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Selective reflection from a vapor of three-level atoms

G. Nienhuis
Huygens Laboratorium, Rijksuniversiteit Leiden, P.O. Box 9504, 2300 RA Leiden, The Netherlands

F. Schuller
Laboratoire de Physique des Lasers, Institut Galilée, Université Paris-Nord, 93430 Villetaneuse, France
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We study internal selective reflection from a dielectric-vapor interface, where three levels of the vapor atoms in a Λ configuration are connected by two light frequencies. The reflectivity contains then both a sub-Doppler structure due to spatial dispersion in the boundary layer and the dark resonance arising from coherent population trapping. We derive analytical expressions for the reflectivity of either one of the two light beams. The contributions from atoms approaching or leaving the interface are strongly different, whereas they are identical for the case of two-state atoms.

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I. INTRODUCTION

Selective reflection of light from the interface between a transparent dielectric and a dilute vapor is caused by the atoms in a thin layer of a few wavelengths near the boundary. When an atom collides with the surface, its internal state is modified and it is generally assumed that it is completely deexcited. The transient behavior of atoms leaving the surface after a collision gives rise to a sub-Doppler structure in the frequency-dependent reflection coefficient [1–3]. For the special case of two-state atoms, this structure has been described theoretically both at low intensity [4] and in the presence of saturation [5,6]. Also the reflection of a weak probe signal in the presence of a stronger pump beam has been studied with various angles of incidence, both experimentally [7] and theoretically [8]. Then Doppler-free structures arise, which are a combination of the effects of the transient behavior of atoms leaving the surface and of nonlinear spectroscopic effects. This Doppler-free technique of selective reflection can be applied to study the atom-wall interactions [9,10].

Situations in which more than two atomic transitions are excited offer an attractive extension of previous work, since they bring in new independent time scales. For a Λ-type transition, the additional time scales are determined by the rate of optical pumping and by the relaxation rate of the lower states. Recently, selective reflection was studied theoretically in the case of a three-state atom with cascade-type excitation [11]. Dark resonances, arising from destructive interference between two excitations have also been observed in selective reflection [12]. In this work, the resonances appeared against a Doppler-broadened background at incidence angles of about 16°. The effect of the transient behavior is not very pronounced at these angles.

The present paper is a theoretical study of reflection by a vapor of three-state atoms, driven by two light beams in a Λ configuration. The atomic model and the beam geometry are sketched in Fig. 1. We evaluate the reflection of the light beams. The effect of the transient regime is combined with the effects of coherent population trapping. In particular, we compare the contribution to the reflection coefficient from the atoms with positive and negative values of the velocity component normal to the surface. In the special case of two-state atoms and in the absence of saturation, these contributions are known to be identical [6,13]. Reflection studies of the dark resonances have the advantage that they can be performed also in optically dense systems.

II. GENERAL EXPRESSIONS FOR REFLECTIVITY

For later use and in order to fix the notation, we summarize some general expressions for the reflectivity of a light beam normally incident on the interface between a dielectric and a vapor. The dilute atomic vapor fills the half space $z>0$ and the opposite half space with $z<0$ consists of a dielectric with refractive index n. A light beam with frequency ω, incident on the interface, is reflected back into the dielectric. We do not make any restrictive assumptions as to the number of atomic levels and we allow for the presence of other light fields driving atomic transitions. The reflectivity of the light beam under consideration is determined by the dielectric polarization $\mathbf{P}$ that the transmitted light beam induces in the va-

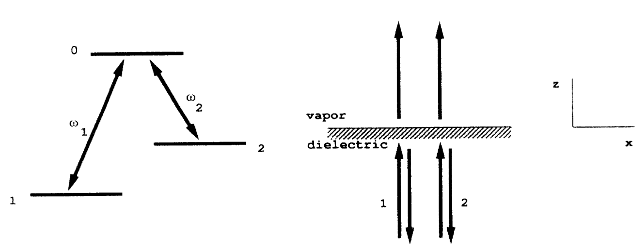

FIG. 1. Geometry of the dielectric and the vapor with two light beams at normal incidence. Also shown is the level scheme of the vapor atoms, driven by the two transmitted beams.

por. At normal incidence, $\mathbf{P}$ is independent of x and y, and we may write

$$\mathbf{P}(z,t)=2\,\mathrm{Re}\mathbf{P}_1(z)e^{ikz-i\omega t}\ , \tag{2.1}$$

with $\mathbf{P}_1$ a z-dependent amplitude. This dielectric polarization is induced by the transmitted electric field, which for $z>0$ takes the form

$$\mathbf{E}(z,t)=2\,\mathrm{Re}\mathbf{E}_1e^{ikz-i\omega t}\ . \tag{2.2}$$

This transmitted field arises from an incident field in the dielectric, described by the amplitude

$$\mathbf{E}_i=(n+1)\mathbf{E}_1/2n\ . \tag{2.3}$$

The reflected field back into the dielectric has the amplitude [6]

$$\mathbf{E}_r=\frac{n-1}{n+1}\mathbf{E}_i+\frac{1}{n+1}\frac{ik}{\varepsilon_0}\int_0^\infty dz\, e^{2ikz}\mathbf{P}_1(z)\ . \tag{2.4}$$

