laser and magnetic field.

Of course detailed calculations must show whether this classical picture holds. However, calculations on the B dependence of a $J_l = 2 \rightarrow J_u = 3$ laser induced transition of neon [$\text{Ne}(2p^5 3s)P_2 \rightarrow (2p^5 3p)D_3$] have already shown that this concept is applicable, and that φ can have values up to 20° .⁴

III. THEORY

We use generalized rate equations to describe the time dependence, and to find the stationary state of the density matrices $\rho_l(t)$ and $\rho_u(t)$ for the ground and excited hyperfine level of the atom, respectively. Such a description by means of rate equations can in principle not be used, because the spectral linewidth of our laser is much smaller than the

FIG. 2. Classical picture of the precession of the angular momentum J about the magnetic field vector B . This J is the angular momentum of an atom in the ground state, since there is no exciting laser field.

FIG. 3. Classical description of the behavior of the angular momentum J , when the exciting laser and the magnetic field are both present. This angular momentum of the excited atom precesses about an axis at an angle φ compared to the laser propagation direction, the magnitude of φ depending on the relative strength of laser and magnetic field. With respect to the relative collision velocity direction the projection of J varies between the classical boundaries M^- and M^+ , but now M^- is larger than M^+ .

width of the atomic absorption. However, Nienhuis⁵ has proven that, when only two (degenerate) levels are coupled by the radiation field, the stationary state achieved using rate equations is correct and that, when the laser irradiance is moderate, also the calculated time dependence is reliable in the sense that it correctly averages over the Rabi oscillations (which oscillations are anyway small when the laser irradiance is far from saturation). Furthermore, as far as the magnetic field influence is concerned, we neglect Zeeman sublevel splitting, which means that this splitting must be small compared to the natural linewidth.

The general evolution equations under these conditions have been described by Nienhuis.⁵ Slijkhuys *et al.*⁶ gave a detailed description of the way in which these equations can be rewritten in order to allow an easy numerical treatment. Their result is

$$\begin{aligned} \frac{d\rho_u}{dt} &= -\frac{i}{\hbar} [H_u^{\text{pho}}, \rho_u] - A_{lu}\rho_u \\ &\quad + 3B_{lu}\rho_v (C_\lambda \rho_l C_\lambda^\dagger - \frac{1}{2} C_\lambda^\dagger C_\lambda \rho_u - \frac{1}{2} \rho_u C_\lambda C_\lambda^\dagger), \\ \frac{d\rho_l}{dt} &= -\frac{i}{\hbar} [H_l^{\text{pho}}, \rho_l] + A_{lu} \sum_\lambda C_\lambda^\dagger \rho_u C_\lambda \\ &\quad + 3B_{lu}\rho_v (C_\lambda^\dagger \rho_u C_\lambda - \frac{1}{2} C_\lambda^\dagger C_\lambda \rho_l - \frac{1}{2} \rho_l C_\lambda^\dagger C_\lambda). \end{aligned} \quad (1)$$

The first terms at the right-hand side of the equations describe the free evolution of the atom in the magnetic field, the second ones loss and gain, respectively, due to spontaneous decay processes, and the third ones loss and gain by absorption and stimulated emission. The operators C_λ are defined by their matrix elements:

$$\langle M_u | C_\lambda | M_l \rangle \equiv \langle F_u M_u | F_l M_l; 1\lambda \rangle \quad (2)$$

with $\lambda = +1$ for excitation with σ^+ light.

