

Observation of ground-state Zeeman coherences in the selective reflection from cesium vapor

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We report an observation of $\Delta m = 2$ ground-state coherences in selective-reflection spectra at a glass-metal-vapor interface. The coherence was created by a linearly polarized laser beam tuned to a sub-Doppler resonance in the selective-reflection spectrum of the cesium D_2 line and detected through the magnetorotation of the plane of polarization of the reflected beam. The coherence effect appears as a dispersive shaped resonance of subnatural width. The dependence of the width on the intensity of the optical field shows a saturation behavior. For vanishing light intensity and cesium vapor pressure the linewidth is limited by transverse time-of-flight broadening, whereas its pressure dependence indicates an additional contribution from the finite longitudinal interaction time.

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INTRODUCTION

The behavior of neutral atoms near a surface, e.g., the long-range atom-surface interaction, the quenching of optical excitation, and the relaxation of ground-state coherence by wall collisions or QED effects near a surface are of great interest in many areas of fundamental and applied atomic physics. High-resolution reflection spectroscopy at the interface of a transparent solid and an atomic gas has become a new powerful tool for the study of atom-wall interactions or the investigation of optically dense gas samples. One distinguishes two major high-resolution techniques in reflection spectroscopy, viz., evanescent wave spectroscopy (EWS), when the incidence angle on the interface is close to or larger than the critical angle [1–3], and selective-reflection spectroscopy (SRS), when the incidence angle is close to normal [4–6].

In EWS an evanescent wave propagates along the surface in the medium of lower refractive index, so that due to the Doppler effect, EW spectra mainly reflect the distribution of atomic velocities parallel to the surface. EWS therefore cannot discriminate velocity components normal to the surface. On the other hand, SRS distinguishes the various velocity groups of atoms flying towards the surface or departing from it. Moreover atom-wall collisions lead to the appearance of sub-Doppler resonances in SR spectra [5] which are due to the transient readjustment of the atomic dipole to the driving optical field after a wall collision. Both techniques probe vapor atoms in typical depths corresponding to a reduced wavelength and give complementary information on the properties of atomic or molecular species near a surface.

Linear SRS has been used to study the collisional self-broadening [6] and shift [7] of the Cs D_2 line, the van der Waals interaction between Cs atoms and a glass surface [7], and the influence of the Lorentz local field on the potassium D_1 line [8].

In the last few years nonlinear effects due to optical saturation [9–11] and ground-state optical pumping [12] in Doppler-free SR spectra have been studied. In EWS optical-pumping effects have also been observed [3], and

recently Suter, Abersold, and Mlynek [13] have reported the observation of $\Delta m = 1$ ground-state coherences in EWS. However, no experimental study of ground-state coherences in SRS has been performed so far. Magneto-optical techniques have proven to be powerful for the investigation of such coherences in transmission experiments [14]. To our knowledge the only previous study of magneto-optical effects in SRS was an experiment performed by Stanzel [15], and described theoretically by Schuurmans [16], where excited-state magnetic level crossings were investigated using a broadband light source.

In the present work we have used a sensitive polarization-modulation technique to study the nonlinear magneto-optical activity induced by ground-state coherences in the Doppler-free SR spectra of the Cs D_2 line. In particular we investigated the rotation of the plane of polarization of the reflected beam when the medium is exposed to a longitudinal magnetic field. As was previously observed in transmission experiments [14] $\Delta m = 2$ ground-state coherences induced by a linearly polarized light beam lead to a resonant enhancement of the magnetorotation of the plane of polarization. Here we report on the first observation of similar ground-state level-crossing resonances in SRS and discuss the properties of these resonances for a wide range of experimental parameters.

