Nonlinear selective reflection in cascade three-level atomic systems

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We study the cascade three-level selective-reflection spectroscopy for normal incidence at a dielectric–atomic-vapor interface in a pump-probe scheme. Taking into account the effects of spatial dispersion arising from deexcitation by collisions with the interface and the transient response of atoms leaving the surface, we evaluate the modification of the probe-field reflection induced by the presence of the pump field in terms of a nonlocal nonlinear susceptibility. The dependence of this susceptibility on the probe frequency for various given pump frequencies is calculated. Sub-Doppler structures in the selective reflection spectrum are obtained and analyzed. Potential applications to the study of the interaction of highly excited atomic states with the surface are indicated.

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I. INTRODUCTION

Selective reflection of light at the interface between a dielectric and a dilute atomic vapor has been studied in recent years by several authors for the case of two-level atoms [1–4]. It has been shown that at normal incidence, collisions between the atoms and the wall can give rise to a sub-Doppler structure in the reflection coefficient. This structure is due to the transient behavior of atoms leaving the interface just after having been deexcited at the contact with the dielectric. These effects appear even more strikingly in the case of a pump-probe scheme, where one studies the modification of the reflection coefficient of a weak probe beam induced by the presence of an intense pump beam [5]. In this scheme the sub-Doppler structure is due to both saturation narrowing and transient behavior. Moreover, for nonzero pump-frequency detunings, one predicts extra resonances due exclusively to wall collisions.

In the present paper we extend our previous calculations to the case of a three-level system with cascade type excitation. We consider the two possible cases: (i) the pump is tuned to the frequency of the upper transition and the probe to the frequency of the lower one [see Fig. 1(a); (ii) the inverse case, i.e., the pump tuned to the frequency of the lower transition and the probe to that of the upper one [see Fig. 1(b)].

As in Ref. [5], we study the modification of the reflection coefficient of the probe beam induced by the presence of the pump beam. Experimentally, this contribution is isolated from the main part of the reflection coefficient by standard modulation techniques.

The predicted reflectivity-difference signal is compared with the one expected in a local approach, i.e., in the absence of a transient regime when the steady-state nonlinear susceptibility is proportional to the local field. This approach is directly related to standard three-level spectroscopy in gas cells. In the following we shall restrict our discussion to the first nonvanishing term in the pump-probe interaction, although the case of arbitrary intensities of the pump is covered by our general formalism as well.

II. GENERAL THEORY

In previous studies [1–5] of selective reflection by atomic vapors at a vapor-dielectric interface the following model has been considered: one assumes that the state of atoms moving towards the interface is described by a constant density matrix $\bar{\rho}$ (steady-state regime), whereas atoms moving in the opposite direction are described by a density matrix that varies with position as long as they are sufficiently close to the interface. This comes about because, assuming that all atoms leave the surface in their ground state, excitation takes place only progressively, giving rise to a position-dependent density matrix ρ, which merges into the constant one only at larger distances. As has been pointed out, in the intermediate regime spatial dispersion occurs [1], i.e., the dielectric polarization is no longer proportional to the exciting local field. By choosing a coordinate system with

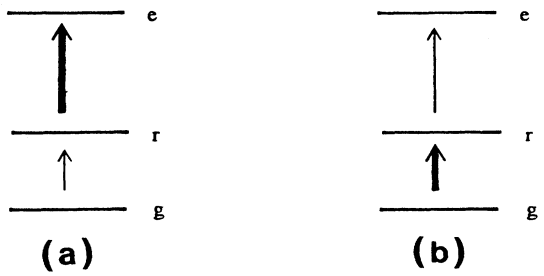

FIG. 1. Two possible pump-probe schemes for a cascade three-level system with g, r, and e the ground, the relay, and the excited level, respectively. The broad arrow represents the transition induced by the pump field, the slim one that induced by the probe field. In (a) the pump field is tuned close to the frequency of the transition $r \rightarrow e$, the probe field close to the transition $g \rightarrow r$, whereas in (b) the roles of the two fields are reversed.

its z axis perpendicular to the interface, the distinction between atoms moving towards the dielectric and those moving away from it amounts to considering separately the cases $v_z<0$ and $v_z>0$ ($z<0$ being the half-space filled by the dielectric, $z>0$ that filled by the vapor).

In Ref. [3] a two-state atom has been studied with g labeling the ground and e the excited state. The off-diagonal element ρ_{eg} of the density matrix has been split into that of a reduced density matrix σ multiplied by a phase factor, according to the relation

$$\rho_{eg}=e^{-i(\omega t-kz)}\sigma_{eg}(v_z,z)\ , \tag{2.1}$$

where σ_{eg} is time independent and ω is the radiation frequency. Light is incident from the dielectric into the vapor. Furthermore, it is assumed that due to the very low vapor density, each atom is submitted only to the field that would exist if the vapor were replaced by vacuum. Considering the stationary value $\bar{\sigma}_{eg}(v_z)$ together with the Laplace-transformed quantity

$$\hat{\sigma}_{eg}(v_z,p)=\int_0^{+\infty}dz\,e^{-pz}\sigma_{eg}(v_z,z)\ , \tag{2.2}$$

it has been shown [3] that selective reflection is governed by a quantity T, which is related to the matrix elements, defined above, by the expression

$$T=-\frac{\pi N\Omega}{2|E_1|^2}\int_{-\infty}^{+\infty}dv_z\,W(v_z)[\Theta(-v_z)\bar{\sigma}_{eg}(v_z)+\Theta(v_z)(-2ik)\times\hat{\sigma}_{eg}(v_z,-2ik)]\ , \tag{2.3}$$

where the Heaviside functions $\Theta(-v_z)$ and $\Theta(v_z)$ ensure the separation of contributions coming from atoms with $v_z<0$ and $v_z>0$, respectively. $W(v_z)$ is a normalized Maxwell-Boltzmann distribution, N the number density of atoms, E_1 the amplitude of the radiation field propagating through the vapor, and Ω the Rabi frequency, defined as

$$\Omega=\frac{2}{\hbar}\mu_{eg}\cdot E_1\ , \tag{2.4}$$

with μ_{eg} the transition dipole moment. It should be noted that we are specializing the results of [3] for normal incidence. In terms of the quantity T, the reflection coefficient for radiation incident from the dielectric and reflected at its surface is given by the expression

$$R=\left[\frac{n-1}{n+1}\right]^2+\frac{4n(n-1)}{(n+1)^3}\,\mathrm{Re}T\ , \tag{2.5}$$

n being the refraction index of the dielectric. Here only the second term on the right-hand side (rhs) is relevant for our problem, since it represents the contribution due to the presence of the atomic vapor. Note that $(-T)$ can be interpreted as an effective complex vapor index change [5].