In Eq. (1) we need the Hamiltonians H_l^{pho} and H_u^{pho} of the interaction due to the magnetic field with respect to a coordinate frame with the z axis parallel to the laser beam

direction, the so-called σ -photon frame, shown in Figs. 1–3. With respect to the magnetic field ($\mathbf{B}$) direction these Hamiltonians are (for $F_l = 2$ and $F_u = 3$ levels):

$$H_l^B = \mu_B g_F B \begin{pmatrix} 2 & & & \\ & 1 & 0 & \\ & & 0 & -1 \\ & & & -2 \end{pmatrix},$$

$$H_u^B = \mu_B g_F B \begin{pmatrix} 3 & & & \\ & 2 & & 0 \\ & & 1 & \\ & & & 0 & -1 \\ & & & & -2 & -3 \end{pmatrix}, \quad (3)$$

where μ_B is the Bohr magneton and g_F the Landé factor for the specific level.

We can obtain the H_l^{pho} and H_u^{pho} from Eqs. (3) by a rotation defined by

$$H_l^{\text{pho}} = D^2(\text{pho} \rightarrow B) H_l^B D^{2\dagger}(\text{pho} \rightarrow B),$$

$$H_u^{\text{pho}} = D^3(\text{pho} \rightarrow B) H_u^B D^{3\dagger}(\text{pho} \rightarrow B), \quad (4)$$

where “pho $\rightarrow$ B” stands for the appropriate Euler angles $\alpha\beta\gamma$. The rotation operators D can be found in Brink and Satchler.⁷ In our case, with the coordinate frame shown in Fig. 3 and using σ^+ light, $(\alpha\beta\gamma) = (0^\circ, -90^\circ, 0^\circ)$, and D reduces to $d(-90^\circ)$. Finally A_{lu} and B_{lu} are the Einstein coefficients and ρ_ν is the spectral energy density of the laser⁸:

$$\rho_\nu = \frac{4}{cA_{lu}} E \quad (5)$$

with E the laser irradiance (dimension power/area).

IV. NUMERICAL CALCULATIONS

Equations (1) were derived explicitly for the $F_l = 2 \rightarrow F_u = 3$ hyperfine component of the $\text{Na}(3^2S_{1/2} \rightarrow 3^2P_{3/2})$ transition we are interested in. Then we solved the equations numerically for a few laser irradiances and a series of magnetic fields ranging from 0 to 2 G. We calculated the time behavior of the two density matrices up to 8 μs , the average passage time through the spot in our experiment. For the irradiance we normally use in our collision experiments (8 mW/cm^2) it appeared that indeed a stationary state was reached within 1 μs . This stationary state can in principle be characterized by the concept of the angular momentum precessing around an axis at an angle φ with respect to the laser propagation direction (Sec. II). However, this characterization cannot be done exactly. Therefore we will not use a characterization by means of φ . Instead we characterize the atom by the two density matrices describing the lower and the upper level. These matrices are averages of eight intermediate ones, calculated at time intervals of 1 μs .

FIG. 4. (A) Schematic overview of the apparatus. Two counterrunning thermal beams of Na atoms are intersected at nearly right angles (87°) by laser light from a cw-dye laser, tuned to the $F_l = 2 \rightarrow F_u = 3$ component of the $\text{Na}(3s)^2S_{1/2} \rightarrow \text{Na}(3p)^2P_{3/2}$ transition. The laser beam is reflected into its own path. In this arrangement, atoms from beam 2 are only excited by the direct laser beam, atoms from beam 1 only by the reflected laser beam. Furthermore, the excitation process becomes velocity selective, which enables us to control the collision velocity. All ions created in the interaction region are extracted by a weak electric field, and are counted by a high current channeltron. The fluorescence light is monitored by five photodiodes, at various angles. (B) The position of the three photodiodes used in this experiment with respect to the σ photon frame.

