Selective reflection from an atomic vapor in a pump-probe scheme

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We evaluate the emission into a dielectric induced by the dipole polarization of an atomic vapor, in the case where two light beams are incident on the dielectric-vapor interface. These beams are a weak signal beam and a pump beam. Effects of spatial dispersion, arising from deexcitation of the vapor particles at collisions with the interface, are accounted for. The reflected radiation contains sub-Doppler structures arising from combinations of the effects of velocity-dependent transient behavior and the nonlinear response of the vapor. We express the modification of the signal-field reflection due to the pump field in terms of an effective susceptibility of the signal field. This susceptibility explicitly depends on the incidence angles and the frequencies of both beams. We present explicit simplified expressions for this effective susceptibility, valid in special cases. We also give some numerical results.

I. INTRODUCTION

The reflection of light from the interface between a dielectric and a vapor originates in a layer with a thickness of a few wavelengths. This allows us to use reflection spectroscopy as a probe of the properties of the boundary layer. It is known that the transient behavior of the atoms leaving the wall can give rise to a sub-Doppler structure in the reflection coefficient.[1–6] This structure arises from the stepwise discontinuity in the velocity dependence of the internal state. This effect has been studied for near-normal incidence,[2,3] for large-angle incidence in the region of total internal reflection,[5] and for large-density vapors, where frustrated total internal reflection may arise.[7]

A more sensitive probing of the velocity dependence of the internal state may be expected in the case of a pump-probe scheme in selective reflection.[8] This situation corresponds to the case where two nearly resonant light beams are incident on the interface. The reflection properties of one of the beams will then be modified by the presence of the other one. A typical feature of selective reflection spectroscopy as compared with absorption spectroscopy is that this modification is expected to be appreciable even when the two incident beams are not parallel or antiparallel, provided that their crossing coincides with the interface.

In a previous paper,[9] we have evaluated the reflectivity of a single incident beam with arbitrary intensity and arbitrary incidence angle. In the present paper, we consider the modification of the internal reflection of a weak signal field, due to the presence of a pump field. Both fields are incident on the interface between a dielectric with a real refractive index n and an atomic vapor. We derive explicit expressions for the dipole polarization of the vapor to first order in the signal field, as modified by the pump field. This dipole polarization, in turn, determines the contribution of the vapor to the beams reflected back into the dielectric. We explicitly study these reflected signals both for nearly normal incidence, and for incidence near the critical angle for total internal reflection.

The broadening of the absorptive and the dispersive nonlinear response of a medium as a result of the atomic motion and relaxation is a crucial feature in the theory of gas lasers.[10,11] In the present paper, the relaxation effect of the surface of the dielectric is explicitly included. Furthermore, it is known that at an interface between a dielectric and a nonlinear medium the reflectivity can switch between a state of total internal reflection and a state of transmission.[12,13] This effect requires an optically dense medium, and therefore it does not arise in the case of weak absorption considered here.

II. OPTICAL BLOCH EQUATIONS

We consider a vapor of two-state atoms[14] with ground state $|g\rangle$ and excited state $|e\rangle$. This vapor fills the half-space with $z>0$, and the xy plane is the interface between the vapor and a dielectric with the refractive index n. The vapor is driven by a pump field with frequency ω_p, and a weak signal field with frequency ω_s. Both fields correspond to a beam incident on the interface from the dielectric, and refracted into the vapor, as sketched in Fig. 1. Both incident plane waves have the same plane of incidence, which is taken as the xz plane.

The state of the atomic vapor is described by a local density matrix $\rho(\mathbf{r},\mathbf{v},t)$, with $\mathbf{r}$ a point in space, and $\mathbf{v}$ the

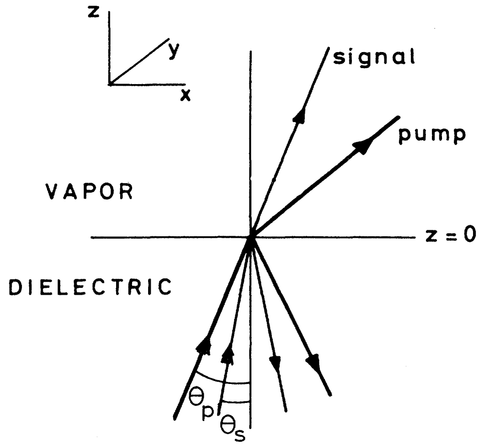

FIG. 1. Geometry of the interface between the dielectric ($z<0$), the vapor ($z>0$), and the two incident beams. The interface is the xy plane, and all wave vectors fall in the xz plane.

velocity of atoms passing that point at time t. We neglect velocity-changing collisions between atoms. Then the density matrix obeys the Liouville equation:

$$D\rho=-\frac{i}{\hbar}[H,\rho]-\Gamma\rho \ , \tag{2.1}$$

with

$$D=\frac{\partial}{\partial t}+\mathbf{v}\cdot\nabla \ . \tag{2.2}$$

The diagonal elements of the Hamiltonian H are the energies E_e and E_g of the free atoms, and the off-diagonal elements,

$$H_{eg}=-\boldsymbol{\mu}_{eg}\cdot\mathbf{E}_+(\mathbf{r},t), \quad H_{ge}=H_{eg}^* \ , \tag{2.3}$$

represent the interaction with the external radiation field, which has $\mathbf{E}_+$ as its positive-frequency part. The term $\Gamma\rho$ describes spontaneous decay; its diagonal elements are $A\rho_{ee}$, $-A\rho_{ee}$, and the off-diagonal terms are $(A/2)\rho_{eg}$, $(A/2)\rho_{ge}$. We assume that the atoms move in an external field that would exist in the half-space with $z>0$ in vacuum. This is justified when the effective refractive index of the vapor deviates little from unity, unless the angles of incidence are very close to the critical angle.

A. Transmission

When both incidence angles θ_p and θ_s are less than the critical angle θ_c, defined by $n\sin\theta_c=1$, the pump field and the signal field are refracted into the vapor, where they propagate with wave vectors $\mathbf{k}_p$ and $\mathbf{k}_s$. In order to eliminate the rapid oscillations imposed by the pump field, we introduce a new density matrix σ, with off-diagonal elements defined by

$$\begin{aligned}\rho_{eg}&=\sigma_{eg}e^{i\mathbf{k}_p\cdot\mathbf{r}-i\omega_p t} \ ,\\ \rho_{ge}&=\sigma_{ge}e^{-i\mathbf{k}_p\cdot\mathbf{r}+i\omega_p t} \ ,\end{aligned} \tag{2.4}$$

and with no change in the diagonal elements. From (2.1) we obtain the evolution equation:

$$\begin{aligned}D\sigma_{ee}&=\frac{i\Omega_p}{2}(\sigma_{ge}-\sigma_{eg})\\ &\quad+\frac{i\Omega_s}{2}(e^{-i\mathbf{q}\cdot\mathbf{r}+i\delta t}\sigma_{ge}-e^{i\mathbf{q}\cdot\mathbf{r}-i\delta t}\sigma_{eg})-A\sigma_{ee}\\ &=-D\sigma_{gg}\\ D\sigma_{eg}&=-\left[\frac{A}{2}-i\Delta_p\right]\sigma_{eg}+\frac{i\Omega_p}{2}(\sigma_{gg}-\sigma_{ee})\\ &\quad-\frac{i\Omega_s}{2}e^{-i\mathbf{q}\cdot\mathbf{r}+i\delta t}(\sigma_{gg}-\sigma_{ee}) \ ,\end{aligned} \tag{2.5}$$

where Ω_p and Ω_s are the Rabi frequencies, and

$$\Delta_p=\omega_p-\omega_0-\mathbf{k}_p\cdot\mathbf{v}, \quad \Delta_s=\omega_s-\omega_0-\mathbf{k}_s\cdot\mathbf{v} \ ,$$