In this paper we study the case of a pump-probe scheme in cascade three-level systems. The lowest and highest level are labeled as g and e, respectively, and the intermediate (relay) level as r. The system is driven by an intense pump field E_p with frequency ω_p which influences the selective reflection of a weak probe (or signal) field E_s with frequency ω_s. The following two situations are considered: (i) the frequency ω_p is close to the transition frequency between levels r and e, whereas ω_s is close to the transition frequency between levels g and r [Fig. 1(a)]; (ii) the inverse case with ω_p close to the transition frequency between g and r and ω_s close to the transition frequency between r and e [Fig. 1(b)].

The selective reflection coefficient of the probe field can be derived from Eq. (2.3) if the pair of states eg is replaced by the relevant one, i.e., gr for case (i) and re for case (ii). As mentioned above, the quantity of interest is the modification of the reflection coefficient induced by the presence of the pump field. This modification arises because the density matrix σ depends on both the probe and the pump field. It has to be determined by solving the optical Bloch equations for an atom submitted to the simultaneous action of the two radiation fields.

III. THE OPTICAL BLOCH EQUATIONS

The state of the atoms in the vapor is determined by the density matrix $\rho(v_z,z,t)$. Note that we are considering here a local density matrix, i.e., one that describes the state of the vapor at a given point in space at time t. Then this quantity obeys the Liouville equation

$$\left[\frac{\partial}{\partial t}+v_z\frac{\partial}{\partial z}\right]\rho=-\frac{i}{\hbar}[H,\rho]-\Gamma\rho\ . \tag{3.1}$$

The diagonal elements of the Hamiltonian H are the energies E_g, E_r, and E_e of the free atom, whereas the off-diagonal ones represent the interaction energies with the radiation field.

Consider first the situation represented in Fig. 1(a). Then in the rotating-wave approximation, these off-diagonal elements are given by the expressions

$$\begin{aligned}&H_{rg}=-\frac{\hbar\Omega_s}{2}e^{-i(\omega_s t-k_s z)},\quad H_{gr}=H_{rg}^*\ ,\\ &H_{er}=-\frac{\hbar\Omega_p}{2}e^{-i(\omega_p t-k_p z)},\quad H_{re}=H_{er}^*\ ,\\ &H_{eg}=H_{ge}=0\ ,\end{aligned} \tag{3.2}$$

where Ω_s and Ω_p are the usual Rabi frequencies defined as the scalar products of either the signal field E_s or the pump field E_p with the relevant dipole transition matrix elements. The term $-\Gamma\rho$ in (3.1) describes spontaneous decay in a closed three-level system. It is represented by the following matrix elements:

$$\begin{aligned}&-(\Gamma\rho)_{ee}=-A_2\rho_{ee}\ ,\\ &-(\Gamma\rho)_{rr}=-A_1\rho_{rr}+A_2\rho_{ee}\ ,\\ &-(\Gamma\rho)_{gg}=A_1\rho_{rr}\ ,\end{aligned} \tag{3.3}$$

and

$$\begin{aligned}&-(\Gamma\rho)_{eg}=-\tfrac{1}{2}A_2\rho_{eg}\ ,\\ &-(\Gamma\rho)_{er}=-\tfrac{1}{2}(A_1+A_2)\rho_{er}\ ,\\ &-(\Gamma\rho)_{rg}=-\tfrac{1}{2}A_1\rho_{rg}\ ,\end{aligned} \tag{3.4}$$

where the two decay constants A_1 and A_2 refer to the transitions $r \rightarrow g$ and $e \rightarrow r$, respectively (cf. [6]). With these definitions (3.1) can be written out explicitly in terms of matrix elements of ρ yielding a system of coupled differential equations. These can be written in a time-independent form if we introduce a reduced density matrix σ obtained by means of the following transformations similar to that of Eq. (2.1):

$$\begin{aligned} \rho_{er} &= e^{-i(\omega_p t - k_p z)} \sigma_{er} , \\ \rho_{rg} &= e^{-i(\omega_s t - k_s z)} \sigma_{rg} , \\ \rho_{eg} &= e^{-i[(\omega_p + \omega_s)t - (k_p + k_s)z]} \sigma_{eg} , \end{aligned} \tag{3.5}$$

together with the corresponding Hermitian conjugate expressions. k_s and k_p are the wave numbers of the signal and pump field, respectively. A positive value of k_p implies pump propagation in the positive direction, i.e., from the dielectric towards the vapor, while a negative value implies propagation in the opposite direction; k_s is always positive. For the matrix elements of σ we then obtain the equations

$$\begin{aligned} v_z \frac{d}{dz}\sigma_{ee} &= \frac{i\Omega_p}{2}(\sigma_{re} - \sigma_{er}) - A_2 \sigma_{ee} , \\ v_z \frac{d}{dz}\sigma_{rr} &= \frac{i\Omega_p}{2}(\sigma_{er} - \sigma_{re}) \\ &\quad + \frac{i\Omega_s}{2}(\sigma_{gr} - \sigma_{rg}) - A_1 \sigma_{rr} + A_2 \sigma_{ee} , \\ v_z \frac{d}{dz}\sigma_{gg} &= \frac{i\Omega_s}{2}(\sigma_{rg} - \sigma_{gr}) + A_1 \sigma_{rr} , \\ v_z \frac{d}{dz}\sigma_{eg} &= i\tilde{\Delta}_{eg}\sigma_{eg} + \frac{i\Omega_p}{2}\sigma_{rg} - \frac{i\Omega_s}{2}\sigma_{er} - \tfrac{1}{2}A_2 \sigma_{eg} , \\ v_z \frac{d}{dz}\sigma_{er} &= i\tilde{\Delta}_{er}\sigma_{er} + \frac{i\Omega_p}{2}(\sigma_{rr} - \sigma_{ee}) - \frac{i\Omega_s}{2}\sigma_{eg} \\ &\quad - \tfrac{1}{2}(A_1 + A_2)\sigma_{er} , \\ v_z \frac{d}{dz}\sigma_{rg} &= i\tilde{\Delta}_{rg}\sigma_{rg} + \frac{i\Omega_p}{2}\sigma_{eg} \\ &\quad + \frac{i\Omega_s}{2}(\sigma_{gg} - \sigma_{rr}) - \tfrac{1}{2}A_1 \sigma_{rg} , \end{aligned} \tag{3.6}$$

where we have introduced Doppler-shifted frequency detunings defined as

$$\begin{aligned} \tilde{\Delta}_{rg} &= \omega_s - \omega_{rg} - k_s v_z = \Delta_s - k_s v_z , \\ \tilde{\Delta}_{er} &= \omega_p - \omega_{er} - k_p v_z = \Delta_p - k_p v_z , \\ \tilde{\Delta}_{eg} &= \tilde{\Delta}_{rg} + \tilde{\Delta}_{er} = \omega_p + \omega_s - (\omega_{rg} + \omega_{er}) - (k_p + k_s)v_z \\ &= \Delta_p + \Delta_s - (k_p + k_s)v_z , \end{aligned} \tag{3.7}$$

with ω_{rg} and ω_{er} the transition frequencies between the corresponding levels. In (3.6) we assume that the interaction between the electromagnetic fields and the atomic vapor has reached a steady-state regime, i.e., that σ depends on z and v_z only.

