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# Optical pumping saturation effect in selective reflection

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## Abstract

The reflection from a dielectric-atomic vapor interface displays resonance behavior in the vicinity of an atomic line (selective reflection). At normal incidence, a sub-Doppler structure arises which is due to the symmetry-breaking presence of the interface. The optical saturation of the selective reflection signal for the cesium D<sub>1</sub> line is investigated and a different saturation behavior for atoms leaving the surface and atoms moving toward the surface is found. The experimental results agree with reasonable accuracy with the predictions derived from a three-level atomic model. The saturation effect may be important for the correct interpretation of long-range atom-wall interaction and of collision-induced broadening measurements, for which selective reflection spectroscopy is particularly suitable.

## 1. Introduction

The diffuse scattering of resonance radiation (fluorescence) at a dielectric-vapor interface is accompanied by selective reflection (SR) [1]. The reflection originates in a thin vapor layer with a thickness of the order of a reduced wavelength. This opens a way to use SR spectroscopy for the study of the optical properties of the vapor boundary layer. At normal incidence, a sub-Doppler structure is observed in SR spectra [2]. This structure is due to symmetry breaking in the velocity distribution of optically polarized atoms near the interface. In Ref. [3] a frequency-modulation (FM) technique was proposed which allows to eliminate the Doppler background in SR spectra. Doppler-free FM SR spectroscopy has been used for the measurement of the collision-induced broadening and shift of atomic resonance lines [3-

6], and for the investigation of long-range atom-wall interactions [7,8].

A first investigation of nonlinear SR was performed by Vartanjan [9], who studied the optical saturation of single-beam SR for a dilute gas of two-level atoms<sup>#1</sup>. At arbitrary incidence angle the nonlinear reflection from two-level atoms in single-beam and pump-probe schemes was studied theoretically in Refs. [9,10]. Nonlinear SR in cascade three-level systems has been analyzed by Schuller et al. [12].

Saturation of SR resonances was first observed in total reflection [13] and in near-normal incidence SR experiments [14] by using a pump-probe scheme. The two-beam scheme has also been used for the detection of ground state population trapping [15] in SR experiments. In the single-beam scheme optical

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<sup>#1</sup> Here a dilute gas means that the collision-induced broadening of the atomic line is smaller than the Doppler width. For high vapor pressures, when the thermal motion of the atoms is negligible, the nonlinear effects in SR have been studied in Refs. [18,19] and references therein.

saturation of SR resonances has been first observed in Ref. [16]. In Refs. [5,15] it was noted that at low vapor pressure ground state optical pumping may contribute to the saturation coefficient. Recently nonlinear magnetic rotation due to a ground state Zeeman coherence has been observed by Weis et al. [17].

In this work we investigate the optical saturation of selective reflection due to ground state optical pumping. The experiment was performed on the cesium  $D_1$  line (894 nm), where different hyperfine (hf) SR resonances are spectrally isolated due to a large hyperfine splitting.

## 2. Theory. Nonlinear SR in the three-level atom scheme

In this paper we study the reflection of monochromatic radiation from a transparent dielectric-atomic vapor interface at normal incidence (Fig. 1) and use the theoretical approach developed in Refs. [10-12]. We consider the case of three-level atoms with ground state  $g$ , excited state  $e$  and non-absorbing state  $n$  (Fig. 2). A monochromatic optical field  $E(t, z)$  excites only

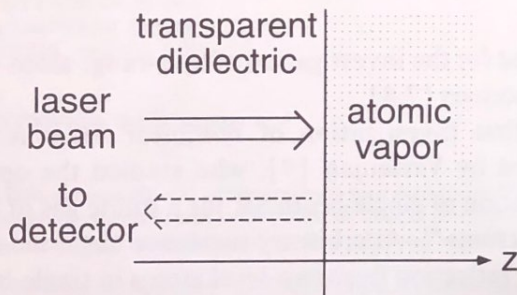


Fig. 1. Basic set-up for selective-reflection spectroscopy.