are the Doppler-shifted detunings from resonance for the pump and signal field. Furthermore, we introduced

$$\mathbf{q}=\mathbf{k}_p-\mathbf{k}_s, \quad \delta=\omega_p-\omega_s \ .$$

B. Total reflection

In the oppposite case, when both angles of incidence are larger than the critical angle θ_c, the x components of the wave vectors k_{px} and k_{sx} are larger than ω_p/c and ω_s/c, and the z components $k_{pz}=i\kappa_p$, and $k_{sz}=i\kappa_s$, become imaginary. The fields

$$\begin{aligned}\mathbf{E}_p&=2\,\mathrm{Re}(\mathbf{E}_{p+}e^{ik_{px}x-\kappa_p z}) \ ,\\ \mathbf{E}_s&=2\,\mathrm{Re}(\mathbf{E}_{s+})e^{ik_{sx}x-\kappa_s z} \ ,\end{aligned} \tag{2.6}$$

are evanescent waves. We introduce the z-dependent Rabi frequencies

$$\begin{aligned}\Omega_p(z)&=\frac{2}{\hbar}\boldsymbol{\mu}_{eg}\cdot\mathbf{E}_{p+}e^{-\kappa_p z}=\Omega_p e^{-\kappa_p z} \ ,\\ \Omega_s(z)&=\frac{2}{\hbar}\boldsymbol{\mu}_{eg}\cdot\mathbf{E}_{s+}e^{-\kappa_s z}=\Omega_s e^{-\kappa_s z} \ ,\end{aligned} \tag{2.7}$$

and we define a modified density matrix σ' by

$$\begin{aligned}\rho_{eg}&=\sigma'_{eg}e^{ik_{px}x-i\omega_p t} \ ,\\ \rho_{ge}&=\sigma'_{ge}e^{-ik_{px}x+i\omega_p t} \ .\end{aligned} \tag{2.8}$$

This leads to the evolution equations:

$$\begin{aligned}D\sigma'_{ee}&=\frac{i\Omega_p(z)}{2}(\sigma'_{ge}-\sigma'_{eg})\\ &\quad+\frac{i\Omega_s(z)}{2}(e^{-iq_x x+i\delta t}\sigma'_{ge}-e^{iq_x x-i\delta t}\sigma'_{eg})-A\sigma'_{ee}\\ &=-D\sigma'_{gg} \ ,\\ D\sigma'_{eg}&=-\left[\frac{A}{2}-i\Delta'_p\right]\sigma'_{eg}+\frac{i\Omega_p(z)}{2}(\sigma'_{gg}-\sigma'_{ee})\\ &\quad+\frac{i\Omega_s(z)}{2}e^{-iq_x x+i\delta t}(\sigma'_{gg}-\sigma'_{ee}) \ ,\end{aligned} \tag{2.9}$$

where

$$\Delta'_p=\omega_p-\omega_0-k_{px}v_x, \quad \Delta'_s=\omega_s-\omega_0-k_{sx}v_x \ . \tag{2.10}$$

III. DIPOLE POLARIZATION IN WEAK-PROBE LIMIT

In order to evaluate the signals emitted back into the dielectric by the vapor, we calculate the dipole polarization of the vapor to the first order in the signal-field Rabi frequency Ω_s. Since the dipole polarization of two-state atoms contains only odd powers in the total field, the lowest nonvanishing term in the pump field is of the second order.

A. Transmission

We wish to derive the steady-state solution of the evolution equation (2.5) to first order in the signal field, with boundary conditions corresponding to the geometry and the properties of the interface. To zeroth order in Ω_s, the steady-state solutions of (2.5) vary only with z and v_z, since the geometry of the system is translationally invariant in the xy plane, and since the zeroth-order evolution is independent of x, y, and t. Since the first-order evolution contains terms proportional to $\exp[\pm i(\mathbf{q}\cdot\mathbf{r}-\delta t)]$, also the first order can have only a dependence on x, y, and t proportional to these terms. Hence, up to first order in Ω_s, the steady-state solution of (2.5) takes the form

$$\sigma(\mathbf{r},\mathbf{v},t)=S(z,\mathbf{v})+F(z,\mathbf{v})e^{-i\mathbf{q}\cdot\mathbf{r}+i\delta t}+G(z,\mathbf{v})e^{i\mathbf{q}\cdot\mathbf{r}-i\delta t} . \tag{3.1}$$

Here, S is the zeroth-order solution, and F and G give the first-order contribution to σ. Since σ is Hermitian, we obtain the relations

$$S^\dagger=S, \quad F^\dagger=G . \tag{3.2}$$

For a normalized solution of (2.5), we find that

$$\mathrm{Tr}S=1, \quad \mathrm{Tr}F=\mathrm{Tr}G=0 . \tag{3.3}$$

After substituting (3.1) into (2.5), we obtain the equations determining S and F in the form

$$\begin{aligned}\left[A+v_z\frac{d}{dz}\right]S_{ee}&=\tfrac{1}{2}i\Omega_p(S_{ge}-S_{eg}) ,\\ \left[\tfrac{1}{2}A-i\Delta_p+v_z\frac{d}{dz}\right]S_{eg}&=\tfrac{1}{2}i\Omega_p(1-2S_{ee}) ,\end{aligned} \tag{3.4}$$

and

$$\begin{aligned}\left[A+i(\Delta_p-\Delta_s)+v_z\frac{d}{dz}\right]F_{ee}&=\tfrac{1}{2}i\Omega_p(F_{ge}-F_{eg})+\tfrac{1}{2}i\Omega_s S_{ge},\\ \left[\tfrac{1}{2}A-i\Delta_s+v_z\frac{d}{dz}\right]F_{eg}&=-i\Omega_p F_{ee}+\tfrac{1}{2}i\Omega_s(1-2S_{ee}) ,\\ \left[\tfrac{1}{2}A+i(2\Delta_p-\Delta_s)+v_z\frac{d}{dz}\right]F_{ge}&=i\Omega_p F_{ee} .\end{aligned} \tag{3.5}$$

According to (3.2), G is determined by F.

We now proceed to solve Eqs. (3.4) and (3.5). First, we consider the case that $v_z>0$. We assume that the particles leave the surface at $z=0$ in the ground state, so that F_{ee}, F_{eg}, and F_{ge} disappear for $z=0$, $v_z>0$. We define the Laplace transform,

$$f(p,v_z)=\int_0^\infty dz\, e^{-pz}F(z,v_z) , \tag{3.6}$$

and, likewise, we define $s(p,v_z)$ and $g(p,v_z)$. Then, we obtain the algebraic equations:

$$\begin{aligned}(pv_z+A)s_{ee}&=\tfrac{1}{2}i\Omega_p(s_{ge}-s_{eg}) ,\\ (pv_z+\tfrac{1}{2}A-i\Delta_p)s_{eg}&=\tfrac{1}{2}i\Omega_p\left[\frac{1}{p}-2s_{ee}\right] ,\end{aligned} \tag{3.7}$$

and

$$\begin{aligned}[pv_z+A+i(\Delta_p-\Delta_s)]f_{ee}&=\tfrac{1}{2}i\Omega_p(f_{ge}-f_{eg})+\tfrac{1}{2}i\Omega_s s_{ge} ,\\ (pv_z+\tfrac{1}{2}A-i\Delta_s)f_{eg}&=-i\Omega_p f_{ee}+\tfrac{1}{2}i\Omega_s\left[\frac{1}{p}-2s_{ee}\right] ,\\ [pv_z+\tfrac{1}{2}A+i(2\Delta_p-\Delta_s)]f_{ge}&=i\Omega_p f_{ee} .\end{aligned} \tag{3.8}$$

