PHYSICAL REVIEW A VOLUME 38, NUMBER 10 NOVEMBER 15, 1988

Nonlinear selective reflection from an atomic vapor at arbitrary incidence angle

G. Nienhuis
Fysisch Laboratorium, Rijksuniversiteit Utrecht, Postbus 80 000, 3508 TA Utrecht, The Netherlands
and Huygens Laboratorium, Rijksuniversiteit Leiden, Postbus 9504, 2300 RA Leiden, The Netherlands

F. Schuller and M. Ducloy
Laboratoire de Physique des Lasers, Université Paris-Nord, 93430 Villetaneuse, France
(Received 11 February 1988; revised manuscript received 6 June 1988)

We calculate the reflection coefficient for a light beam incident on the interface between a dielectric and a resonant atomic vapor to first order in the dipole polarization in the vapor. The angle of incidence and the polarization direction are chosen arbitrarily, and saturation is fully accounted for. The atoms are supposed to get deexcited at collisions with the surface. The resulting transient behavior of atoms leaving the surface is responsible for a nonlocal response. This spatial dispersion near the surface is known to produce a natural-linewidth-limited resonance in the vapor reflection coefficient at normal incidence. Our analysis shows that this sub-Doppler structure is broadened by the residual Doppler effect for non-normal incidence. This structure disappears at the critical angle for total internal reflection, where one predicts a conventional Voigt-type dispersion response, based on the complex-refractive-index approach. We also calculate the saturation broadening of the atomic response for large intensities. Beyond the critical angle, the spectral response suddenly shifts from dispersion to absorption line shapes. In the case of total internal reflection, the spatial dispersion leads to an additional Lorentzian broadening, which results from an effective imaginary Doppler shift at passage through the evanescent wave (transit-time broadening).

I. INTRODUCTION

Selective reflection of light from the interface between a dielectric and an atomic vapor is a spectroscopic tool that differs in various respects from ordinary absorption spectroscopy. First, the reflection is mainly sensitive to a thin layer near the interface, so that one may expect to observe effects of collisions of the vapor atoms with the wall. At normal incidence, wall collisions can give rise to sub-Doppler structure in the reflection coefficient.[1-3] This structure is due to the transient behavior of atoms leaving the wall just after a collision.[4] Even though the field in the gas may be considered a plane wave, the atomic polarization is not uniform in the boundary region, which reflects the nonlocal relation between the applied field and the polarization. This spatial dispersion results from the combination of the atomic motion and the symmetry-breaking effect of the wall.

Another possibility of selective reflection is offered in the case of total internal reflection, where the evanescent wave penetrating into the vapor by a few wavelengths gives rise to absorption, and thereby to a decrease of the reflection. This situation has the additional advantage that the absorption takes place exclusively in a narrow boundary region.[5] More recently, saturated reflection spectroscopy has been applied using a pump-probe scheme, both in near-normal incidence,[6] and in total reflection.[7] Saturation effects in single-beam selective reflection have been studied theoretically for normal incidence.[8,9] The sub-Doppler structure displays saturation broadening,[8] and the high-intensity corrections have a longer transient time than the first-order term.[9]

A complementary method to study the difference in radiative behavior of atoms approaching and leaving the surface is the observation of the fluorescence excitation spectrum. By selecting the fluorescence emitted into the dielectric under an angle beyond the critical angle for total reflection, one effectively selects the emitted evanescent waves from the atoms near the surface. This method has been adopted by Burgmans *et al.*[10] A sharp decrease of the fluorescence intensity for an excitation frequency in the blue Doppler wings then indicates that atoms leaving the surface contribute less to the fluorescence than atoms in the steady state.

In this paper we present a general theoretical description of selective reflection, for arbitrary angle of incidence, arbitrary intensity, and for various directions of polarization. For clarity of the presentation, we separately discuss the evaluation of the polarization of the vapor, and the reflected fields as a function of this polarization. We distinguish the case of a polarization direction perpendicular to or in the plane of incidence.

II. REFLECTION FOR GIVEN DIPOLE POLARIZATION

We consider the interface between a dielectric with a real refractive index n, and an atomic vapor, which may be embedded in a buffer gas of nonactive molecules. The interface is taken as the xy plane, and the dielectric fills the half space with $z<0$. A plane wave with wave vector $\mathbf{k}_0$ is incident on the interface, and it is reflected back into the gas, as sketched in Fig. 1. The refracted field in the gas drives the active atoms, thereby creating a dipole po-

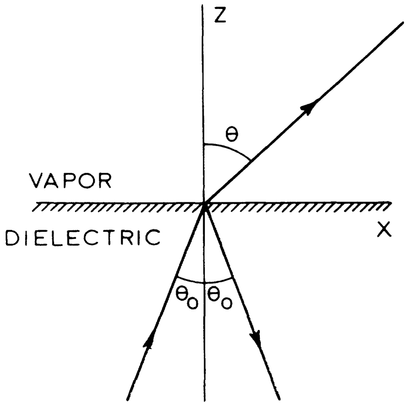

FIG. 1. Geometry of the incident, the refracted, and the reflected wave. The xy plane is the plane of the surface and the wave vectors fall in the xz plane.

larization $\mathbf{P}(\mathbf{r})$. This polarization emits radiation in the direction of the reflected wave, thereby modifying the reflection coefficient. The field and the polarization are supposed to be monochromatic with frequency ω, and we consider the positive-frequency part $\mathbf{E}(\mathbf{r})$ and $\mathbf{P}(\mathbf{r})$, such that

$$\mathbf{E}(\mathbf{r},t)=2\,\mathrm{Re}\mathbf{E}(\mathbf{r})e^{-i\omega t}\,,\quad \mathbf{P}(\mathbf{r},t)=2\,\mathrm{Re}\mathbf{P}(\mathbf{r})e^{-i\omega t}\,. \tag{2.1}$$

