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Observation of collisional modification of the Zeeman effect in a high-density atomic vapor

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We have studied the Zeeman effect of the resonance transition of atomic potassium vapor in a density range where the collision time is comparable with the time in between collisions. We find that the Zeeman splitting in this density range changes significantly as compared to the value for a free atom. In particular, we observe that the Zeeman effect of the two resonance *D* lines become equal at the highest densities that we have studied. The modification of the Zeeman splitting is attributed to resonant dipole-dipole collisions between ground-state and excited-state atoms. [S1050-2947(97)08907-5]

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I. INTRODUCTION

When a dense atomic vapor interacts with a resonant laser field the interaction between the atoms is dominated by the resonant dipole-dipole interaction between ground-state and excited-state atoms. Such systems have recently received renewed attention, in particular, in connection with their nonlinear optical response. A keypoint in these studies is that the *local* field rather than the external optical driving field governs the atomic response; this gives rise to a variety of nonlinear optical phenomena such as optical bistability [1,2], self-induced transparency [3], piezophotonic switching [4], etc. Local-field effects have also been shown to be important in the context of lasers without inversion [4], a subject that has been studied extensively over the last few years. In all these studies it is usually assumed that the response of the dense vapor to the optical field is well described by a Lorentzian line with a width given by the self-broadened linewidth Γ_{self} and a line shift $\Delta\omega$ that is dominated by the local-field shift $\Delta\omega_{\mathrm{L}}$ [5–7]. This assumption is well established for the density regime where the collisions are binary, i.e., where a probe atom interacts with one perturber at a time.

At low densities the duration of a collison is much shorter than the time in between collisions which implies that the impact model for the collisions applies [8]. When one increases the density the collisions are no longer well separated in time and the impact model loses its validity. One may say that an atom is permanently in a state of collision. For a short time it forms a quasimolecule with a neighbor, exchanging partners in rapid succession. This is the quasimolecular density regime, which has recently been shown to be full of surprises [9–11]. Upon further increase of the density the binary collision approximation fails and one enters the regime where multiperturber effects dominate. A description of the atomic response in this last density regime is considered to be prohibitively complicated. It should be noted that it has recently been argued that for the dipole-dipole interaction many-body effects are not only important at very high densities but are manifest at all densities [12].

Our interest lies in the density regime where quasimolecular effects become important because this allows us to study the onset of the transition from a gas of free atoms to ultimately, the condensed phase. The quasimolecular regime can be seen as the density range where atoms still interact pairwise but where the symmetry of the atomic wave function is broken due to the rapid succession of two-body collisions. The onset can be characterized by the condition:

$$\rho_{\mathrm{w}}/\overline{v} \approx (N\pi\rho_{\mathrm{w}}^{2}\overline{v})^{-1}, \tag{1}$$

where ρ_{w} is the Weisskopf radius, the impact parameter that gives unity optical phase shift in the impact limit. Equation (1) equates the time of a collision $\rho_{\mathrm{w}}/\overline{v}$ with the inverse of the collision frequency, where N is the atomic density and $\overline{v}$ the mean relative speed of the atoms in the vapor. Note that Eq. (1) can be rewritten as

$$\pi N\rho_{\mathrm{w}}^{3} \approx 1, \tag{2}$$

which indicates that on average there is one atom in a sphere of radius ρ_{w} around our probe atom. One can easily show that under these conditions a non-negligible fraction (≈ 20%) of the atoms has more than one collision partner within the Weisskopf radius at a time. This means that one cannot clearly distinguish between the density range where very frequent pairwise collisions dominate and the density where multiperturber effects start to become important.

In a series of experiments on absorption spectra and magneto-optic spectra performed in our laboratory, it was shown that in the quasimolecular density regime the impact description for the line core of the spectral line breaks down and the full line shape must be explained in terms of a quasistatic picture. In particular, the Faraday spectrum of Rb atoms immersed in a high density xenon gas [9,13] was measured for densities up to $N\rho_{\mathrm{w}}^{3} \approx 1$. Later, the Faraday experiments on Rb in Xe were extended to densities where $N\rho_{\mathrm{w}}^{3} \approx 4$; in this case the description, that was successful at $N\rho_{\mathrm{w}}^{3} \approx 1$, was shown to break down. This was interpreted as indicative for the onset of multiperturber effects [10]. Also, the excitation-modulated reflectivity spectrum of a pure Rb gas at densities where $N\rho_{\mathrm{w}}^{3} \approx 0.5$ has been investigated experimentally [11].