The emission into the dielectric is determined by the optical coherences s_{eg}, f_{eg}, and g_{eg}. These quantities are easily evaluated from (3.7) and (3.8), to all orders in Ω_p, and to first order in Ω_s. In the present paper, we consider only the lowest nonvanishing contribution of the pump to the emission induced by the probe field. Therefore we evaluate f_{eg} and g_{eg} to second order in Ω_p. For abbreviation, we introduce the notation of

$$\Lambda_p=\frac{A}{2}-i\Delta_p, \quad \Lambda_s=\frac{A}{2}-i\Delta_s . \tag{3.9}$$

We obtain

$$f_{eg}(p)=\tfrac{1}{2}i\Omega_s\frac{1}{p(pv_z+\Lambda_s)}\left\{1-\tfrac{1}{2}\Omega_p^2\left[\frac{1}{pv_z+\Lambda_p^*+\Lambda_s}\left[\frac{1}{pv_z+\Lambda_s}+\frac{1}{pv_z+\Lambda_p^*}\right]+\frac{1}{pv_z+A}\left[\frac{1}{pv_z+\Lambda_p}+\frac{1}{pv_z+\Lambda_p^*}\right]\right]\right\} . \tag{3.10}$$

The expression for g_{eg} follows from that for f_{ge}, if we use the relation

$$g_{eg}(p)=[f_{ge}(p^*)]^* , \tag{3.11}$$

and we find that

$$g_{eg}=-\tfrac{1}{4}i\Omega_s\Omega_p^2\frac{1}{p(pv_z+\Lambda_p+\Lambda_s^*)(pv_z+2\Lambda_p-\Lambda_s)}\times\left[\frac{1}{pv_z+\Lambda_s^*}+\frac{1}{pv_z+\Lambda_p}\right] . \tag{3.12}$$

In the case of $v_z<0$, the particles are in their stationary state, so that S, F, and G are independent of z. The resulting expressions to the second order in Ω_p are

$$f_{eg}=\tfrac{1}{2}i\Omega_s\frac{1}{p\Lambda_s}\left[1-\frac{\Omega_p^2}{2\Lambda_p^*}\left[\frac{1}{\Lambda_s}+\frac{1}{\Lambda_p}\right]\right] \tag{3.10'}$$

and

$$g_{eg}=-\tfrac{1}{4}i\Omega_s\Omega_p^2\frac{1}{p(2\Lambda_p-\Lambda_s)\Lambda_p\Lambda_s^*} . \tag{3.12'}$$

B. Total reflection

In the case of incidence angles larger than the critical angle, we have to solve Eqs. (2.9). To the first order in Ω_s, the dependence on x and t may be eliminated by substitution:

$$\sigma'(\mathbf{r},\mathbf{v},t)=S'(z,\mathbf{v})+F'(z,\mathbf{v})e^{-iq_xx+i\delta t}+G'(z,\mathbf{v})e^{iq_xx-i\delta t} . \tag{3.13}$$

After defining the Laplace transforms s', f', and g' analogous to (3.6), we obtain the algebraic equations equivalent to (2.9) in the case that $v_z>0$:

$$(pv_z+A)s'_{ee}(p)=\tfrac{1}{2}i\Omega_p[s'_{ge}(p+\kappa_p)-s'_{eg}(p+\kappa_p)] ,$$
$$(pv_z+\tfrac{1}{2}A-i\Delta_p)s'_{eg}(p)=\tfrac{1}{2}i\Omega_p\left[\frac{1}{p+\kappa_p}-2s'_{ee}(p+\kappa_p)\right] , \tag{3.14}$$

and

$$\begin{aligned}[pv_z+A+i(\Delta_p'-\Delta_s')]f'_{ee}(p)&=\tfrac{1}{2}i\Omega_p[f'_{ge}(p+\kappa_p)-f'_{eg}(p+\kappa_p)]+\tfrac{1}{2}i\Omega_s s'_{ge}(p+\kappa_s) ,\\ (pv_z+\tfrac{1}{2}A-i\Delta_s')f'_{eg}(p)&=-i\Omega_p f'_{ee}(p+\kappa_p)+\tfrac{1}{2}i\Omega_s\left[\frac{1}{p+\kappa_s}-2s'_{ee}(p+\kappa_s)\right] ,\\ [pv_z+\tfrac{1}{2}A+i(2\Delta_p'-\Delta_s')]f'_{ge}(p)&=i\Omega_p f'_{ee}(p+\kappa_p) .\end{aligned} \tag{3.15}$$

Again, g' is determined by f', according to (3.11). Note that these equations contain the Laplace transforms at displaced values, as a result of the exponential decrease of the Rabi frequencies.

Equations (3.14) and (3.15) can be solved by an expansion in Ω_p, and we obtain to the second order for $v_z>0$ the following:

$$\begin{aligned}f'_{eg}(p)=\tfrac{1}{2}i\Omega_s\frac{1}{pv_z+\Lambda_s'}\Bigg\{&\frac{1}{p+\kappa_s}-\tfrac{1}{2}\Omega_p^2\frac{1}{p+\kappa_s+2\kappa_p}\\ &\times\Bigg[\frac{1}{(p+\kappa_p)v_z+(\Lambda_p')^*+\Lambda_s'}\left[\frac{1}{(p+2\kappa_p)v_z+\Lambda_s'}+\frac{1}{(p+\kappa_p+\kappa_s)v_z+(\Lambda_p')^*}\right]\\ &+\frac{1}{(p+\kappa_s)v_z+A}\left[\frac{1}{(p+\kappa_s+\kappa_p)v_z+\Lambda_p'}+\frac{1}{(p+\kappa_s+\kappa_p)v_z+(\Lambda_p')^*}\right]\Bigg]\Bigg\} ,\end{aligned} \tag{3.16}$$

and

$$g'_{eg}(p)=-\tfrac{1}{4}i\Omega_s\Omega_p^2\frac{1}{p+2\kappa_p+\kappa_s}\frac{1}{pv_z+2\Lambda_p'-\Lambda_s'}\frac{1}{(p+\kappa_p)v_z+\Lambda_p'+(\Lambda_s')^*}\left[\frac{1}{(p+2\kappa_p)v_z+(\Lambda_s')^*}+\frac{1}{(p+\kappa_p+\kappa_s)v_z+\Lambda_p'}\right] , \tag{3.17}$$

with

$$\Lambda_p'=\frac{A}{2}-i\Delta_p', \quad \Lambda_s'=\frac{A}{2}-i\Delta_s' . \tag{3.18}$$

For $v_z<0$, the particles are approaching the interface while entering the evanescent waves, and they experience an exponentially increasing intensity of both fields. When we expand the matrices S' and F' in powers of Ω_p, the second-order contribution to F'_{eg} and F'_{ge} are proportional to $\exp[-(2\kappa_p+\kappa_s)v_z]$, while the first-order contribution to F'_{ee} varies as $\exp[-(\kappa_p+\kappa_s)v_z]$. Likewise, the second-order contributions to S'_{ee} and S'_{ge} vary as $\exp(-2\kappa_p z)$ and $\exp(-\kappa_p z)$. Hence the z derivatives in the expansion of the evolution equations for S' and F' can be directly substituted. In this way, we readily obtain explicit results for F'_{eg} and G'_{eg} to second order in Ω_p, with the Laplace transforms:

$$f'_{eg}=\tfrac{1}{2}i\Omega_s\left\{\frac{1}{(p+\kappa_s)(\Lambda'_s-\kappa_p v_z)}-\tfrac{1}{2}\Omega_p^2\frac{1}{p+2\kappa_p+\kappa_s}\frac{1}{\Lambda'_s-(2\kappa_p+\kappa_s)v_z}\right.$$
$$\times\left[\frac{1}{(\Lambda'_p)^*+\Lambda'_s-(\kappa_p+\kappa_s)v_z}\left[\frac{1}{\Lambda'_s-\kappa_s v_z}+\frac{1}{(\Lambda'_p)^*-\kappa_p v_z}\right]\right.$$
$$\left.\left.+\frac{1}{A-2\kappa_p v_z}\left[\frac{1}{\Lambda'_p-\kappa_p v_z}+\frac{1}{(\Lambda'_p)^*-\kappa_p v_z}\right]\right]\right\}, \qquad (3.16')$$

and

$$g'_{eg}=-\tfrac{1}{4}i\Omega_s\Omega_p^2\frac{1}{p+2\kappa_p+\kappa_s}\frac{1}{2\Lambda'_p-\Lambda'_s-(2\kappa_p+\kappa_s)v_z}\frac{1}{\Lambda'_p+(\Lambda'_s)^*-(\kappa_p+\kappa_s)v_z}\left[\frac{1}{(\Lambda'_s)^*-\kappa_s v_z}+\frac{1}{\Lambda'_p-\kappa_p v_z}\right]. \qquad (3.17')$$