The incident wave has the wave vector

$$k_0=k_0(a\hat{\mathbf{x}}+b\hat{\mathbf{z}})\,, \tag{2.2}$$

with $k_0=nk=n\omega/c$, and

$$a=\sin\theta_0,\quad b=\cos\theta_0 \tag{2.3}$$

with θ_0 the angle of incidence. In the dielectric, the electric field is

$$\mathbf{E}(\mathbf{r},t)=[\mathbf{E}_0\exp(ibkz)+\mathbf{E}_2\exp(-ibkz)]\exp(iakx)\,, \tag{2.4}$$

with $\mathbf{E}_0$ the amplitude of the incident field, and $\mathbf{E}_2$ the amplitude of the reflected field. Then the variation of the fields and the induced polarization with the coordinates x and y must be given by $\exp(iak_0x)$. For convenience, we introduce the parameter

$$\alpha=an\,, \tag{2.5}$$

which equals $\sin\theta$ with θ the refraction angle into vacuum. The dipole polarization is written as

$$\mathbf{P}(\mathbf{r})=\mathbf{P}_0(z)\exp(i\alpha kx),\quad z>0\,. \tag{2.6}$$

The purpose of this section is to give expressions for the contribution to the reflected field due to an arbitrary dipole polarization of the form (2.6). We start from the wave equation for the vector potential $\mathbf{A}$ in the Lorentz gauge, in the absence of free charges and currents

$$(\nabla^2+k^2)\mathbf{A}=i\omega\mu_0\mathbf{P}\,. \tag{2.7}$$

We have used the fact that the vector potential is monochromatic, and that the time derivative of $\mathbf{P}$ is an effective current density. We introduce a Green's function

$$\phi(r)=\frac{1}{2i\beta k}\exp(i\beta k\,|z-z_0|+i\alpha kx)\,, \tag{2.8}$$

which is the solution of the wave equation

$$(\nabla^2+k^2)\phi=\delta(z-z_0)\exp(i\alpha kx)\,, \tag{2.9}$$

with β determined by

$$\alpha^2+\beta^2=1\,. \tag{2.10}$$

When $\alpha>1$, we shall take

$$\beta=i\eta \tag{2.11}$$

imaginary, with $\eta>0$. It is now an easy matter to evaluate the electric field due to the polarization (2.6) that would exist in the region with $z<0$ in the absence of the dielectric. Each layer parallel to the xy plane contributes a plane wave with the wave vector

$$\mathbf{K}=k(\alpha\hat{\mathbf{x}}-\beta\hat{\mathbf{z}}) \tag{2.12}$$

for $z<0<z_0$. The vector potential that solves (2.7) is a simple superposition of solutions (2.8), and we find

$$\mathbf{A}(\mathbf{r})=\frac{\omega\mu_0}{2\beta k}\exp(i\mathbf{K}\cdot\mathbf{r})\int_0^\infty dz_0\mathbf{P}_0(z_0)\exp(i\beta kz_0)\,. \tag{2.13}$$

According to Maxwell's equations, we have the equality

$$\mathbf{E}=\frac{-1}{i\omega\epsilon_0\mu_0}\nabla\times\nabla\times\mathbf{A} \tag{2.14}$$

in the region of vanishing polarization. Hence we obtain

$$\mathbf{E}(\mathbf{r})=\frac{1}{2\beta\epsilon_0 k}\exp(i\mathbf{K}\cdot\mathbf{r})\int_0^\infty dz_0(k^2-\mathbf{K}\mathbf{K}\cdot)\mathbf{P}_0(z_0)\times\exp(i\beta kz_0)\,. \tag{2.15}$$

In the presence of the dielectric, (2.15) gives the plane wave emitted by the dipole polarization, and incident on the surface. A part of this field is reflected back into the region with $z>0$, and a part is refracted into the dielectric, as described by the laws of Fresnel and Snellius. It is this refracted part that gives the contribution of the dipole polarization to the reflection coefficient. We shall now consider separately the cases where the incident field is normal to the plane of incidence, or in this plane.

A. Polarization normal to the plane of incidence

When the incident field $\mathbf{E}_0$ and the dipole polarization $\mathbf{P}_0$ are directed normal to the plane of incidence, the same is true for the reflected field $\mathbf{E}_2$, and we write

$$\mathbf{E}_0=\hat{\mathbf{y}}E_0,\quad \mathbf{P}_0(z)=\hat{\mathbf{y}}P_0(z),\quad \mathbf{E}_2=\hat{\mathbf{y}}E_2\,. \tag{2.16}$$

The reflected field amplitude E_2 consists of two parts, and we write

$$E_2=E_{2v}+E_{2p}\,. \tag{2.17}$$

The first part, E_{2v}, is the amplitude in the absence of the dipole polarization, and it corresponds to the internal reflection when the half space $z>0$ is vacuum. According to Fresnel's laws, this contribution is

$$E_{2v}=\frac{nb-\beta}{nb+\beta}E_0 \; . \tag{2.18}$$

The contribution E_{2p} due to the dipole polarization is likewise calculated from Fresnel's laws in the case that the field (2.15) incident on the surface is refracted into the dielectric. In the present case of polarization in the y direction this gives

$$E_{2p}=\frac{S}{nb+\beta} \; , \tag{2.19}$$

with

$$S=\frac{ik}{\epsilon_0}\int_0^\infty dz_0 \exp(i\beta k z_0)P_0(z_0) \; . \tag{2.20}$$

B. Polarization in the plane of incidence

Now we consider the situation that the incident field and the dipole polarization are directed in the plane of incidence. Then the same must hold true for the reflected field $\mathbf{E}_2$, and we write

$$\mathbf{E}_0=E_0(-b\hat{\mathbf{x}}+a\hat{\mathbf{z}}), \quad \mathbf{E}_2=E_2(-b\hat{\mathbf{x}}-a\hat{\mathbf{z}}) \; . \tag{2.21}$$

To be specific, we assume that the polarization $\mathbf{P}$ is parallel to the electric field that would exist in the half space with $z>0$ in the absence of the vapor, and we write

$$\mathbf{P}_0(z)=P_0(z)(-\beta\hat{\mathbf{x}}+\alpha\hat{\mathbf{z}}), \quad z>0 \; . \tag{2.22}$$

The reflected amplitude E_2 consists again of two parts as in (2.17). The vacuum contribution is now

$$E_{2v}=\frac{n\beta-b}{n\beta+b}E_0 \; , \tag{2.23}$$

according to Fresnel's laws. The contribution E_{2p} due to the polarization is obtained in two steps. First we substitute (2.22) into (2.15) in order to obtain the field emitted by the polarization. Then we treat this field as the incident field on the surface of the dielectric, and we evaluate the refraction into the dielectric, which is the contribution E_{2p}. We obtain

$$E_{2p}=\frac{\beta^2-\alpha^2}{n\beta+b}S \; . \tag{2.24}$$

This result vanishes when $\alpha=\beta=1/\sqrt{2}$. This is understandable, since then the propagation direction of the emitted field is parallel to the direction of the dipole polarization, and dipole emission is zero along the dipole direction.