In the present paper we show experimental results for the density dependence of the optical response of a pure potas-

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sium vapor in a magnetic field. In particular, we show that, by performing a magneto-optic experiment, we sensitively probe how *resonant dipole-dipole collisions modify the magnetic response of the atoms*. Since a theoretical description of collisional modifications of the magnetic response of an atom is complicated, the discussion of our experimental results will be qualitative.

II. QUASIMOLECULAR PROBES

Linear spectroscopy is not a suitable tool to probe the character of the spectral line in a dipole-dipole broadened system [12]. This is because for this type of broadening the absorption line is Lorentzian shaped (except for the very far wings) in both the impact [8] and quasistatic [14] regimes, with comparable widths. Within the binary approximation it is not possible to determine from the line shape whether the impact or a quasistatic model applies. For example, a study on high-density potassium vapor showed a linear relation between the self-broadened linewidth and density when proceeding from the impact limit ($N\rho_{\mathrm{w}}^3 \ll 1$) to the regime where $N\rho_{\mathrm{w}}^3 \approx 1$ [5].

However, nonlinear spectroscopy or, equivalently, linear spectroscopy in the presence of an external static field, provides more information. For example, it has been observed that the spectral shape of the resonance line in dense rubidium vapor is modified when it is partially excited [11]. In this experiment the density was such that the Weisskopf radius was of the order of the interatomic distance, i.e., the experiment probed the limit of the binary impact-collision approximation. The dielectric function of the dense vapor was found to be well represented by the dielectric function for a dilute vapor including an excitation dependent linewidth and shift. The results for the excitation dependence of the linewidth agreed with an estimate based on a quasistatic picture of the collisions.

Kristensen *et al.* [9,13] have shown how linear spectroscopy in an external magnetic field can provide a handle on the collision physics. In their work the Faraday spectrum, i.e., the rotation of the polarization of the transmitted laser light as a function of frequency, was studied for rubidium atoms immersed in a very high-density Xe gas in a static magnetic field. For low perturber densities the Faraday spectrum is, up to a scale factor, identical to the dispersion $dn/d\omega$, a relationship established by Woerdman, Nienhuis, and Kuščer [15]. The deviations that were observed between the Faraday and dispersion spectrum were attributed to quasimolecular effects, i.e., the creation of an additional axis during the collision that perturbs the free precession of the atomic magnetic moment in the static magnetic field.

In the present experiment we study the collision physics in a pure, dense atomic vapor using a different variant of magneto-optic spectroscopy. Because the vapor is optically very thick (extinction length $\ell \approx \lambda_0/2\pi$ with λ_0 the resonance optical wavelength) we measure its reflection spectrum. In particular, we measure the magnetic-field induced difference in reflectivity for σ^+-polarized and σ^--polarized light with the incident light propagating parallel to the direction of the externally applied magnetic field. The Zeeman effect lies at the heart of this reflectivity difference.