For the situation represented on Fig. 1(b), the roles of the signal and the pump are reversed. Therefore, the differential equations valid for this case are easily obtained from (3.6) and (3.7) by simply interchanging the indices s and p.

In order to solve the set of equations (3.6) we introduce a Laplace transformation by which we define a quantity

$$\hat{\sigma}(v_z, p) = \int_0^{+\infty} dz\, e^{-pz}\sigma(v_z, z) , \tag{3.8}$$

already mentioned in the preceding section. The derivatives on the left-hand side (lhs) of (3.6) are then easily obtained from the relation

$$\int_0^{+\infty} dz\, e^{-pz}\frac{d}{dz}\sigma = -\sigma(z=0) + p\hat{\sigma} . \tag{3.9}$$

Remembering that, according to our assumption of complete deexcitation of all atoms on the surface, the only nonzero element of $\sigma(z=0)$ is σ_{gg}, it follows that taking the Laplace transform on both sides of Eq. (3.6) amounts to replacing on the lhs $d\sigma_{ee}/dz$, $d\sigma_{rr}/dz$, $d\sigma_{eg}/dz$, $d\sigma_{er}/dz$, $d\sigma_{rg}/dz$ by $p\hat{\sigma}_{ee}$, etc., and $d\sigma_{gg}/dz$ by $-1 + p\hat{\sigma}_{gg}$. On the rhs it is sufficient to put a caret on all matrix elements of σ. Finally, the stationary value of σ, which is also needed for the calculation of the reflection coefficient, is obtained from the relation

$$\bar{\sigma} = \lim_{p \rightarrow 0} p\hat{\sigma}(v_z, p) . \tag{3.10}$$

IV. THE WEAK-PROBE LIMIT

Let us first consider the situation represented in Fig. 1(a). Then the transition associated with the probe field is the one from g to r. Therefore, the relevant quantity entering the general equation (2.3) is the matrix element $\hat{\sigma}_{rg}$. Consequently, since we assume that the intensity of the probe field is very weak, we derive an expression for this quantity representing the first-order term of its expansion with respect to Ω_s. To do so, we start from the Laplace-transformed version of Eqs. (3.6), where we retain only terms of first order in Ω_s. In fact, only two of these equations are needed for the calculation of $\hat{\sigma}_{rg}$, so that we have

$$\begin{aligned} &(v_z p - i\tilde{\Delta}_{eg} + \tfrac{1}{2}A_2)\hat{\sigma}^1_{eg} = i\frac{\Omega_p}{2}\hat{\sigma}^1_{rg} \\ &\text{and} \\ &(v_z p - i\tilde{\Delta}_{rg} + \tfrac{1}{2}A_1)\hat{\sigma}^1_{rg} = i\frac{\Omega_p}{2}\hat{\sigma}^1_{eg} + i\frac{\Omega_s}{2}\hat{\sigma}^0_{gg} . \end{aligned} \tag{4.1}$$

Here the superscripts 0 and 1 refer to the terms which are probe independent and linear in probe amplitude, respectively. Hence these equations relate the required quantity $\hat{\sigma}^1_{rg}$ to $\hat{\sigma}^0_{gg} = 1/p$, the only nonzero element of the zeroth-order terms $\hat{\sigma}^0$. Thus only first-order matrix elements appear in (4.1). The relevant solution is given by the expression

$$\hat{\sigma}^1_{rg}=\frac{i\frac{\Omega_s}{2}\frac{1}{p}(v_zp-i\tilde{\Delta}_{eg}+\frac{1}{2}A_2)}{(v_zp-i\tilde{\Delta}_{rg}+\frac{1}{2}A_1)(v_zp-i\tilde{\Delta}_{eg}+\frac{1}{2}A_2)+\frac{\Omega_p^2}{4}} . \tag{4.2}$$

By expanding this expression, which is valid for arbitrary pump field intensities, we obtain for the first nonvanishing interaction term the result

$$\hat{\sigma}^1_{rg}=\frac{i\frac{\Omega_s\Omega_p^2}{8}}{p(v_zp-i\tilde{\Delta}_{rg}+\frac{1}{2}A_1)^2(v_zp-i\tilde{\Delta}_{eg}+\frac{1}{2}A_2)} . \tag{4.3}$$

Let us now consider the situation represented in Fig. 1(b). As already mentioned, in this case the optical Bloch equations are obtained from (3.6) by interchanging the indices p and s. Here the quantity of interest is the first-order matrix element $\hat{\sigma}^1_{er}$. Again, we only briefly describe the method that is used to obtain this solution. If in the Laplace-transformed equations which are now valid only terms of first order in Ω_s are retained, then there will appear an independent set of two equations which will yield the required solution. These equations are

$$\begin{aligned}&(v_zp-i\tilde{\Delta}_{eg}+\tfrac{1}{2}A_2)\hat{\sigma}^1_{eg}+i\frac{\Omega_p}{2}\hat{\sigma}^1_{er}=i\frac{\Omega_s}{2}\hat{\sigma}^0_{rg} ,\\ &[v_zp-i\tilde{\Delta}_{er}+\tfrac{1}{2}(A_1+A_2)]\hat{\sigma}^1_{er}+i\frac{\Omega_p}{2}\hat{\sigma}^1_{eg}=i\frac{\Omega_s}{2}\hat{\sigma}^0_{rr} .\end{aligned} \tag{4.4}$$

As will be seen below, we have $\hat{\sigma}^0_{ee}=0$. Equations (4.4) relate the required quantity $\hat{\sigma}^1_{er}$ to the zeroth-order terms $\hat{\sigma}^0_{rg}$ and $\hat{\sigma}^0_{rr}$, which have to be calculated separately as function of Ω_p. By solving (4.4) we thus obtain

$$\hat{\sigma}^1_{er}=i\frac{\Omega_s}{2}[(v_zp+\tfrac{1}{2}A_2-i\tilde{\Delta}_{eg})\hat{\sigma}^0_{rr}-i\frac{\Omega_p}{2}\hat{\sigma}^0_{rg}]/D , \tag{4.5}$$

with

$$D=(v_zp+\tfrac{1}{2}A_2-i\tilde{\Delta}_{eg})[v_zp+\tfrac{1}{2}(A_1+A_2)-i\tilde{\Delta}_{er}]+\frac{\Omega_p^2}{4} . \tag{4.6}$$