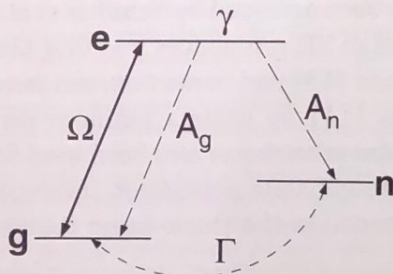


Fig. 2. Three-level atom with two ground states  $g$ ,  $n$  and an excited state  $e$ . The incident light field is resonant with the  $g \rightarrow e$  transition, but not with the  $n \rightarrow e$  transition. The ground state relaxation rate is  $\Gamma$ .

the transition  $g \rightarrow e$ . An atom in the excited state  $e$  can decay into ground state  $g$  and non-absorbing state  $n$  with decay rates  $A_g$  and  $A_n$ , respectively.  $A = A_g + A_n$  is the spontaneous decay rate of level  $e$ , while  $\gamma$  is the homogeneous (collisional and radiative) line width of the  $g \rightarrow e$  transition. Furthermore we introduce the relaxation rate  $\Gamma$  ( $\Gamma \ll \gamma, A, A_n$ ), which characterizes the decay of the  $n$  and  $g$  states.

A coordinate system is chosen with its origin at the surface and with its  $z$  axis perpendicular to the surface, such that the atoms moving towards the surface have negative  $v_z$  and the atoms leaving the surface have positive  $v_z$ . The optical field in the atomic vapor,  $E(t, z) = E_0 \exp(-i\omega t + ikz)$ , induces an atomic polarization  $p$ , which can be written as  $p(t, z, v_z) = Nd_{ge}\rho_+(t, z, v_z)$ . Here  $N$  is the number density of atoms and  $d_{ge}$  is the electric dipole moment of the optical transition  $g \rightarrow e$ . The time and spatial dependence of the incident electric field can be separated from the density matrix element  $\rho_+$  according to

$$\rho_+(t, z, v_z) = \exp(-i\omega t + ikz) \xi_+(z, v_z), \quad (1)$$

where the off-diagonal reduced matrix element  $\xi_+(z, v_z)$  is time independent. We assume complete decay of all atoms at the surface ( $z=0$ ). The optical polarization of atoms with  $v_z < 0$  can be described by a position-independent density matrix  $\xi_+(v_z < 0)$  (steady state regime), while atoms with  $v_z > 0$  are described by a density matrix  $\xi_+(z, v_z > 0)$  which depends on  $z$  (non-local optical polarization) [10].

By considering the reduced density matrix element  $\xi_+(z, v_z)$  together with its Laplace transform

$$\xi_+(p, v_z) = \int_0^\infty \exp(-pz) \xi_+(z, v_z) dz, \quad (2)$$

it was found in Ref. [10] that SR is described in the general case by the effective refraction index

$$T = 1 + T_1 + T_2, \quad (3)$$

which comprises a contribution from atoms with  $v_z < 0$ ,

$$T_1 = \frac{4\pi d_{ge} N}{E_0} \int_{-\infty}^0 W(v_z) \xi_+(v_z < 0) dv_z \quad (4)$$

and a contribution from atoms with  $v_z > 0$

$$T_2 = \frac{4\pi d_{ge} N}{E_0} \int_0^{\infty} W(v_z) (-2ik) \xi_+ (-2ik, v_z > 0) dv_z, \quad (5)$$

where  $W(v_z)$  is the normalized Maxwell velocity distribution.

The relative change in the reflection coefficient due to the presence of a dilute vapor ( $|T-1| \ll 1$ ) is given by the expression

$$\Delta R = \frac{R-R_0}{R_0} = \Delta R_1 + \Delta R_2 = -\frac{4n_0}{n_0^2-1} \text{Re}(T_1 + T_2), \quad (6)$$

where  $n_0$  is the refraction index of the transparent dielectric and  $R_0 = |n_0-1|^2/|n_0+1|^2$  is the reflection coefficient of a dielectric–vacuum interface.