The results of this section remain valid when $\alpha=an$ is larger than 1. In this domain of total internal reflection, β is imaginary, as in (2.11). Note, however, that in this case P_0 is not the absolute value of $\mathbf{P}_0$. Equations (2.19) and (2.24) are the main results of this section. They give the emission into the dielectric resulting from a given dipole polarization as in (2.16) or (2.22), and thereby the contribution to the reflection coefficient due to the polarization of the vapor.

III. REFLECTION COEFFICIENT FOR ARBITRARY INTENSITY

We wish to evaluate the polarization of the gas while assuming that the atoms move in the external field $\mathbf{E}_1(\mathbf{r})$ that would exist in the half plane with $z>0$ in a vacuum. This means that we neglect the back reaction of the gas polarization on the field that the atoms experience. This common assumption implies that the reflection is evaluated to first order in the gas density, which is justified as long as the refractive index of the gas deviates negligibly from unity. From the Fresnel conditions one directly obtains the well-known expression for the refracted field for $z>0$

$$\mathbf{E}(\mathbf{r})=\boldsymbol{\epsilon}_1 E_1 \exp(i\alpha kx+i\beta kz) \; . \tag{3.1}$$

For a normal polarization direction $\boldsymbol{\epsilon}_1=\hat{\mathbf{y}}$, the field amplitude is

$$E_1=\frac{2nb}{nb+\beta}E_0 \; . \tag{3.2}$$

For a polarization in the plane of incidence, the refracted field has the polarization direction

$$\boldsymbol{\epsilon}_1=-\beta\hat{\mathbf{x}}+\alpha\hat{\mathbf{z}} \; , \tag{3.3}$$

and the field amplitude is

$$E_1=\frac{2nb}{n\beta+b}E_0 \; . \tag{3.4}$$

A. Refraction

First we consider the case of refraction, where $\alpha<1$, so that the field in the gas is a plane wave. We describe the gas as a system of quantum-mechanical two-state atoms, with ground state $|g\rangle$ and excited state $|e\rangle$. The dipole polarization $\mathbf{P}(\mathbf{r})$ is determined by the expectation value of the dipole moment of the atoms at position $\mathbf{r}$, averaged over the velocity distribution. This dipole moment is determined by the optical Bloch equations, with appropriate initial conditions. It is reasonable to assume that atoms get deexcited at a collision with the surface. Therefore we assume that the atoms are in the ground state $|g\rangle$ at the instant that they leave the surface after a collision. This implies that the average dipole moment is zero at this instant. However, this assumption not only implies a loss of phase memory of the dipole moment, but also the population of the excited state is put equal to zero. The velocity distribution of the atoms leaving the surface is taken to be Maxwellian.

In order to evaluate the dipole polarization $\mathbf{P}$, it is convenient to introduce the quantity $\mathbf{p}(\mathbf{r},\mathbf{v},t)$ as the positive-frequency part of the dipole moment of a two-state atom at position $\mathbf{r}$ and velocity $\mathbf{v}$ that has entered the plane-wave field in its ground state a time t ago. This quantity obeys the relation

$$\mathbf{p}(\mathbf{r},\mathbf{v},t)=\boldsymbol{\mu}_{ge}\exp[i(\alpha kx+\beta kz-\phi)]\sigma_+(\mathbf{v},t) \; , \tag{3.5}$$

with σ_+ the off-diagonal (raising) part of the normalized density matrix that solves the optical Bloch equations

$$\frac{\partial}{\partial t}\sigma_{ee} = -A\sigma_{ee} + \frac{i}{2}\Omega(\sigma_- - \sigma_+) = -\frac{\partial}{\partial t}\sigma_{gg} \,,$$
$$\frac{\partial}{\partial t}\sigma_+ = -[\gamma - i(\Delta - \mathbf{k}\cdot\mathbf{v})]\sigma_+ + \frac{i}{2}\Omega(\sigma_{gg} - \sigma_{ee}) \,,$$
$$\frac{\partial}{\partial t}\sigma_- = -[\gamma + i(\Delta - \mathbf{k}\cdot\mathbf{v})]\sigma_- + \frac{i}{2}\Omega(\sigma_{ee} - \sigma_{gg}) \,. \tag{3.6}$$

The phase ϕ arises from the atom-field coupling

$$\boldsymbol{\mu}_{eg}\cdot\boldsymbol{\epsilon}_1 E_1 = \tfrac{1}{2}\hbar\Omega e^{-i\phi} \,, \tag{3.7}$$

with $\boldsymbol{\mu}_{eg}$ a dipole matrix element, and Ω the real Rabi frequency. A well-known closed expression for σ_+ is easily found from the Bloch equations for the Laplace transform[11]

$$\hat{\sigma}_+(\mathbf{v},s) = \int_0^\infty dt\, e^{-st}\sigma_+(\mathbf{v},t) \,. \tag{3.8}$$

The result is

$$\hat{\sigma}_+(\mathbf{v},s) = \frac{i\Omega(A+s)}{2s}\,\frac{\gamma + s + i(\Delta - \mathbf{k}\cdot\mathbf{v})}{(A+s)[(\gamma+s)^2 + (\Delta - \mathbf{k}\cdot\mathbf{v})^2] + \Omega^2(\gamma+s)} \,, \tag{3.9}$$

where Δ is the detuning of the light frequency from resonance, A is the spontaneous decay rate, and γ is the homogeneous (collisional and radiative) linewidth. The velocity $\mathbf{v}$ enters only through the Doppler shift $\mathbf{k}\cdot\mathbf{v}$.