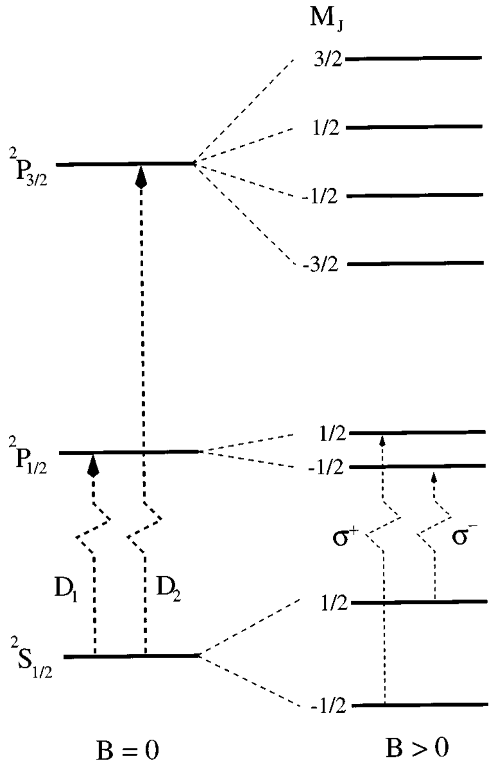

FIG. 1. Fine-structure levels of the $4S$-$4P$ transition in potassium showing the magnitude of the Zeeman splittings in the various states.

Potassium is eminently suited for the study of dipole-dipole effects for the following reasons: The hyperfine splittings in the ground $4s^2S_{1/2}$ and excited $4p^2P_{1/2,3/2}$ states are relatively small ($<$ 0.5 GHz). The resonance lines of potassium have a self-broadening coefficient $\Gamma_{\mathrm{self}}/N = 2\pi\times 7\times 10^{-8}\ \mathrm{cm}^3\,\mathrm{s}^{-1}$ for the D_1 line and $\Gamma_{\mathrm{self}}/N = 2\pi\times 10\times 10^{-8}\ \mathrm{cm}^3\,\mathrm{s}^{-1}$ for the D_2 line [8]; here Γ_{self} represents the full width at half maximum of the self-broadened line. In the density range of interest ($N = 0.2\times 10^{17}\mathrm{cm}^{-3}$–$5\times 10^{17}\mathrm{cm}^{-3}$) the pressure broadened width is then appreciably larger than the hyperfine splitting and the Doppler width (1 GHz at 600 K); both effects will therefore be neglected. In addition, the fine-structure splitting is very large (1700 GHz) so that even at the highest densities the two resonance lines (D_1 and D_2) are very well separated. This all implies that we can discuss the Zeeman effect in terms of fine-structure states.

The energy shift ΔE_i of an atomic state i with angular momentum J_i and magnetic quantum number m induced by a magnetic field B is given by

$$\Delta E_{\mathrm{i}} = g_{\mathrm{J_i}}\mu_B B m, \tag{3}$$

with $g_{\mathrm{J_i}}$ the Landé factor for the level i and μ_B the Bohr magneton ($\approx$ 1.4 MHz/G). In Fig. 1 the fine-structure levels of potassium split by the Zeeman effect are shown.

In our experiment we effectively determine the frequency difference between the atomic responses for two different polarizations (σ^+ and σ^-) of the incident light. This frequency difference between transitions induced by $\sigma^+(\Delta m_J = +1)$ and $\sigma^-(\Delta m_J = -1)$ light can be expressed as $2\alpha\mu_B B/h$ with h Planck's constant (Fig. 1). In the collisionless regime the Zeeman transition parameter α for the

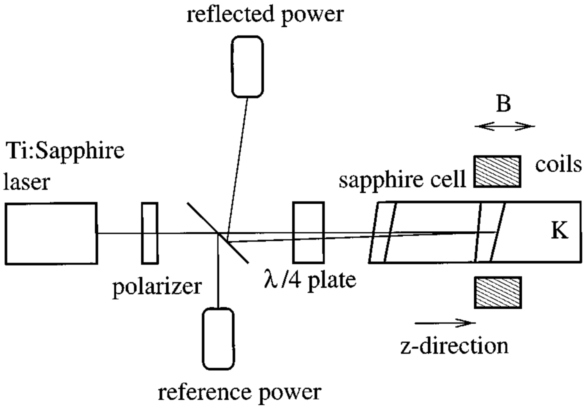

FIG. 2. Experimental setup to measure the magneto-optic spectrum of a high-density potassium vapor.

D_1 transition equals half the sum of the single atom Landé factors of the ground-state ($^2S_{1/2}$) and excited-state ($^2P_{1/2}$): $\alpha = 4/3$. For the D_2 transition a more elaborate procedure is necessary. For each polarization component two contributions with their appropriate intensities have to be taken into account. This yields $\alpha = 7/6$ [15,16]. The idea of the experiment is to measure the Zeeman transition parameter α as a function of density in order to probe the onset of quasimolecular effects.