By taking into account the relaxation rate  $\Gamma$  of the ground state the optical Bloch equations for the reduced density matrix  $\xi(v_z, z)$  can be written in the form:

$$\begin{aligned} v_z \partial \xi_{ee} / \partial z &= -A \xi_{ee} + i\Omega(\xi_- - \xi_+) / 2, \\ v_z \partial \xi_{gg} / \partial z &= +A_g \xi_{ee} - i\Omega(\xi_- - \xi_+) / 2 - \Gamma(\xi_{gg} - \xi_{gg}^0), \\ v_z \partial \xi_{nn} / \partial z &= +A_n \xi_{ee} - \Gamma(\xi_{nn} - \xi_{nn}^0), \\ v_z \partial \xi_+ / \partial z &= -(\gamma - i\delta) \xi_+ + i\Omega(\xi_{gg} - \xi_{ee}) / 2, \\ v_z \partial \xi_- / \partial z &= -(\gamma + i\delta) \xi_- - i\Omega(\xi_{gg} - \xi_{ee}) / 2, \\ \xi_{ee} + \xi_{gg} + \xi_{nn} &= 1, \end{aligned} \quad (7)$$

with

$$\xi_{gg}^0 = 1/2, \quad \xi_{nn}^0 = 1/2, \quad \xi_{ee}^0 = 0, \quad (8)$$

where  $\delta = \omega_0 - \omega - kv_z = \Delta - kv_z$  and  $\Omega$  is the Rabi frequency defined as

$$\Omega = E_0 d_{ge} / 2\hbar. \quad (9)$$

By using the Laplace transformation (2) and taking into account the boundary condition (decay of atoms at the surface [10–12]), we can write

$$\begin{aligned} \xi_+(p, v_z) &= (i\Omega/4p)(\Gamma + v_z p)(A + v_z p)(\gamma + v_z p + i\delta) \\ &\times \{(\Gamma + v_z p)(A + v_z p)[(\gamma + v_z p)^2 + \delta^2] \\ &+ \Omega^2(\gamma + v_z p)(v_z p + A_n/2)\}^{-1}. \end{aligned} \quad (10)$$

For very low intensity (linear reflection) the arriving and departing atoms give the same contribution to the reflectivity:

$$T_1 = T_2 = -\frac{\pi N d_{ge}^2}{\hbar} \int_{-\infty}^0 \frac{W(v_z)}{\delta + i\gamma} dv_z. \quad (11)$$

Here we would like to note that for small detuning,  $|\omega_0 - \omega| = |\Delta| \ll \Delta_D$ , ( $\Delta_D$  Doppler width), from Eqs. (11) and (6) we find that the frequency derivative of the SR coefficient takes the form

$$\partial R / \partial \omega \propto \Delta / (\Delta^2 + \gamma^2), \quad (12)$$

with the dispersive lorentzian line shape. The spectral interval between the maximum and the minimum of the curve equals  $2\gamma$ . In Ref. [4] the FM technique was applied to carry out the frequency derivation of the SR coefficient and the relation (12) was used for the estimation of the cesium D<sub>2</sub> line self-broadening coefficient.

In a two-level system ( $A_n=0, \Gamma=0$ ) we immediately obtain the same expression for the matrix element  $\xi_+$  as in Ref. [10].

The stationary value  $\xi_+(v_z < 0)$ , which is necessary for the calculation of the term  $T_1$ , is obtained from the Laplace transform relation

$$\xi_+(v_z < 0) = \lim_{p \rightarrow 0} p \xi_+(p, v_z < 0). \quad (13)$$

Therefore the value  $T_1$  is given by the expression

$$T_1 = -\frac{\pi N d_{ge}^2}{\hbar} \int_{-\infty}^0 dv_z \frac{W(v_z)(\delta - i\gamma)}{\delta^2 + \gamma^2(1+G)}, \quad (14)$$

where

$$G = (\Omega^2 / 2\Gamma\gamma) A_n / A \quad (15)$$

is a saturation parameter typical of the nonlinearity induced by optical pumping [17,20]. For atoms departing from the surface ( $v_z > 0$ ), the expression for  $T_2$ , which is given by Eqs. (5) and (10), is more complicated, but even without explicit calculation we can predict that the contributions to the reflectivity  $\Delta R_1$  and  $\Delta R_2$  can exhibit different nonlinear properties, for instance, the saturation intensities can differ. We remind that SR originates in a very thin vapor layer and atoms with  $v_z > 0$  do not spend enough time in this volume for effective optical pumping. Therefore the saturation intensity will be higher for  $\Delta R_2$  than for  $\Delta R_1$ .

