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Selective reflection from a dense atomic vapor

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A theory of selective reflection (SR) of a light beam from the interface of a dielectric medium and a dense atomic vapor is presented. Following another earlier work on this subject [J. Guo *et al.*, Opt. Commun. **110**, 732 (1994)], we calculate the atomic density matrix and the transmitted field in the vapor self-consistently. We extend our previous theory to include an attractive atom-dielectric wall interaction and multiple atomic transitions. We examine the limits at which the conventional SR theories, which are based on the assumption of a plane-wave field in the vapor, start to fail. It is shown that at or above vapor densities of order $10^{17}/\mathrm{cm}^3$, the frequency shifts of the SR line shape due to the local-field correction (Lorentz-Lorenz shift), the atom-wall interaction, and the nonexponential attenuation of the field in the vapor can have comparable orders of magnitude, and the proper determination of all these effects is essential for calculating the correct SR signals. A comparison of the theoretical SR spectra with an experimental result for a Rb vapor with a density of order $10^{17}/\mathrm{cm}^3$ is also presented.

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I. INTRODUCTION

When a light beam is incident on the interface between a dielectric medium and an atomic vapor, the reflectivity signal of the light exhibits resonance behavior as the light frequency is tuned across an atomic transition frequency. Selective reflection (SR) spectroscopy, which is based on this type of resonance phenomenon, can be used for studying the width and frequency of an atomic or molecular transition [1–3], as well as the interaction between the atoms and the dielectric surface [4,5]. In contrast to conventional absorption spectroscopy, which measures the intensity of a transmitted light, the reflected field is monitored in SR spectroscopy, which makes it especially attractive in the case of an optically thick medium, where probe transmission techniques are not applicable. For example, a number of SR experiments [6–9] have been carried out, in efforts to measure resonance line broadening and shift of certain atomic transitions, which become significant only in the limit of dense atomic vapors.

Modern theories of SR spectroscopy have been emerging during the last few decades [5,10–14]. The majority of these theories are in a low-density limit, as may be defined by $Nk^{-3} \ll 1$, where N is the atomic vapor density, $k = 2\pi/\lambda$ is the laser wave vector, and λ is the laser wavelength. To give an idea of the order of magnitude of the vapor density, $Nk^{-3} = 1$ corresponds to a density N of order $10^{15}/\mathrm{cm}^3$ for alkali atoms. In the low-density limit, one calculates the SR signals to first order in Nk^{-3}. An underlying assumption of these SR theories is that the transmitted field in the atomic vapor is a plane wave of constant or exponentially attenuating amplitude. In the case of an exponentially attenuating plane-wave field, the attenuation constant may be given by the steady-state absorption coefficient of the field in the vapor. This assumption may be true at sufficiently low vapor densities. At moderate to high vapor densities such that $Nk^{-3} \geqslant 1$, however, the transmitted field must be evaluated more accurately. By calculating the field and the atomic density matrix self-consistently, we have shown recently [15] that the transmitted field attenuates in a nonexponential fashion in the case of a dense vapor.

The nonexponential attenuation of the field is related to a transient component of the polarization density of the atomic vapor. At thermal temperatures, the atom-wall collisions are diffuse in nature, which completely quench the atomic excitation and return the atoms to their ground states. As an atom moves away from the dielectric wall following a collision, it interacts with the field in a transient manner, and the density matrix of the atom possesses a transient component, which decays away after a certain time scale (or a certain distance from the dielectric wall). This transient aspect of the atom-field interaction is known to lead to a sub-Doppler structure in the SR line shape at sufficiently low vapor densities [10]. Owing to the same transient component in the polarization density, the field in the vapor need not decay exponentially as in the steady state. Since the field in the vapor depends on the polarization density of the vapor, it is necessary to calculate the transmitted field and atomic density matrix self-consistently in this case. For vapor densities such that $Nk^{-3} \geqslant 1$, it was shown in Ref. [15] that the amplitude of the field can exhibit an oscillatory behavior near the wall, and that there exists a line shift (called the "transient shift" below) between the SR signals calculated self-consistently and that calculated by assuming a plane-wave field in the vapor. This transient shift has a magnitude which can be comparable with, for example, the well-known Lorentz-Lorenz shift [16], and it increases approximately linearly with the vapor density N.

In this paper, we generalize our previous theory [15] to include the long-range atom-wall interaction, and to include multiple atomic transitions which may exist in a realistic atomic vapor with hyperfine structure, and which may overlap with each other in the case of a dense vapor due to large

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resonance line broadening. We make a detailed comparison between the results obtained respectively by assuming an exponentially attenuating field and by solving the field self-consistently at various vapor densities. A major purpose of this paper is to show that at high vapor densities such that $Nk^{-3} \gg 1$, the line shifts due to the Lorentz-Lorenz correction, the atom-dielectric wall interaction, and the nonexponential decay of the field in the vapor are all of comparable orders of magnitude, and one must evaluate them all properly in order to determine, for example, the resonance shift from the SR signal of a dense atomic vapor.

This paper is organized as follows. In Sec. II, we outline our self-consistent calculation of the SR signal, including the Lorentz-Lorenz corrections, the atom-wall interaction, and multiple atomic transitions. In Sec. III, we discuss the numerical implementation of the theory, and introduce a simplification procedure based on a Taylor expansion that facilitates numerical computations at vapor densities of order $Nk^{-3} \geqslant 100$. In Sec. IV, the effects of nonexponential attenuation of the field, the atom-wall interaction, and the Lorentz-Lorenz correction are analyzed respectively, and modifications to the SR line shape introduced by these effects are addressed separately. A preliminary comparison between different theoretical spectra and an experimental result of a Rb vapor with a density $N = 1.53 \times 10^{17}/\mathrm{cm}^3$ is also presented. We conclude in Sec. V with a discussion of the validity conditions of the earlier theories.