III. EXPERIMENTAL TECHNIQUE

The experiments are performed using a single-mode Ti:sapphire laser beam, attenuated to $P = 10$ mW, that is incident onto a potassium vapor cell as a left-circular polarized beam. The spotsize is of the order of a few mm so the beam intensity is well below the saturating value which is of the order of 1W/cm^2 at a potassium density $N = 2\times10^{16}\text{cm}^{-3}$ [17]. The cell is made completely of sapphire and consists of two parts. The rear section is heated to the desired temperature and contains the potassium vapor; the front section sticks out of the oven and is evacuated to avoid turbulence (see Fig. 2). The two sections of the cell are separated by a wedged sapphire window so the reflection from the air-sapphire and sapphire-vapor interfaces can easily be distinguished. To avoid effects of birefringence the optical axis of the window lies perpendicular to its plane and the laser beam is reflected at near-normal angle of incidence. The reflected beam is measured with a photodetector and its signal is proportional to the reflected power

$$P_{\text{refl}} = P_{\text{in}} R(\omega - \omega_0), \tag{4}$$

where P_{in} is the power of the incident laser beam and

$$R(\omega-\omega_0) = \left| \frac{\eta - \sqrt{\epsilon(\omega-\omega_0)}}{\eta + \sqrt{\epsilon(\omega-\omega_0)}} \right|^2, \tag{5}$$

the interface reflectivity between the sapphire window and the K vapor. In Eq. (5) η is the refractive index of the sapphire window ($\eta = 1.76$ for $\lambda \approx 770$ nm) and $\epsilon(\omega)$ is the dielectric function of the potassium vapor.

In order to measure the magneto-optic spectrum as described in the preceding section the cell is placed in a homogeneous, longitudinal magnetic field supplied by two coils. We modulate the magnetic-field amplitude according to $B_z = B_0 \sin(\Omega t)$ with $B_0 = 0.01$ T and $\Omega = 2\pi\times10$ Hz. The magnetic field is determined with a calibrated magnetometer but we cannot probe the field exactly at the position of the cell window because of the geometry of the oven. The signal from the photodetector is demodulated and we record the amplitude of the demodulated signal as a function of the frequency ω of the laser. This approach, where we use a left-circular polarized laser beam and periodically switch the direction of the longitudinal magnetic field, is equivalent to a situation where the magnetic field is stationary and we modulate the polarization of the incident light from left circular to right circular. Modulation of the magnetic field implies that we modulate the atomic resonance frequency around its zero-field value ω_0. It then becomes clear that for the magnitude of our demodulated signal $S^{\text{mag}}(\omega-\omega_0)$ we have

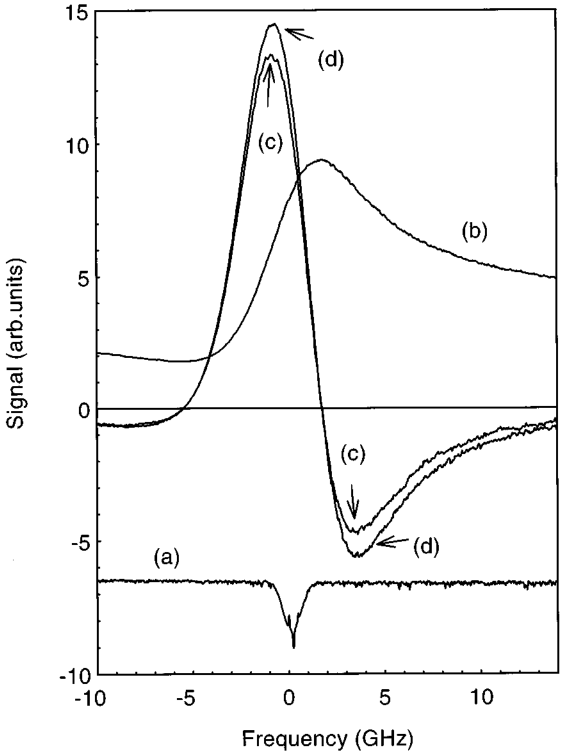

FIG. 3. Reflectivity spectra on the D_1 line for potassium vapor ($N = 0.8\times10^{17}$ cm^{-3}). For details see text.