### 3. Experimental arrangement

In our experiment the optical saturation of Doppler-free SR resonances is studied by means of the FM technique [4]. We record the SR FM signal of the  $6S_{1/2}(F_g=3)-6P_{1/2}(F_e=4)$  hyperfine transition at different laser intensities  $I$ . The set-up is presented in Ref. [6]. The energy level diagram of the cesium  $D_1$  line is shown in Fig. 3. In the experiment a tunable single-mode titan-sapphire laser is used which is frequency-locked to the resonance of a confocal reference cavity. The laser line width is less than 1 MHz. We modulate the laser frequency with 1 kHz and the FM-technique is used for the detection of Doppler-free SR resonances.

The linearly polarized light is sent onto a reflection glass cell which has a wedged window to avoid interference from the inner and outer surfaces. The cell is heated to a variable temperature and the corresponding atomic density  $N$  is calculated from the temperature of the coldest spot of the cell by means of the Langmuir–Taylor relation. The estimated residual Doppler broadening of the SR resonances due to nonzero incidence angle and diffraction divergence of the laser beam does not exceed 2 MHz. The laser light intensity (beam diameter 6 mm) is varied with calibrated neutral filters.

The light reflected from the glass–cesium vapor interface is detected with a photodiode and a lock-in amplifier. To obtain a frequency reference, we simultaneously record a saturation spectrum in a second cesium cell at low pressure ( $p=10^{-6}$  Torr). For frequency calibration we use two parallel probe beams that are frequency shifted relative to each other by

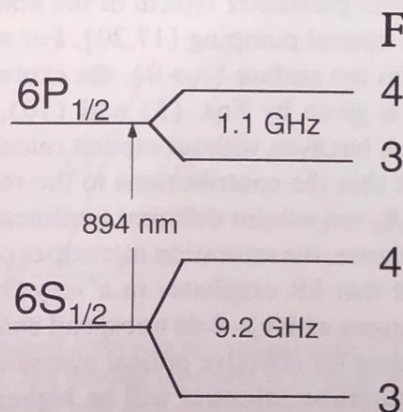


Fig. 3. Level scheme of the cesium  $D_1$  transition.

means of an acousto-optic modulator. Thus we obtain two identical saturation resonances with a well defined frequency spacing.

### 4. Experimental results and discussion

A typical Doppler-free SR resonance at very low laser intensity (linear interaction regime) is presented in Fig. 4. The observed line shape is similar to the dispersive Lorentzian line shape (12) in the central region but it is slightly asymmetrical. The shape of Doppler-free SR resonances has recently been investigated by Ducloy and Fichet in a theoretical work [7] where they took into account surface-induced long-range atom–surface interactions. They obtained line shapes that can be characterized by the homogeneous width  $\gamma$  and a dimensionless coupling constant  $A$ , which represents the surface-induced shift in units of the homogeneous line width. For our experimental conditions with  $2 \times 10^{13} \text{ cm}^{-3} \leq N \leq 5 \times 10^{13} \text{ cm}^{-3}$  in the linear interaction regime by comparison with the calculated line shapes we estimate  $A \leq 0.05$  [6]. The difference between the full width of the observed Doppler-free resonance, defined as the spectral interval between the two extrema of the curve, and the homogeneous fwhm  $2\gamma$  of the theoretical line shape functions is less than 4%, which stays within the limits of our measurement accuracy. For nonlinear reflection a theory which includes long-range atom–surface interactions is yet to be developed. To interpret the measured intensity dependence of the SR line width we do not take into account the possible contributions from long-range atom–surface interactions.

In Fig. 5 the square of the SR line width  $\Delta\nu_{\text{SR}}$  is plotted against the incident intensity for two differ-

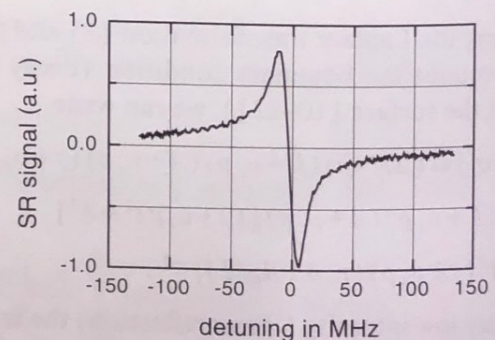


Fig. 4. Typical FM SR spectrum recorded at  $N=2 \times 10^{13} \text{ cm}^{-3}$  at low laser intensity.