The calculation of the Ω_s-independent terms $\hat{\sigma}^0_{rg}$ and $\hat{\sigma}^0_{rr}$ from the Laplace-transformed system (3.6) is somewhat lengthy and shall therefore be presented in a condensed form. At the beginning one easily shows that $\hat{\sigma}^0_{eg}=\hat{\sigma}^0_{er}=\hat{\sigma}^0_{ee}=0$ and $\hat{\sigma}^0_{gg}+\hat{\sigma}^0_{rr}=1/p$. Note that the latter relation follows also from the normalization condition $\sigma_{gg}+\sigma_{rr}=1$. Hence one has $\hat{\sigma}^0_{gg}-\hat{\sigma}^0_{rr}=1/p-2\hat{\sigma}^0_{rr}$. Then one derives from the second and the sixth of the equations (3.6) the relations

$$\begin{aligned}&(v_zp+A_1)\hat{\sigma}^0_{rr}=i\frac{\Omega_p}{2}(\hat{\sigma}^0_{gr}-\hat{\sigma}^0_{rg}) ,\\ &(v_zp+\tfrac{1}{2}A_1-i\tilde{\Delta}_{rg})\hat{\sigma}^0_{rg}=i\frac{\Omega_p}{2}\left[\frac{1}{p}-2\hat{\sigma}^0_{rr}\right] .\end{aligned} \tag{4.7}$$

Taking the Hermitian conjugate of the latter equation, we also have

$$(v_zp+\tfrac{1}{2}A_1+i\tilde{\Delta}_{rg})\hat{\sigma}^0_{gr}=-i\frac{\Omega_p}{2}\left[\frac{1}{p}-2\hat{\sigma}^0_{rr}\right] . \tag{4.8}$$

We thus arrive at a system of three equations for the three unknown quantities $\hat{\sigma}^0_{gr}$, $\hat{\sigma}^0_{rg}$, and $\hat{\sigma}^0_{rr}$. After solving this system, we obtain for the required quantities $\hat{\sigma}^0_{rg}$ and $\hat{\sigma}^0_{rr}$ the following expressions:

$$\begin{aligned}&\hat{\sigma}^0_{rg}=-\tfrac{1}{2}\Omega_p(v_zp+A_1)[\tilde{\Delta}_{rg}-i(v_zp+\tfrac{1}{2}A_1)]/(pD_0) ,\\ &\hat{\sigma}^0_{rr}=\frac{\Omega_p^2}{2}(v_zp+\tfrac{1}{2}A_1)\Big/(pD_0) ,\end{aligned} \tag{4.9}$$

with

$$D_0=(v_zp+A_1)[(v_zp+\tfrac{1}{2}A_1)^2+\tilde{\Delta}_{rg}^2]+(v_zp+\tfrac{1}{2}A_1)\Omega_p^2 . \tag{4.10}$$

If these expressions are substituted into (4.5) we obtain for the first-order matrix element of σ a result which is valid for arbitrary intensities of the pump field. However, in practice, one is mostly interested in the first nonvanishing term of the expansion related to nonlinear-optical effects due to the pump-probe interaction. This term is obtained by replacing (4.9) and (4.10) by the simplified expressions

$$\begin{aligned}&\hat{\sigma}^0_{rg}=\left[i\frac{\Omega_p}{2}\right]\Big/[p(v_zp+\tfrac{1}{2}A_1-i\tilde{\Delta}_{rg})] ,\\ &\hat{\sigma}^0_{rr}=\left[\frac{\Omega_p^2}{4}(v_zp+\tfrac{1}{2}A_1)\right]\Big/\{p(v_zp+A_1)[(v_zp+\tfrac{1}{2}A_1)^2+\tilde{\Delta}_{rg}^2]\} ,\end{aligned} \tag{4.11}$$

which lead to the final result

$$\hat{\sigma}^1_{er}=\frac{i\frac{\Omega_s\Omega_p^2}{4}}{p[v_zp+\frac{1}{2}(A_1+A_2)-i\tilde{\Delta}_{er}](v_zp+\frac{1}{2}A_1-i\tilde{\Delta}_{rg})}\left[\frac{v_zp+\frac{1}{2}A_1}{(v_zp+A_1)(v_zp+\frac{1}{2}A_1+i\tilde{\Delta}_{rg})}+\frac{\frac{1}{2}}{v_zp+\frac{1}{2}A_2-i\tilde{\Delta}_{eg}}\right] . \tag{4.12}$$

The stationary values of matrix elements of σ, which are also needed according to Eq. (2.3), are obtained by applying the defining relation (3.10) to Eqs. (4.3) and (4.12). The results are thus given by

$$\bar{\sigma}^1_{rg}=\frac{i\frac{\Omega_s\Omega_p^2}{8}}{(\frac{1}{2}A_1-i\tilde{\Delta}_{rg})^2(\frac{1}{2}A_2-i\tilde{\Delta}_{eg})} \tag{4.13}$$

for the case of Fig. 1(a) and

$$\bar{\sigma}^1_{rg}=\frac{i\Omega_s\frac{\Omega_p^2}{4}}{[\frac{1}{2}(A_1+A_2)-i\tilde{\Delta}_{er}](\frac{1}{2}A_1-i\tilde{\Delta}_{rg})\times[\frac{1}{2}/(\frac{1}{2}A_1+i\tilde{\Delta}_{rg})+\frac{1}{2}/(\frac{1}{2}A_2-i\tilde{\Delta}_{eg})]} \tag{4.14}$$

for the case of Fig. 1(b).

V. RESULTS AND DISCUSSION

A. General expressions

The nonlinear contribution to the reflection coefficient can be calculated by means of a formula analogous to (2.3). We assume that the velocity distribution function is Maxwellian for all v_z:

$$W(v_z)=\frac{1}{\sqrt{\pi}v_0}\exp\left[-\frac{v_z^2}{v_0^2}\right], \tag{5.1}$$

with v_0 the average thermal velocity of the atoms. The quantities Ω and k in (2.3) are replaced by those corresponding to the probe beam, i.e., Ω_s and k_s, and E_1 also changes its significance accordingly. The factor ReT, which is the quantity relevant for our calculation, can now be expressed in the form

$$\mathrm{Re}T=\mathrm{Re}\frac{-\sqrt{\pi}N\Omega_s}{2|E_s|^2v_0}\int_{-\infty}^{+\infty}dv_z\exp\left[-\frac{v_z^2}{v_0^2}\right][\Theta(-v_z)\bar{\sigma}_{ab}(v_z)+\Theta(v_z)(-2ik_s)\hat{\sigma}_{ab}(v_z,-2ik_s)], \tag{5.2}$$