An atom with a positive value of v_z at the position $\mathbf{r}$ has left the surface a time z/v_z ago in its ground state so that the positive-frequency part of the dipole moment is $\mathbf{p}(\mathbf{r},\mathbf{v},z/v_z)$. On the other hand, when v_z is negative, the atom has traveled many lifetimes in the field, so that it will have reached its steady state, and $\mathbf{p}$ has the value

$$\bar{\mathbf{p}}(\mathbf{r},\mathbf{v}) = \boldsymbol{\mu}_{ge}\exp[i(\alpha kx + \beta kz - \phi)]\bar{\sigma}_+(\mathbf{v}) \,, \tag{3.10}$$

with

$$\bar{\sigma}_+(\mathbf{v}) = \lim_{s\to 0} s\hat{\sigma}_+(\mathbf{v},s) = \frac{\frac{1}{2}i\Omega A[\gamma + i(\Delta - \mathbf{k}\cdot\mathbf{v})]}{A[\gamma^2 + (\Delta - \mathbf{k}\cdot\mathbf{v})^2] + \gamma\Omega^2} \tag{3.11}$$

the steady-state value of σ_+.

Now that we have specified $\mathbf{p}$ both for negative and positive values of v_z, we also have determined the dipole polarization

$$\mathbf{P}(\mathbf{r}) = N\int d\mathbf{v}\, W(\mathbf{v})[\Theta(v_z)\mathbf{p}(\mathbf{r},\mathbf{v},z/v_z) + \Theta(-v_z)\bar{\mathbf{p}}(\mathbf{r},\mathbf{v})] \,, \tag{3.12}$$

with W the normalized Maxwell distribution, Θ the Heaviside step function, and N the density of active atoms. The scalar polarization $P_0(z)$ is then easily found to obey the relation

$$P_0(z)/\epsilon_0 E_1 = \frac{\hbar\Omega N}{I_1}\int d\mathbf{v}\, W(v)e^{i\beta kz}[\Theta(v_z)\sigma_+(\mathbf{v},z/v_z) + \Theta(-v_z)\bar{\sigma}_+(\mathbf{v})] \,, \tag{3.13}$$

with

$$I_1 = 2\epsilon_0 |\epsilon_1 E_1|^2 \,, \tag{3.14}$$

the energy density of the radiation field in the vapor. If we substitute the expression (3.13) into (2.20), the integration over z leads to an expression for S in terms of the Laplace transform of σ_+. For later convenience, we give the expression for the dimensionless quantity $T = S/E_1$, and we obtain

$$T = -\frac{N\hbar\Omega}{2\beta I_1}\int d\mathbf{v}\, W(\mathbf{v})[\Theta(-v_z)\bar{\sigma}_+(\mathbf{v}) + \Theta(v_z)(-2i\beta kv_z) \times \hat{\sigma}_+(\mathbf{v}, -2i\beta kv_z)] \,. \tag{3.15}$$

This equation (3.15) gives an explicit expression for the quantity T (or S), where $\hat{\sigma}_+$ and $\bar{\sigma}_+$ are given in (3.9) and (3.11). The Maxwellian average is the only remaining integration. When the atoms move by much less than a wavelength in a radiative lifetime, the quantity $-2i\beta kv_z$ may be set equal to zero, and the integrand in (3.15) is simply $W(\mathbf{v})\bar{\sigma}_+(\mathbf{v})$. This corresponds to a saturated analogue of Voigt's theory of the complex refractive index.[12] Then T reduces to the local-response contribution

$$T = \frac{N\hbar\Omega}{2\beta I_1}\int d\mathbf{v}\, W(\mathbf{v})\bar{\sigma}_+(\mathbf{v}) = -\delta n/\beta \,, \tag{3.16}$$

with δn the resonant refractive index variation of the vapor. It is the deviation of $-2i\beta kv_z\hat{\sigma}_+(-2i\beta kv_z)$ from the stationary limit $\bar{\sigma}_+$ that reflects the transient behavior of atoms in the boundary region leaving the wall. This effect arises since, due to the atomic motion, the polarization (3.13) is not just determined by the local field. The relative effect of the spatial dispersion decreases for decreasing β, which means that the refraction angle θ approaches $\pi/2$, or that the angle of incidence approaches the critical angle. The effect of spatial dispersion decreases also at high intensities when Ω gets larger than the homogeneous linewidth, since then the approach to the steady state takes place more rapidly.

One should note that very close to the critical angle, where

$$\beta = \cos\theta \ll \gamma/kv_0 \,, \tag{3.17}$$

the effect of the transient behavior becomes negligible, since the optical phase does not vary appreciably over the distance that an atom travels during a lifetime. At the same time, this fact gives us a clue as to how to handle the divergence of the atomic response at the critical angle, where $\beta = 0$. This divergence, that shows up in (3.15) [and in (3.16)], is an artifact resulting from the expansion to first order in the atomic dipole polarization. This expansion is justified when

$$\beta \gg |\delta n| \ .$$

Since the effect of spatial dispersion is negligible, we can simply apply the local complex index theory in the neighborhood of the critical angle.

The quantity (3.15) directly determines the amplitude refraction coefficient

$$r = E_2/E_0 \ . \tag{3.18}$$

By using the relations (3.2) and (3.4) between the refracted field E_1 and the incident field E_0, we find from (2.18) and (2.19) for a polarization normal to the plane of incidence

$$r_\perp = \frac{nb - \beta}{nb + \beta} + \frac{2nb}{(nb+\beta)^2} T \ . \tag{3.19}$$

The intensity reflection coefficient

$$R = |r|^2 \tag{3.20}$$

is then to first order in the atom density N

$$R_\perp = \left[\frac{nb - \beta}{nb + \beta} \right]^2 + \frac{4nb(nb-\beta)}{(nb+\beta)^3} \mathrm{Re} T \ . \tag{3.21}$$

For a polarization parallel to the plane of incidence, we find from (2.23) and (2.24)

$$r_\parallel = \frac{n\beta - b}{n\beta + b} + (\beta^2 - \alpha^2) \frac{2nb}{(n\beta+b)^2} T \ , \tag{3.22}$$

which leads us to the expression

$$R_\parallel = \left[\frac{n\beta - b}{n\beta + b} \right]^2 + (\beta^2 - \alpha^2) \frac{4nb(n\beta - b)}{(n\beta+b)^3} \mathrm{Re} T \tag{3.23}$$

for the intensity reflection coefficient.