The first term on the right-hand side of (2.4) represents the reflected field in the absence of the vapor and the second term gives the contribution to the reflection due to the dielectric polarization. We may assume that the polarization $\mathbf{P}_1$ is parallel to the field $\mathbf{E}_1$. Then, slightly generalizing the treatment in Ref. [6], we can express the integral in (2.4) as

$$\frac{ik}{\varepsilon_0}\int_0^\infty dz\, e^{2ikz}\mathbf{P}_1(z)=T\mathbf{E}_1\ , \tag{2.5}$$

where the dimensionless quantity T determines the contribution of the vapor to the reflectivity. Notice that in the intensity region where the response of the vapor to the field $\mathbf{E}_1$ is linear, T does not depend on the intensity. With (2.3) and (2.5) the reflected field (2.4) may be put in the form

$$\mathbf{E}_r=\frac{n-1}{2n}\mathbf{E}_1\left[1+\frac{2n}{n^2-1}T\right]\ . \tag{2.6}$$

The intensity reflection coefficient $R=|\mathbf{E}_r|^2/|\mathbf{E}_i|^2$ to first order in the vapor density is equal to

$$R=\left[\frac{n-1}{n+1}\right]^2\left[1+\frac{4n}{n^2-1}\mathrm{Re}T\right]\ . \tag{2.7}$$

It is useful to note that $1-T$ has the physical significance of the complex refractive index of the vapor [6,8].

For a dilute vapor, the dielectric polarization is expressed in terms of the dipole moment of the individual atoms in the vapor. These atomic dipole moments depend on the velocity component v of the atoms in the z direction and on the distance z to the surface. Hence we write

$$\mathbf{P}_1(z)=N\int dv\,\mathbf{p}_1(z,v)\ , \tag{2.8}$$

where N is the density of active atoms and $N\mathbf{p}_1(z,v)dv$ is the contribution to $\mathbf{P}_1(z)$ from the atoms with velocity between v and $v+dv$. We assume that the rate of velocity-changing collisions is small compared with the Doppler width and with the internal relaxation rate. This implies that an atom has time to reach its internal steady state in between two velocity-changing collisions. Then the atoms with negative velocity, which are approaching the surface, will be in their internal steady state, so that their optical coherences are independent of z. The corresponding contribution to $\mathbf{p}_1(z,v)$ is thus equal to $\bar{\mathbf{p}}_1(v)$. In contrast, the atoms with a positive velocity v have just left the surface and they need a transient time before reaching their steady state. When the internal density matrix immediately after leaving the surface at $z=0$ is known, the value of $\mathbf{p}_1(z,v)$ is determined by the optical Bloch equations valid for the specific atomic model.

The Laplace transform is denoted as

$$\hat{\mathbf{p}}_1(s,v)=\int_0^\infty dz\, e^{-sz}\mathbf{p}_1(z,v)\ . \tag{2.9}$$

Then Eq. (2.5) can be expressed as

$$T\mathbf{E}_1=-\frac{N}{2\varepsilon_0}\int dv[\Theta(-v)\bar{\mathbf{p}}_1(v) +\Theta(v)(-2ik)\hat{\mathbf{p}}_1(-2ik,v)]\ , \tag{2.10}$$

where Θ denotes the Heaviside step function. This result generalizes an expression derived before for two-state atoms [6]. Hence the explicit evaluation of the reflection coefficient requires only the Laplace transform of the density matrix for positive velocities and the steady-state density matrix of the atoms at negative velocities.

III. ATOMIC MODEL SYSTEM

We consider the model system sketched in Fig. 1. The active atoms have a single upper state $|0\rangle$, which is coupled by two monochromatic light fields to the lower states $|1\rangle$ and $|2\rangle$. The light beams at frequencies ω_1 and ω_2 are incident normally to the interface at $z=0$ between the dielectric and the vapor. The two fields give the two Rabi frequencies Ω_1 and Ω_2, which may be assumed to be real. We introduce the density matrix $\rho(z,v,t)$, which describes both the distribution over the internal states, over the z component v of the velocity, and over the position. For instance, $\rho_{00}(z,v,t)dv$ is the density of atoms in the excited state $|0\rangle$, with velocity between v and $v+dv$, at a distance z from the interface. The standard transformation to a rotating frame is performed if we introduce the density matrix σ by

$$\begin{aligned}\rho_{01}&=\sigma_{01}e^{ik_1z-i\omega_1t}\ ,\\ \rho_{02}&=\sigma_{02}e^{ik_2z-i\omega_2t}\ ,\\ \rho_{21}&=\sigma_{21}e^{i(k_1-k_2)z-i(\omega_1-\omega_2)t}\ .\end{aligned} \tag{3.1}$$