II. THE MAXWELL-BLOCH EQUATIONS

We assume that a linearly polarized laser field of amplitude E_i is propagating in the positive z direction, and it is normally incident on the interface between a dielectric medium of refractive index n_g and an atomic vapor of density N. The atoms are assumed to have multiple resonance transition frequencies (due usually to hyperfine structures), and each optical transition is modeled as two-level with the ground and excited states denoted by $|1\rangle$ and $|2\rangle$, respectively. The transmitted field in the vapor, $E(z)$, is assumed to be sufficiently weak such that

$$\chi \ll \Gamma, \quad \Gamma_h = \chi^2/\Gamma \ll \Gamma_g, \tag{1}$$

where $\chi = E(z)d/\hbar$ is the Rabi frequency and d is the atomic dipole moment along the linear polarization of the field. $\Gamma = 4d^2k^3/\hbar$ is the atomic excited state decay rate. Γ_h is the on-resonance optical pumping rate between the hyperfine ground states, and Γ_g is a characteristic relaxation rate for the atomic ground state at thermal equilibrium. In this weak-field limit, one can neglect the effects of saturation and the modification to the atomic ground-state populations due to optical pumping, and one is left with the equation for the off-diagonal density-matrix element ρ_{21} (or ρ_{12}) only. Moreover, given a transmitted field $E(z)$, the multiple atomic transitions can be treated independently, and the overall polarization density of the vapor is simply given by a linear superposition of the polarizations corresponding to the individual transitions.

The steady-state (i.e., $\partial/\partial t = 0$) equation for the off-diagonal density-matrix element ρ_{21}^j of the jth atomic transition is given by

$$v\frac{d}{dz}\rho_{21}^j(z,v) = -\left[\gamma_{21} - i\Delta_j + i\frac{C\gamma}{(kz)^3}\right]\rho_{21}^j(z,v) - i\eta_j\chi_{loc}(z)w(v), \tag{2}$$

where v is the atomic velocity in the z direction, and $\Delta_j = \omega - \omega_j$ is the detuning of the incident field frequency ω from the jth atomic transition frequency ω_j. The left-hand side of Eq. (2) is equivalent to the Doppler effects. The rate γ_{21} represents the homogeneous linewidth of the atomic transition, given by

$$\gamma_{21} = \frac{\Gamma + \Gamma_c}{2}, \tag{3}$$

where Γ_c is the resonance broadening [17]. In our calculation, it is assumed that Γ_c is given by

$$\Gamma_c = 5.6(Nk^{-3})\Gamma, \tag{4}$$

which corresponds to the resonance broadening of the rubidium D1 line [18]. The constant η_j in Eq. (2) represents the fraction of atomic population participating in the jth transition at thermal equilibrium (as given by the statistical weight factor), multiplied by the Clebsh-Gordan coefficient associated with this transition. $w(v)$ is the thermal velocity distribution function of the vapor, assumed to be a Maxwellian with a most probable speed u (ku is the Doppler width). The effective Rabi frequency, $\chi_{loc}(z) = E_{loc}(z)d/\hbar$, is related to the local field $E_{loc}(z)$, which is given by [16]

$$E_{loc}(z) = E(z) + \frac{4\pi}{3}P(z), \tag{5}$$

where $P(z)$ is the polarization density of the medium, given in terms of ρ_{21}^j as

$$P(z) = -Nd\left\langle \sum_j \rho_{21}^j(z,v)\right\rangle_v . \tag{6}$$

We have also included in Eq. (2) a long-range Van der Waals interaction potential between the individual atoms and the dielectric wall [5], which is represented by the frequency shift

$$\delta_{VW} = -\frac{C\gamma}{(kz)^3}, \tag{7}$$

where $\gamma = \Gamma/2$, and the dimensionless coefficient C represents the strength of the atom-wall interaction.

The solutions to Eq. (2), with a boundary condition at $z = 0$ due to diffuse atom-wall collisions, and at $z = \infty$ as

$$\rho_{21}^j(0,v>0) = 0\,, \quad \rho_{21}^j(\infty,v<0) = 0\,, \tag{8}$$

are given by

$$\rho_{21}^j(z,v>0) = -i\frac{\eta_j d}{\hbar}\int_0^z \exp\left[-\frac{\gamma_{21} - i\Delta_j}{v}(z-z') - \frac{iC\gamma}{2k^3v}\left(\frac{1}{z^2} - \frac{1}{z'^2}\right)\right]\frac{W(v)}{v}E_{loc}(z')dz', \tag{9}$$

$$\rho_{21}^{j}(z,v<0) = -i\frac{\eta_j d}{\hbar}\int_{\infty}^{z}\exp\left[-\frac{\gamma_{21}-i\Delta_j}{v}(z-z') - \frac{iC\gamma}{2k^3 v}\left(\frac{1}{z^2}-\frac{1}{z'^2}\right)\right]\frac{W(v)}{v}E_{loc}(z')dz'. \tag{10}$$

The propagation of the transmitted field is determined by the polarization density of the vapor $P(z)$. As shown in Ref. [15], the transmitted field can be solved from the following integral equation:

$$E(z) = \frac{2n_g}{n_g+1}E_i e^{ikz} - \frac{n_g-1}{n_g+1}2\pi ik\int_0^{\infty}P(z')e^{ik(z+z')}dz' + 2\pi ik\int_0^{\infty}P(z')e^{ik|z-z'|}dz', \tag{11}$$

which is formally equivalent to the Maxwell equations. The reflected field in the dielectric is also found to be given by

$$E_r = \frac{n_g-1}{n_g+1}E_i + \frac{4\pi ik}{n_g+1}\int_0^{\infty}P(z')e^{ikz'}dz'. \tag{12}$$

The reflection coefficient, R, is then given by

$$R = \left|\frac{E_r}{E_i}\right|^2. \tag{13}$$

III. NUMERICAL IMPLEMENTATION

As described in Ref. [15], one can substitute Eqs. (9) and (10) into Eq. (11), and obtain a linear set of equations for the values of the local field

$$E_{loc}(i) = E(z_i) + \frac{4\pi}{3}P(z_i) \tag{14}$$

on a given spatial grid $z_i = ih$, $i = 0,1,\ldots,N$. In order for the result to fully converge, the step size of the grid, h, has to be sufficiently small to resolve the rapid spatial oscillation of the integrand in Eqs. (9) and (10). A rough estimate shows that the step size h should be chosen such that $kh \ll ku/\max(\gamma_{21},|\Delta_j|)$. In the case of high vapor densities such that $\gamma_{21} \gg ku$, the required small grid size h results in a large number of grid points, and makes it computationally extensive to solve the linear equations for the field at the grid points.