EXPERIMENT

Figure 1 shows the experimental setup. The experiments were performed using a single-mode laser diode. The reflecting interface was formed by a window and saturated Cs vapor contained in a heated glass cell. A temperature gradient prevented cesium from condensing on the window and the vapor pressure was inferred from the temperature of the coldest spot of the cell by using the expression of Taylor and Langmuir [17]. The window was wedged, so that the reflections from the two surfaces could be easily distinguished. A homogeneous magnetic field was provided by a pair of Helmholtz coils. The laser

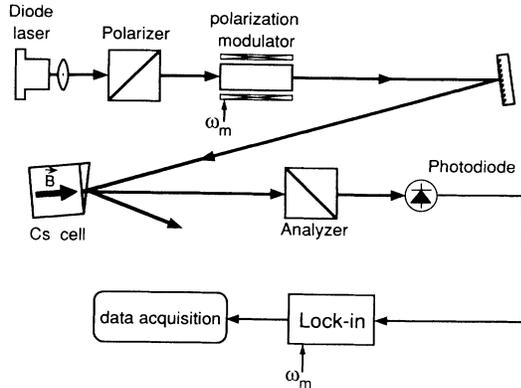


FIG. 1. Experimental setup. The polarization modulator uses the Faraday effect in lead glass to modulate the orientation of the linear polarization at frequency $\nu_m = 80$ Hz.

power P was measured with a calibrated photodiode and the laser intensity I calculated according to $I = 2P / \pi w_x w_y$, where the orthogonal beam diameters w_x and w_y take the slight astigmatism of the transverse intensity distribution into account. w_x and w_y were measured using a linear photodiode array.

The incident light beam was linearly polarized by a Glan prism and the polarization of the beam reflected from the Cs-glass interface was analyzed with a crossed polarizer. When the interface is exposed to a magnetic field the reflected beam becomes in general elliptically polarized.

In analogy with transmission experiments we shall in the following refer to the orientation of the polarization ellipse and its ellipticity as Faraday rotation angle Φ_F and circular dichroism, respectively. The total intensity I_t transmitted by the analyzer depends on both the Faraday rotation and the circular dichroism. However, if one modulates [18] the orientation of the incident polarization periodically at frequency ω_m , it is easy to show that the Fourier component of the intensity I_t at ω_m depends on the rotation angle Φ_F only.

Using a polarization modulator and phase-sensitive detection with a lock-in amplifier we thus obtain a sign-sensitive signal proportional to the Faraday rotation angle Φ_F . The lock-in signal is digitized and recorded with a personal computer for off-line analysis.

RESULTS

Figure 2 shows the spectral dependence of the Faraday angle for the $6S_{1/2}(F=4) \rightarrow 6P_{3/2}$ transition. The spectrum shows three sub-Doppler resonances, corresponding to the $F=4 \rightarrow F'=3, 4, 5$ hyperfine transitions superposed on a small Doppler-broadened background. The spectrum was recorded with low light intensities in a magnetic field B of 6.4 G and represents thus a purely linear effect as will become apparent later. In large ($B \gg 1$ G) magnetic fields the spectral line shapes were found to be independent of the magnetic field, whereas in lower fields ($B \leq 1$ G) we found that the relative amplitudes of the

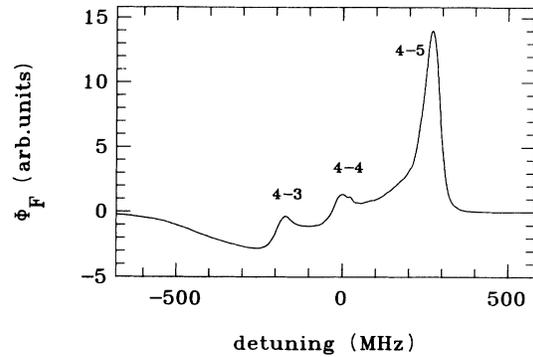


FIG. 2. Faraday rotation spectrum of the $6S_{1/2}(F=4) \rightarrow 6P_{3/2}(F')$ transition recorded in a field of 6.4 G. The sub-Doppler resonances correspond to the $F=4 \rightarrow F'=3, 4, 5$ hf components.

three hyperfine (hf) resonances depended strongly on the intensity of the incident light. In order to investigate this in more detail we proceeded as follows: we tuned the laser frequency to the center of the $4 \rightarrow 4$ transition and studied the dependence of the Faraday angle Φ_F on the strength of the magnetic field B for different cesium densities N_{Cs} and laser intensities I .