$$S^{\text{mag}}(\omega-\omega_0) \propto \frac{\partial R(\omega-\omega_0)}{\partial\omega_0} \Delta\omega_0, \tag{6}$$

with $\Delta\omega_0$ the modulation depth of the atomic resonance frequency. For the latter we can write

$$\Delta\omega_0 = \frac{\alpha\mu_B B_0}{\hbar}, \tag{7}$$

with B_0 the magnetic-field modulation depth.

Instead of trying to model the modulated reflection spectrum and compare it with the experimental spectrum in order to determine a value of the Zeeman transition parameter α we compare the magnetic-field modulated spectrum with the

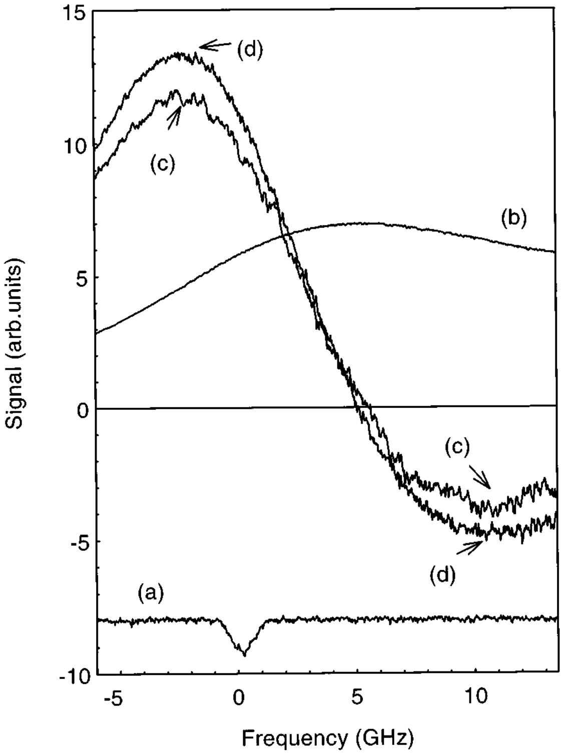

FIG. 4. Reflectivity spectra on the D_1 line for potassium vapor ($N = 2.6\times 10^{17}\,\mathrm{cm}^{-3}$). For details see text.

reflection spectrum that we obtain by modulating the laser frequency by an amount $\Delta\omega_F$. It can easily be shown that for the frequency modulated signal we have

$$S^{\mathrm{freq}}(\omega-\omega_0) \propto \frac{\partial R(\omega-\omega_0)}{\partial\omega}\Delta\omega_F. \tag{8}$$

In this way we avoid errors that can arise because we do not know *a priori* the exact frequency dependence of the dielectric function $\epsilon(\omega-\omega_0)$. Comparison of the amplitudes of the two modulated reflection spectra together with knowledge of the modulation depths then yields a value for α. It is obvious that the magnetic-field modulation depth $(\alpha\mu_B B/\hbar)$ and the frequency modulation depth $(\Delta\omega_F)$, which are both of the order of 100 MHz, should be much smaller than the width of the reflection spectrum (order of GHz).

IV. RESULTS AND DISCUSSION

We have investigated the magneto-optic spectrum in the density range from $N = 0.2\times 10^{17}$ cm^{-3} to $N = 4.8\times 10^{17}$ cm^{-3}. Typical experimental results for the D_1 transition in potassium for densities $N = 0.8\times 10^{17}$ and $N = 2.6\times 10^{17}$ cm^{-3} are shown in Figs. 3 and 4, respectively.