V. COMPARISON WITH EXPERIMENTAL FLUORESCENCE MEASUREMENTS

Before we use the calculated density matrices for an analysis of collision experiments we want to check the validity and accuracy of the description presented in the previous section. Therefore we used the experimental setup, shown in Fig. 4(A). It has been described in Refs. 9, 10, and 11 and in much detail in Ref. 8. We measured, at three different angles, the fluorescence from the $\text{Na } 3^2P_{3/2}$, ($F_u = 3$) excited state, pumped with σ^+ light, as a function of $\mathbf{B}$ for several irradiances, and compared these results with theory. The positions of the three photodiodes, important in this experiment, are shown in Fig. 4(B) in the σ photon frame. The magnetic field is in the x direction. We calibrated our magnetic field with a Gaussmeter. We determined “zero magnetic field” to less than 5 mG by using the $\text{Na } 3^2S_{1/2} \rightarrow 3^2P_{1/2}$, $F_l = 2 \rightarrow F_u = 1$ transition, which is very sensitive to magnetic fields (see the Appendix). The comparison between theory and experiment is shown in Fig. 5 for irradiances 8, 3, and 0.8 mW/cm^2 . Since the relative sensitivity of the photodiodes

FIG. 5. Fluorescence monitored by three different photodiodes at three different angles, as a function of the magnetic field. The full curves are the theoretical predictions, in which we have to use the experimentally determined irradiance E of the laser. In (A) this was 8 mW/cm^2 , in (B) 3 mW/cm^2 , and in (C) 0.8 mW/cm^2 . With a lower laser irradiance, already weaker magnetic fields have an important influence on the fluorescence, and thus on the excited state population. Agreement between theory and experiment is rather good, the largest uncertainty being the error in the determination of the laser irradiance ($\pm 20\%$). Therefore, in (A) theoretical calculations using $E = 10 \text{ mW/cm}^2$ are also shown (dashed curves). Then agreement is even better.

was not known, the experimental points at zero magnetic field are normalized to the theoretical curves. Overall agreement is good, but the experimental curves show a somewhat weaker dependence on B than the theoretical ones. This can be due to the fact that the experimentally determined laser

irradiance is somewhat too small. In Fig. 5(A) also the theoretical results with $E = 10 \text{ mW/cm}^2$ are shown, with better agreement.

Photodiode 2, at 55° off the z axis in the x - z plane, is at the magic angle where the emitted light intensity is propor-

tional to the total emitted intensity (independent of the polarization of the excited state). Therefore in the absence of a magnetic field its signal is a direct measure for the excited Na density. Theory shows that even with a magnetic field present, this is still the case within 3% up to 1500 mG. Thus the plots in Fig. 5 clearly show that the density of excited atoms decreases considerably when a magnetic field is applied. Even a magnetic field of 500 mG ($\approx B_{\text{earth}}$) causes a significant reduction of excited atom density. This effect is caused by the fact that in the $F_l = 2 \rightarrow F_u = 3$ transition pumped with σ^+ light the $M_l = 2 \rightarrow M_u = 3$ has the largest partial Einstein coefficient ($b_{23} = 3 B_{lu}$, compared with $b_{12} = 2 B_{lu}$, $b_{01} = 1.2 B_{lu}$, $b_{-10} = 0.6 B_{lu}$, and $b_{-2-1} = 0.2 B_{lu}$). In the absence of a magnetic field this transition is pumped exclusively in stationary state, while with a magnetic field due to the Larmor precession this transition is reduced in favor of the other ones with a lower b value.

In view of the good agreement between calculations and experiments, we expect that using the magnetic field dependence as an additional tool in polarization dependence studies can provide us with additional information about the processes.

VI. DEPENDENCE OF ASSOCIATIVE IONIZATION ON MAGNETIC FIELD

We performed measurements on the associative ionization process:

at different magnetic fields in our counterpropagating beam arrangement with the laser beam intersecting the Na beams at 87° . We compared the magnetic field dependence of collisions where both atoms were excited with left-hand circularly polarized (σ^+) light (with the ion signal R^{++}), to the case in which one atom was σ^+ excited and the other σ^- excited (ion signal R^{+-}). We measured both cases in one measuring series, by exciting one atomic beam with σ^+ light, and the other one with the reflected laser beam, which either stayed σ^+ polarized (with respect to the same coordinate frame) or was changed to σ^- by putting a $\lambda/4$ plate into the laser beam (for a detailed description see Refs. 8 and 11). We monitored the excited state density by means of the signal of the photodiode at approximately the magic angle (and corrected this value for the theoretically expected deviation at higher magnetic fields). This procedure was repeated at various collision velocities. The results for one collision velocity are shown in Fig. 6. Shown is the normalized ion signal, i.e., the ion signal corrected for the (magnetic field dependent) density of excited atoms. There appears to be a very pronounced dependence of the R^{++} ion signal on magnetic field, while hardly anything happens in the R^{+-} case.