Figure 3 shows a typical dependence of Φ_F on the magnetic field strength B obtained with cesium number density $N_{Cs} = 3.3 \times 10^{13} \text{ cm}^{-3}$ and $I = 30 \text{ mW/cm}^2$. The nonlinear resonance appears as a dispersive shaped structure on a linear background. The amplitude of this resonance (approximately 0.5 mrad in Fig. 3) strongly depends on the intensity I and vanishes for low intensities. The solid line in Fig. 3 is a least-squares fit where a dispersive Lorentzian of half-width ΔB ,

$$\Phi_F \propto \frac{B}{B^2 + \Delta B^2}, \quad (1)$$

gives a satisfactory description of the resonance line shape. The resonance shows considerable power broadening. For every setting of the Cs number density N_{Cs} we therefore measured the intensity dependence of

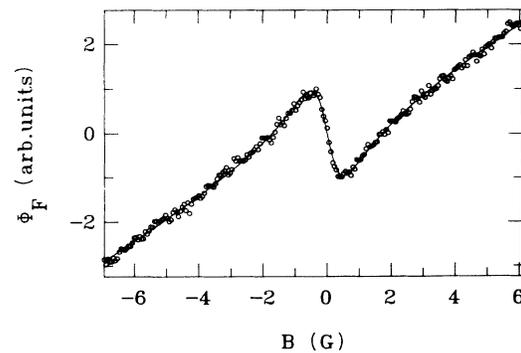


FIG. 3. Nonlinear Faraday effect of the $F=4 \rightarrow F'=4$ hyperfine component. The solid line is a fitted dispersive Lorentzian on a linear slope.

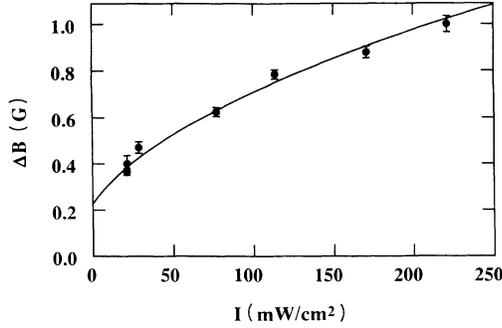


FIG. 4. Dependence of the width ΔB of the nonlinear Faraday resonance on laser intensity I for cesium number density $N_{\text{Cs}} = 1.4 \times 10^{13} \text{ cm}^{-3}$. The solid line is the fitted function $\Delta B = \Delta B_0(1 + I/I_S)^{1/2}$ with $I_S = 11(3) \text{ mW/cm}^2$.

the width ΔB . A typical curve is shown in Fig. 4. It is well described by the fitted function

$$\Delta B = \Delta B_0(1 + I/I_S)^{1/2}. \quad (2)$$

We shall discuss later the significance of the saturation intensity I_S . The extrapolated ($I \rightarrow 0$) width ΔB_0 shows a linear dependence on the Cs number density N_{Cs} (Fig. 5):

$$\Delta B_0 = \Delta B_{00} + kN_{\text{Cs}}, \quad (3)$$

where ΔB_{00} is the extrapolated width of the nonlinear resonance when both $I \rightarrow 0$ and $N_{\text{Cs}} \rightarrow 0$.

DISCUSSION

To our knowledge there has been so far no theoretical investigation of nonlinear magneto-optical effects in SR spectroscopy. In the following we shall therefore use some theoretical results obtained for the description of the nonlinear Faraday effect in transmission experiments. We stress that we apply these results only by analogy to the effects investigated here. The dependence of the Faraday rotation angle on the magnetic field in transmission experiments has various contributions [19]: at very low light intensities, the effect is linear, i.e., it is independent of the light intensity I and shows a linear B -field

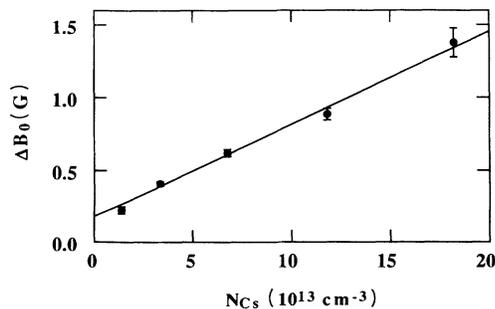


FIG. 5. Dependence of the extrapolated ($I \rightarrow 0$) width ΔB_0 of the nonlinear Faraday resonance on cesium number density. The solid line represents a least-squares fit.