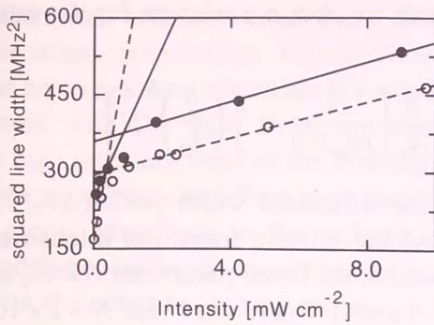


Fig. 5. Square of the measured line width plotted against the incident intensity. (○:  $N=2 \times 10^{13} \text{ cm}^{-3}$ , ●:  $N=5 \times 10^{13} \text{ cm}^{-3}$ ). Two regions at low and high intensity corresponding to two different saturation intensities can be distinguished.

Table 1  
Saturation intensities  $I_1$  and  $I_2$  for the low and high intensity regions, respectively, as obtained from the experimental results by fitting the expression  $\Delta\nu_{\text{SR}}^2 = \Delta\nu_0^2(1+I/I_k)$ ,  $k = 1, 2$ .

$N[10^{13}\text{cm}^{-3}]$	$\Delta\nu_0[\text{MHz}]$	$I_1[\text{mW cm}^{-2}]$	$I_2[\text{mW cm}^{-2}]$
2	12.8 (0.1)	0.42 (0.04)	16.7 (0.1)
5	15.8 (0.1)	1.7 (0.8)	18.6 (0.1)

ent cesium densities  $N$ . It is not possible to fit all experimental curves with only one simple saturation formula. Instead, we use the expressions

$$\Delta\nu_{\text{SR}}^2 = \Delta\nu_0^2(1+I/I_i), \quad i=1,2, \quad (16)$$

where  $I_1$  is a parameter for the low intensity region ( $I < 1 \text{ mW/cm}^2$ ) and  $I_2$  is valid for higher laser intensities  $I$ . The fitting parameters were calculated and the results are presented in Table 1. The SR resonance width  $2\pi\Delta\nu_0$  includes the natural line width  $2\gamma_{\text{nat}}=A=2\pi \times 5.3 \text{ MHz}$ , the collisional broadening induced by a constant buffer gas  $2\gamma_c=2\pi \times 4 \text{ MHz}$ , the residual Doppler broadening  $2\pi \times 2 \text{ MHz}$  and the self-broadening  $2\gamma_{\text{Cs}}=kN=2\pi \times 0.75(11) \cdot 10^{-13} \text{ N cm}^3 \text{ MHz}$  [6].

At low intensities the saturation value  $I_1$  is typical for ground state optical pumping, and at high intensities  $I_2$  is typical of the optical saturation of cycling transitions in alkaline atoms. The existence of two distinct regions with different saturation intensities in Fig. 4 can be explained by the difference in the saturation behavior of atoms arriving onto the surface ( $v_z < 0$ ) and atoms departing from the surface ( $v_z > 0$ ). Complete quenching of the atomic dipole was found for collisions of cesium atoms with un-

coated glass surfaces [21]. At low intensity the arriving atoms are optically pumped since they spend some time interacting with the light field before they reach the layer adjoining the interface where they contribute to the reflected field. The departing atoms, on the other hand, have no time for several optical cycles before they leave the dielectric–vapor boundary region and there is only the small saturation due to the excited state population process. The saturation of the SR signal in the low intensity region is governed by the optical pumping of the arriving atoms and a correspondingly small saturation intensity  $I_1$ . At high laser intensity most of the arriving atoms have already been pumped into the non-absorbing state before they reach the boundary region and only atoms with  $v_z > 0$  give rise to the SR signal.