C. Dipole polarization

Now that we have obtained explicit expressions for the atomic optical coherences to first order in the signal-field amplitude, we are ready to give the dipole polarization of the vapor, at a density N, with the velocities $\mathbf{v}$ described by the Maxwell distribution $W(\mathbf{v})$. When both angles of incidence are below the critical angle, we find that

$$\mathbf{P}(\mathbf{r},t)=\hat{y}N\mu_{ge}2\,\mathrm{Re}\left[\int d\mathbf{v}\,W(\mathbf{v})[F_{eg}(z,v_z)e^{i\mathbf{k}_s\cdot\mathbf{r}-i\omega_s t}+G_{eg}(z,v_z)e^{i(2\mathbf{k}_p-\mathbf{k}_s)\cdot\mathbf{r}-i(2\omega_p-\omega_s)t}]\right], \qquad (3.19)$$

where the Laplace transforms of F_{eg} and G_{eg} are given in Eqs. (3.10) and (3.12) for $v_z>0$, and in Eqs. (3.10′) and (3.12′) for $v_z<0$. In the domain of total internal reflection for both incident beams, the dipole polarization is

$$\mathbf{P}(\mathbf{r},t)=\hat{y}N\mu_{ge}2\,\mathrm{Re}\left[\int d\mathbf{v}\,W(\mathbf{v})[F'_{eg}(z,v_z)e^{ik_{sx}x-i\omega_s t}+G'_{eg}(z,v_z)e^{i(2k_{px}-k_{sx})x-i(2\omega_p-\omega_s)t}]\right]. \qquad (3.20)$$

The Laplace transforms of F'_{eg} and G'_{eg} are specified in (3.16) and (3.17) for $v_z>0$, and in (3.16′) and (3.17′) for $v_z<0$. The first terms in (3.19) and (3.20) represent the polarization of the vapor at the frequency of the signal field. This polarization has a strength that is modified by the pump field. The second terms in (3.19) and (3.20) describe a polarization at the frequency $2\omega_p-\omega_s$, resulting from nonlinear mixing of the two incident frequencies. Of course, there is also a contribution to $\mathbf{P}$ to zeroth order in Ω_s. This is merely the dipole polarization for a single incident beam, which has been studied in a previous paper.[9]

In the next section we give the expressions determining the emission into the dielectric.

IV. EMISSION BY DIPOLE POLARIZATION

A. Emission amplitudes

The reflected fields into the dielectric can be separated according to

$$\mathbf{E}_r(\mathbf{r},t)=\mathbf{E}_{r0}(\mathbf{r},t)+\mathbf{E}_{rd}(t)\,. \qquad (4.1)$$

Here, $\mathbf{E}_{r0}$ gives the reflected field that would be present in the absence of the atomic vapor, corresponding to internal reflection at a dielectric-vacuum interface. The field $\mathbf{E}_{rd}$ is the emission back into the dielectric, due to the polarization of the vapor. As we demonstrated in Sec. III, in the present case of a single plane of incidence chosen as the xz plane, the polarization $\mathbf{P}(\mathbf{r})$ may be separated into various terms that are characterized by a frequency and an oscillatory variation with s. Hence we may generally write

$$\mathbf{P}(\mathbf{r},t)=\hat{y}\sum_\alpha 2\,\mathrm{Re}[P_\alpha(z)e^{i\xi_\alpha x-i\omega_\alpha t}]\,. \qquad (4.2)$$

The first-order contribution in Ω_s, given in (3.19) and (3.20), contains a term $\alpha=s$ at the signal-field frequency ω_s, and $\xi_s=k_{xs}$, and a term $\alpha=n$, with the nonlinear frequency combination $\omega_n=2\omega_p-\omega_s$, and $\xi_n=2k_{px}-k_{sx}$. In a previous paper, we derived an expression for the radiation emitted into the dielectric by a dipole polarization of the form of the summand in (4.2). Each layer of polarization at a given value of z emits radiation to opposite sides, with wave vectors (ξ_α, 0, $\pm\zeta_\alpha$), with

$$\zeta_\alpha=(k_\alpha^2-\xi_\alpha^2)^{1/2}\,, \qquad (4.3)$$

with $k_\alpha=\omega_\alpha/c$. The radiation emitted toward the interface is refracted into the dielectric and propagates there with the wave vector $\mathbf{K}_\alpha$, with

$$K_{\alpha x}=\xi_\alpha,\quad K_{\alpha z}=-(n^2k_\alpha^2-\xi_\alpha^2)^{1/2}\,. \qquad (4.4)$$

We specify the emission angles in the vapor and the dielectric by specifying their cosine as

$$\beta_\alpha=\zeta_\alpha/k_\alpha,\quad b_\alpha=|K_{\alpha z}|/nk_\alpha\,. \qquad (4.5)$$

From the results of our previous paper,[9] it follows that each summand in (4.2) gives a separate contribution to the emission, and that the electric field of the radiation emitted into the dielectric by the dipole polarization (4.2) is

$$\mathbf{E}_{rd}(\mathbf{r},t)=\hat{y}\sum_{\alpha} 2\mathrm{Re}(E_\alpha e^{i\mathbf{K}_\alpha\cdot\mathbf{r}-i\omega_\alpha t}) , \tag{4.6}$$

with amplitude

$$E_\alpha=\frac{1}{nb_\alpha+\beta_\alpha}\frac{ik_\alpha}{\epsilon_0}\int_0^\infty dz\, e^{i\beta_\alpha k_\alpha z}P_\alpha(z) . \tag{4.7}$$

This amplitude is determined by the Laplace transform of $P_\alpha(z)$.