where the quantities $\hat{\sigma}_{ab}$ and $\bar{\sigma}_{ab}$ are those given by Eqs. (4.3), (4.12), (4.13), and (4.14), respectively, and with indices a and b corresponding to either er or rg. We recall that the notation used in (5.2) means that the parameter p appearing in the results of the previous section [Eqs. (4.3) and (4.12)] has to be given the value $-2ik_s$. For the situation of Fig. 1(a) the explicit form of Eq. (5.2) is given by

$$\mathrm{Re}T=C\,\mathrm{Im}\int_{-\infty}^{+\infty}dv_z\frac{1}{2}\exp\left[-\frac{v_z^2}{v_0^2}\right]\left[\Theta(v_z)\frac{1}{[\frac{1}{2}A_2-i(\Delta_s+\Delta_p)+i(k_p-k_s)v_z](\frac{1}{2}A_1-i\Delta_s-ik_sv_z)^2}+\Theta(-v_z)\frac{1}{[\frac{1}{2}A_2-i(\Delta_s+\Delta_p)+i(k_p+k_s)v_z](\frac{1}{2}A_1-i\Delta_s+ik_sv_z)^2}\right], \tag{5.3}$$

where the definitions (3.7) have been used and where the constant C is given by

$$C=\frac{\sqrt{\pi}}{2}\frac{N}{|E_s|^2v_0}\frac{\Omega_s^2\Omega_p^2}{4}. \tag{5.4}$$

When one changes the sign of k_p, i.e., if the propagation of the pump beam is reversed, the contributions to the integral (5.3) of atoms with $v_z>0$ and of those with $v_z<0$ are exchanged. Notice that this invariance only holds if the distribution of atomic velocities is symmetric with respect to v_z. In the counterpropagating geometry, we will neglect the effect of the reflected pump beam, which is a fair approximation from the experimental point of view, since the dielectric reflects only a few percent of the incident light. Moreover, in the Doppler limit, when the decay rates A_1 and A_2 and both detunings Δ_p and Δ_s are small compared with the Doppler width kv_0, the main contribution to the integral (5.3) arises from atoms with velocity small compared to v_0. Thus, in this limiting case, $W(v)$ can be replaced by $W(0)$, and the remaining integral can be performed analytically, yielding results given in the Appendix.

For the situation of Fig. 1(b) we obtain

$$\begin{aligned}\mathrm{Re}T=C\,\mathrm{Im}\int_{-\infty}^{+\infty}dv_z\exp\left[-\frac{v_z^2}{v_0^2}\right]\times\left\{\left[\Theta(v_z)\frac{1}{[\frac{1}{2}(A_1+A_2)-i\Delta_s-ik_sv_z][\frac{1}{2}A_1-i\Delta_p+i(k_p-2k_s)v_z]}\right]\right.\\ \times\left[\frac{\frac{1}{2}A_1-2ik_sv_z}{A_1-2ik_sv_z}\frac{1}{\frac{1}{2}A_1+i\Delta_p-i(k_p+2k_s)v_z}+\frac{1}{2}\frac{1}{\frac{1}{2}A_2-i(\Delta_p+\Delta_s)+i(k_p-k_s)v_z}\right]\\ +\left[\Theta(-v_z)\frac{1}{(\frac{1}{2}(A_1+A_2)-i\Delta_s+ik_sv_z)(\frac{1}{2}A_1-i\Delta_p+ik_pv_z)}\right.\\ \left.\left.\times(\frac{1}{2})\left[\frac{1}{\frac{1}{2}A_1+i\Delta_p-ik_pv_z}+\frac{1}{\frac{1}{2}A_2-i(\Delta_p+\Delta_s)+i(k_p+k_s)v_z}\right]\right]\right\}.\end{aligned} \tag{5.5}$$

Let us recall that now the definitions (3.7) have to be used with indices s and p exchanged. Note that in this case there is no symmetry associated with the change $k_p \rightarrow -k_p$.

We have evaluated the expressions (5.3) and (5.5) by numerical integration. The parameters are chosen to match roughly the values corresponding to the transitions $6s_{1/2} \rightarrow 6p_{3/2}$ (852 nm) and $6p_{3/2} \rightarrow 9d_{5/2}$ (585 nm) in cesium, which are of interest from the experimental point of view [7,8]. According to this choice, we have taken for the ratio between the frequency of the transition $g \rightarrow r$ and that of the transition $r \rightarrow e$ the value 0.7, while for the damping constants we have set $A_1/A_2 = 10$. In the numerical analysis we use the values $A_1 = 0.1kv_0$ and $A_2 = 0.01kv_0$, where we define $k = (k_s + k_p)/2$; these values are several times larger than the actual ones. Clearly, this latter fact will not affect the conclusions concerning the features of the spectra, whereas it greatly facilitates numerical computation and graphical representation of the results.

We consider curves representing the values given, respectively, by expressions (5.3) and (5.5) as functions of the frequency detunings Δ_s of the probe field for several fixed values of the detuning Δ_p of the pump field with respect to the corresponding resonances. Moreover, we compare these results with those of the local-response approach in which the atomic response is supposed to be in the steady-state regime, and spatial dispersion does not