The two Doppler-shifted detunings from resonance are introduced by

$$\tilde{\Delta}_1=\Delta_1-k_1v\ ,\quad \tilde{\Delta}_2=\Delta_2-k_2v\ , \tag{3.2}$$

with $\Delta_1=\omega_1-\omega_{01}$ and $\Delta_2=\omega_2-\omega_{02}$. Then the equations of motion for the populations are given by

$$D\sigma_{00}=-\Gamma\sigma_{00}+\frac{i}{2}\Omega_1(\sigma_{10}-\sigma_{01})+\frac{i}{2}\frac{\Omega}{2}(\sigma_{20}-\sigma_{02}) ,$$
$$D\sigma_{11}=\Gamma_1\sigma_{00}+\frac{i}{2}\Omega_1(\sigma_{01}-\sigma_{10})-\tfrac{1}{2}f(\sigma_{11}-\sigma_{22}) , \quad (3.3)$$
$$D\sigma_{22}=\Gamma_2\sigma_{00}+\frac{i}{2}\Omega_2(\sigma_{02}-\sigma_{20})+\tfrac{1}{2}f(\sigma_{11}-\sigma_{22}) ,$$

and the coherences obey the equations

$$D\sigma_{01}=-(\gamma_1-i\tilde{\Delta}_1)\sigma_{01}+\frac{i}{2}\Omega_1(\sigma_{11}-\sigma_{00})+\frac{i}{2}\Omega_2\sigma_{21} ,$$
$$D\sigma_{02}=-(\gamma_2-i\tilde{\Delta}_2)\sigma_{02}+\frac{i}{2}\Omega_2(\sigma_{22}-\sigma_{00})+\frac{i}{2}\Omega_1\sigma_{12} , \quad (3.4)$$
$$D\sigma_{21}=i(\tilde{\Delta}_1-\tilde{\Delta}_2)\sigma_{21}+\frac{i}{2}(\Omega_2\sigma_{01}-\Omega_1\sigma_{20})-f'\sigma_{21} .$$

Here D denotes the total derivative

$$D=\frac{\partial}{\partial t}+v\frac{\partial}{\partial z} \quad (3.5)$$

and

$$\Gamma=\Gamma_1+\Gamma_2 \quad (3.6)$$

is the total decay rate of the excited state to the two lower states. The decay rates of the optical coherences are γ_1 and γ_2, and the rates f and f' describe the relaxation of the lower-state populations and coherences. Since the evolution operator for σ does not depend on time, σ approaches a constant value in the steady state.

The reflection of beam 1 is determined by the velocity-dependent dipole density

$$\mathbf{p}_1(z,v)=\boldsymbol{\mu}_{10}\sigma_{01}(z,v) , \quad (3.7)$$

with $\boldsymbol{\mu}$ the dipole vector operator. This dipole density determines the quantity T, according to (2.10)–(2.12), and thereby the reflectivity (2.7). Likewise, the reflectivity of beam 2 is determined by the dipole density $\mathbf{p}_2(z,v)$, which obeys an equation similar to (3.7). Obviously, the states $|1\rangle$ and $|2\rangle$ play a fully symmetric role in the problem, so that it is sufficient to discuss explicitly the reflection of beam 1 only. Expressions pertaining to beam 2 are obtained by simply interchanging the indices 1 and 2.

As explained in Sec. II, the dipole density $\mathbf{p}_1$ for negative values of v is determined by the internal steady state. This state is obtained by setting the right-hand side of Eqs. (3.3) and (3.4) equal to zero. The resulting solution $\bar{\sigma}(v)$ does not depend on the position z. It must be normalized to the Maxwell distribution $W(z)$, so that

$$\mathrm{Tr}\bar{\sigma}=W(v) . \quad (3.8)$$

The Laplace transform of the dipole density for positive velocity v follows from the equality

$$\hat{\mathbf{p}}_1(s,v)=\boldsymbol{\mu}_{10}\hat{\sigma}_{01}(s,v) , \quad (3.9)$$

with $\hat{\sigma}$ the Laplace transform of the density matrix. The equations determining $\hat{\sigma}$ are obtained by taking the Laplace transform of Eqs. (3.3) and (3.4), while omitting the time derivatives. We adopt the usual assumption that atoms leave the surface with only the two lower states populated, so that for $v>0$ one may take