In order to overcome this practical difficulty, one can improve the above straightforward procedure as follows. In the integral expressions Eqs. (9) and (10) for ρ_{21}, one recognizes that in the limit of $\gamma_{21} \gg ku$, the exponential function $\exp[-\gamma_{21}(z-z')/v]$ in the integrand represents approximately a δ function centered at $z=z'$. On the other hand, the transmitted field $E(z)$ is a relative smooth function of z, and only varies appreciably over a length comparable with $\lambda/2\pi$. Therefore, in the case of dense vapors such that $\gamma_{21} \gg ku$, one can expand the field $E(z')$ in the integrals of Eqs. (9) and (10) around z. Up to second order in the Taylor expansion, one arrives at the following expression for the velocity-averaged atomic coherence $\langle \Sigma_j \rho_{21}^j(z,v)\rangle_v$:

$$\left\langle \sum_j \rho_{21}^j(z,v)\right\rangle_v = \xi^{(0)}(z)E_{loc}(z) + \xi^{(1)}(z)E'_{loc}(z) + \xi^{(2)}(z)E''_{loc}(z), \tag{15}$$

where the expansion coefficients $\xi^{(m)}(z)$, $m=0,1,2$, are given by

$$\xi^{(m)}(z) = \sum_j \frac{-i\eta_j d}{\hbar}\left(\int_0^{\infty}dv\int_0^z dz' + \int_{-\infty}^{0}dv\int_{\infty}^{z}dz'\right) \times \exp\left[-\frac{\gamma_{21}-i\Delta_j}{v}(z-z') - i\frac{C}{2v}\left(\frac{1}{z^2}-\frac{1}{z'^2}\right)\right] \times \frac{W(v)}{v}f^{(m)}(z-z'), \tag{16}$$

where

$$f^{(0)}(z-z') = 1, \quad f^{(1)}(z-z') = (z-z'),$$
$$f^{(2)}(z-z') = \frac{(z-z')^2}{2}. \tag{17}$$

The above coefficients can be calculated numerically, with sufficiently small spatial step size h_z for the integration in order to evaluate the approximate δ function in Eqs. (9) and (10). However, the grid size h used for calculating the field can be much greater than h_z, since the field varies smoothly over a fraction of a wavelength. The first- and second-order derivatives in Eq. (15), $E'_{loc}(i)$ and $E''_{loc}(i)$, can be evaluated according to the standard central difference rules:

$$E_{loc}'(i) = \frac{E_{loc}(i+1)-E_{loc}(i-1)}{2h},$$
$$E''_{loc}(i) = \frac{E_{loc}(i+1)+E_{loc}(i-1)-2E_{loc}(i)}{h^2}. \tag{18}$$

Upon substitution of Eqs. (15) and (18) into Eq. (11), one obtains a linear set of equations for $E_{loc}(i)$, the solutions of which lead to the reflected field E_r through Eqs. (9), (10), and (12).

IV. RESULTS AND ANALYSIS

In the following, we analyze the SR signal calculated self-consistently by solving Eq. (11) for the field and Eqs. (9) and (10) for the density matrix simultaneously. The SR signal is calculated as the relative change in the reflectivity coefficient R, i.e., $(R-R_0)/R_0$, where $R_0=(n_g-1)^2/(n_g+1)^2$, is the reflectivity of light in the absence of the atomic vapor. The refractive index n_g is assumed to be 1.76 for the results given below, which corresponds to that of a sapphire dielectric medium.

A. Effects of the nonexponential attenuation of the field: The transient shift

In this subsection, we consider the case of a single atomic resonant transition, and the detuning between the field and the atomic transition is simply denoted by Δ. As shown in

Ref. [15], for moderately dense vapors ($Nk^{-3} \geqslant 1$), the transmitted field attenuates non-exponentially in the vapor, whose amplitude $|E(z)|$ can exhibit a spatial oscillation with an oscillation period of order λ. In fact, the first- and second-order derivatives of $|E(z)|$ with respect to z are comparable with each other in magnitude, indicating the slowly varying envelope approximation is not applicable in this case. The nonexponential decay of the field is directly related to the transient response of the atoms to the driving field after their diffuse collisions with the dielectric wall at thermal velocities. After each collision with the wall, the atoms are assumed to be completely deexcited. As they leave the wall with velocity $v > 0$, the atoms undergo transient Rabi oscillations under the influence of the transmitted field $E(z)$. Due to the existence of the transient terms in the polarization density of the vapor, the field itself does not attenuate exponentially.

In the absence of the diffuse wall collisions, the atoms respond to the transmitted field in a steady-state fashion, and the field $E(z)$ attenuates according to the well-known exponential Beer's law, given by

$$E(z) = E_0 e^{ikz - \alpha z}, \tag{19}$$

where the linear absorption coefficient α is given by

$$\alpha = \pi^{3/2}\left(\frac{\Gamma}{2ku}\right)(Nk^{-3}) I\left(\frac{i\gamma_{21} + \Delta}{ku}\right)k, \tag{20}$$

where the function $I(z)$ is defined as

$$I(z) = \frac{i}{\pi}\int_{-\infty}^{\infty} \frac{\exp(-t^2)}{z - t} dt, \tag{21}$$

which represents the familiar Voigt profile of linear absorption. In the presence of diffuse atom-wall collisions, the density matrix of the atoms leaving the wall exhibit transient Rabi oscillations due to the exchange of energy between the field and the atoms, which results in an oscillatory pattern of the field itself. The field attenuation patterns in the vapor are shown in Fig. 1 for several different vapor densities. The field amplitude $|E(z)|$ in Fig. 1 is in units of $E_s = \hbar\gamma/d$. The oscillatory behavior is most apparent at moderate vapor densities where Nk^{-3} is of order unity, but the nonexponential behavior persists.