We compare our theoretical results and experimental findings for the saturation intensity. For low laser light intensity, the Rabi frequency  $\Omega$  is much smaller than  $\gamma$  or  $A$  and by using the relation (14) for the arriving atoms ( $v_z < 0$ ), the linear response (11) for the departing atoms ( $v_z > 0$ ) and the asymptotic expressions for the derivatives, one arrives at an expression for the Doppler-free FM SR resonance

$$\frac{\partial R}{\partial \omega} \propto \text{Re}\left(\frac{\partial(T_1+T_2)}{\partial \omega}\right) \propto \left(\frac{A}{A^2+\gamma^2(1+G)} + \frac{A}{A^2+\gamma^2}\right), \quad (17)$$

with a saturation parameter

$$G = \frac{\Omega^2 A_n}{2\Gamma\gamma A} = \frac{I}{I_s}, \quad (18)$$

where the light intensity is  $I=cE^2/8\pi$ .

In the limit of small saturation,  $G \ll 1$ , the spectral width of the SR resonance can be written as

$$\Delta_{\text{SR}}^2 \approx 4\gamma^2(1+G/2). \quad (19)$$

By using a well-known relation for the absorption cross-section at a hyperfine transition  $F_g \rightarrow F_e$  [20,22]

$$\sigma_{\text{ge}}(\gamma) = \frac{4\pi\omega d_{\text{ge}}^2}{hc\gamma} = \frac{\lambda^2}{4\pi} g_{F_g F_e} \frac{2J_e+1}{2J_g+1} \frac{A}{\gamma}, \quad (20)$$

the saturation intensity can be expressed as

$$I_s = \frac{\hbar\omega}{\sigma_{\text{ge}}(\gamma)} \frac{A}{A_n} \Gamma. \quad (21)$$

In these formulae

$$g_{FF'} = \frac{(2F+1)(2F'+1)}{2I+1} \begin{Bmatrix} F & J & I \\ J' & F' & 1 \end{Bmatrix}^2 \quad (22)$$

is the normalized relative intensity of the  $F \rightarrow F'$  hyperfine component and

$$A/A_n = (g_{F_e F_g} + g_{F_e F_n}) / g_{F_e F_n} \quad (23)$$

is the branching ratio [22].

The relaxation rate of the atomic ground states in the general case is given by

$$\Gamma = \Gamma_{\text{col}} + \Gamma_{\text{int}} = \Gamma_{\text{col}} + \Gamma_{\text{trans}} + \Gamma_{\text{long}}, \quad (24)$$

where  $\Gamma_{\text{col}}$  is collisional broadening relaxation rate and  $\Gamma_{\text{int}} = \Gamma_{\text{trans}} + \Gamma_{\text{long}}$  is due to the finite interaction time of the atoms with the laser beam. The relaxation rate  $\Gamma_{\text{int}}$  is governed by the escape rate from a cylinder of absorption length  $l_{\text{abs}}$  and laser beam radius  $r$  [17].

The relaxation rate due to the transverse time of flight is given by  $\Gamma_{\text{trans}} = v_{\text{th}}/r$  [20], where  $v_{\text{th}}$  is the average thermal atomic velocity.

To calculate the longitudinal interaction time we need to know the effective longitudinal atomic velocity  $v_{\text{eff}}$  and the absorption length  $l_{\text{abs}}$ . On resonance the optical field excites mainly atoms with average velocity

$$v_{\text{eff}} = (\pi/2)^{1/2} v_{\text{th}} \gamma / \Delta_D, \quad (25)$$

with Doppler half-width  $\Delta_D = \omega (\ln 2)^{1/2} (k_B T \ln 2 / m_{\text{Cs}} c^2)^{1/2}$ .

The relaxation rate due to the longitudinal interaction time is given by the relation

$$\Gamma_{\text{long}} = v_{\text{eff}} / l_{\text{abs}} = v_{\text{eff}} \sigma_{\text{ge}}(\Delta_D) N, \quad (26)$$

where

$$\sigma_{\text{ge}}(\Delta_D) = \sigma_{\text{ge}}(\gamma) (\pi/2)^{1/2} \gamma / \Delta_D \quad (27)$$

is the peak absorption cross section of the Doppler broadened  $F_g \rightarrow F_e$  transition.