VII. COMPARISON WITH THEORY

We have to describe the density matrix ρ on an $|LM_L\rangle$ basis with the z axis in the direction of the relative collision velocity ("collision frame"), in order to be able to compare our results with theoretical potential curve calculations. Applying the theory of Nienhuis,¹² we expand the density matrix ρ into spherical tensors T_{kq} , defined with respect to the collision frame:

FIG. 6. Example of the dependence of the associative ionization on magnetic field. Shown is the normalized ion signal, that is, the ion signal corrected for the (magnetic field dependent) density of excited atoms as a function of the magnetic field. The R^{++} signal is very dependent on B , whereas the R^{+-} signal hardly changes.

$$\rho = \sum_{kq} a_{kq} T_{kq}^\dagger \quad (7)$$

and

$$a_{kq} = \text{Tr } \rho T_{kq} = \sum_i \sum_j \rho_{ij}^{\text{coll}} (T_{kq})_{ji}. \quad (8)$$

In order to calculate the a_{kq} we need to know the density matrix ρ^{coll} , with respect to the collision frame, whereas the numerical calculations (Secs. III and IV) give us ρ^{pho} , with respect to the photon frame. The relation between the matrices ρ^{coll} and ρ^{pho} is

$$\rho_{MM'}^{\text{coll}} = \sum_{\mu\mu'} D_{M\mu}^1(\alpha\beta\gamma) \rho_{\mu\mu'}^{\text{pho}} (D_{\mu'M'}^1(\alpha\beta\gamma))^\dagger \quad (9)$$

with $(\alpha\beta\gamma)$ the Euler angles for the rotation from the collision frame to the photon frame. If we take into account that the laser intersects the atomic beams at 87° (which is important here, because it acts as a positive offset for the angle φ , see Sec. II), we get $(\alpha, \beta, \gamma) = (-90^\circ, 93^\circ, 90^\circ)$. Then the D matrices can be found in Ref. 7.

In our experiment we compare two cases: (i) both atoms excited with σ^+ light, with density matrices $\rho^{(++)}$ and expansion coefficients $a_{kq}^{(++)}$ and (ii) one atom excited with σ^+ light, the other with σ^- light ($\rho^{(+-)}$ and $a_{kq}^{(+-)}$).

The relation between $\rho^{(+)}$ and $\rho^{(-)}$ is simply

$$\rho_{mn}^{(+)} = \rho_{-m-n}^{(-)} \quad (10)$$

So

$$a_{kq}^{(+)} = \text{Tr } \rho^{(+)} T_{kq} = \sum_i \sum_j \rho_{ij}^{(+)} (T_{kq})_{ji}, \quad (11)$$

$$\begin{aligned} a_{kq}^{(-)} &= \text{Tr } \rho^{(-)} T_{kq} \\ &= \sum_i \sum_j \rho_{ij}^{(-)} (T_{kq})_{ji} = \sum_i \sum_j \rho_{-i-j}^{(+)} (T_{kq})_{ji}. \end{aligned} \quad (12)$$

With

$$(T_{kq})_{ji} = (-)^k (T_{k-q})_{-j-i} \quad (13)$$

this leads to

$$a_{kq}^{(-)} = (-)^k \sum_i \sum_j \rho_{ij}^{(+)} (T_{k-q})_{ji} = (-)^k a_{k-q}^{(+)} \quad (14)$$