dependence; at higher intensities one notes the appearance of nonlinear contributions which have their origin in the creation of sublevel coherences and population redistributions in the excited and ground states. These effects manifest themselves as dispersive shaped resonances, the widths of which reflect the different coherence relaxation rates in the two levels. As is typical for magnetic level crossing experiments the width ΔB of the observed resonance is related to the coherence relaxation time τ by

$$2g_F\mu_B\Delta B\tau/\hbar \equiv \Delta\Omega\tau = 1, \quad (4a)$$

where

$$\Omega = 2g_F\mu_B B/\hbar \quad (4b)$$

is the precession frequency of the sublevel coherence in the state with angular momentum F . The coefficient 2 reflects the fact that the linearly polarized light field couples only sublevels with magnetic quantum numbers differing by $\Delta m_F = 2$. In the following we shall use ΔB or equivalently $\Delta\Omega$ to refer to the resonance linewidth.

The natural width of the Cs D_2 line is $\gamma = 1/\tau \approx 2\pi \times 5$ MHz, so that one expects a width ΔB of the nonlinear resonance of 7 G. Note that this value represents only a lower limit as the lines are broadened by collisions. The typical linewidths observed in our experiments were less than 1 G. We therefore conclude that their origin is in the creation of ground-state coherences.

In order to get a better understanding of our results, we compare in the following our experimental findings to theoretical predictions obtained for the nonlinear Faraday effect in transmission experiments. Schuller, Macpherson, and Stacey [20] have calculated the Faraday rotation angle for a $J = 1 \rightarrow J' = 0$ transition in arbitrary light intensities. From their results, valid for monochromatic, resonant excitation, we obtain the asymptotic formula

$$\Phi_F \propto \frac{\Omega}{\Omega^2 + \Delta\Omega^2}, \quad (5)$$

with

$$\Delta\Omega = \Gamma_0(1 + G)^{1/2}, \quad (6)$$

where

$$G = \frac{\beta^2}{\gamma\Gamma_0} \quad (7)$$

is a saturation parameter typical for optical-pumping processes [21]. We assumed that the ground-state relaxation rate $\Gamma_0(N_{\text{Cs}})$ is small compared to the excited-state relaxation rate γ , and that the Rabi frequency $\beta = ex_{ge}E/\hbar \ll \gamma$, so that optical saturation effects may be neglected.

Note that one can write the saturation parameter as $G = I/I_S$, where $I = cE^2/8\pi$ is the laser intensity and $I_S = \hbar\gamma\Gamma_0/4\pi\alpha x_{ge}^2$ is the saturation intensity. Here α is the fine-structure constant. The observed resonances are dispersive Lorentzians with intensity-dependent widths in good agreement with the experimental findings shown in Figs. 3 and 4.

For a quantitative analysis of the saturation behavior of the nonlinear resonance we use a slightly modified expression for the saturation parameter by defining $G \equiv \beta^2/\Delta\omega\Gamma_0$. We took the finite linewidth of the laser $\Delta\omega_L$ as well as the collisionally broadened homogeneous linewidth $\Delta\omega_c$ into account by introducing an effective spectral width $\Delta\omega = \Delta\omega_L + \Delta\omega_c$ and replacing the optical excitation rate β^2/γ in Eq. (7) by $\beta^2/\Delta\omega$. Note that all data were recorded under conditions where $\beta \ll \Delta\omega$, i.e., for negligible optical saturation. Defining the Rabi frequency of the $F \rightarrow F'$ transition by $\beta_{FF'} = ex_{FF'}E/\hbar$ the optical-pumping saturation intensity becomes

$$I_S = \frac{\hbar}{4\pi\alpha} \frac{\Delta\omega\Gamma_0}{x_{FF'}^2} \quad (8)$$

with the electric dipole matrix element

$$\begin{aligned} x_{FF'}^2 &= \sum_{M, M'} |\langle 6P_{3/2} FM | x | 6S_{1/2} F' M' \rangle|^2 \\ &= \frac{g_{FF'}}{3} |\langle 6P_{3/2} || \mathbf{r} || 6S_{1/2} \rangle|^2, \end{aligned}$$

where

$$g_{FF'} = \frac{(2F+1)(2F'+1)}{2I+1} \begin{Bmatrix} F & J & I \\ J' & F' & 1 \end{Bmatrix}^2 \quad (9)$$

is the normalized relative intensity of the $F \rightarrow F'$ hyperfine component [22].