Hence we can write

$$\Gamma_{\text{long}} = (\pi/2) v_{\text{th}} \sigma_{\text{ge}}(\gamma) (\gamma / \Delta_D)^2. \quad (28)$$

Under our experimental conditions (average velocity  $v_{\text{th}} = 2 \times 10^4$  cm/s, beam radius  $r = 0.3$  cm, cesium density  $2 \times 10^{13} \text{ cm}^{-3} \leq N \leq 5 \times 10^{13} \text{ cm}^{-3}$ ), the contribution from cesium–cesium spin-exchange collisions [23]  $\Gamma_{\text{col}} \sim 2\pi \times 10^{-16} \text{ N MHz cm}^{-3}$  is negligible compared to  $\Gamma_{\text{int}}$ .

As a result we obtain a relation for the saturation intensity

$$I_s = \hbar \omega v_{\text{th}} \frac{A}{A_n} \left[ \frac{1}{r \sigma_{\text{ge}}(\gamma)} + \frac{\pi}{2} \left( \frac{\gamma}{\Delta_D} \right)^2 N \right]. \quad (29)$$

By using the measured width  $\gamma = 2\pi(\Delta\nu_0/2)$  from Table 1 and the relative transition intensities  $g_{34} = 0.33$ ,  $g_{44} = 0.23$ , the fitted parameter  $I_1 = 2I_s$  is calculated as 0.6 and 1.9 mW cm<sup>-2</sup> for  $N = 2 \times 10^{13}$  and  $5 \times 10^{13} \text{ cm}^{-3}$ , respectively. These estimated saturation intensities are in a good agreement with the experimental data observed at low laser intensity. The saturation of atoms with  $v_z > 0$  can be roughly estimated from the effective interaction time  $\Delta t$ . We remind that SR is formed in a thin vapor layer of the order of a reduced wavelength  $\lambda/2\pi$ . On resonance the interaction time is approximately

$$\Delta t \approx \frac{\lambda}{2\pi v_{\text{eff}}} \approx \frac{1}{\gamma}. \quad (30)$$

The saturation intensity for  $\Delta R_2$  is hence given by

$$I_s \approx \frac{\hbar \omega}{\sigma_{\text{ge}}(\gamma)} \gamma. \quad (31)$$

With the measured width  $\gamma = 2\pi \times 3$  MHz we estimate  $I_s \approx 10$  mW cm<sup>-2</sup>, which is in qualitative agreement with our fitting values for  $I_2$  at high laser intensities.

The results confirm the prediction that at low light intensities the saturation of SR spectra is mainly associated with ground state optical pumping.

## 5. Conclusions

The earlier developed theory [9,10] of nonlinear selective reflection for two-level atoms cannot explain the observed saturation behavior of the reflection from a dielectric–atomic vapor interface at low and high laser intensity, as observed for cesium vapor. In this paper we propose a straightforward theoretical model for the nonlinear reflection which takes into account optical pumping effects in three-level atomic systems.

The observed different saturation behavior at low and high laser intensities can be explained by the different optical saturation of atoms moving onto the glass surface and atoms moving away from the glass

surface. Atom–wall collisions randomize the orientation of the atomic polarization. Hence at low intensity only the arriving atoms, which are in a steady state of interaction with the light field, are saturated, whereas at high intensity most of the arriving atoms are pumped into the non-absorbing state and only the departing atoms contribute to the reflectivity. We have derived formulae for the saturation intensity associated with the optical pumping process in SR. By using asymptotic relations for the SR resonance width reasonable agreement between the theoretical predictions and the experimental data has been achieved.

The results obtained here may be used to estimate the optical saturation of SR in a near linear regime which might be important for the correct interpretation of the collision-induced broadening and shift and also of long-range atom–wall interaction measurements [4–8]. Additional information could be obtained by using an intensity modulation (IM) technique. In this case at low laser intensity the saturation is mainly due to optical pumping and it will be possible to observe only the contribution from atoms moving onto the uncoated glass surface. The optical saturation effect in SR could also be applied to the study of the disorientation of atomic dipoles at coated surfaces (silan or metal coating [24]).

For future studies a more detailed description of the SR line shapes taking into account a theoretical model of long-range atom–wall interactions will be desirable. This interaction is responsible for the distortion and the frequency shift of SR resonances [7,8].

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