B. Effective susceptibility

The emission by the polarization at the frequency ω_s contributes to the reflection of the incident signal field, and it is modified by the pump field. It is natural to express this emission by an effective susceptibility χ_s of the vapor. This susceptibility is defined as the macroscopic susceptibility of a fictitious macroscopic medium that would reproduce the reflectivity of the signal field. The dipole polarization in this fictitious medium would be given by

$$\mathbf{P}_{\rm fic}(\mathbf{r},t)=\hat{y}\,2\,\mathrm{Re}\left[\epsilon_0\chi_s\frac{\hbar}{2\mu_{eg}}\Omega_s e^{i\mathbf{k}_s\cdot\mathbf{r}-i\omega_s t}\right] . \tag{4.8}$$

This polarization leads to the same integral as that in (4.7) for $\alpha=s$, provided that

$$\chi_s=\frac{-2ik_{sz}}{\epsilon_0}\frac{2\mu_{eg}}{\hbar\Omega_s}\int_0^\infty dz\, e^{ik_{sz}z}P_s(z) . \tag{4.9}$$

This expression is also valid for total reflection of the signal beam, but then we have to replace ik_{sz} by $-\kappa_s$. An explicit expression for $P_s(z)$ is obtained if we compare (3.19) or (3.20) with the general form (4.2), and if we identify the term with $\alpha=s$ with the contribution of F_{eg} and F'_{eg}. Hence we find

$$P_s(z)=N\mu_{ge}\int d\mathbf{v}\, W(\mathbf{v})F_{eg}(z,\mathbf{v})e^{ik_{sz}z} . \tag{4.10}$$

The effective susceptibility is therefore

$$\chi_s=\frac{-2ik_{sz}}{\epsilon_0}\frac{2|\mu_{eg}|^2N}{\hbar\Omega_s}\int d\mathbf{v}\, W(\mathbf{v})f_{eg}(-2ik_{sz},\mathbf{v})$$
$$=\chi_{s0}+\Omega_p^2\chi_{s1} \tag{4.11}$$

in the transmission case. In the situation of total reflection, we find that

$$P_s(z)=N\mu_{ge}\int d\mathbf{v}\, W(\mathbf{v})F'_{eg}(z,\mathbf{v}) , \tag{4.12}$$

and

$$\chi_s=\frac{2\kappa_s}{\epsilon_0}\frac{2|\mu_{eg}|N^2}{\hbar\Omega_s}\int d\mathbf{v}\, W(\mathbf{v})f'_{eg}(\kappa_s,\mathbf{v})$$
$$=\chi_{s0}+\Omega_p^2\chi_{s1} . \tag{4.13}$$

The slight difference in form (4.11) and (4.13) is a direct consequence of the different definition of F and F', according to (3.1) and (3.13). In fact, the effective susceptibility χ_s varies continuously when the signal-field incidence angle passes the critical angle. One should notice that χ_s as given by (4.11) or (4.13) is independent of the signal-field Rabi frequency Ω_s.

In the transmission case, when the angle of incidence of the signal field is near the critical angle, so that

$$k_{sz}v_0 \ll A \tag{4.14}$$

for typical thermal velocities v_0, the effective susceptibility (4.11) takes the simple form

$$\bar{\chi}_s=\frac{i|\mu_{eg}|^2N}{\hbar\epsilon_0}\int d\mathbf{v}\, W(\mathbf{v})\frac{1}{\Lambda_s}\left[1-\tfrac{1}{2}\Omega_p^2\frac{1}{\Lambda_p^*}\left[\frac{1}{\Lambda_s}+\frac{1}{\Lambda_p}\right]\right]$$
$$=\bar{\chi}_{s0}+\Omega_p^2\,\bar{\chi}_{s1} . \tag{4.15}$$

This is just the local susceptibility for the signal field, as modified by the pump. The effect of the boundary layer has disappeared.

On the other hand, when both angles of incidence are just beyond the critical angle, so that (4.13) applies with

$$\kappa_s v_z,\quad \kappa_p v_z \ll A , \tag{4.16}$$

then we obtain

$$\chi_s=\bar{\chi}_{s0}+\frac{\kappa_s}{\kappa_s+\kappa_p}\Omega_p^2\,\bar{\chi}_{s1} . \tag{4.17}$$

This demonstrates that the modifying effect of the pump field on the reflection of the signal field vanishes at the critical angle ($\kappa_s=0$), when the incidence angle of the pump field is beyond the critical angle ($\kappa_p>0$). This is due to the fact that the critical signal-field reflection builds up over a large depth into the vapor, whereas the pump field extends only over a finite depth.

We emphasize that the effective susceptibility χ_s is just a characterization of the integral in (4.7), for the term $\alpha=s$. Its introduction implies no additional approximation. On the other hand, when χ_s varies negligibly with the incidence angle θ_s of the signal beam in the region where β_s is not large compared with χ_s, we can use this value of χ_s as a macroscopic susceptibility that determines the continuous transition of the reflectivity between the case of transmission and the case of total reflection. In this way, we avoid the singularity in f_{eg} or f'_{eg} at the p value indicated in (4.11) and (4.13) and which does not show up in χ_s.

C. Nonlinear emission

The term in (3.19) or (3.20) at frequency $2\omega_p-\omega_s$ is labeled by $\alpha=n$ in (4.2). This term gives rise to an emission at this same frequency into the dielectric with electric field as indicated in (4.6). This term actually corresponds to a four-wave mixing process, in which absorption of two photons from the pump beam is accompanied by emission of a photon into the signal beam and by emission of a photon with wave vector $2\mathbf{k}_p-\mathbf{k}_s$. The nonlinear polarization at this wave vector then leads to coherent emission into the dielectric. This emission has recently been observed in the case of small incidence angles.[15] The emission angle has the cosine

$$b_n = \left[1 - \frac{c^2}{n^2} \left[\frac{2k_{px} - k_{sx}}{2\omega_p - \omega_s} \right]^2 \right]^{1/2} . \tag{4.18}$$

Only when this cosine term is real does the emission lead to propagation in the dielectric. Otherwise, the polarization term P_n gives rise to an evanescent wave in the dielectric. According to (4.7), in the transmission case, the amplitude of the emission is determined by the Laplace transform $g_{eg}(p)$ for $p = i\zeta_n - i(2k_{pz} - k_{sz})$, with

$$\zeta_n = \left[\left[\frac{2\omega_p - \omega_s}{c} \right]^2 - (2k_{px} - k_{sx})^2 \right]^{1/2} . \tag{4.19}$$

In the case of total reflection, the emission amplitude is determined by $g'_{eg}(p)$ for $p = -i\zeta_n$, which is real and positive when the argument under the square root in (4.19) is negative.

Other schemes of four-wave mixing in the presence of a standing pump wave may lead to spatial instabilities.[16,17] This type of effect can only compete with selective reflection to higher order in the signal-field amplitude.

V. REFLECTIVITY AND SUSCEPTIBILITY NEAR THE CRITICAL ANGLE

The effective susceptibility χ_s, given by (4.9), is defined in such a way that it reproduces the correct reflection of the signal beam. This implies that the field amplitude (4.7) for $\alpha = s$, which is the contribution to the reflected signal field from the vapor polarization, coincides with the result of the macroscopic Fresnel method for the reflected field to first order in a given complex susceptibility. This linearization of the reflected field in the susceptibility is not justified in the transition region between transmission and total internal reflection, where

$$k_{sz}/k_s, \kappa_s/k_s \lesssim \chi_s . \tag{5.1}$$

However, the macroscopic theory of reflection also gives an explicit expression for the transmission region (5.1), where the incidence angle θ_s of the signal beam is very near the critical angle θ_c. Hence we may employ this macroscopic theory in the transition region, provided that the effective susceptibility χ_s is constant in this region. This is the case only when the condition (4.14) is valid over the transition region (5.1). This is only true provided that

$$\chi_s \ll A/k_s v_0 , \tag{5.1$'$}$$

which implies that the susceptibility χ_s is much smaller than the ratio between the natural width and the Doppler width. Furthermore, the incidence angle θ_p of the pump field must lie outside the transition region.