The nonexponential character of the field attenuation shown in Fig. 1 leads to a line shift in the corresponding SR signal. In Figs. 2 and 3, we show the SR line shapes calculated self-consistently (SC) according to the formalism outlined in the preceding section, but without the inclusion of the local field correction and the wall shift, for different vapor densities in the ranges $Nk^{-3} < 1$ and $Nk^{-3} \geqslant 100$, respectively. As a comparison, we also show by the dashed line the results calculated by assuming an exponentially attenuating field in the vapor given by Eq. (19) at the corresponding densities. For the smallest vapor density $Nk^{-3} = 0.2$ shown in Fig. 2, the line shapes calculated using these two methods largely agree with each other. For higher vapor densities, however, these two results start to differ from each other. We denote the frequencies at which $R = R_0$ as δ_0^{SC} and δ_0^{EXP}, which are calculated self-consistently and by assuming an exponentially attenuating field, respectively. In Fig. 4, the difference between the two frequencies, $\delta_0^{SC} - \delta_0^{EXP}$, is plotted as a function of Nk^{-3} for $Nk^{-3} \geqslant 100$. As can be seen from Fig. 4, in this limit of high vapor densities, the relative shift between δ_0^{SC} and δ_0^{EXP} scales linearly with Nk^{-3}, i.e., $\delta_0^{SC} - \delta_0^{EXP} = A_{tr}(Nk^{-3})\gamma$, where the coefficient A_{tr} is found numerically to have a value 1.04 for the range of vapor density presented in Fig. 4.

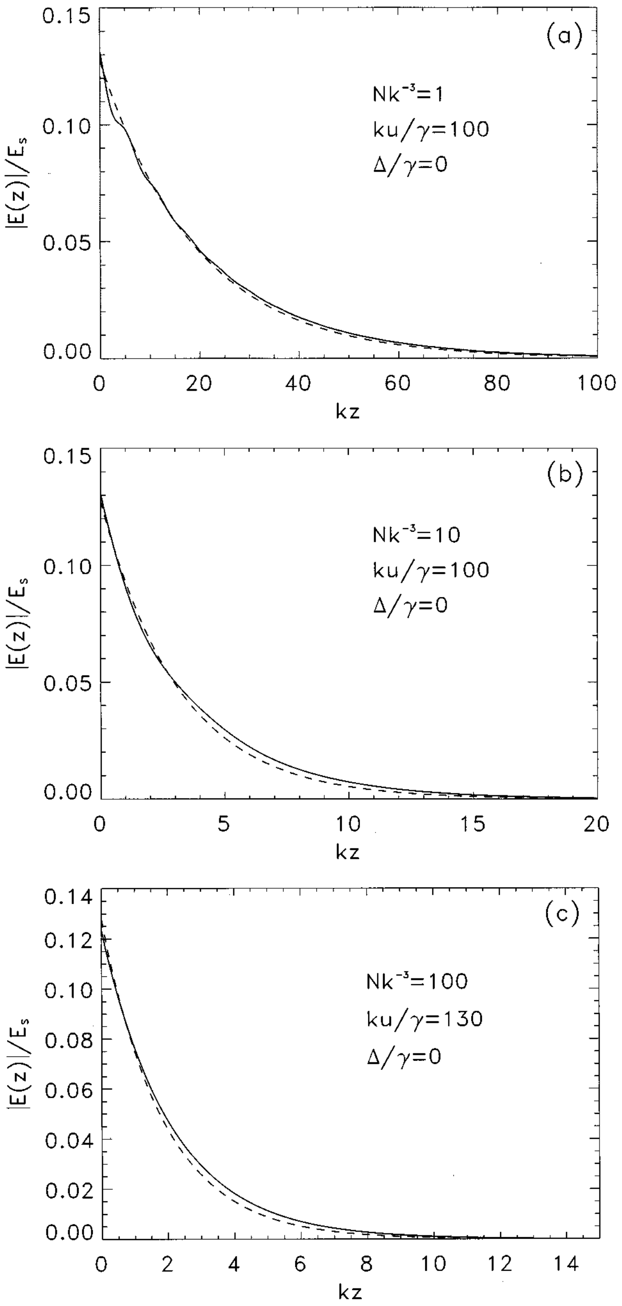

FIG. 1. The nonexponential attenuation of the field E_z in the vapor. The vapor densities are given by (a) $Nk^{-3} = 1$, (b) $Nk^{-3} = 10$, and (c) $Nk^{-3} = 100$, respectively. The exponentially decaying fields given by Eq. (19) are also plotted as dashed lines for comparison.