Furthermore, we can use the fact that the terms of ρ_{ij} are either real ($i+j$ even) or purely imaginary ($i+j$ odd):

$$\rho_{ij} = \rho_{ji}^* = (-)^{i+j} \rho_{ji} \quad (15)$$

and use

$$(T_{kq})_{ij} = (-)^q (T_{k-q})_{ji}. \quad (16)$$

Then we get

$$a_{k-q}^{(\pm)} = \sum_i \sum_j (-)^{i+j} \rho_{ji}^{(\pm)} (-)^q (T_{kq})_{ij}. \quad (17)$$

A spherical tensor has only nonzero elements at positions (i, j) with $(i+j)$ odd, when q is odd, or with $(i+j)$ even, when q is even. So we get

$$a_{k-q}^{(+)} = a_{kq}^{(+)}, \quad a_{k-q}^{(-)} = a_{kq}^{(-)} \quad (18)$$

and, with Eq. (10),

$$a_{kq}^{(-)} = (-)^k a_{kq}^{(+)}. \quad (19)$$

The ion production rate of the associative ionization reaction can be written¹²:

$$R = \text{Tr}_A \text{Tr}_B G \rho_A \rho_B \quad (20)$$

with ρ_A and ρ_B the density matrices of the two colliding excited Na atoms, and G the detection operator. This operator contains all information that can be extracted out of a measurement with this specific geometry. For each basis of M states, the diagonal elements of G are connected to cross sections $\sigma(M_A, M_B)$, whereas the off-diagonal ones represent coherence terms. However, there is no principal difference between the diagonal and off-diagonal elements, and they can be transferred into each other by selection of another basis of M states.

The detection operator G can also be expanded in spherical tensors:

$$G = \sum_{kqk'q'} g(kq; k'q') T_{kq} T_{k'q'}. \quad (21)$$

Those components of g , for which $q = q'$, can be expressed in terms of cross sections $\sigma(M_A M_B)$, whereas the other ones represent the off-diagonal or coherence terms.

The ionization signal R is now given by

$$R = \sum_{kq} \sum_{k'q'} g(kq; k'q') a_{kq} b_{k'q'}, \quad (22)$$

where the $b_{k'q'}$ are the expansion coefficients (8) for collision partner B. Since we use the L picture for our description (which means that we neglect possible electron spin effects on the associative ionization reaction), k and k' run from 0 to 2.

Using Eqs. (22) and (19), we find for the difference between the R^{++} and the R^{+-} signal

$$\begin{aligned} \Delta R = R^{++} - R^{+-} &= \sum_{kq} \sum_{k'q'} g(kq; k'q') a_{kq}^{(+)} a_{k'q'}^{(+)} \\ &\quad - \sum_{kq} \sum_{k'q'} g(kq; k'q') a_{kq}^{(+)} (-)^{k'} a_{k'q'}^{(+)}. \end{aligned} \quad (23)$$

Only the $g(kq; k'q')$ with $k' = 1$ contribute:

$$\Delta R = 2a_{10}^{+2} \cdot q + 4a_{11}^{+2} \cdot r \quad (24)$$

with $q = g(10; 10)$ and $r = g(11; 1-1)$.

The coefficients a_{10} and a_{11} are dependent on the magnetic field, and their value and dependence can be extracted from the numerically calculated density matrices, by means of Eq. (8). This allows us to fit the experimental results to the expression

$$\Delta R = q \cdot \alpha(B) + r \cdot \beta(B) \quad (25)$$

with α and β known functions, shown in Fig. 7, and q and r the fit parameters. While the coherence term r is already known,¹⁰ the possibility of fitting q provides us with valuable additional information, since

FIG. 7. The theoretical magnetic field dependence of the coefficients of q (α) and r (β). By means of these two theoretical curves, we can fit Eq. (25) to the data, and thus get values for q and r .