We measured the width $\Delta\omega$ for our experimental conditions using a frequency modulation technique on the SR spectra [6] and found a weak dependence on N_{Cs} . The width $\Delta\omega$ increased from $2\pi \times 50$ MHz to $2\pi \times 70$ MHz in the range $N_{Cs} = 0.14 \times 10^{14}$ to 1.8×10^{14} cm $^{-3}$. This increase is consistent with the previously measured [6] collisional self-broadening parameter. The main contributions to $\Delta\omega$ are the laser linewidth and a residual Doppler width $\Delta\omega_D \sin\theta$ due to the finite reflection angle ($2\theta = 30$ mrad) [6]. Under these conditions we may neglect the pressure dependence of $\Delta\omega$; for the estimations to be given below we shall use $\Delta\omega = 2\pi \times 70$ MHz. The only pressure-dependent quantity in Eq. (8) is then Γ_0 , so that we expect I_S to be proportional to Γ_0 . This is indeed the case as is apparent from Fig. 6, where we plot-

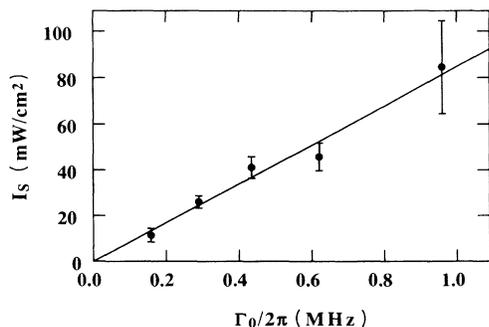


FIG. 6. Test of the predicted [Eq. (8)] linear relation between the values I_S and $\Gamma_0/2\pi = 2g\mu_B\Delta B_0/h$ obtained by fitting the width vs intensity dependences (Fig. 4) for different vapor pressures.

ted the fitted values I_S versus the fitted widths $\Gamma_0/2\pi$. The experimental slope $a = dI_S/d(\Gamma_0/2\pi)$ is $8.5(9) \times 10^{-4}$ J/m 2 . From Eq. (8) we obtain for the theoretical value of the slope

$$a = \frac{dI_S}{d(\Gamma_0/2\pi)} = \frac{\hbar\Delta\omega}{2\alpha x_{FF'}^2} = 5 \times 10^{-4} \text{ J/m}^2,$$

where we used the absorption oscillator strength of the D_2 line [23] to infer the reduced matrix element. The agreement with the experimental value is reasonable considering that the experimental assignment of laser intensities is difficult due to uncertainties in the determination of the transverse intensity distribution of the laser beam.

We next analyze the pressure dependence of ΔB_0 as introduced by Eq. (3). The extrapolated width ($I \rightarrow 0$ and $N_{Cs} \rightarrow 0$) of the nonlinear resonance is $\Gamma_{00} = 2g\mu_B\Delta B_{00}/\hbar = 2\pi \times 0.13(2)$ MHz. In this noncollisional limit the linewidth is determined by relaxations due to the finite interaction time of the atoms with the laser beam [20]. The width due to the *transverse* time of flight may be estimated as

$$\Gamma_{\text{trans}} = \Gamma_{00} = v_{\text{th}}/\langle r \rangle = 2\pi \times 0.1 \text{ MHz},$$

where $v_{\text{th}} = 2 \times 10^4$ cm/s is the average transverse thermal velocity of the atoms at $T = 400$ K, and $\langle r \rangle = 330$ μm is the average beam radius. The estimated width is in good agreement with the measured value. The pressure dependence shown in Fig. 5 has the slope $k' = (2g_F\mu_B/h)k$ of 4.5×10^{-15} MHz cm 3 , which is several orders of magnitude larger than one would expect from spin exchange collisions only [24]. We believe that the pressure broadening in our case is due to the finite *longitudinal* interaction time which is limited by the density-dependent absorption length