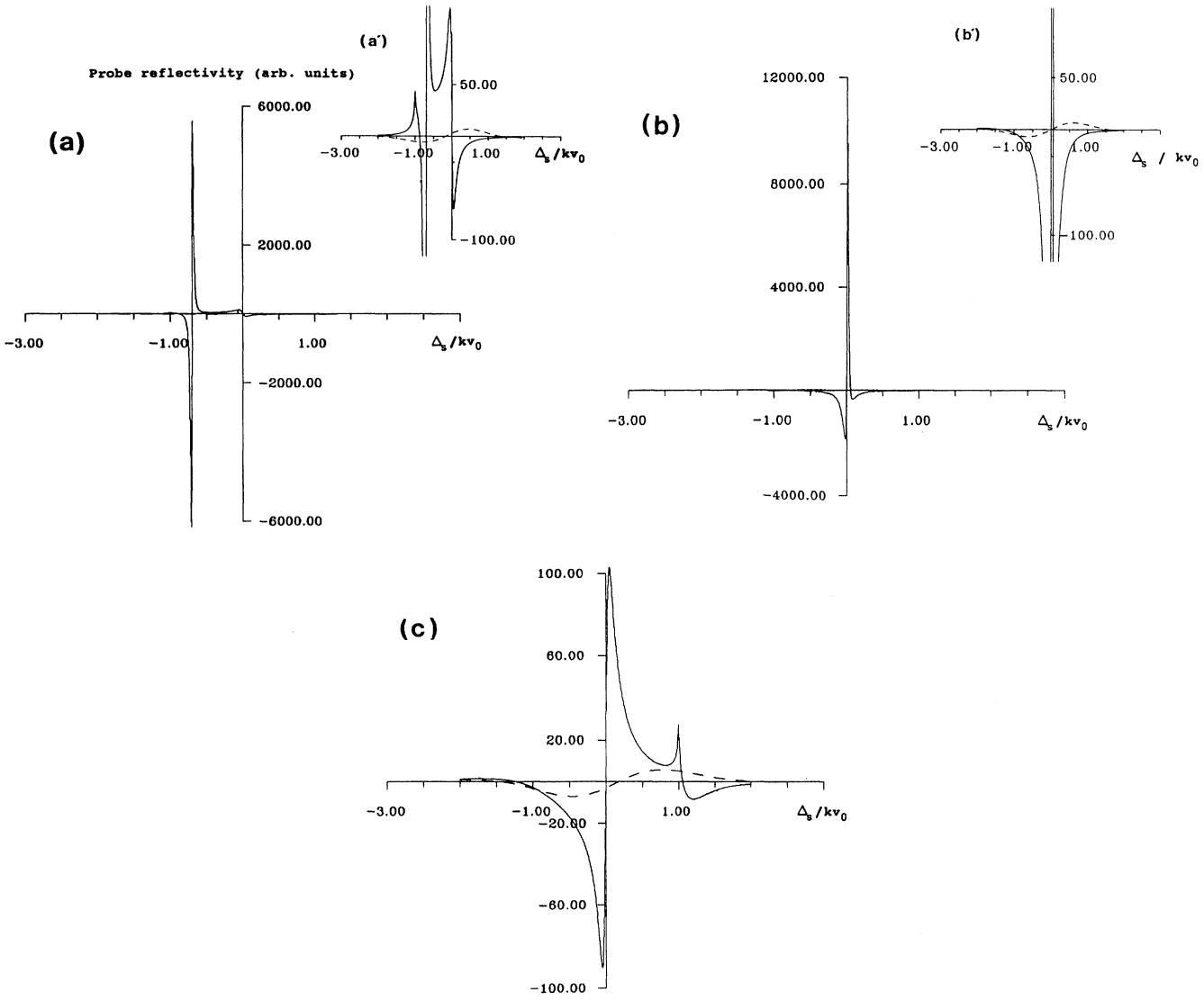

FIG. 2. Spectral variations of the pump-induced probe reflectivity change when the pump drives the upper transition $r \rightarrow e$ (solid lines, nonlocal susceptibility approach; dashed lines, local approach). Pump detuning is positive ($\Delta_p = +kv_0$) in (a), zero in (b), and negative ($\Delta_p = -kv_0$) in (c). Im(T) is in arbitrary units, probe frequency detunings Δ_s in units kv_0 with $k = (k_s + k_p)/2$. (a') is obtained from (a) by modification of the vertical scale to bring out otherwise masked weak structures [the same procedure is used to obtain (b') from (b)].

exist. In that case, the reflection signal is given by

$$\mathrm{Re}T_l=\mathrm{Re}\frac{-\sqrt{\pi}N\Omega_s}{2|E_s|^2v_0}\int_{-\infty}^{+\infty}dv_z\exp\left[-\frac{v_z^2}{v_0^2}\right]\bar{\sigma}_{ab}(v_z) \quad (5.6)$$

instead of being given by (5.2). In this approach, single-dispersion line shapes are predicted from standard three-level spectroscopy [9,10].

B. Pump field driving the upper $r\rightarrow e$ transition

Let us start with the situation represented in Fig. 1(a), where the pump field acts on the transition $r\rightarrow e$ (i.e., between unpopulated states) and the probe field is scanning the transition $g\rightarrow r$. Let us recall that the selective-reflection signal predicted by (5.3) is independent of the direction of propagation of the pump beam ($k_p<0$ or $k_p>0$). This noticeable property differs considerably from the highly *anisotropic* character prevailing in standard three-level spectroscopy [9,10]; for example, in the Doppler limit, there is no signal if the two light beams copropagate in the gas cell. On the other hand, if the two beams are counterpropagating, there is a Doppler-free signal related to two-photon coherence, only if $\omega_p>\omega_s$.

Let us consider the three cases $\Delta_p=0$, $\Delta_p>0$, and $\Delta_p<0$:

(i) If the pump is exactly on resonance with the transition $g\rightarrow r$, i.e., $\Delta_p=0$ [Fig. 2(b)], a narrow asymmetrical structure is present around $\Delta_s=0$. In the local response approach [Eq. (5.6)], a much weaker (more than three orders of magnitude), totally antisymmetric Doppler-broadened dispersion curve is obtained. In Fig. 2(b) the latter is hidden by the trace of the horizontal axis; we thus give in Fig. 2(b′) the same plot using a more appropriate scale. As discussed above, in the Doppler-limit approximation for the local response, there would be no signal at all.

(ii) If the pump is detuned towards the blue by an amount $\Delta_p=+kv_0$ [Fig. 2(a)], a narrow dispersion curve appears around frequency $\Delta_s=-(\omega_s/\omega_p)\Delta_p$. (a) In the *counterpropagating* geometry ($k_p<0$), this signal comes from the permanent response of atoms moving towards the surface with a normal velocity around $v_z^{(0)}=\Delta_p/k_p=-c\Delta_p/\omega_p$. We have checked that this structure is identical to the one given in the local-response approach such as obtained from volume three-level spectroscopy [9]. In the Doppler limit [i.e., when $\exp(-v_z^2/v_0^2)$ is set equal to one] an analytical expression for this structure is obtained from Eqs. (4.13) and (5.6) as

$$\mathrm{Re}T=\frac{C}{2}\mathrm{Im}\int_{-\infty}^{+\infty}\frac{dv_z}{\left[\frac{1}{2}A_2-i(\Delta_s+\Delta_p)+i(\omega_s-\omega_p)\frac{v_z}{c}\right]\left[\frac{1}{2}A_1-i\Delta_s+i\omega_s\frac{v_z}{c}\right]^2}. \quad (5.7)$$

Since we have taken $\omega_p>\omega_s$ and $k_p<0$, the poles of the analytical function to be integrated lie on both sides of the real axis. Using the residue theorem, one obtains

$$\mathrm{Re}T=C\,\mathrm{Im}\frac{-\pi(\omega_p-\omega_s)}{c\omega_p^2\left[\Delta_s+\frac{\omega_s}{\omega_p}\Delta_p+\frac{i}{2}\left[\frac{\omega_s}{\omega_p}A_2+\frac{\omega_p-\omega_s}{\omega_p}A_1\right]\right]^2}. \quad (5.8)$$

The peak-to-peak width of the dispersion structure around $\Delta_s=-(\omega_s/\omega_p)\Delta_p$ is thus given by

$$\Gamma=\frac{1}{\sqrt{3}}\left[\frac{\omega_s}{\omega_p}A_2+\left[\frac{\omega_p-\omega_s}{\omega_p}\right]A_1\right]. \quad (5.9)$$

(b) In the *copropagating* geometry ($k_p>0$), the predicted dispersion signal (identical to the previous one) now comes entirely from the *transient* response of *departing* atoms having velocity around $v_z'^{(0)}=-v_z^{(0)}$. This latter fact deserves special attention, since, to our knowledge, it is the first time that it becomes possible to single out the contribution of atoms moving away from a dielectric-vapor interface with a velocity inside a narrow interval. Velocity distribution of atoms leaving the surface is subject to experimental test by changing the pump-field frequency detuning. As noted above, in this geometry, the steady-state atomic response averages to zero after velocity integration in the Doppler limit.