FIG. 8. Three examples of a fit of Eq. (25) to the experimental data. As explained in the text, discrepancies between experiment and theory between 0 and 1000 mG are understandable at low (A) and high (C) collision velocities, since there a deviation from stationary state conditions exists. In case (B), at intermediate collision velocity, the fit quality is good throughout the plot, as it should, since in this case stationary state situations exist. The difference between the fit curves (full) and the other curves (dashed) is explained in the text.

$$q = \sigma_{11} - \sigma_{1-1}, \quad (26)$$

where $\sigma_{1\pm 1}$ is the associative ionization cross section for a collision between two excited atoms of which one is in the $M_L = +1$ sublevel, and the other in the $M_L = \pm 1$ sublevel. This is the information we were looking for (see Sec. II).

Three examples of a fit of Eq. (25) to the experimental points, at various collision velocities, are shown in Fig. 8. In the fit procedure the point at $B = 0$, and the points beyond $B = 1000$ mG, are considered to be the more reliable ones. In the intermediate range the theoretical results are strongly dependent on laser irradiance, and on small deviations from stationary state (as they occur systematically in our experiment as a function of the selected velocity^{8,11}). Therefore discrepancy between theory and experiment in this range is quite understandable. A clear indication that small deviations from stationary state indeed lead to discrepancies in this region, is the fact that the fit at 1590 m/s [Fig. 8(B)], where stationary state conditions exist, is good throughout this region. Furthermore, the fluorescence measurements, which accompanied the ionization measurements, did also show discrepancies in the intermediate range. This in contrast to the measurements presented in Fig. 5, which were performed with the laser intersecting the atomic beam perpendicularly, thus under stationary state conditions. However, because agreement between theory and experiment at magnetic fields above 1000 mG is again rather good, we can certainly trust that region.

The value of r can be obtained as accurately as the data point at $B = 0$, because at zero magnetic field $\alpha(B) = 0$ and $\beta(B) = -1$. The full curves in the graphs are the best fits. In the dashed curves we use the same r value as in the full ones, but we make $q = 0$. Comparison of the two curves shows that q significantly contributes to the signals.

Figure 9 shows the results for r and q , at the same y scale as used in Refs. 8–11.

VIII. DISCUSSION AND CONCLUSIONS

The values obtained for r agree very well with those presented before.¹⁰ This is to be expected, because the measurements presented there were of the same type as the ones in this paper with $B = 0$.

The absolute value of q is very large compared to the cross sections in Ref. 9. As q is the difference between σ_{11} and σ_{1-1} , and in Ref. 9 $\bar{\sigma}_{11} = (\sigma_{11} + \sigma_{1-1})/2$ is presented, one could calculate these two cross sections separately. One can see immediately that this would lead to a negative value for σ_{11} , especially at lower collision velocity. This could indicate that the L picture is not adequate for the description of this process. Comparison of the fine structure precession time (2×10^{-13} s) to the collision time ($\approx 5 \times 10^{-13}$ s at $v_c = 1500$ m/s) shows that it would by no means be surprising if the L picture were wrong. However, we postpone the discussion until we give a full analysis of the collision process in cross sections and coherence contributions in the L picture.¹¹ The determination of q , as presented here, is the necessary final addition to that analysis, in which all eight cross sections and coherence terms playing a role in the collision process at our geometry are determined. Of course, the mag-

FIG. 9. The final results of our magnetic field analysis, the cross section difference $q = \sigma_{11} - \sigma_{1-1}$ and the coherence contribution r . While r can be easily obtained from measurements without a magnetic field, q cannot.

netic field measurements can also be analyzed in the J picture, taking the electron spin into account. An analysis similar to that presented in Sec. VII shows, however, that in the J picture the difference between R^{++} and R^{+-} depends on six parameters. Clearly, agreement between theory and experiment is not good enough to obtain a reliable six parameter fit.