Equations (3.19) and (3.22), together with (3.15), give the reflection coefficient, for arbitrary intensity and angle of incidence and for the two selected polarization directions. This result generalizes the results of Refs. 4, 8, and 9, where only normal incidence was considered. We notice that the reflection coefficient $R_\parallel$ will show a dispersion curve that changes sign in the case of in-plane polarization when the refraction angle passes 45°, and also when $n\beta = b$, or

$$a = (n^2+1)^{-1/2} = \beta \ . \tag{3.24}$$

This latter angle corresponds to the Brewster angle, where the in-plane polarized field has a vanishing reflection at the dielectric-vacuum interface. Furthermore, the sub-Doppler structure that is due to the discontinuity of the integrand in (3.17) at $v_z = 0$ will be broadened by the width $\alpha k v_0$ for non-normal incidence.

Before giving a numerical result for T, we first extract an overall dimensionless factor by writing

$$\mathrm{Re} T = \frac{N\hbar\Omega^2}{2\pi I_1 k v_0} \Phi \ . \tag{3.25}$$

By employing standard expressions for the spontaneous decay rate A in terms of the dipole matrix element μ_{eg}, it may be shown that the factor multiplying Φ in (3.25) is equal to $NA/(k^4 v_0)$. This determines the number of active atoms in a cubic wavelength, multiplied by the ratio of the natural width and the Doppler width.

In Fig. 2 we display the function Φ determining the saturated reflection in the special case of normal incidences as a function of a reduced detuning δ, and for two values of the Rabi frequency Ω. The sub-Doppler structure at resonance, which is present at low intensities, is obscured by saturation when the Rabi frequency becomes comparable to the Doppler width. On the basis of their calculation of nonlinear effects on the selective reflection to third order in the Rabi frequency, Singh and Agarwal[9] predicted a hole-burning effect near resonance. However, no trace of this effect is found in the present calculation to all orders.

B. Total reflection

When $\alpha = an$ is larger than 1, total internal reflection occurs, and the field (3.1) decays exponentially as a function of z. The general result (3.12) for the polarization of the vapor remains valid, but now the explicit evaluation of $\mathbf{p}$ requires solving the Bloch equations with a Rabi frequency that decays exponentially, according to the equality

$$\mu_{eg} \cdot \epsilon_1 E_1 e^{-\eta k z} = \tfrac{1}{2}\hbar\Omega(z) e^{-i\phi} \ . \tag{3.26}$$

The variation of $\mathbf{p}$ with x is proportional to $\exp(i\alpha k x)$, but the variation with z cannot be given in a simple analytical form, unless saturation effects are neglected. The expression for the quantity $T = S/E$ now takes the form

$$T = \frac{ik\hbar\Omega N}{I_1} \int_0^\infty dz\, e^{-\eta k z} \times \int d\mathbf{v}\, W(\mathbf{v}) [\Theta(v_z)\sigma_+(z,\mathbf{v},z/v_z) + \Theta(-v_z)\bar{\sigma}(z,\mathbf{v})] \ . \tag{3.27}$$

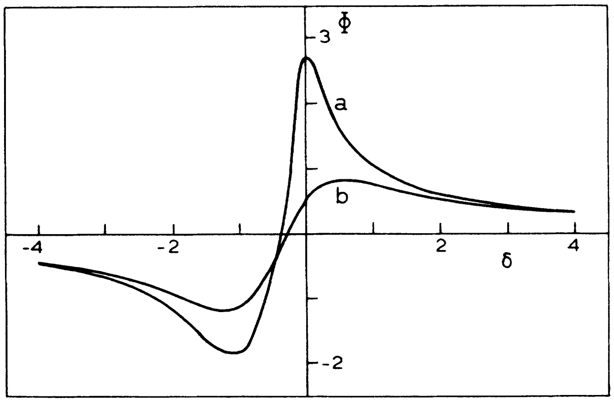

FIG. 2. Plot of the function Φ, for normal incidence and with saturation, as a function of the reduced detuning $\delta = \Delta/kv_0$. The selected values for the linewidths are $\gamma = kv_0/10$, $A = kv_0/5$. The Rabi frequency is a, $\Omega = kv_0/5$; b, $\Omega = kv_0$.

The matrix element $\sigma_+(z,\mathbf{v},z/v_z)$ for $v_z>0$ pertains to the density matrix solving the Bloch equations with a Rabi frequency proportional to $\exp(-\eta k z)$ and gives the dipole moment of an atom at the distance z from the surface that left the surface a time z/v_z ago in the ground state. Likewise, the element $\bar{\sigma}_+(z,\mathbf{v})$ for $v_z<0$ refers to an atom at the distance z from the surface that was in the ground state at an infinitely large distance. Only the x component v_x contributes to the Doppler shift, and the Bloch equations for an atom traversing the exponentially decaying field are

$$\begin{aligned}
&\frac{\partial}{\partial t}\sigma_{ee}=-A\sigma_{ee}+\frac{i}{2}\Omega(z)(\sigma_- -\sigma_+)=-\frac{\partial}{\partial t}\sigma_{gg}\ ,\\
&\frac{\partial}{\partial t}\sigma_+=-[\gamma-i(\Delta-\alpha k v_x)]\sigma_+ +\frac{i}{2}\Omega(z)(\sigma_{gg}-\sigma_{ee})\ ,\\
&\frac{\partial}{\partial t}\sigma_-=-[\gamma+i(\Delta-\alpha k v_x)]\sigma_- +\frac{i}{2}\Omega(z)(\sigma_{ee}-\sigma_{gg})\ ,
\end{aligned} \tag{3.28}$$

with $z(t)=z(0)+v_z t$.