$$\sigma_{11}(0,v)=n_1W(v) , \quad \sigma_{22}(0,v)=n_2W(v) , \quad (3.10)$$

the other matrix elements being zero. Normalization requires that

$$n_1+n_2=1 . \quad (3.11)$$

Then $\hat{\sigma}(s,v)$ is determined by the set of algebraic equations

$$vs\hat{\sigma}_{00}=-\Gamma\hat{\sigma}_{00}+\frac{i}{2}\Omega_1(\hat{\sigma}_{10}-\hat{\sigma}_{01})+\frac{i}{2}\Omega_2(\hat{\sigma}_{20}-\hat{\sigma}_{02}) ,$$
$$vs\hat{\sigma}_{11}-vn_1W(v)=\Gamma_1\hat{\sigma}_{00}+\frac{i}{2}\Omega_1(\hat{\sigma}_{01}-\hat{\sigma}_{10}) -\tfrac{1}{2}f(\hat{\sigma}_{11}-\hat{\sigma}_{22}) ,$$
$$vs\hat{\sigma}_{22}-vn_2W(v)=\Gamma_2\hat{\sigma}_{00}+\frac{i}{2}\Omega_2(\hat{\sigma}_{02}-\hat{\sigma}_{20}) +\tfrac{1}{2}f(\hat{\sigma}_{11}-\hat{\sigma}_{22}) , \quad (3.12)$$
$$vs\hat{\sigma}_{01}=-(\gamma_1-i\tilde{\Delta}_1)\hat{\sigma}_{01}+\frac{i}{2}\Omega_1(\hat{\sigma}_{11}-\hat{\sigma}_{00})+\frac{i}{2}\Omega_2\hat{\sigma}_{21} ,$$
$$vs\hat{\sigma}_{02}=-(\gamma_2-i\tilde{\Delta}_2)\hat{\sigma}_{02}+\frac{i}{2}\Omega_2(\hat{\sigma}_{22}-\hat{\sigma}_{00})+\frac{i}{2}\Omega_1\hat{\sigma}_{12} ,$$
$$vs\hat{\sigma}_{21}=i(\tilde{\Delta}_1-\tilde{\Delta}_2)\hat{\sigma}_{21}+\frac{i}{2}(\Omega_2\hat{\sigma}_{01}-\Omega_1\hat{\sigma}_{20})-f'\hat{\sigma}_{21} .$$

The equations for the matrix elements $\hat{\sigma}_{10}$, $\hat{\sigma}_{20}$, and $\hat{\sigma}_{12}$ follow by taking the complex conjugate of the last three equations of (3.12), for real values of s. Analytic continuation to complex values of s (with $\mathrm{Re}s\geq 0$) can be performed in the final results.

IV. EVALUATION OF THE DENSITY MATRIX

A. Positive velocities

We want to extract from the set of equations (3.12) an expression for the optical coherence $\hat{\sigma}_{01}$, which allows direct numerical computation. The last three equations of (3.12) and their complex conjugates can be used to express the coherences in terms of the populations, with the result

$$\hat{\sigma}_{21}=-\tfrac{1}{4}\Omega_1\Omega_2Q_+[P_{1-}(\hat{\sigma}_{11}-\hat{\sigma}_{00})+P_{2+}(\hat{\sigma}_{22}-\hat{\sigma}_{00})] , \quad (4.1)$$

$$\hat{\sigma}_{01}=\frac{i}{2}\Omega_1P_{1-}[(1-\tfrac{1}{4}\Omega_2^2Q_+P_{1-})(\hat{\sigma}_{11}-\hat{\sigma}_{00}) -\tfrac{1}{4}\Omega_2^2Q_+P_{2+}(\hat{\sigma}_{22}-\hat{\sigma}_{00})] , \quad (4.2)$$

$$\hat{\sigma}_{20}=-\frac{i}{2}\Omega_2P_{2+}[(1-\tfrac{1}{4}\Omega_1^2Q_+P_{2+})(\hat{\sigma}_{22}-\hat{\sigma}_{00}) -\tfrac{1}{4}\Omega_1^2Q_+P_{1-}(\hat{\sigma}_{11}-\hat{\sigma}_{00})] . \quad (4.3)$$

We introduced the quantities

$$P_{i\pm}=(vs+\gamma_i\pm i\tilde{\Delta}_i)^{-1} \quad \text{for } i=1,2 , \quad (4.4)$$

$$Q_{\pm}=[vs+f'\mp i(\tilde{\Delta}_1-\tilde{\Delta}_2)+\tfrac{1}{4}\Omega_1^2P_{2\pm}+\tfrac{1}{4}\Omega_2^2P_{1\mp}]^{-1} . \tag{4.5}$$

The other coherences follow from these expressions after interchanging the labels 1 and 2. Note that at this interchange, Q_+ changes into Q_-. A set of coupled equations for populations only follow if we substitute (4.1)–(4.3) into the second and third equations of (3.12), while introducing the abbreviations

$$W_1=\tfrac{1}{4}\Omega_1^2P_{1+}[1-\tfrac{1}{4}\Omega_2^2P_{1+}Q_-] + \tfrac{1}{4}\Omega_1^2P_{1-}[1-\tfrac{1}{4}\Omega_2^2P_{1-}Q_+] , \tag{4.6}$$

$$W_2=\tfrac{1}{4}\Omega_2^2P_{2+}[1-\tfrac{1}{4}\Omega_1^2P_{2+}Q_+] + \tfrac{1}{4}\Omega_2^2P_{2-}[1-\tfrac{1}{4}\Omega_1^2P_{2-}Q_-] , \tag{4.7}$$

$$U=\tfrac{1}{16}\Omega_1^2\Omega_2^2(P_{1+}P_{2-}Q_-+P_{1-}P_{2+}Q_+) . \tag{4.8}$$