As a conclusion we can say that analyzing magnetic field dependences is not simple, neither are the experiments themselves. However, they can give valuable additional information by breaking symmetries, for which it otherwise would be necessary to change the experimental geometry, thereby hindering comparison of, e.g., cross sections obtained in different geometries. In our case it would have been necessary to vary the angle between laser and atomic beam, which at the same time would change the size of the interaction region, the density of the excited atoms, their velocity, the relative population of the magnetic sublevels, and, above all, would severely change the influence of the nearby $F_1 = 2 \rightarrow F_u = 2$ transition. Therefore to fit data as a function of that angle in a reliable way would be practically impossible. The application of weak magnetic fields appeared to be an effective alternative.

Finally, we showed with this paper that generally in polarization experiments already the magnetic field of the earth in the collision center can influence the polarization considerably. In how far this magnetic field influences the results is dependent on its direction with respect to the polarization axis of the laser. For instance, in Ref. 12 results for

FIG. 10. The relative populations of the magnetic sublevels of the lower and upper state while pumping the $^2S_{1/2}, F_l = 2 \rightarrow ^2P_{1/2}, F_u = 1$ transition with π light. The first picture shows the initial situation. In the absence of a magnetic field, all atoms end up in the $M_l = \pm 2$ substates. With a magnetic field, however, they will be permanently liberated out of that trap, due to the Larmor precession. This causes the number of emitted photons per atom to increase as a function of the magnetic field.

the polarization dependence of the associative ionization are presented, measured both in Utrecht and Berlin. Whereas in neither of the two experiments the terrestrial magnetic field had been compensated, the $\cos 4\theta$ term in Utrecht was significantly larger than in Berlin. This $\cos 4\theta$ term was mainly

FIG. 11. Experimental result of the dependence of the fluorescence signal on the external magnetic field in a certain direction. From this picture, one can see that an external magnetic field of 552 mG will lower the net magnetic field in that direction to 0 ± 3 mG.

caused by the terrestrial magnetic field (cf. Ref. 9), which had a more unfavorable direction in Utrecht than it had in Berlin. On the other hand, in Utrecht the anisotropy of the fluorescence light was not remarkably lowered, whereas, e.g., in Orsay,¹³ this parameter appeared to depend critically on the (compensation of) the terrestrial magnetic field.

APPENDIX: DETERMINATION OF ZERO MAGNETIC FIELD

The $\text{Na } 3^2S_{1/2}, F_l = 2 \rightarrow 3^2P_{1/2}, F_u = 1$ transition is very sensitive to magnetic fields. As shown by Slijkhuys *et al.*⁶ the fluorescence intensities can be varied by a factor of 5 as a function of the strength of the magnetic field. Qualitatively this can be understood very well (see Fig. 10). In the absence of a magnetic field, all atoms will be trapped in the $M_l = \pm 2$ sublevels after a few excitation/decay cycles, because linearly polarized light only induces $\Delta M = 0$ transitions, whereas spontaneous decay obeys the selection rule $\Delta M = 0, \pm 1$ (excitation with circularly polarized light, with $\Delta M = +1$, would drive all the atoms into $M_l = +1$ and $M_l = +2$). Calculations show that at magnetic field $B = 0$ every atom emits on the average 1.2 photons before it is trapped.

When a magnetic field is present with a component perpendicular to the polarization vector, the trapped atoms will be liberated again because of the Larmor precession. When the magnetic field is effective this can increase the average number of emitted photons to six (not to infinity, since there is also a leak to the other hyperfine ground level).

Using this effect as a means to detect very weak magnetic fields (with a properly chosen laser irradiance, between 1 and 10 mW/cm²) we can reduce the net magnetic field perpendicular to the polarization vector. Figure 11 shows the dependence of the fluorescence signal on the magnetic field. By varying the polarization vector direction, the total magnetic field can be made smaller than 5 mG. A final check is the measurement of the anisotropy of the fluorescence light by rotating the polarization vector. In the absence of a magnetic field any anisotropy must be absent.

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