A signal beam with incidence angle θ_s, and frequency ω_s, incident from a dielectric with refractive index n on the interface with a macroscopic medium with complex susceptibility χ_s, gives rise to a field in the medium with a complex z component of the wave vector, which we specify as $B\omega_s/c$. The complex quantity B is determined by the requirement that the wave solves the Maxwell wave equation, and that the x component of the wave vector is the same at both sides of the interface, according to Snell's law. Hence B is specified by the equation

$$B^2 + n^2 \sin^2\theta_s = 1 + \chi_s . \tag{5.2}$$

The amplitudes of the field in the medium and of the reflected field are determined by the Fresnel continuity conditions for the parallel components of the electric and the magnetic fields. In the case considered in this paper, where the polarization direction is normal to the plane of incidence, the intensity reflection coefficient R is found to be

$$R = \left| \frac{nb_s - B}{nb_s + B} \right|^2 , \tag{5.3}$$

with $b_s = \cos\theta_s$. When $|1 - n^2\sin^2\theta_s|$ is much larger than χ_s, we expand R to first order in χ_s. For $\theta_s < \theta_c$, we introduce

$$\beta_s = (1 - n^2 \sin^2\theta_s)^{1/2} , \tag{5.4}$$

and the expansion of (5.3) gives

$$R = \left[\frac{nb_s - \beta_s}{nb + \beta_s} \right]^2 - \frac{2nb_s(nb_s - \beta_s)}{\beta_s(nb_s + \beta_s)} \mathrm{Re}(\chi_s) . \tag{5.5}$$

This corresponds to the case of transmission. In the opposite case of total internal reflection, when $\theta_s > \theta_c$, we introduce

$$\eta_s = (n^2 \sin^2\theta_s - 1)^{1/2} , \tag{5.6}$$

and we obtain from (5.3), to first order in χ_s,

$$R = 1 - \frac{2nb_s}{(nb_s)^2 + \eta_s^2} \frac{1}{\eta_s} \mathrm{Im}(\chi_s) . \tag{5.7}$$

These reflection coefficients correspond exactly to the polarization contribution (4.7) for $\alpha = s$.

In the transition region, where $\theta_s - \theta_c$ is very small, also β_s or η_s are close to zero, and the expansion is no longer justified. However, in this transition region, we can expand B^2 to first order in $\theta_s - \theta_c$, with the result that

$$B^2 = \chi_s - 2nb_c(\theta_s - \theta_c) , \tag{5.8}$$

with $b_c = \cos\theta_c = (1 - n^{-2})^{1/2}$. Note that $|B| \ll 1$ in the transition region. In order to obtain an approximate expression for R, we use the additional expansion[18]

$$nb_s = nb_c - (\theta_s - \theta_c) . \tag{5.9}$$

If we substitute (5.8) and (5.9) in (5.3), we find to first order in B that

$$R = 1 - \frac{4}{nb_c} \mathrm{Re}[\chi_s - 2nb_c(\theta_s - \theta_c)]^{1/2} . \tag{5.10}$$

For $\chi_s \ll 1$, the validity conditions $\beta_s, \eta_s \gg \chi_s$ for (5.3) and (5.5), and $\theta_s - \theta_c \ll 1$ for (5.10) cover all possible values of θ_s. Hence (5.10) describes the transition between the validity region of (5.3), and that of (5.5).

When the incidence angle θ_p of the pump field is smaller than θ_c, so that the pump is in the transmission regime, we can apply the transition equation (5.10) with χ_s given by (4.15). The effect of the spatial dispersion in the boundary disappears in this case.

When the pump is in the region of total reflection, $\theta_p > \theta_c$, the transition region for the signal reflection is described by (5.10), with $\chi_s = \chi_{s0}$. The modification by the pump field disappears in this case, and the reflection in the critical region, $|\theta_s - \theta_c| \lesssim \chi_s$, is determined by the single-field reflectivity without spatial dispersion.

VI. APPROXIMATE ANALYTICAL INTEGRATION

In Sec. V we showed how the reflection coefficient R of the signal field is determined by the effective susceptibility χ_s. This relation between R and χ_s is given by (5.5) in the transmission case, when $\theta_s < \theta_c$. Then, χ_s is given by (4.11), with f_{eg} specified in (3.10). In the case of total reflection, when $\theta_s > \theta_c$, R is determined by χ_s according to (5.7), with χ_s given in (4.13) in terms of f'_{eg}, which in turn is specified in (3.16). In the transition region, where $2nb_c|\theta_s - \theta_c|$ is of the same order as χ_s, the relation between R and χ_s is given in (5.10).

In all these cases, the modification due to the pump field of the signal-field reflectivity is determined by the terms proportional to Ω_p^2 in (3.10) or (3.16). This pump contribution has the form of a Maxwellian average of products of three denominators. It is noteworthy that these averages involve only a single velocity component, both in the case of normal incidence [when (3.10) and (3.16) depend only on v_z], and in the situation of critical incidence [when (3.10) and (3.16) exclusively depend on v_x]. In the Doppler limit, when A, $\omega_s - \omega_0$, and $\omega_p - \omega_0$ are small compared with the Doppler width kv_0, the integrals are mainly determined by the atoms with velocity component zero. Then we may replace the one-dimensional Maxwellians $W_0(v_z)$ or $W_0(v_x)$ by $W_0(0)$. The remaining integration can be performed analytically.

A. Normal incidence

For normal incidence, the pump contribution to χ_s takes the form obtained from (4.11) and (3.10):

$$\chi_{s1} = -\frac{i|\mu_{eg}|^2 N}{2\epsilon_0 \hbar} \int dv\, W_0(v) \times \left\{ \Theta(v) \frac{1}{A/2 - i\delta_s - ikv} \left[\frac{1}{A + i\delta - 2ikv} \left[\frac{1}{A/2 - i\delta_s - ikv} + \frac{1}{A/2 + i\delta_p - 3ikv} \right] + \frac{1}{A - 2ikv} \left[\frac{1}{A/2 - i\delta_p - ikv} + \frac{1}{A/2 + i\delta_p - 3ikv} \right] \right] + \Theta(-v) \frac{1}{A/2 - i\delta_s + ikv} \frac{1}{A/2 + i\delta_p - ikv} \left[\frac{1}{A/2 - i\delta_s + ikv} + \frac{1}{A/2 - i\delta_p + ikv} \right] \right\}, \tag{6.1}$$

with $\delta_s = \omega_s - \omega_0$, $\delta_p = \omega_p - \omega_0$, $\delta = \delta_p - \delta_s$, and Θ the step function. When δ_s and δ_p are both small compared with the Doppler width, the integral arises from the velocity group with $v \cong 0$, and we may replace $W_0(v)$ by $W_0(0)$. The remaining integration can be performed, with the result that

$$\chi_{s1} = -\frac{|\mu_{eg}|^2 N}{2\epsilon_0 \hbar k} W_0(0) \times \left\{ \left[\tfrac{1}{2}\ln \left[1 + \frac{4\delta_s^2}{A^2} \right] - i \arctan \left[\frac{2\delta_s}{A} \right] \right] \left[\frac{\delta_p + 5\delta_s + 2iA}{(\delta_s + \delta_p)^2 (\delta_p + 3\delta_s + iA)} + \frac{4\delta_s + iA}{2\delta_s \delta (\delta_p + 3\delta_s + iA)} - \frac{iA}{\delta(\delta - iA)^2} \right] + \left[\tfrac{1}{2}\ln \left[1 + \frac{\delta^2}{A^2} \right] + i \arctan \left[\frac{\delta}{A} \right] \right] \frac{-4(\delta - iA)}{(\delta_s + \delta_p)^2 (\delta_p - 3\delta_s - 2iA)} + \left[\tfrac{1}{2}\ln \left[1 + \frac{4\delta_p^2}{A^2} \right] + i \arctan \left[\frac{2\delta_p}{A} \right] - \ln 3 \right] \frac{3(\delta_p + 3\delta_s + 4iA)}{2(\delta_p + iA)(\delta_p + 3\delta_s + iA)(\delta_p - 3\delta_s - 2iA)} + \left[\tfrac{1}{2}\ln \left[1 + \frac{4\delta_p^2}{A^2} \right] + i \arctan \left[\frac{2\delta_p}{A} \right] - i\pi \right] \frac{-(\delta - 2iA)}{iA(\delta - iA)^2} + \left[\tfrac{1}{2}\ln \left[1 + \frac{4\delta_p^2}{A^2} \right] - i \arctan \left[\frac{2\delta_p}{A} \right] \right] \frac{\delta_p - iA/2}{iA\delta_p \delta} \right\}. \tag{6.2}$$