In both figures curve (a) shows the low-density ($N \approx 10^{11}\,\mathrm{cm}^{-3}$) absorption spectrum of potassium as measured in a separate cell; it serves as an absolute frequency reference. The reflection spectra of the high density K vapor near the sapphire interface are depicted in curves (b). The reflection curve is a dispersionlike shaped curve centered around the resonance frequency. Far in the wing the reflectivity has a value of 7.6% corresponding to the sapphire-vacuum interface. The reflectivity of the vapor at densities of about $N = 3\times 10^{17}\,\mathrm{cm}^{-3}$ varies between $\approx$ 20 % and $\approx$ 3 %. Curves (c) and (d) show the high-density frequency and magnetic-field modulated reflection spectra, respectively.

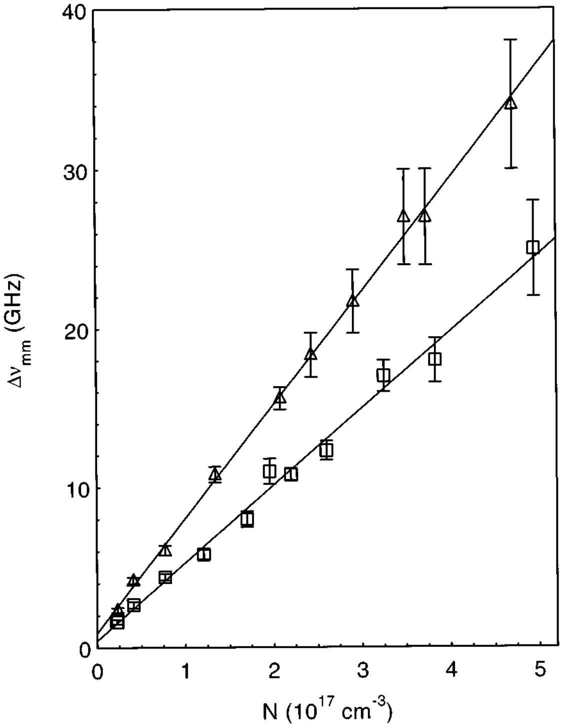

FIG. 5. Frequency difference $\Delta\nu_{mm}$ between the maximum and minimum signal of the frequency modulated-reflection spectrum for the D_1 (□) and D_2 (△) lines of potassium as a function of atomic density.

An important parameter that can be extracted from these spectra is the self-broadened linewidth Γ_{self}. Experimentally it is tedious to spectrally cover a larger frequency range than roughly 25 GHz which is the electronic tuning range of the Ti:sapphire laser; we have not attempted to do this. We therefore cannot extract the self-broadened linewidth Γ_{self} directly from the reflection spectrum at high densities. However, for a Lorentzian line Γ_{self} can be directly deduced from the frequency difference $\Delta\nu_{mm}$ between the maximum and minimum of the frequency modulated-reflection spectrum. This frequency difference $\Delta\nu_{mm}$ is smaller than 25 GHz for densities up to $N = 5\times 10^{17}\,\mathrm{cm}^{-3}$. Assuming the absorption line shape to be Lorentzian one can show that for the D_1 line of potassium $\Delta\nu_{mm} = 0.81\times\Gamma_{\mathrm{self}}$ while for the D_2 line $\Delta\nu_{mm} = 0.87\times\Gamma_{\mathrm{self}}$. In Fig. 5 the frequency difference $\Delta\nu_{mm}$ is depicted as a function of vapor density. From our data we can extract a value for the self-broadening coefficient for the D_1 line: $\Gamma_{\mathrm{self}}/N = 2\pi\times 5.9\times 10^{-8}$ s^{-1}, in good agreement with the result found by Maki *et al.* [5].