An explicit expression for T will be given for the case with negligible saturation in Sec. IV. Note that (3.2)–(3.4) as well as the results (3.19) and (3.22) remain valid for total reflection, where $\beta=i\eta$ is imaginary. For a dipole polarization in the y direction, the intensity reflection coefficient is

$$R_\perp=1+\frac{4nb}{(nb)^2+\eta^2}\mathrm{Re}T\ . \tag{3.29}$$

For a polarization parallel to the plane of incidence we find

$$R_\parallel=1+(\alpha^2+\eta^2)\frac{4nb}{(n\eta)^2+b^2}\mathrm{Re}T\ . \tag{3.30}$$

The second terms on the right-hand sides of (3.29) and (3.30) are always negative, and absorption of the evanescent wave makes the reflection coefficient R less than unity. This decrease of the reflection is larger for polarization parallel to the incidence plane than for polarization in the y direction.

The integrated dipole polarization, as expressed in the quantity T, Eq. (3.27), modifies the amplitude E_2 of the reflected wave, even in the domain of total internal reflection. The loss of power at reflection must correspond to the absorption of the evanescent wave, or, equivalently, to the fluorescent emission in the vapor near the boundary. The energy balance can be checked explicitly by evaluating the lost power per unit area

$$Q=2\epsilon_0 ncb(|E_0|^2-|E_2|^2)\ . \tag{3.31}$$

In the case of a polarization direction normal to the plane of incidence, we substitute (2.17)–(2.19), while using (3.2), with the replacement $\beta=i\eta$. For a polarization direction in the plane of incidence, (2.23) and (2.24) apply. To first order in the vapor density, i.e., to first order in T, we obtain in both cases

$$Q=-cI_1\mathrm{Re}T\ . \tag{3.32}$$

This is equal to the power that the evanescent field exerts on the gas per unit area of the surface

$$Q=\int_0^\infty dz\,\mathbf{E}(\mathbf{r},t)\frac{\partial}{\partial t}\mathbf{P}(\mathbf{r},t) \tag{3.33}$$

as follows from the definitions (2.1), (3.1), and (2.20), with $T=S/E_1$.

IV. WEAK-FIELD LIMIT AND POLARIZATION SPECTROSCOPY

We have demonstrated that for an incident field with a polarization in one of the principal directions, the intensity reflection coefficient R is fully determined by the real part of $T=S/E_1$. This is obvious from Eqs. (3.21), (3.23), and (3.29), (3.30). In this section we demonstrate that also the imaginary part of T is accessible to observation, provided that only the reflected radiation with a selected polarization direction is observed. In order to avoid unnecessary complications, we study the polarization properties of the reflected field in the absence of saturation.

A. Transmission case

In the situation when $\alpha=an<1$, the reflection coefficient r is given by (3.19) and (3.22) for the two polarization directions. The quantity T is specified by (3.27), with (3.19) and (3.11) for the matrix elements. To lowest order in Ω, we can neglect the terms proportional to Ω^2 in the denominators of (3.9) and (3.11). If we substitute these low-intensity limits into (3.15), we find that the contribution to the integral due to the atom with $v_z>0$ is equal to the integral for $v_z<0$. For the case of normal incidence, this remarkable feature was obtained previously.[13] The explicit result takes the form

$$\begin{aligned}
T=&\frac{-iN}{\beta\epsilon_0\hbar}|\boldsymbol{\mu}_{eg}\cdot\boldsymbol{\epsilon}_1|^2\\
&\times\int d\mathbf{v}\,W(\mathbf{v})\Theta(v_z)[\gamma-i(\Delta-\alpha k v_x+\beta k v_z)]^{-1}\ .
\end{aligned} \tag{4.1}$$

If we substitute (4.1) into (3.21) and (3.23), we recover the well-known result that the intensity reflection coefficient R is modified near resonance by a dispersion curve that is convoluted by half of the Maxwell distribution. For non-normal incidence, the distribution over v_x gives an additional broadening by a full Doppler profile with width $\alpha k v_0$. The validity condition for the result (4.1) without saturation is

$$\Omega^2\ll\gamma A\ . \tag{4.2}$$

The real part of T is determined by the factor Φ, as in (3.25). In Fig. 3 we plot this quantity Φ as a function of the reduced detuning and for various angles of incidence. The contribution of the vapor to the reflectivity increases with the angle of incidence. This is due to the fact that the volume where the reflection originates gets thicker when the normal component of the wave vector decreases. This increase of the vapor contribution was also observed in the experiment of Woerdman and Schuurmans.[2] We also plot the function Φ_0 pertaining to the

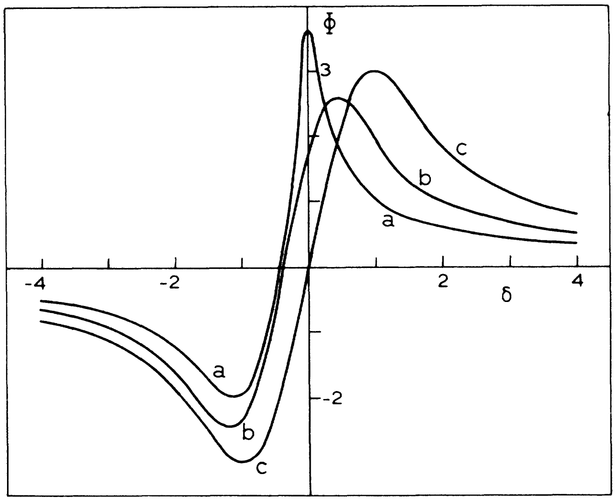

FIG. 3. Plot of the function Φ, for $\gamma = kv_0/10$, as a function of the reduced detuning δ, in the case of refraction with no saturation. *a*, normal incidence; *b*, refraction angle $\theta = 45°$; *c*, $2\times\Phi_0$, i.e., twice the function Φ in the absence of spatial dispersion, for normal incidence.

case without spatial dispersion. The effect of spatial dispersion decreases at larger angles of incidence. The main reason is that the variation of the phase of the field with z is slower. This in turn slows down the variation of the integrand of (4.1) with v_z. Moreover, the sub-Doppler structure gets broadened by the Doppler shift resulting from the transverse velocity v_x.