Eliminating $\hat{\sigma}_{00}$ by using the normalization leads to linear equations for $\hat{\sigma}_{11}$ and $\hat{\sigma}_{22}$,

$$X_1\hat{\sigma}_{11}-Y_1\hat{\sigma}_{22}=M_1 , \quad X_2\hat{\sigma}_{22}-Y_2\hat{\sigma}_{11}=M_2 , \tag{4.9}$$

with

$$X_i=vs+\Gamma_i+2W_i+\tfrac{1}{2}f-U , \tag{4.10}$$

$$Y_i=\tfrac{1}{2}f+2U-\Gamma_i-W_i , \tag{4.11}$$

$$M_i=\left[vn_i+\frac{1}{s}(\Gamma_i+W_i-U)\right]W(v) . \tag{4.12}$$

Explicit expressions for the populations $\hat{\sigma}_{00}$, $\hat{\sigma}_{11}$, and $\hat{\sigma}_{22}$ follow immediately, which can be used to evaluate the optical coherence $\hat{\sigma}_{01}$ by (4.2). Since the expression is a bit lengthy, we do not write it out explicitly.

Equations (4.2) and (4.10)–(4.12) can be used directly for a numerical evaluation of the reflectivity of beam 1. One notices that if s would be real, the quantities W_i and U would be real as well, and so would be the Laplace-transformed populations $\hat{\sigma}_{11}$ and $\hat{\sigma}_{22}$. However, we recall that according to Eq. (2.10), the reflectivity depends on the density matrix $\hat{\sigma}(s)$ with $s=-2ik$. At this imaginary value of s, $\hat{\sigma}_{11}$ and $\hat{\sigma}_{22}$ are complex.

It is important to note that for large values of s, the Laplace transformed coherence $\hat{\sigma}_{01}$ can be approximated by a very simple expression, which is based on the limiting behavior of σ_{01} for small times after a collision with the surface. When v/z is large compared with the rates of relaxation and of absorption in the state $|1\rangle$, the value of $\sigma_{01}(z,v)$ is well approximated by

$$\sigma_{01}(z,v)\cong\frac{1}{v}\int_0^\infty dz'\exp\left[-\frac{z-z'}{v}(\gamma_1-i\tilde{\Delta}_1)\right] \times\frac{i}{2}\Omega_1 n_1 W(v) , \tag{4.13}$$

since only the matrix element $\sigma_{11}(0,v)$ serves as a source for σ_{01} [see the first Equation of (3.4)]. This leads to a good approximation of the Laplace transform $\hat{\sigma}_{01}(s,v)$ when sv is large compared with the rate of depletion of state $|1\rangle$. Then we obtain

$$\hat{\sigma}_{01}(s,v)\cong\frac{1}{s(sv+\gamma_1-i\tilde{\Delta}_1)}\frac{i}{2}\Omega_1 n_1 W(v) . \tag{4.14}$$

This result should give a reasonable approximation for the contribution T_+ of the atoms with positive velocity to the reflectivity of beam 1, at moderate values of the intensity. Then this contribution is basically the same as in the case of two-state atoms [6]. Physically, this means that the reflection occurs in a narrow region near the boundary, where the atoms departing from the surface have had no time to experience coupling between their two lower states. We shall present a numerical test of this approximation in Sec. V.

B. Negative velocities

Atoms at negative velocities, which are approaching the surface, can be supposed to have reached their stationary internal state. Therefore, we may write

$$\sigma(z,v)=\bar{\sigma}(v) \tag{4.15}$$

for $v<0$. This velocity-dependent density matrix can be obtained by taking the formal limit of the Laplace transform

$$\bar{\sigma}(v)=\lim_{s\searrow 0}s\hat{\sigma}(s,v) , \tag{4.16}$$

or it can be derived by setting the right-hand sides of (3.3) equal to zero. The calculation is just a simplified version of the evaluation of the Laplace transform $\hat{\sigma}$. The coherences can be expressed in the populations in a way very similar to Eqs. (4.1)–(4.3), the main difference being that one has to substitute $s=0$. The quantities $P_{i\pm}$ and $Q_\pm$ are then replaced by

$$p_{i\pm}=(\gamma_i\pm i\tilde{\Delta}_i)^{-1} \text{ for } i=1,2 , \tag{4.17a}$$

$$q_\pm=[f'\mp i(\tilde{\Delta}_1-\tilde{\Delta}_2)+\tfrac{1}{4}\Omega_1^2p_{2\pm}+\tfrac{1}{4}\Omega_2^2p_{1\mp}]^{-1} . \tag{4.17b}$$