The experimentally observed linear dependence of the width versus density shows that for all measured densities N, i.e., covering the impact and quasimolecular regime as well as the regime where multiperturber collisions are expected to become dominant, the ratio of collisional width and density is constant. This observation is in agreement with our argument that the linewidth itself does not give information

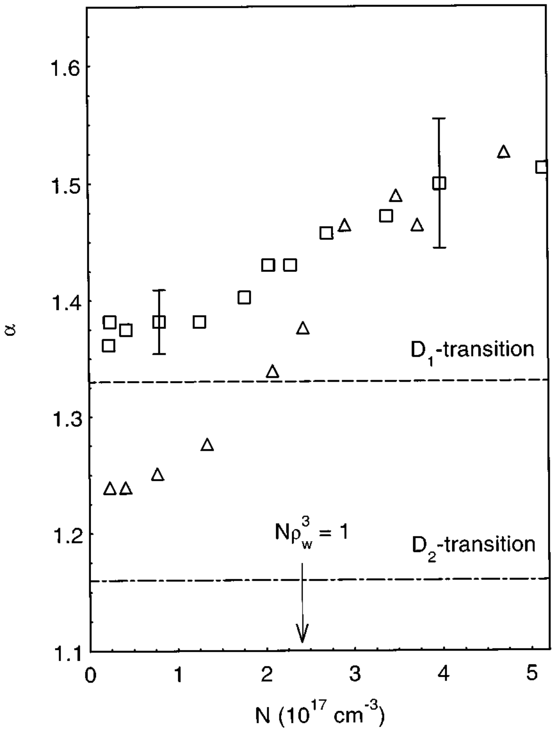

FIG. 6. The Zeeman transition parameter α as a function of density N for the D_1 (□) and D_2 (△) lines in K vapor. The horizontal dashed lines give values for α for a free atom. Typical error bars on either side of the density range are indicated.

about the nature of the collisions. However, as will be shown below, the magneto-optic spectrum *does* change significantly at densities where the impact approximation breaks down.

As explained in the preceding section we can condense all the amplitude information contained in the modulated reflectivity spectra into the Zeeman transition parameter α which we determine as a function of density. In Fig. 6 we show a clear density dependence of this parameter for both the D_1 and D_2 transitions. The density dependence becomes apparent already at densities for which $N\rho_w^3 \gtrsim 1$. This corresponds to the onset of the regime where the time of a collision equals the time in between the collisions. In this density regime multiperturber effects may be of importance. At the highest densities measured we observe that α converges to the same value for both the D_1 and D_2 transitions.

At low densities ($N \leqslant 1\times10^{17}\,\mathrm{cm}^{-3}$) the Zeeman transition parameter α is found to be independent of density. This is not surprising since the atom is free most of the time and the symmetry of the atomic wave function is conserved: α should be equal to the free atom value. Note that the measured values for α at low densities slightly deviate from the values for a free atom (4/3 for the D_1 and 7/6 for the D_2 transition). This is ascribed to the uncertainty in the determination of the magnetic field that has been discussed before; however the *ratio* of the α parameters for the D_1 and D_2 transition (0.89) is in good agreement with the free atom value (0.88).

We will now discuss the higher-density regime where the collision time becomes comparable with the time in between collisions. An excited atom experiences a rapid succession of collisions with different ground-state partners forming a quasimolecule during the collision. Since an atom in the ground state has a much smaller polarizability than an atom in the excited state the effect of the collision on the excited atom Zeeman splitting is much larger than that on the Zeeman splitting of an atom in the ground state. This implies that we only need to consider the effect of collisons on the Zeeman effect of the ^2P_J states. The evolution of an excited atom alternates then between that of a more or less free atom with its free-atom Zeeman precession frequency and one forming a quasimolecule with a collision partner with a modified precession frequency. Averaged over many collisions this may lead to a precession frequency different from that of a free atom and thus to modified Zeeman splittings.

A quantitative description of this effect is very difficult since there is an extremely rapid succession of collisions with partners from all directions. In each collision the interatomic distance and therefore the coupling strength changes on even faster time scales. In a molecular picture one would need to calculate the Zeeman effect dynamically, taking into account all the directions of the collision axes relative to the external magnetic field and various Hund's coupling cases [18]. In many of these collisions interatomic distances are probed where the dipole-dipole interaction is much larger than the spin-orbit interaction. This implies that the total electronic angular momentum of each atom is not conserved anymore. The immediate consequence of this uncoupling phenomenon is again a modified Zeeman precession frequency (compare with the Paschen-Back effect).