B. Critical incidence

Now, we consider the case that the incidence angles θ_p and θ_s are opposite and near the critical angle θ_c, so that $k_{px}=-k_{sx}\cong k$. First, we consider the transmission case, so that the z components of the wave vectors are real, and we write

$$k_{pz}=k_{sz}=\beta k \ , \tag{6.3}$$

with $\beta \ll 1$. Then the pump contribution to the effective susceptibility is given by (4.11) and (3.10). In the Doppler limit, only the velocity group with small values of v_x contributes. In the Maxwellian, the x component of the velocity may be put equal to zero. Then the integration over v_x may be performed analytically, where only a single term from (3.10) and from (3.10′) contributes. In the resulting expression, the only remaining integration is over v_z. We find, after a simple contour integration, that

$$\chi_{s1}=\frac{-i\pi|\mu_{eg}|^2N}{\epsilon_0\hbar k}W_0(0) \times \int dv\, W_0(v) \times \left[\Theta(v)\frac{1}{A-i\delta_s-i\delta_p-2i\beta kv}\frac{1}{A-2i\beta kv} +\Theta(-v)\frac{1}{A-i\delta_s-i\delta_p+2i\beta kv}\frac{1}{A}\right] . \tag{6.4}$$

The corresponding contribution to the signal reflection is an almost Doppler-free dispersion curve.

Next, we consider the case of incidence slightly beyond the critical angle, so that

$$\kappa_p=\kappa_s=\eta k \ , \tag{6.5}$$

with $\eta \ll 1$. Then, the effective susceptibility is given by (4.13) and (3.16). In the Dopper limit, the integration over v_x is performed analytically. The contribution from positive and negative values of v_z turns out to be equal, and we obtain the result

$$\chi_{s1}=\frac{-i\pi|\mu_{eg}|^2N}{\epsilon_0\hbar k}W_0(0) \times \int_0^\infty dv\, W_0(v)\frac{1}{A+2\eta kv} \times \frac{1}{A+4\eta kv-i\delta_s-i\delta_p} . \tag{6.6}$$

The remaining integrations in (6.4) and (6.6) must be performed numerically. The contribution to the signal reflection is a Doppler-free absorption profile that is broadened by the finite transit time of atoms passing the evanescent wave of the pump field.

VII. NUMERICAL RESULTS

We present some examples of numerical evaluations of the modification of the signal-field reflectivity due to the pump. We use the reduced frequencies f_s and f_p, with

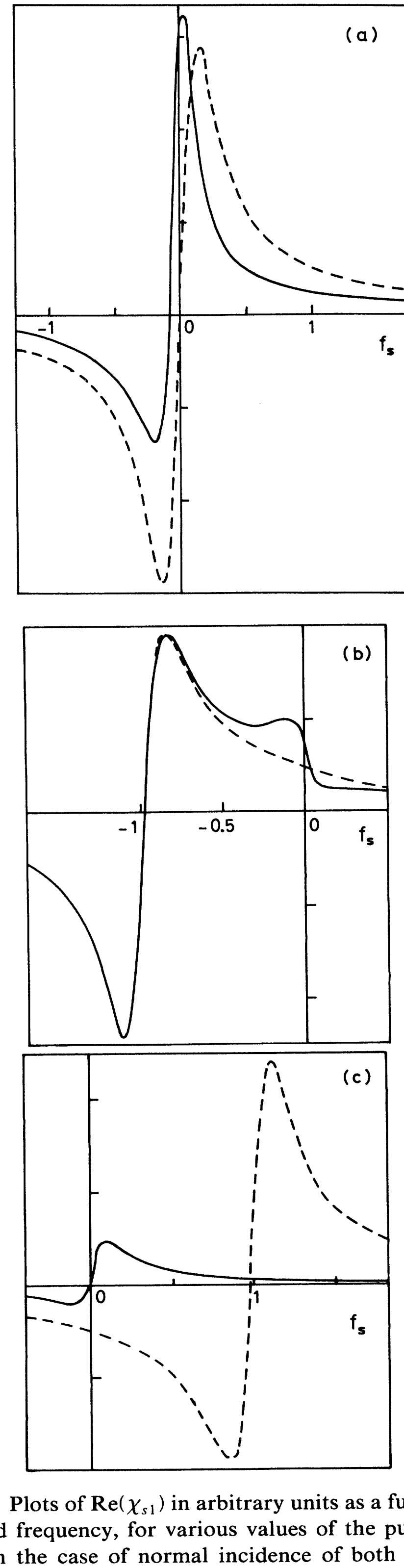

FIG. 2. Plots of Re(χ_{s1}) in arbitrary units as a function of the signal-field frequency, for various values of the pump-field frequency, in the case of normal incidence of both beams. This quantity determines the modification of the signal reflectivity due to the pump. The solid curves correspond to Eq. (4.11), and the dotted curves give Re($\overline{\chi}_{s1}$) as specified by (4.15), where spatial dispersion is neglected. In this and the following figures we have taken $kv_0=5A$. (a) $f_p=0$; (b) $f_p=-1$; (c) $f_p=1$.

$$f_s = \delta_s / k v_0, \quad f_p = \delta_p / k v_0 , \tag{7.1}$$

with $v_0 = (2k_B T/m)^{1/2}$ a typical thermal velocity. We consider the case that $2kv_0/A = 10$, so that the Doppler width is ten times larger than the natural width.

A. Normal incidence

In Fig. 2 we present results for the situation of normal incidence, so that $k_{px} = k_{sx} = 0$, for various values of the pump frequency. We plot $\text{Re}(\chi_{s1})$, defined in (4.11), as a function of f_s. Notice that according to (5.5), the effect on the reflectivity is proportional to $-\text{Re}(\chi_{s1})$.

Figure 2(a) displays $\text{Re}(\chi_{s1})$ for $f_p = 0$. Then the pump affects mainly the atoms with small values of v_z, and the pump modification is centered at values $f_s = 0$. The dispersion curve is not fully antisymmetric, due to the spatial dispersion in the boundary layer. This is obvious if we compare this curve with $\text{Re}(\bar{\chi}_{s1})$, indicated by the dotted curve in Fig. 2(a), for the same value $f_p = 0$ of the pump frequency. This latter plot, which represents Eq. (4.15) with the k vector chosen in the z direction, is perfectly antisymmetric around $f_s = 0$.

The transient behavior in the boundary region is more pronounced in the case of a pump frequency in the red Doppler wing. Figure 2(b) shows $\text{Re}(\chi_{s1})$ as a function of f_s for $f_p = -1$. Then the pump is resonant for particles approaching the surface. The dotted curve in Fig. 2(b) represents $\text{Re}(\bar{\chi}_{s1})$, which is the local susceptibility without spatial dispersion. One notices that the effect of the transient behavior of the particles leaving the interface is mainly a dispersion curve at resonance. This additional dispersive resonance is induced by the velocity-changing collisions with the interface.

The effect of the boundary layer on the pump contribution to the signal susceptibility is most dramatic for positive values of f_p. Then, the pump is on resonance for particles leaving the interface, which are in their transient regime after deexcitation at the interface. Comparing the curves for $\text{Re}(\chi_{s1})$ and for $\text{Re}(\bar{\chi}_{s1})$ in Fig. 2(c), one notices that the deexcitation at the boundary strongly diminishes the pump contribution to $\text{Re}(\chi_{s1})$. The deexcitation at the interface destroys the phase variation of the pump contribution to the z-dependent polarization, thereby diminishing the integral in (4.9). The result is that $\text{Re}(\chi_{s1})$ merely displays a weak dispersion curve at resonance.