In addition, in both geometries, a weaker dispersion structure which has the width A_1 of the lower transition is present around the frequency $\Delta_s=0$. A narrow asymmetrical peak is also present around the frequency $\Delta_s=-\Delta_p$ [see Fig. 2(a′)].

We have carried out the numerical integration of Eq. (5.3) in a cascade system for which $\omega_p<\omega_s$ (i.e., $\omega_{re}<\omega_{gr}$). The strong dispersion structure around $\Delta_s=-(\omega_s/\omega_p)\Delta_p$ is then totally washed out. This is not surprising, since in volume three-level spectroscopy the corresponding structure is also known to vanish [9].

(iii) If the pump is detuned towards the red by an amount $\Delta_p=-kv_0$ [Fig. 2(c)] the spectrum is much less intense than in cases (i) and (ii). It contains a dispersion structure with frequency Δ_s around 0 having a width A_1 and a narrow asymmetrical peak around $\Delta_s=-\Delta_p$ similar to case (ii).

In all of the above cases, the structures around $\Delta_s=0$ and $\Delta_s=-\Delta_p$ are related to the presence of the surface and represent the contribution of atoms with small v_z. The resonance around $\Delta_s=0$ comes from pump-induced modifications of the logarithmic singularity known to appear in linear reflection [1–3]. On the other hand, the structures around $\Delta_s=-\Delta_p$ are characteristic of two-photon resonances. This structure is also a sub-Doppler

logarithmic singularity, which now originates in the *transient response of two-photon coherence* (which is maximum at $v_z \simeq 0$). This interpretation is corroborated by analytical expressions which are obtained in the Doppler limit (see the Appendix).

C. Pump field driving the lower $g \rightarrow r$ transition

We now turn to the situation represented in Fig. 1(b), where the pump field drives the lower transition $g \rightarrow r$ and the probe field is scanning the transition $r \rightarrow e$. For this configuration, Doppler-free signals centered at $\Delta_s = k_s/k_p \Delta_p$ are predicted in standard three-level spectroscopy independently of the geometry and of the ω_s/ω_p ratio. As previously, three cases are considered (with $k_p > 0$):

(i) The pump is tuned exactly to the frequency of the transition $g \rightarrow r$, i.e., $\Delta_p = 0$ [Fig. 3(b)]; then a narrow asymmetrical structure appears around $\Delta_s = 0$. In the local response case, one obtains an antisymmetrical dispersion curve whose width is mainly determined by the decay rate A_1 of the lower transition. The peak height of the nonlocal response result is one order of magnitude larger than that of the local one.

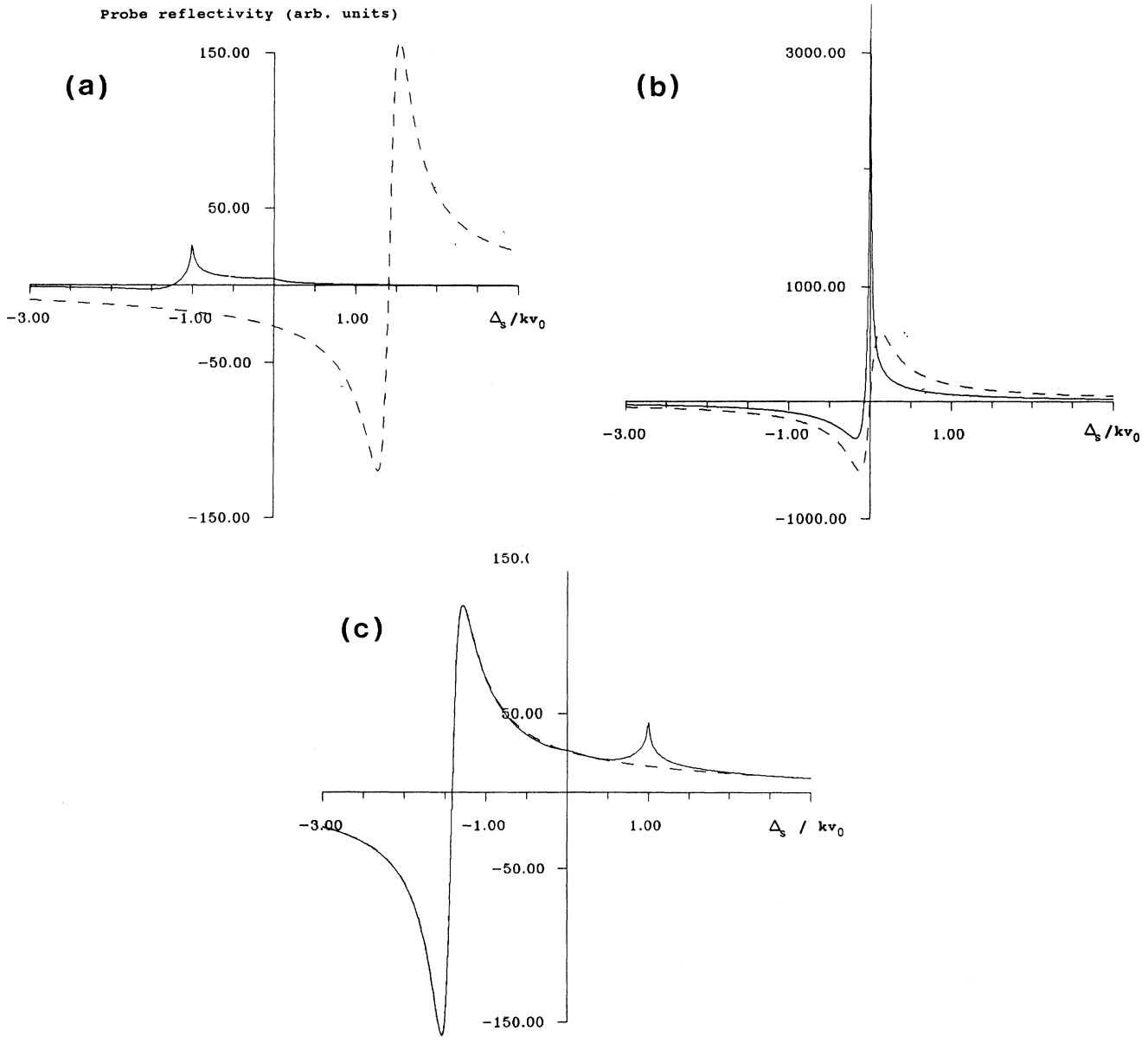

FIG. 3. Spectral variations of the probe reflectivity when the pump drives the lower transition $g \rightarrow r$ (solid lines, nonlocal susceptibility approach; dashed lines, local approach). Pump detuning is positive ($\Delta_p = +kv_0$) in (a), zero in (b), and negative ($\Delta_p = -kv_0$) in (c). Im(T) is in arbitrary units, probe frequency detunings Δ_s in units kv_0 with $k = (k_s + k_p)/2$.