It is interesting to notice that the function Φ_0, corresponding to neglecting spatial dispersion, is precisely equal to the antisymmetric part of Φ. The symmetric part may thus be identified as the result of spatial dispersion.

B. Total reflection

In the case of total reflection, the illuminated region has a width of the order of a wavelength, which a typical atom traverses in a time of the order of an inverse Doppler width $(kv_0)^{-1}$. Saturation effects are negligible when the condition

$$\Omega \ll kv_0 \tag{4.3}$$

is satisfied, provided that the incidence angle is not very close to the critical angle. Then we may evaluate the matrix elements in (3.27) to first order in $\Omega(0)$, with the result

$$\sigma_+(z,\mathbf{v},z/v_z) = \frac{i\Omega(0)}{2[\gamma-\eta kv_z - i(\Delta-\alpha kv_x)]} \times \left(e^{-\eta kz} - e^{-z[\gamma-i(\Delta-\alpha kv_x)]/v_z}\right) \tag{4.4}$$

for $v_z > 0$, and

$$\bar{\sigma}_+(z,\mathbf{v}) = [i\Omega(0)/2][\gamma-\eta kv_z - i(\Delta-\alpha kv_x)]^{-1}e^{-\eta kz} \tag{4.5}$$

for $v_z < 0$. If we substitute these expressions into (3.27), we find again that the atoms with $v_z > 0$ give exactly the same contribution as the atoms with $v_z < 0$. The final result is

$$T = -\frac{N}{\eta\epsilon_0\hbar}\frac{|\mu_{eg}\cdot\epsilon_1|^2}{|\epsilon_1|^2} \times \int d\mathbf{v}\, W(\mathbf{v})\Theta(v_z)[\gamma+\eta kv_z - i(\Delta-\alpha kv_x)]^{-1} . \tag{4.6}$$

This result is almost exactly the same as in (4.1) for the refraction case with the substitution $\beta = i\eta$. Note that the factor $|\epsilon_1|^2$ in the denominator of (4.6) is not unity for a polarization direction in the plane of incidence.

In Fig. 4 we display the quantity Φ as defined in (3.25) with T given in (4.6), for the case $\eta = \frac{3}{4}$. Also the corresponding result Φ_0 in the absence of spatial dispersion is displayed. One notices that the absorption dip in the reflectivity is broadened by spatial dispersion. This broadening has two causes. First, the spatial dispersion gives an additional Lorentzian broadening, which may be viewed as transit-time broadening in the evanescent wave. Second, the component of the evanescent wave vector along the surface is larger than in the propagation case, which enhances Doppler broadening.

C. Polarization-dependent reflection

We introduce the notation

$$u_{0\parallel} = -b\hat{\mathbf{x}} + a\hat{\mathbf{z}}, \quad u_{2\parallel} = -b\hat{\mathbf{x}} - a\hat{\mathbf{z}} \tag{4.7}$$

for the normalized polarization vectors parallel to the plane of incidence, for the incident wave and for the refracted wave. For an arbitrary polarization direction, the incident field can always be decomposed as

$$\mathbf{E}_0(\mathbf{r}) = (E_{0\perp}\hat{\mathbf{y}} + E_{0\parallel}\hat{\mathbf{u}}_{0\parallel})\exp[ik_0(ax+bz)] , \tag{4.8}$$

with possibly complex components $E_{0\perp}$ and $E_{0\parallel}$. For low

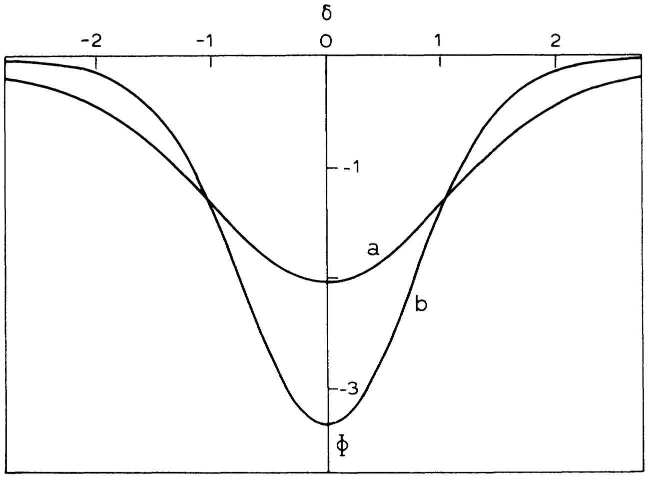

FIG. 4. Plot of the function Φ, for $\gamma = kv_0/10$, in the case of total reflection and without saturation for $\eta = \frac{3}{4}$ (and therefore $\alpha = \frac{5}{4}$). *a*, with spatial dispersion; *b*, spatial dispersion neglected.

intensities, the reflected field is found by multiplying the two components by the amplitude reflection coefficients r, and we obtain

$$\mathbf{E}_2(\mathbf{r})=(r_\perp E_{0\perp}\hat{\mathbf{y}}+r_\parallel E_{0\parallel}\hat{\mathbf{u}}_{2\parallel})\exp[ik_0(ax-bz)] \ . \qquad (4.9)$$

If the reflected field is passed through a polarization analyzer that transmits the polarization ϵ, the natural definition of the polarization-dependent reflection coefficient is

$$R(\epsilon)=|\epsilon^*\cdot\mathbf{E}_2|^2/|\mathbf{E}_0|^2 \ , \qquad (4.10)$$

with $\mathbf{E}_0$ and $\mathbf{E}_2$ given in (4.8) and (4.9). According to the expressions for $r_\perp$ and $r_\parallel$ derived in Sec. III, we may write

$$r_\perp=r_\perp^{(0)}(1+s_\perp T), \quad r_\parallel=r_\parallel^{(0)}(1+s_\parallel T) \qquad (4.11)$$

with $r_\perp^{(0)}$ and $r_\parallel^{(0)}$ the first terms on the right-hand sides of (3.19) and (3.22). The factors s are given by

$$s_\perp=\frac{2nb}{(nb)^2-\beta^2}, \quad s_\parallel=\frac{2nb}{b^2-(n\beta)^2}(\alpha^2-\beta^2) \ . \qquad (4.12)$$

In the case of total internal reflection, the value of β is imaginary. Note that the factors s are real both in the case of refraction and the case of total reflection. Since we assumed negligible saturation, the quantity T is independent of the incident intensity, according to Eqs. (4.1) and (4.6).