At the highest densities ($N \approx 4\times10^{17}\,\mathrm{cm}^{-3}$) the atom is permanently in a state of collision and the coupled representation is not valid at any time. It is then very plausible that the Zeeman effect for the $^2P_{1/2}$ and $^2P_{3/2}$ excited states are equal in this limit. This agrees with our observation that the two curves for the D_1 and D_2 line converge at high densities. It is important to realize that at these densities the two resonance lines are still very well separated; for our highest densities the linewidth is a small fraction ($\approx$ 2%) of their frequency separation.

V. CONCLUDING REMARKS

In conclusion, we have experimentally shown that for a dense potassium vapor ($N \geqslant 1\times10^{17}\,\mathrm{cm}^{-3}$) the Zeeman precession frequency of atoms in the excited P state is modified as the result of dipole-dipole collisions with ground-state atoms. These modifications first appear at densities where the collision time is no longer short as compared with the time in between collisions. In the density range where the binary-collision approximation loses its validity the Zeeman splittings of the two resonance lines become equal. This implies a breakdown of the quantum number associated with the electronic angular momentum of an excited P atom. Since our results give new insight into the resonant dipole-dipole collision physics in dense atomic vapors, a well-built theory would be very valuable to the field of nonlinear optics.

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[1] Y. Ben-Aryeh, C. M. Bowden, and J. C. Englund, Phys. Rev. A **34**, 3917 (1986).

[2] M. P. Hehlen, H. U. Güdel, Q. Shu, J. Rai, S. Rai, and S. C. Rand, Phys. Rev. Lett. **73**, 1103 (1994).

[3] C. R. Stroud and C. M. Bowden, Opt. Commun. **67**, 387 (1988).

[4] A. S. Manka, J. P. Dowling, C. M. Bowden, and M. Fleischhauer, Quantum Opt. **6**, 371 (1994).

[5] J. J. Maki, M. S. Malcuit, J. E. Sipe, and R. W. Boyd, Phys. Rev. Lett. **67**, 972 (1991).

[6] R. Friedberg, S. R. Hartmann, and J. T. Manassah, Phys. Rep. **7**, 101 (1973); Phys. Rev. A **39**, 3444 (1989); **40**, 2446 (1989); **42**, 5573 (1990).

[7] F. A. Hopf, C. M. Bowden, and W. H. Louisell, Phys. Rev. A **29**, 2591 (1984).

[8] E. L. Lewis, Phys. Rep. **58**, 1 (1980).

[9] M. Kristensen, F. J. Blok, M. A. van Eijkelenborg, G. Nienhuis, and J. P. Woerdman, Phys. Rev. A **51**, 1085 (1995).

[10] J. P. Woerdman, F. J. Blok, M. Kristensen, and C. A. Schrama, Phys. Rev. A **53**, 1183 (1996).

[11] V. A. Sautenkov, H. van Kampen, E. R. Eliel, and J. P. Woerdman, Phys. Rev. Lett. **77**, 3327 (1996).

[12] J. A. Leegwater and S. Mukamel, Phys. Rev. A **49**, 146 (1994).

[13] M. Kristensen, M. A. van Eijkelenborg, and J. P. Woerdman, Phys. Rev. Lett. **72**, 2155 (1994).

[14] K. Niemax and G. Pichler, J. Phys. B **8**, 179 (1975); K. Niemax, M. Movre, and G. Pichler, *ibid.* **12**, 3503 (1979).

[15] H. Beaguerel, C. R. Acad. Sci. **125**, 679 (1897); J. P. Woerdman, G. Nienhuis, and I. Kuščer, Opt. Commun. **93**, 135 (1992).

[16] G. Nienhuis, J. P. Woerdman, and I. Kuščer, Phys. Rev. A **46**, 7079 (1992).

[17] J. J. Maki, W. V. Davis, and R. W. Boyd, Phys. Rev. A **46**, 7155 (1992).

[18] G. Herzberg, *Spectra of Diatomic Molecules* (Van Nostrand Reinhold, Princeton, NJ, 1957).