Likewise, we introduce the simplified real quantities

$$w_1=\tfrac{1}{4}\Omega_1^2p_{1+}[1-\tfrac{1}{4}\Omega_2^2p_{1+}q_-]+\tfrac{1}{4}\Omega_1^2p_{1-}[1-\tfrac{1}{4}\Omega_2^2p_{1-}q_+], \tag{4.18}$$

$$w_2=\tfrac{1}{4}\Omega_2^2p_{2+}[1-\tfrac{1}{4}\Omega_1^2p_{2+}q_+]+\tfrac{1}{4}\Omega_2^2p_{2-}[1-\tfrac{1}{4}\Omega_1^2p_{2-}q_-], \tag{4.19}$$

$$u=\tfrac{1}{16}\Omega_1^2\Omega_2^2(p_{1+}p_{2-}q_-+p_{1-}p_{2+}q_+) , \tag{4.20}$$

in analogy to (4.6)–(4.8). In terms of these quantities, the excited-state population in the steady state is

$$\bar{\sigma}_{00}(v)=\frac{1}{K}[\tfrac{1}{2}f(w_1+w_2-2u)+w_1w_2-u^2]W(v) \tag{4.21}$$

and the two population differences that determine the coherence $\bar{\sigma}_{01}$ are

$$\bar{\sigma}_{11}(v)-\bar{\sigma}_{00}(v)=\frac{1}{K}[\tfrac{1}{2}f\Gamma+\Gamma_2u+\Gamma_1w_2]W(v) , \tag{4.22}$$

$$\bar{\sigma}_{22}(v)-\bar{\sigma}_{00}(v)=\frac{1}{K}[\tfrac{1}{2}f\Gamma+\Gamma_1u+\Gamma_2w_1]W(v) . \tag{4.23}$$

We introduced the denominator

$$K=\tfrac{1}{2}f(2\Gamma+3w_1+3w_2-6u)$$
$$+\Gamma_1 w_2+\Gamma_2 w_1+\Gamma u+w_1 w_2-3u^2 . \quad (4.24)$$

Finally, the optical coherence determining the reflection of beam 1 by the atoms with negative velocity is

$$\overline{\sigma}_{01}(v)=\frac{i}{2K}\Omega_1 p_{1-}[(1-\tfrac{1}{4}\Omega_2^2 p_{1-}q_+)(\tfrac{1}{2}f\Gamma+\Gamma_2 u+\Gamma_1 w_2)$$
$$-\tfrac{1}{4}\Omega_2^2 p_{2+}q_+(\tfrac{1}{2}f\Gamma+\Gamma_1 u+\Gamma_2 w_1)]$$
$$\times W(v) . \quad (4.25)$$

Both the excited-state population (4.21) and the optical coherences vanish exactly at the Raman resonance condition $\tilde{\Delta}_1=\tilde{\Delta}_2$, provided that the ground-state relaxation rates f and f' are negligible. This is the well-known dark resonance, which arises due to destructive interference of the two excitation paths [14]. The atom is then in a pure state $\sim\Omega_2|1\rangle-\Omega_1|2\rangle$. The occurrence of this dark resonance is easily checked in (4.21) and (4.25) if one uses the identities

$$1-\tfrac{1}{4}\Omega_2^2 p_{1-}q_+=[f'-i(\tilde{\Delta}_1-\tilde{\Delta}_2)+\tfrac{1}{4}\Omega_1^2 p_{2+}]q_+ , \quad (4.26)$$

$$w_1=\{\tfrac{1}{4}\Omega_1^2 p_{1+}[f'+i(\tilde{\Delta}_1-\tilde{\Delta}_2)]q_- +\text{c.c.}\}+\Omega_1^2\Omega_2^{-2}u , \quad (4.27)$$

$$w_2=\{\tfrac{1}{4}\Omega_2^2 p_{2+}[f'-i(\tilde{\Delta}_1-\tilde{\Delta}_2)]q_+ +\text{c.c.}\}+\Omega_2^2\Omega_1^{-2}u . \quad (4.28)$$

Hence, when f' is neglected, under the Raman condition $\tilde{\Delta}_1=\tilde{\Delta}_2$ one finds that $w_1/u=u/w_2=\Omega_1^2/\Omega_2^2$.

V. NUMERICAL RESULTS

The reflectivity R is expressed in terms of the real part of T by (2.7). By using Eqs. (3.7) and (3.9), we find from (2.10) the result

$$\text{Re}T=-\frac{N\hbar\Omega_1}{2I_1}\phi , \quad (5.1)$$

with $I_1=2\varepsilon_0|E_1|^2$ the intensity of the transmitted field. The quantity

$$\phi=\phi_+ +\phi_- \quad (5.2)$$

is the sum of the real dimensionless quantities ϕ_+ and ϕ_-, defined by

$$\phi_+=\text{Re}\int_0^\infty dv(-2ik)\hat{\sigma}_{01}(-2ik,v) ,$$
$$\phi_-=\text{Re}\int_{-\infty}^0 dv\,\overline{\sigma}_{01}(v) . \quad (5.3)$$

These integrated optical coherences represent the contribution to the reflectivity from atoms with positive and negative velocities. The behavior of ϕ_+ and ϕ_- as a function of Δ_1 are represented in Figs. 2–4 in a few cases. The decay rates $\Gamma_1=\Gamma_2$ are taken to be equal to one-tenth of the Doppler width kv_0, so that the total linewidths are dominated by Doppler broadening.