B. Nearly critical incidence

We consider the situation where both beams have the same frequency $\omega_p = \omega_s = \omega$, and incidence angles near the critical angle, but with opposite values of k_x. Then, the refracted waves in the vapor are nearly counterpropagating, and one expects sub-Doppler structures in the response of the vapor at resonance. Furthermore, according to the results of Sec. IV, the effect of the boundary layer on the signal reflection is expected to disappear when we approach the critical angle θ_c.

In Fig. 3 we plot the pump contribution to the reflective part of the susceptibility $\text{Re}(\chi_{s1})$, in the case that $\theta_p = -\theta_s \lesssim \theta_c$, as a function of $f = f_p = f_s$, for various values of $\beta_s = \beta_p = \beta$, which is the cosine of the refraction angles. We notice that, for increasing values of β, the displacement of the dispersion curves increases. This indicates an increasing deviation from the fully antisymmetric curve pertaining to $\text{Re}(\bar{\chi}_{s1})$, the real part of the susceptibility without spatial dispersion. Furthermore, the curves are increasingly broadened for increasing values of β, because of the increasing disalignment of the two refracted beams, resulting in an increasing Doppler broadening.

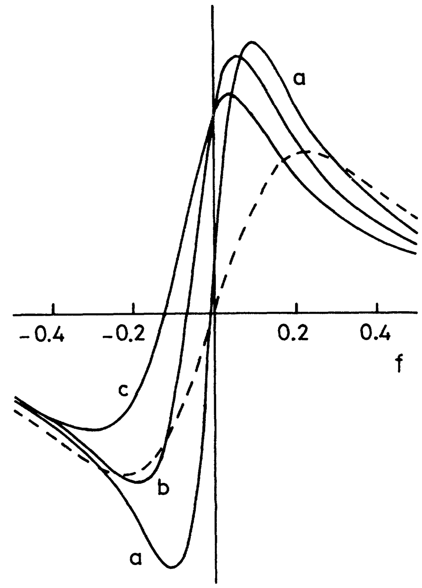

FIG. 3. Plots of $\text{Re}(\chi_{s1})$ in arbitrary units as a function of the reduced frequencies $f_s = f_p = f$, for various incidence angles just below the critical angle. The signal and the pump have opposite incidence angles. The solid curves correspond to $\beta = 0.01$ for curve a, $\beta = 0.1$ for curve b, and $\beta = 0.2$ for curve c, with $\beta_s = \beta_p = \beta$ the cosines of the refraction angles, as defined by (5.4). The dotted curve represents $\text{Re}(\bar{\chi}_{s1})$, corresponding to neglected spatial dispersion, in the case $\beta = 0.2$.

In Fig. 4 we demonstrate the pump effect on the signal reflectivity when the two identical incidence angles, $\theta = \theta_p = -\theta_s$, are slightly larger than θ_c. The plots display $\text{Im}(\chi_{s1})$, which according to Eq. (5.7) contributes to the reflectivity in the regime of total internal reflection. The negative values of $\text{Im}(\chi_{s1})$ indicate a decrease of the signal absorption as a result of the pump field. We display the curves for various values of $\eta_s = \eta_p = \eta$, defined in Eq. (5.6). Since $\eta_s k_s = \kappa_s$, an increase in η corresponds to evanescent waves of shorter range. Increasing η values give rise to broader curves, which may be understood as an increasing imaginary Doppler shift, or equivalently, an increasing transient broadening. Moreover, the values of $\text{Im}(\chi_{s1})$ are seen to diminish for increasing η.

The variation in the width and the shift of the dispersion curves of Fig. 3 and the width of the absorption curves of Fig. 4 are illustrated in Fig. 5. One sees that the width of the dispersion curves with spatial dispersion

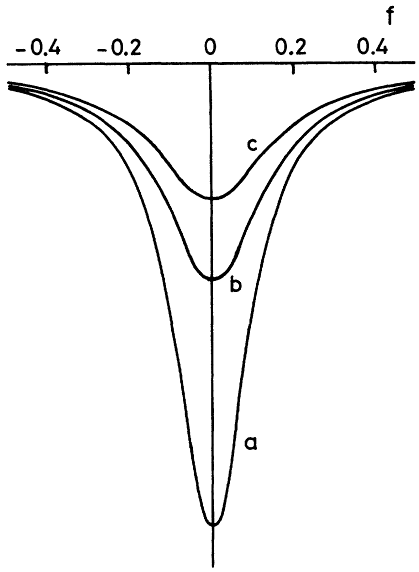

FIG. 4. Plots of $\mathrm{Im}(\chi_{s1})$ in arbitrary units as a function of the reduced frequencies $f_s=f_p=f$, for incidence angle just beyond the critical angle. These curves represent the pump modification of the signal reflectivity for opposite incidence angles of the two beams. The curves correspond to $\eta=0.01$ for curve a, $\eta=0.1$ for curve b, and $\eta=0.2$ for curve c, with $\eta_s=\eta_p=\eta$ defined as in (5.6).

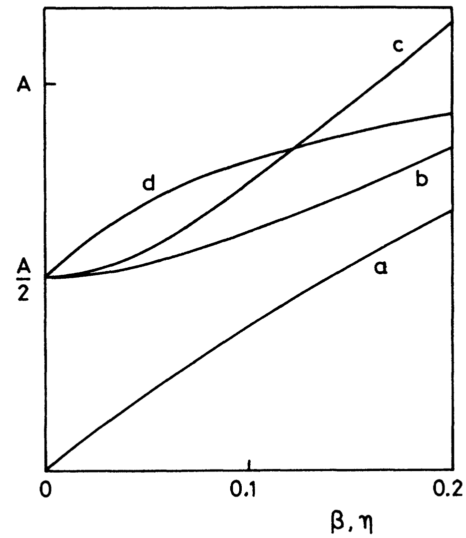

FIG. 5. Curve a represents the variation of the negative average shift of the dispersion curves of Fig. 3 with β. Curve b gives the width of these curves as measured by half of the frequency difference of the maximum and the minimum, and curve c gives the corresponding width calculated while neglecting spatial dispersion. Curve d finally gives the half width at half maximum of the absorption curves of Fig. 4 as a function of η.

is considerably larger than the width determined by the local susceptibility.

VIII. CONCLUSIONS

In this paper we have studied the nonlinear effects of a weak signal field with frequency ω_s and a pump field with frequency ω_p on reflection at the interface of a dielectric and an atomic vapor. The method consists of solving the nonlinear Bloch equations for the velocity-dependent atomic density matrix, while accounting for the transient behavior of the atoms leaving the interface. This leads to explicit expressions for the various frequency components of the dipole polarization, which allows us to evaluate the reflected intensities.

One component is the nonlinear emission at the frequency $2\omega_p-\omega_s$. This emission results from a four-wave mixing process, and it has recently been observed experimentally.[15] It corresponds to the terms G_{eg} and G'_{eg} in Eqs. (3.19) and (3.20) for the dipole polarization. The Laplace transforms of these quantities, which are given in (3.12) and (3.17), determine the electric-field amplitude of this emission, as indicated in (4.7), and discussed in Sec. IV C.

Furthermore, we demonstrate that the reflectivity of the signal field as modified by the pump field can be expressed in terms of an effective susceptibility χ_s, as indicated by Eq. (5.5) for a signal field refracted into the vapor, and by Eq. (5.7) when the incidence angle θ_s of the signal field is larger than the critical angle θ_c for total internal reflection. In the transition region around θ_c, Eq. (5.10) applies. The effective susceptibility χ_s is determined by Eq. (4.11) or (4.13), with f_{eg} and f'_{eg} given by (3.10) and (3.16).

ACKNOWLEDGMENTS

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