The reflection coefficient $R(\epsilon)$ is directly expressed in the field components

$$F_\perp=\epsilon^*\cdot\hat{\mathbf{y}}r_\perp^{(0)}E_{0\perp}, \quad F_\parallel=\epsilon^*\cdot\hat{\mathbf{u}}_{2\parallel}r_\parallel^{(0)}E_{0\parallel} \ . \qquad (4.13)$$

To first order in T we obtain the result

$$R(\epsilon)=\frac{|F_\perp+F_\parallel|^2+2\,\mathrm{Re}(F_\perp^*+F_\parallel^*)(s_\perp F_\perp+s_\parallel F_\parallel)T}{|E_{0\perp}|^2+|E_{0\parallel}|^2} \ . \qquad (4.14)$$

This result demonstrates that by measuring the intensity of the reflected field with a selected polarization ϵ it is possible to attain information both on the real and the imaginary part of T. However, (4.14) is independent of $\mathrm{Im}T$ when $F_\perp$ and $F_\parallel$ have the same phase, and also when $s_\perp$ and $s_\parallel$ are equal. Therefore, in the case of refraction, where $r_\perp^{(0)}$ and $r_\parallel^{(0)}$ are real, R contains no information on $\mathrm{Im}T$ when the incident and the selected polarizations are linear. For total reflection, $\mathrm{Im}T$ can be monitored even in the case of linear polarizations, provided that the polarization directions are not along one of the principal directions.

V. CONCLUSIONS

In this paper we have derived general expressions for the reflection coefficient of light from the interface between a dielectric medium and a resonant atomic vapor. This reflection is affected by the transient behavior of the atoms after deexciting collisions with the surface. We allow for an arbitrary angle of incidence, below or above the critical angle for total internal reflection, and saturation effects are taken into account. The results generalize previous work, which mainly dealt with normal incidence, and low or at most moderate field intensities.

We separate the evaluation in two parts. In Sec. II we derive a functional dependence of the reflected field on the dipole polarization in the vapor. The result is expressed in Eqs. (2.17)–(2.19) for a polarization direction normal to the plane of incidence, and in (2.23) and (2.24) for a polarization parallel to this plane. The quantity S occurring in these equations is defined in (2.20). In Sec. III we evaluate the dipole polarization by solving the optical Bloch equations with the appropriate boundary conditions at the surface for an angle of incidence below or above the critical angle for total reflection. The quantity $S=E_1T$ in these two cases is expressed in Eqs. (3.15) and (3.27). The corresponding expressions for the intensity reflection coefficients for the two principal polarization directions are given in (3.21) and (3.23), or (3.29) and (3.30).

The reflected intensity near resonance is determined by the real part of T, which has a deformed dispersion-type shape for the case of refraction, and an absorption profile for total internal reflection. We point out that the imaginary part of T is accessible to observation when one measures the reflected intensity with a specific selected polarization. Finally, we present numerical calculations of the contribution of the vapor to the reflection coefficient in several specific cases. We compare these results with the values that would arise in the absence of spatial dispersion, when the atoms would have reached their stationary state at each position in the vapor.

The transient approach of atoms towards their stationary state occurs only for atoms leaving the surface, i.e., with $v_z>0$. The resulting step-function behavior of the velocity integrand gives rise to the sub-Doppler structure that has been studied before for normal incidence. This narrow structure gets weaker in the case of non-normal incidence, and it suffers an additional broadening due to the transverse Doppler effect. Furthermore, it is broadened by saturation. The sub-Doppler structure disappears completely close to the critical angle for total internal reflection. On the other hand, the contribution of the vapor to the reflectivity increases with the angle of incidence, in accordance with experiment.[2] This increase reflects that the reflectivity is determined in a layer of thickness of the order of the inverse normal component of the wave vector. In the region of total reflection, the reflection coefficient displays a dip due to absorption of the evanescent wave. The transient effect gives rise to an inhomogeneous Lorentz broadening or, equivalently an imaginary Doppler broadening. The same method developed in this paper can also be used in more complicated cases, corresponding to reflection experiments with a pump-probe configuration.

ACKNOWLEDGMENTS

This work has been supported by a travel grant from the Netherlands Organization for the Advancement of Research (NWO), and the Centre National de la Recherche Scientifique (CNRS). Unité associée au CNRS No. 282.

[1] J. L. Cojan, Ann. Phys. (Paris) **9**, 385 (1954).

[2] J. P. Woerdman and M. F. H. Schuurmans, Opt. Commun. **16**, 248 (1975).

[3] A. L. J. Burgmans and J. P. Woerdman, J. Phys. (Paris) **37**, 677 (1976).

[4] M. F. H. Schuurmans, J. Phys. (Paris) **37**, 469 (1976).

[5] P. Boissel and F. Kerherve, Opt. Commun. **37**, 397 (1981).

[6] S. Le Boiteux, P. Simoneau, D. Bloch, and M. Ducloy, J. Phys. B **20**, L149 (1987).

[7] P. Simoneau, S. Le Boiteux, C. B. de Araujo, D. Bloch, J. R. R. Leite, and M. Ducloy, Opt. Commun. **59**, 103 (1986).

[8] T. A. Vartanyan, Zh. Eksp. Teor. Fiz. **88**, 1147 (1985) [Sov. Phys.—JETP **61**, 674 (1985)].

[9] S. Singh and G. S. Agarwal, Opt. Commun. **59**, 107 (1986).

[10] A. L. J. Burgmans, M. F. H. Schuurmans, and B. Bölger, Phys. Rev. A **16**, 2002 (1977).

[11] D. V. Widder, *The Laplace Transform* (Princeton University Press, Princeton, NJ, 1946).

[12] M. Born, *Optik* (Springer, Berlin, 1946).

[13] M. F. H. Schuurmans, Contemp. Phys. **21**, 463 (1980).