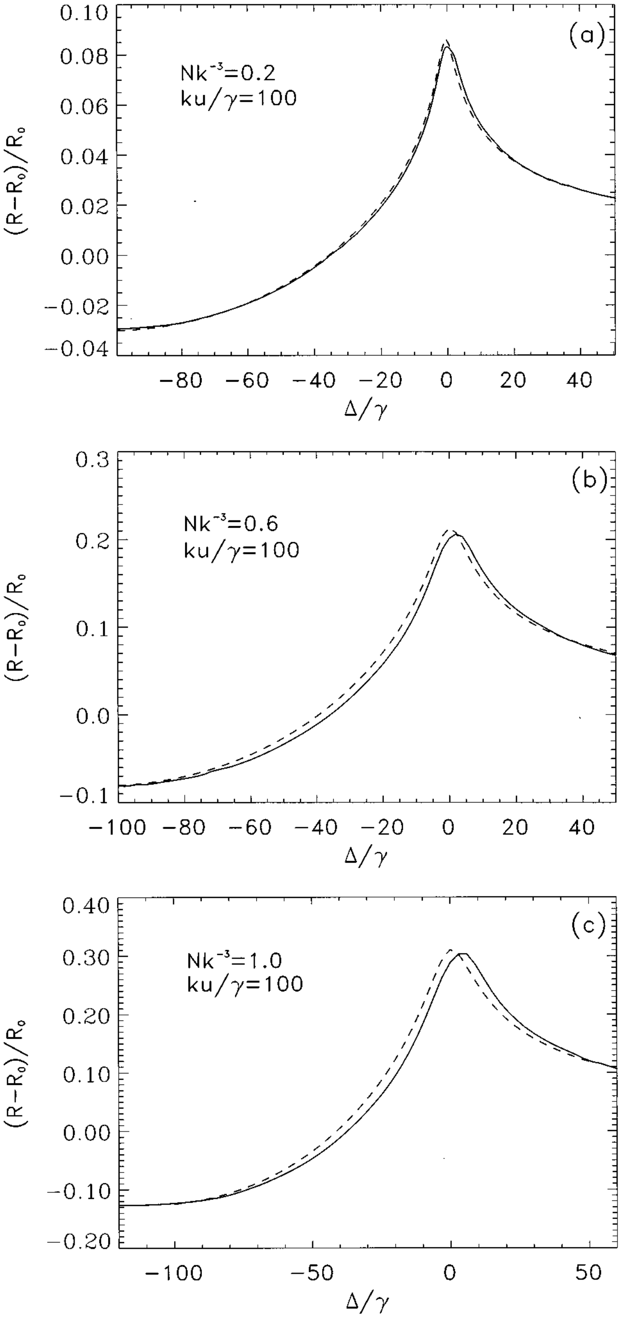

FIG. 2. SR signals calculated self-consistently (solid line) and by assuming an exponentially decaying field (dashed line) for (a) $Nk^{-3}=0.2$, (b) $Nk^{-3}=0.6$, and (c) $Nk^{-3}=1$.

The fact that the result obtained by assuming a plane-wave field in the vapor remains accurate in the limit of $Nk^{-3} \ll 1$, as is evident in Fig. 2(a), can be understood as follows. For sufficiently low vapor densities, the transient component of the atomic polarization density decays after a characteristic distance $l_t = u/\gamma$ away from the wall, as is evident from Eq. (9). The absorption length of the field, which is on the order of $l_a = u/(Nk^{-3}\gamma)$ for low vapor densities, is much longer than l_t. As a result, the transient polarization component has negligible effect, and the field attenuates exponentially in the vapor as in the steady-state case. For higher densities such that $Nk^{-3} \geqslant 1$ and $\gamma_{21} \approx \Gamma_c/2$, the distance l_t, which is now proportional to $u/(Nk^{-3}\gamma)$, is comparable in magnitude with the absorption length l_a, and the transient polarization component becomes significant for the propagation of the field. In this case, a self-consistent calculation of the field and the polarization is necessary.

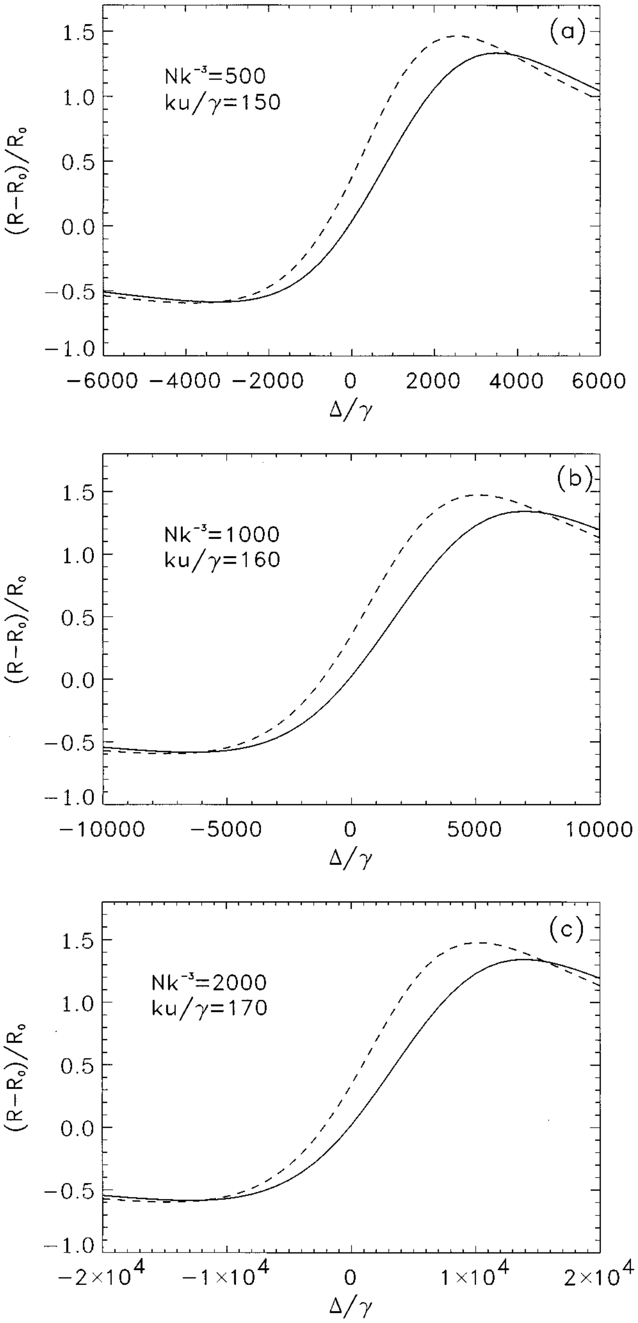

FIG. 3. SR signals calculated self-consistently (solid line) and by assuming an exponentially decaying field (dashed line) for (a) $Nk^{-3}=500$, $ku/\gamma=150$, (b) $Nk^{-3}=1000$, $ku/\gamma=160$, and (c) $Nk^{-3}=2000$, $ku/\gamma=170$.

B. Effects of van der Waals interaction between the atoms and the dielectric: The wall shift

In the presence of the dielectric wall, each atom in the vapor can interact with its own image induced by the wall. This van der Waals interaction leads to a shift in the resonance frequencies of the atomic transitions, which varies as

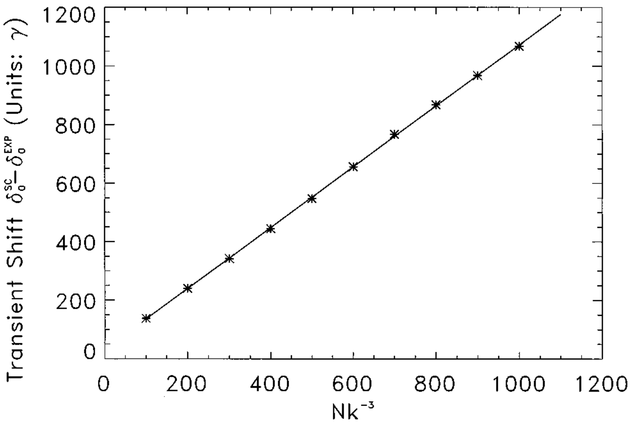

FIG. 4. The relative shift between the two frequencies δ_0^{SC} and δ_0^{EXP} as a function of N. The solid line is a linear fit $\delta_0^{SC}-\delta_0^{EXP}=1.04(Nk^{-3})\gamma$.