(ii) The pump is detuned towards the blue with $\Delta_p = kv_0$ [Fig. 3(a)]. There is a narrow asymmetrical peak around $\Delta_s = -\Delta_p$ due to two-photon resonances in atoms with small v_z. Furthermore, a shallow structure around $\Delta_s = 0$ is present. In the local-response case, one predicts a dispersion curve centered around $\Delta_s = (k_s/k_p)\Delta_p$, due to the response of atoms with $v_z > 0$. In the nonlocal approach, these atoms exhibit a transient behavior while leaving the surface, and thus do not contribute to the signal. Therefore, in contrast to the previous case (i), the nonlocal response result is less intense than the corresponding "local" one by a factor of 5.

(iii) The pump is detuned towards the red with $\Delta_p = -kv_0$ [Fig. 3(c)]. One has a dispersion structure at $\Delta_s = (k_s/k_p)\Delta_p$, which matches the signal predicted in the local-response approach, since it comes from the steady-state response of atoms moving towards the surface. In addition, as previously, one has a narrow peak around $\Delta_s = -\Delta_p$ due to two-photon resonances.

VI. CONCLUSIONS

In this paper, we have considered nonlinear selective reflection spectroscopy from a cascade three-level atomic system. Transient response of atoms leaving the dielectric-vapor interface as well as steady-state response of atoms moving towards it were taken into account by solving the corresponding optical Bloch equations for the velocity-dependent atomic density matrix. Analytical expressions valid for arbitrary pump-field intensity have been derived for the relevant matrix elements. In the lowest nonvanishing order in the pump intensity, integration over the velocity distribution was performed numerically (and analytically in the Doppler limit for one of the two possible pump-probe configurations). Sub-Doppler structures in the selective reflection spectra are predicted and related to various specific processes involving atoms with definite velocity. The position and width of these structures were related to energy level separations and decay rates. Selective-reflection spectroscopy in three-level cascade systems should allow one to monitor the response of atoms leaving the surface and probe their velocity distribution. On the other hand, it yields a powerful tool to study the properties of highly excited atoms near a dielectric surface. In particular, Doppler-free two-photon reflection signals, originating in the response of $v_z = 0$ atoms, may provide a high-resolution optical probe of long-range atom-surface interactions [4,8] for highly excited states.

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APPENDIX

Integration over the velocity distribution in Eqs. (5.3) and (5.5) can be carried out analytically in the Doppler limit, i.e., under the assumption A_1, A_2, $|\Delta_p|$, and $|\Delta_s| \ll kv_0$. In this appendix we give the result for Eq. (5.3), i.e., for the case represented in Fig. 1(a) and discussed in Sec. V B, where the pump field drives the upper (transparent) $r \rightarrow e$ transition, while the weak probe field scans the lower $g \rightarrow r$ transition. The analytical results allow a more detailed discussion of the phenomena under study.

The relevant quantity $\mathrm{Re}T$ in Eq. (5.3) can be given a simplified expression if one takes into account the following property: since the velocity distribution is an even function, the integrand for negative velocity is equal to that for positive velocity if one changes the sign of k_p. Thus one has to calculate

$$I_+ = \int_0^{+\infty} dv \frac{\exp\left[-\dfrac{v^2}{v_0^2}\right]}{\left[\dfrac{A_2}{2} - i(\Delta_s + \Delta_p) + i(k_p - k_s)v\right]\left[\dfrac{A_1}{2} - i\Delta_s - ik_s v\right]^2} . \tag{A1}$$

When detunings and decay rates are small compared to the Doppler width, resonances occur only close to the center of the Gaussian function, so that in the integrand this function is replaced by 1 and the integration can be carried out analytically. One obtains

$$I_+ = \frac{i}{\left[\dfrac{A_1}{2} + i\Delta_s\right]\left[\dfrac{A_2 k_s + A_1(k_p - k_s)}{2} - i(k_s\Delta_p + k_p\Delta_s)\right]} + \frac{(k_p - k_s)}{\left[\dfrac{k_s A_2 + (k_p - k_s)A_1}{2} - i(k_s\Delta_p + k_p\Delta_s)\right]^2} N , \tag{A2}$$

where

$$N = \frac{\pi}{2}\left[1 + \frac{|k_p - k_s|}{k_p - k_s}\right] + \arctan\left[\frac{\Delta_s + \Delta_p}{A_2/2}\right] - \arctan\left[\frac{\Delta_s}{A_1/2}\right] + i \ln\left[\frac{|k_s|}{|k_p - k_s|}\right] + \frac{i}{2}\ln\left[\frac{(A_2/2)^2 + (\Delta_p + \Delta_s)^2}{(A_1/2)^2 + \Delta_s^2}\right] . \tag{A3}$$

The final result in the Doppler limit is given by

$$\mathrm{Re}T=\frac{C}{2}\mathrm{Im}(I_+ + I_-)\ , \tag{A4}$$

where I_- is obtained from I_+ [Eqs. (A2) and (A3)] by the substitution $k_p \rightarrow -k_p$. Plots of the numerical values given by (A4) as functions of the probe detunings Δ_s for fixed values of the pump detuning, i.e., similar to those given in Sec. V B, reproduce qualitatively the main features of the latter.

If the pump is detuned towards the blue by an amount Δ_p, a weak dispersion structure at Δ_s around zero and of the width A_1 arises from the first term in Eq. (A2) and from the corresponding term in I_-. For $\omega_p > \omega_s$, a narrow dispersion curve appears at $\Delta_s = -(k_s/k_p)\Delta_p$ due to the denominators in Eqs. (A2) and (A3), where the combination $(\Delta_p k_s + \Delta_s k_p)$ is present.

If the pump is detuned towards the red by an amount $\Delta_p > A_1$, the structure near zero is still present, and an asymmetrical structure around $\Delta_s = -\Delta_p$ arises from the last term in Eq. (A3). The characteristic width of this structure is given by $\sqrt{\Delta_p A_2}$ and is thus dependent on the pump detuning. This structure can be related to the two-photon coherence transient response for atoms moving along the surface, i.e., atoms with small v_z. It should be noticed that this contribution is also present in the previously discussed case where the pump detuning is positive but then the strong dispersion curve masks this shallow structure.

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