In Fig. 2 we plot ϕ_+ as a function of the detuning Δ_1/kv_0 of beam 1 from resonance, for various values of the Rabi frequencies $\Omega_1=\Omega_2$, and we compare the result with the approximate result based on (4.14). The frequency ω_2 of the second field is on resonance. Only the sub-Doppler structure near resonance is shown. It is noticed that the approximation (4.14) is quite good for Rabi frequencies that are not larger than the homogeneous widths $\Gamma_1=\Gamma_2$. This means that the contribution from the departing atoms to the reflection of beam 1 deviates only slightly from the situation of a vapor of two-state atoms. Only when ω_1 is very close to resonance does a discrepancy arise. This discrepancy displays the onset of optical pumping in the boundary layer where the reflection originates. When the Rabi frequencies are larger than the decay rates Γ_1 and Γ_2, the deviation is ap-

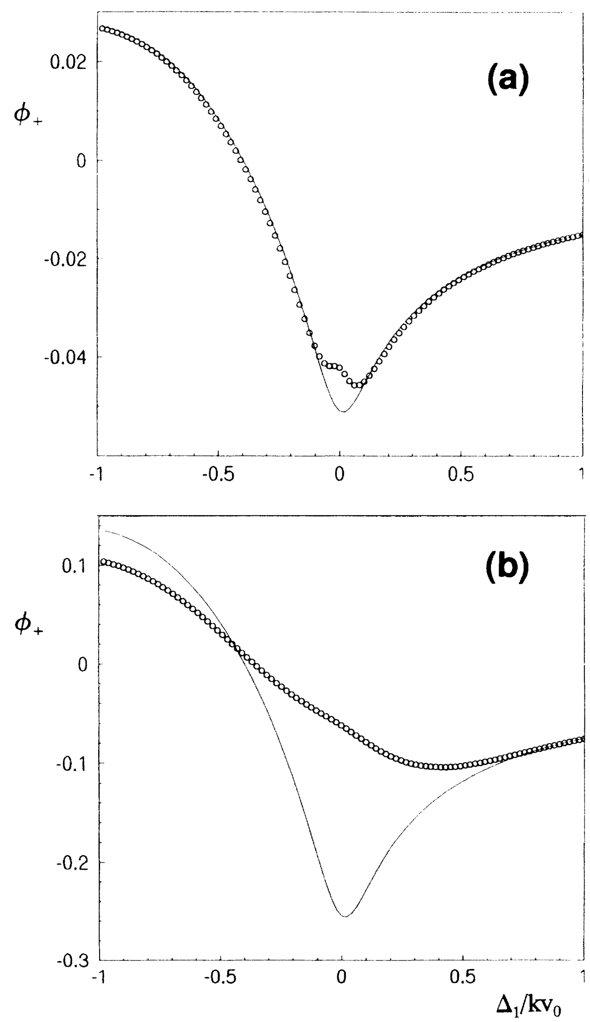

FIG. 2. Plot of the quantity ϕ_+, as a function of Δ_1/kv_0, for various values of the two Rabi frequencies. This quantity determines the contribution of the atoms with positive velocity (departing from the interface) to the reflectivity of beam 1. The ground-state relaxation rates f and f' and the detuning Δ_2 are taken zero, and the decay rates $\Gamma_1=\Gamma_2=0.1kv_0$. The solid lines indicate the approximation (4.14) and the circles represent exact calculations. The two Rabi frequencies $\Omega_1=\Omega_2=\Omega$ are (a) $\Omega=0.1kv_0$ and (b) $\Omega=0.5kv_0$.

preciable within the Doppler width.

In Fig. 3 the effect of the ground-state relaxation rates f and f' for low intensities is demonstrated. The solid lines represent the contribution ϕ_- of the atoms with negative velocity and the circles indicate the total ϕ. The complementary contribution ϕ_+ from the atoms with positive velocity can be read off by taking the difference between ϕ and ϕ_-. For $f=f'=0$, ϕ_- displays a narrow resonance at the Raman condition $\Delta_1=\Delta_2$, with a width determined by the Rabi frequencies. This dark resonance is not visible in the complementary contribution ϕ_+. A very small value of the relaxation rates f and f' already gives an appreciable broadening of the dark resonance.

Figure 4 displays the behavior of ϕ_- and the total ϕ for large values of the detuning Δ_2. Both for a strong blue and a strong red detuning the contributions from the arriving and the departing atoms are comparable in magnitude. But only the contribution ϕ_- displays narrow structures, which correspond to the dark resonance. The shape of the resonance depends significantly on the frequency. The structure is richer in the case of red detuning, where the fields are resonant for the atoms approaching the interface. This frequency-dependent shape of the dark resonance in selective reflection has recently been observed [15].