$1/(kz)^3$ when the atoms are within a distance $\lambda/2\pi$ of the wall. In the low-density regime of $Nk^{-3}\ll 1$, Ducloy *et al.* [5] have shown that this atom-wall interaction leads to both a distortion and a frequency shift of the SR line shape, which can be measured to determine the characteristics of the atom-wall interaction.

It has been speculated that the relative importance of the atom-wall interaction would decrease with the increase of vapor density [7], since the resonance broadening and shift of the atomic transition increase linearly with N, and they are expected to become the dominant spectral features in the limit of dense vapors. However, as we show below, even at the limit of $Nk^{-3}\gg 100$, the line shift due to the atom-wall interaction can be comparable with the resonance shift, and must be included when calculating the SR signal.

For illustration, we consider the case of a single resonance transition and study the effect of the atom-wall interaction. The atom-wall interaction coefficient C in Eq. (7) is given a value of $C=0.2$, which is a typical value for alkali atoms. In Fig. 5, we show the SR line shapes calculated self-consistently with and without the atom-wall interaction at vapor densities given by $Nk^{-3}=500$, 1000, and 2000, respectively. The results in Fig. 5 are independent of ku since $\Gamma_c\gg ku$. At these high vapor densities, which in the case of Rb atoms correspond to number densities N of order $10^{17}-10^{18}/\mathrm{cm}^3$, the effect of the atom-wall interaction remains significant, and it results in both a frequency shift and a slight distortion of the line shapes, as are evident in Fig. 5. The wall shift δ_{wall}, defined as the shift of the frequency at which $R=R_0$ with the addition of the atom-wall interaction, is found to be approximately proportional to the vapor density N, as shown in Fig. 6. It can be fitted by a linear function $\delta_{wall}=A_{wall}(Nk^{-3})\gamma$, where the coefficient A_{wall} has the value of -0.6 for $Nk^{-3}\gg 100$.

It may be somewhat surprising that the wall shift remains important at high vapor densities. As the atom density increases, the effective atom-field interaction region comes closer to the wall, and the strength of the wall shift, which varies as $1/(kz)^3$, also becomes greater. When the vapor density is sufficiently high, the resonantly broadened width γ_{21} of the atomic transition becomes much greater than the Doppler width ku, and the attenuation length of the field in the vapor is expected to reach a minimum value. Therefore one may expect the wall shift to remain unchanged thereafter. Such an argument is not necessarily true, however. One notices that the atom-wall interaction terms in Eqs. (9) and (10) introduce an extra phase shift to the polarization density $P(z)$. When substituted into Eq. (12), this extra phase factor results in the wall shift in the SR line shape. Since the polarization density $P(z)$ is proportional to the vapor density N, it can be expected that the wall shift also increases with N in the calculation of the transmitted and reflected fields. As a result, it is necessary to include the atom-wall interaction

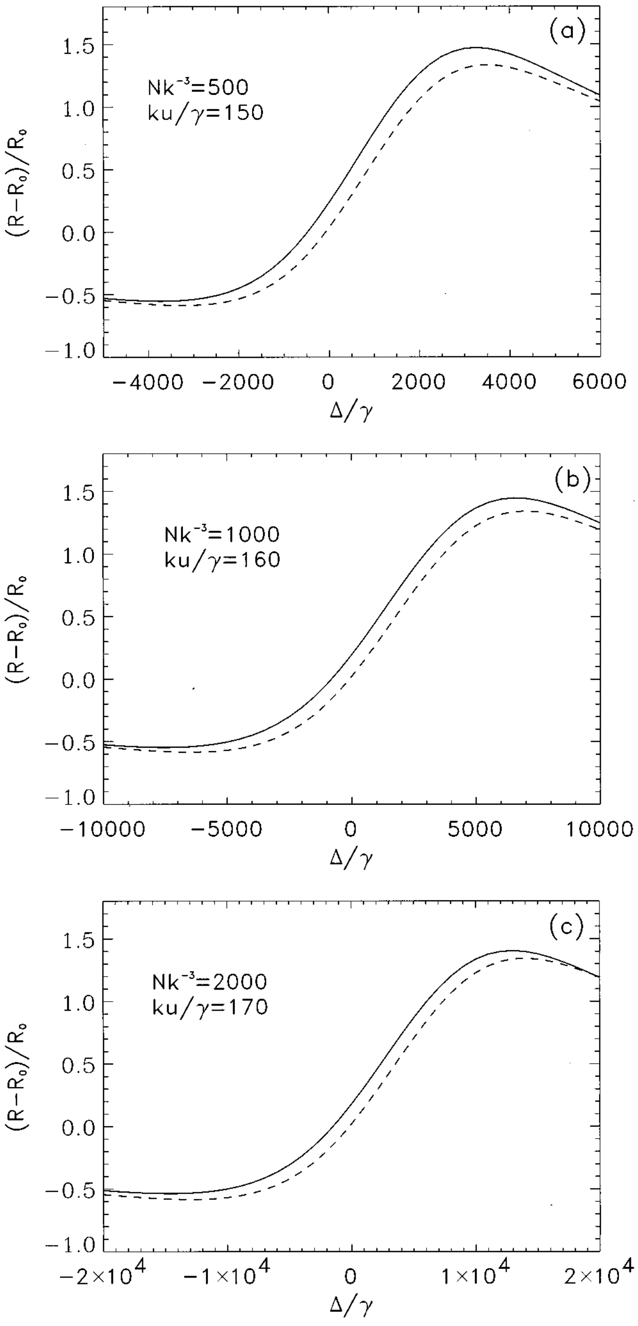

FIG. 5. Effect of the atom-wall interaction. The solid and the dashed lines represents results calculated with and without the atom-wall interaction, respectively. The vapor densities are given by (a) $Nk^{-3}=500$, (b) $Nk^{-3}=1000$, and (c) $Nk^{-3}=2000$.