$$l_{\text{abs}} = \frac{1}{\sigma_{FF'}N_{Cs}},$$

where $\sigma_{FF'}$ is the peak absorption cross section of the $F \rightarrow F'$ hyperfine component. On resonance the laser beam excites mainly atoms going along the surface with average thermal velocity v_{th} . Due to the finite effective spectral width $\Delta\omega$ the beam interacts as well with atoms having an average longitudinal velocity component

$$v_{\text{eff}} = v_{\text{th}}(\pi/2)^{1/2}\Delta\omega/\Gamma_D,$$

where $\Gamma_D = \omega_0(kT/m_{Cs}c^2)^{1/2}$ is the Doppler width. In this case the relaxation is governed by the escape from a cylinder of length l_{abs} and radius $\langle r \rangle$, so that we may write the linewidth as

$$\Gamma_0 = \Gamma_{\text{trans}} + \Gamma_{\text{long}}, \quad (10)$$

where $\Gamma_{\text{long}} = v_{\text{eff}}/l_{\text{abs}}$. The slope of the density dependence in Fig. 5 may thus be expressed as

$$k' = \frac{d(\Gamma_0/2\pi)}{dN_{Cs}} = \frac{\sigma_{FF'}v_{\text{eff}}}{2\pi},$$

where the peak absorption cross section $\sigma_{FF'}$ is given (in the Doppler limit) by

$$\sigma_{FF'} = (8\pi)^{-1/2} g_{FF'} \lambda^2 \gamma / \Gamma_D .$$

Here γ is the natural width and $g_{FF'}$ the relative intensity of the $F \rightarrow F'$ hyperfine component given by Eq. (9). For our conditions we have $g_{44} = 0.164$, $\sigma_{FF'} = 6.4 \times 10^{-12} \text{ cm}^2$, and $v_{\text{eff}} = 4.7 \times 10^3 \text{ cm/s}$, so that we may estimate the slope as $k' = 5 \times 10^{-15} \text{ MHz cm}^3$, in good agreement with the experimental value $k' = 4.5 \times 10^{-15} \text{ MHz cm}^3$.

Due to optical pumping and to saturation the absorption length l_{abs} and hence the longitudinal relaxation rate Γ_0 [Eq. (10)] could depend on the light intensity. In our simple model we neglected these effects and obtained nevertheless reasonable agreement with numerical estimations.

CONCLUSION

We have studied the magneto-optical activity in the selective reflection from a glass-Cs vapor interface. When scanning the longitudinal magnetic field subnatural nonlinear resonances in the magnetorotation angle have been observed. The shape, power, and pressure broadening of these resonances were investigated. Our experimental results are in good agreement with theoretical predictions obtained for the nonlinear Faraday effect in transmission experiments when we consider longitudinal as well as transverse interaction times as a source of relaxation. The transverse and longitudinal relaxation rates were found to be proportional to the light-beam diameter and the absorption length, respectively.

On one hand it is well known [25] that the magneto-optical rotation of the plane of polarization in transmission experiments is governed by dispersion $\Phi_F \propto \text{Re}(n_+ - n_-)$, whereas in reflection experiments, it

is dominated by absorption $\Phi_F \propto \text{Im}(n_+ - n_-)$; on the other hand, we found that the observed magneto-optical resonances in our reflection experiment show the same behavior as the nonlinear Faraday resonances induced by ground-state Zeeman coherences in transmission experiments. A complete theory of the effects reported here is therefore definitely required.

From our results one readily estimates the contribution of optical-pumping effects in SR experiments on alkali-metal-element vapors, e.g., in order to realize a *linear* selective-reflection experiment using a narrow-band laser ($\Delta\omega_L < \gamma$) at a vapor pressure of a few hundred μTorr , where the absorption length is comparable to the laser beam diameter (typically 1 mm) requires laser powers less than a few μW .

The possibility to observe ground-state coherences in SR experiments opens a new way to study atom-surface interactions. For instance, one could use a pump-probe technique for studying relaxation processes near a surface [26,27]. The pump laser beam, tuned to the low-frequency wing of the atomic line, could create a ground-state coherence and a weak probe beam could detect the frequency (velocity) dependence of the atomic ground-state coherence before and after the wall collision.

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