VI. CONCLUSION

The general conclusion is that for these three-state atoms there is a strong difference between the contribution from arriving and departing atoms to the reflection. Nevertheless, these contributions have a similar magnitude in general. Recall that for two-state atoms and a weak field these contributions are equal. For moderate intensities, the reflectivity arising from the departing atoms is the same as for two-state atoms, except at the very center of the sub-Doppler structure. The dark resonance, which occurs for $\Delta_1=\Delta_2$, is appreciably only in

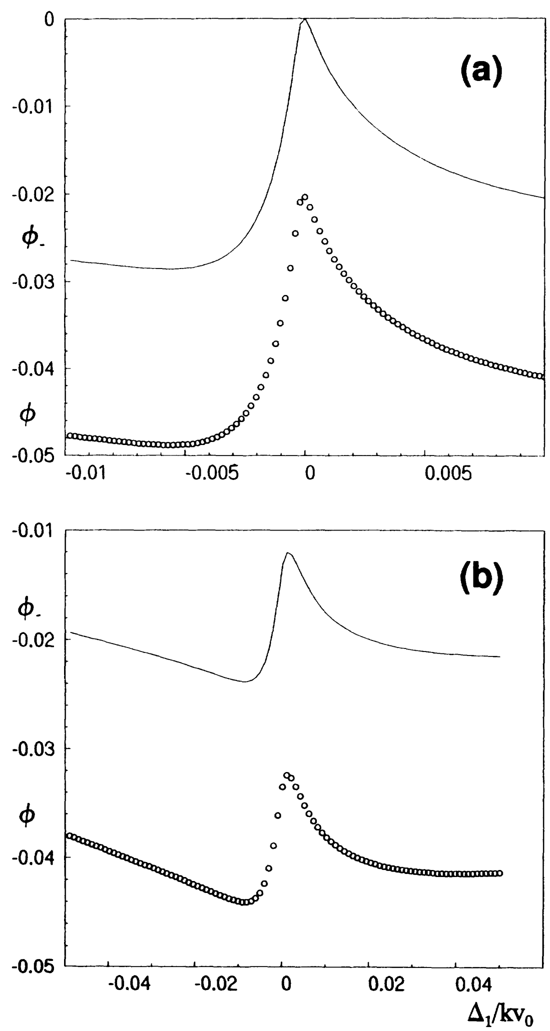

FIG. 3. Plot of the quantity ϕ_0 and of ϕ, as a function of Δ_1/kv_0, with or without ground-state relaxation. The various parameters are $\Delta_2=0$, $\Omega_2=0.02kv_0$, and $\Omega_1=0.04kv_0$. (a) $f=f'=0$, and (b) $f=f'=0.002kv_0$. The solid line gives ϕ_- and the circles represent ϕ.

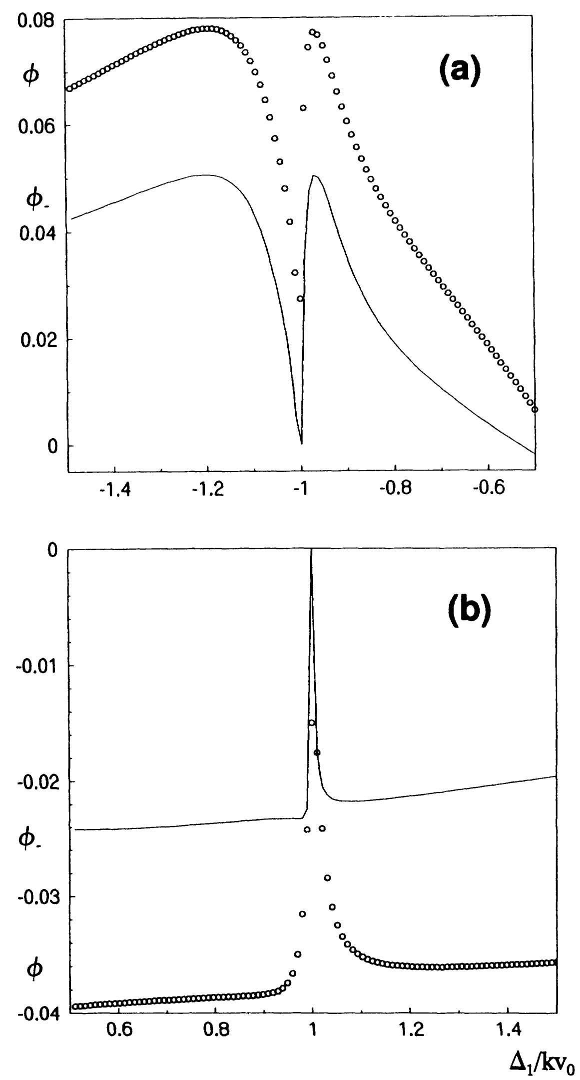

FIG. 4. Plot of the quantity ϕ_- and of ϕ, as a function of Δ_1/kv_0, for large detuning Δ_2 and vanishing ground-state relaxation rates f and f'. The Rabi frequencies are $\Omega_1=\Omega_2=0.1kv_0$. (a) Red detuning $\Delta_2=-kv_0$ and (b) blue detuning $\Delta_2=kv_0$. The solid line gives ϕ_- and the circles represent ϕ.

the contribution from the arriving atoms. Its strength is very sensitive to the relaxation of the ground state, which can arise both from transient effects and from collisions. Furthermore, the shape of the resonance in reflection depends strongly on the frequency of the light fields.

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