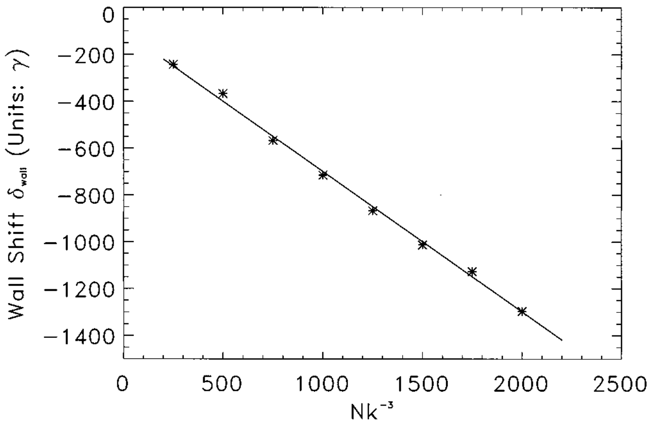

FIG. 6. The wall shift δ_{wall} as a function of vapor density N. The solid line is a linear fit $\delta_{wall} = -0.6(Nk^{-3})\gamma$.

term even in the limit of high vapor densities ($Nk^{-3} \gg 100$).

C. Effects of the local field correction: The Lorentz-Lorenz shift

The atoms in a dense vapor interact with both the macroscopic field E and the scattered field from other atoms, which is included as a local field correction in Eq. (5). The addition of the local field correction, or the Lorentz-Lorenz term in the Bloch equations, results in merely a redshift of the SR line shape, or Lorentz-Lorenz shift, in the conventional SR theories for a homogeneously broadened medium, which is given by [16]

$$\delta'_{LL} = -\frac{2\pi}{3}(Nk^{-3})\gamma. \tag{22}$$

However, when the field and the atomic density matrix are calculated self-consistently as in this case, it is not obvious that such a conclusion will still hold.

To investigate the effects of the local field correction, we again consider the case of a single resonance transition. In Fig. 7, we plot the SR spectra calculated self-consistently with and without the Lorentz-Lorenz term, respectively, for a vapor density given by $Nk^{-3} = 500$. As shown in Fig. 7, the main effect of the Lorentz-Lorenz correction is to shift the line shape towards the red side, by an amount given approximately by Eq. (22). One also notices that the peak height of the signal with the local field correction is slightly smaller than that calculated without this correction.

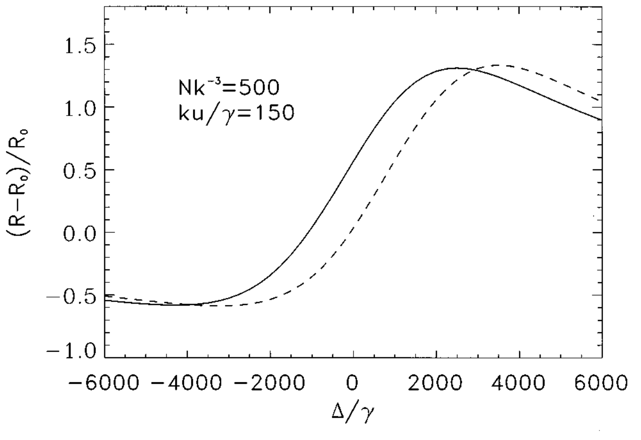

FIG. 7. SR signal with (solid line) and without (dashed line) the local field correction. The vapor density is given by $Nk^{-3} = 500$.

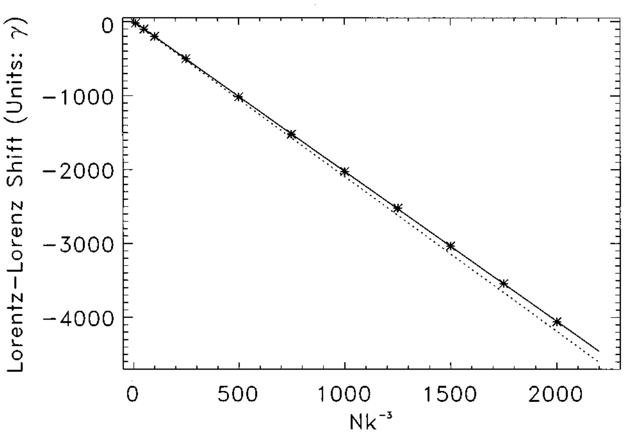

FIG. 8. The Lorentz-Lorenz shift as a function of the vapor density N. The solid line is a linear fit of the data, and the dotted line represents the shift given by Eq. (22).

In Fig. 8, we plot the Lorentz-Lorenz shift δ_{LL} calculated self-consistently for vapor densities in the range between $Nk^{-3} = 10$ and $Nk^{-3} = 2000$, corresponding to $10^{15}/\text{cm}^3 \leq N \leq 10^{18}/\text{cm}^3$ for the case of rubidium atoms. Also plotted for comparison is the shift δ'_{LL} given by Eq. (22). As can be seen from Fig. 8, the calculated Lorentz-Lorenz shift is linearly proportional to N, but the slope of increase is smaller in magnitude by about 3% than that predicted by Eq. (22) based on analysis in the case of a plane-wave field in the vapor.

One also notices from Fig. 8, that the calculated linear dependence of the shift δ_{LL} on N extends to the intermediate density range where the inhomogeneous broadening ku can be greater than the homogeneous width γ_{21}. In fact, we have calculated the self-consistent SR signals with and without the local field correction term at densities such that $Nk^{-3} < 1$, where γ_{21} is only a few percent of ku. Very surprisingly, we find that the Lorentz-Lorenz shift resulting from the inclusion of the local-field correction term is still quite accurately given by Eq. (22). One might, at first sight, expect that the Lorentz-Lorenz shift would be negligible in the limit that $\gamma_{21}/ku \ll 1$ [8], based on the fact that the local field correction, which is proportional to the polarization density of the medium $P(z)$, is reduced by a factor of γ_{21}/ku in this limit. Our finding above clearly contradicts such a rather common belief, indicating that one cannot neglect the Lorentz-Lorenz term when considering the line shift even at low vapor densities. More details on the Lorentz-Lorenz shift in an inhomogeneously broadened medium will be published elsewhere.

Finally, the three line shifts studied above, i.e., the transient shift, the wall shift, and the Lorentz-Lorenz shift, are found to be additive within numerical errors. In other words, the frequency shift between the line shape calculated self-

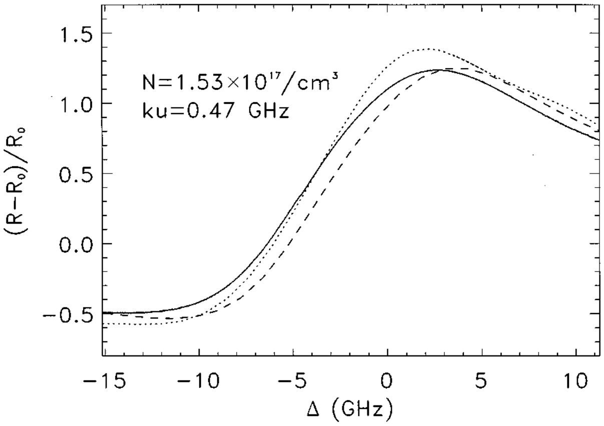

FIG. 9. Comparison between theoretical and experimental SR line shapes. The solid line is the experimental SR signal of a rubidium vapor with a density of $N=1.53\times10^{17}/\mathrm{cm}^3$, and the dashed and dotted lines represent the theoretical spectra obtained self-consistently and by assuming an exponentially decaying field, respectively.

consistently including the atom-wall interaction and the local field correction, and that calculated by assuming a plane-wave field in the vapor without the atom-wall interaction and the local field correction, is found to be given by the sum of δ_{tr}, δ_{wall}, and δ_{LL}.

D. Preliminary comparison between theories and experiment

Based on the self-consistent method outlined in the preceding sections, we are ready to calculate the SR signal which can be compared with experimental results in a dense atomic vapor. Currently, SR experiments with dense rubidium vapors are being conducted at JILA [19]. There are two rubidium isotopes (Rb85 and Rb87) in the vapors, and a total of eight hyperfine transitions for the D1 line can contribute to the SR signal. At low to moderate vapor densities ($N\leqslant10^{15}/\mathrm{cm}^3$), these eight lines are sufficiently separated to allow a comparison of the measurement with calculations on a single resonance transition. However, when the vapor density increases to a level where the different resonance transitions start to overlap with each other, it is no longer possible to separate different hyperfine components, and a calculation including all eight transitions must be carried out, as already described in Sec. II.

In Fig. 9, we plot the theoretical and experimental spectra for a vapor density $N=1.53\times10^{17}/\mathrm{cm}^3$. The laser detuning Δ is measured from the the $F=2\rightarrow F'=3$ hyperfine transition of the Rb85 D1 line. The wall-shift coefficient C for the rubidium atoms is found to be given approximately by $C=0.2$ [19]. In calculating the spectra, we have included the atom-wall interaction, the Lorentz-Lorenz shift, and all eight atomic resonance transitions. The difference of the two theoretical curves lies in the way the transmitted field in the vapor is evaluated. The dashed curve represents the self-consistent result when the field and the atomic density matrix are calculated simultaneously, while the dotted curve is obtained by assuming an exponentially attenuating field as given by Eq. (19).

As can be seen from Fig. 9, the line shape of the self-consistent result closely resembles that of the experimental curve, except that it is shifted relative to the experimental curve to the blue side by approximately 1 GHz. If we assume that this residual shift is entirely due to the collisional shift, $\delta_c/2\pi=A_cN$, not included in the theory, this observation would give a collisional shift coefficient $A_c=-6.5\times10^{-9}$ cm^3 Hz. This corresponds to a resonance shift/broadening ratio of approximately -0.1 for this rubidium D1 transition. An early calculation [20] gave a value of -0.015 for the shift/width ratio due to collisional transfer for a pure resonance r^{-3} interaction.

In comparison, the approximate theoretical line shape given by a dotted line is quite different from the experimental curve. In particular, the calculated signal has a magnitude significantly greater than that of the experimental result. This difference shows that the assumption of an exponential attenuating plane-wave field in the vapor is not valid at high vapor densities, and a self-consistent evaluation of both the field and the atomic density matrix is necessary to calculate the SR signal.

V. SUMMARY

We have presented a theoretical framework for calculating the SR signal of a low-intensity light beam from an arbitrarily dense atomic vapor. The key distinction between our theory and the earlier theoretical calculations is that we calculate the atomic density matrix and the transmitted light field in the atomic vapor self-consistently, while in many previous theories the transmitted field in the vapor is assumed to be a plane wave or an exponentially attenuating plane wave.

The assumption of an exponentially decaying plane-wave field in the vapor is valid as long as the atomic density in the vapor is sufficiently low such that $Nk^{-3}\ll1$, which, in the case of alkali atoms, corresponds to densities $N\ll10^{15}/\mathrm{cm}^3$ for a single hyperfine transition. For higher atomic densities such that $Nk^{-3}\geqslant1$, however, a self-consistent evaluation of the field and the density matrix of the atoms is necessary, and the calculated field exhibits a nonexponential decaying pattern. The SR signals calculated self-consistently and calculated by assuming a plane-wave field differ considerably in the limit of dense vapors both in terms of the line shape and the center frequency of the signal. A comparison between the results of these two calculations and the experimental curve in a dense rubidium vapor support our self-consistent calculation.

The theory presented in this paper also includes multiple resonant transitions and a long-range Van der Waals interaction between the individual vapor atoms and the dielectric wall. The atom-wall interaction remains significant even at very high vapor densities ($Nk^{-3}\gg100$), and it results in a frequency shift of the SR line shape which is comparable in magnitude with the Lorentz-Lorenz shift or the resonance collision shift. It is also found that the Lorentz-Lorenz shift is not significantly affected by the Doppler effect, contrary to previous speculations [8].

In the future, it may be worthwhile to consider generalizing the present theory to the nonlinear regime, and to include effects such as saturation and optical pumping in the limit of high vapor densities.

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