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► **To cite this version:**

M. Ducloy, M. Fichet. General theory of frequency modulated selective reflection. Influence of atom surface interactions. *Journal de Physique II*, 1991, 1 (12), pp.1429-1446. 10.1051/jp2:1991160 . jpa-00247601

**HAL Id: jpa-00247601**

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Submitted on 4 Feb 2008

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Classification

Physics Abstracts

32.70 — 42.50 — 32.80

## General theory of frequency modulated selective reflection. Influence of atom surface interactions

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(Received 29 July 1991, accepted 11 September 1991)

**Résumé.** — On calcule le coefficient de réflexion d'un faisceau lumineux, modulé en fréquence, incident sur une interface entre un milieu diélectrique et une vapeur atomique. Cette vapeur est décrite comme un ensemble de systèmes à deux niveaux, présentant un élargissement Doppler, et dont la fréquence de transition et la largeur de raie sont supposées dépendre de la distance au milieu diélectrique. On suppose par ailleurs que les atomes sont déexcités sur la paroi. La réponse transitoire des atomes est analysée au premier ordre en fonction du champ électromagnétique incident. Du coefficient de réflexion calculé linéairement en fonction de la polarisation atomique, on déduit une expression-formelle de la modulation de réflectivité. On analyse le signal de modulation, en l'absence d'interactions atome-paroi, pour des fréquences de modulation arbitraires. A grande fréquence, ce signal permet d'explorer à la fois l'absorption et la dispersion de la vapeur. La théorie générale est ensuite appliquée à l'étude de la réflectivité pour des interactions atome-paroi de type Van der Waals-London. Le déplacement et les déformations de raie produits par ces interactions sont analysés pour des incidences normale ou oblique du faisceau lumineux.

**Abstract.** — We calculate the modulation of the reflection coefficient for a frequency-modulated (FM) light beam incident on the interface between a dielectric and an atomic vapor. The vapor is described as a gas of resonant, Doppler-broadened, two-level systems, with transition frequency and linewidth arbitrarily depending on the atom-dielectric distance. The atoms are supposed to get deexcited at collisions with the surface. The transient atomic response is calculated to first order in the incident field, for both incoming and desorbed atoms. The reflection coefficient, evaluated to first order in the vapor dipole polarization, leads to a formal expression of the reflectivity modulation, valid for arbitrary atom-surface interaction potentials. One first discusses the reflection signal in absence of wall interactions, for arbitrary modulation frequencies. At large frequencies, it allows one to monitor both vapor absorption and dispersion. Second, the formal theory is applied to the case of a Van der Waals-London surface attraction exerted on the atomic vapor. Both normal and oblique beam incidences are considered. One shows how the vapor dispersion signal is red-shifted and strongly distorted by the appearance of vapor-surface long-range interactions, and how it can be used to monitor these interactions. At non-normal incidences, the lineshapes get Doppler-broadened.

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(\*) U.A. 282 du C.N.R.S.

## 1. Introduction.

The reflection of a light beam at an interface between a dielectric window and an atomic vapor is altered whenever the incident frequency is tuned into resonance with an atomic absorption line. This effect called « Selective reflection » has first been observed by Wood in 1909 [1], and may be straightforwardly assigned to the resonant variations of the real part of the vapor refractive index. A local index approach to selective reflection (SR), based on a *local* relationship between the incident electromagnetic (e.m.) field and the induced dipole polarization, and making use of Fresnel reflection formulae, predicts that SR spectra simply consist of Doppler-broadened dispersion resonances. However, at normal beam incidence, deviations from these predictions have been observed by Cojan [2], who related the appearance of sub-Doppler features to the effect of wall collisions which perturb the interaction between the atomic dipole and the e.m. field. The advent of tunable laser sources allowed Woerdman, Schuurmans and coworkers [3, 4] to make detailed studies of the sub-Doppler resonance appearing in SR spectra, and to demonstrate that they originate in the interruption of the optical dipole phase by atom-wall collisions, and to the subsequent transient response of ground-state atoms departing from the surface [3, 4].

An elegant way of getting rid of the Doppler-broadened part of SR spectra is to use frequency-modulation techniques. When low frequency modulation (FM) is applied to the incident light beam, the amplitude modulation induced on the reflected beam yields the frequency derivative of the SR lineshape: the broad Doppler resonance vanishes, and the sub-Doppler contribution turns into a Doppler-free dispersion lineshape. Indeed, as discussed below in more detail, the FM selective reflection signal becomes solely sensitive to atoms flying parallel to the surface, essentially because for such atoms, the atom-field interaction is not interrupted by wall collisions and they provide the dominant contribution. At normal beam incidence, the Doppler shift is thus cancelled. Such a FM technique has been used by Velichanskii and co-workers [5] to perform, Doppler-free, collisional studies of the Cs resonance line. An important characteristics of SR spectrum is that the signal originates in a vapor layer of extension  $\approx \lambda$ . Thus SR spectroscopy yields high-resolution Doppler-free diagnostics of wavelength-confined vapor-phase atoms, and allows one to perform quantum optics in a kind of 1-D micro-cavity. This feature has been used recently by Oria *et al.* [6] to monitor the spectral signature of the surface-induced long-range attraction potential on vapor-phase atoms. Red shift and line asymmetries have been observed on SR spectra for the two first resonance lines of Cs ( $6S_{1/2} - 6P_{3/2}$  [6] and  $6S_{1/2} - 7P_{3/2}$  [7]).

Theoretical studies of selective reflection have been previously performed [4, 8-12]. In this article, one focuses on the applications of FM techniques to SR spectroscopy. After a general presentation of SR theoretical background (Sect. 2), section 3 considers the case when long-range atom-surface interactions are negligible. Angular dependence of the SR signal and high frequency modulation techniques, known to afford shot-noise-limited sensitivity, will be examined. The last part of this article is devoted to analyzing the influence of long-range atom-surface interactions on SR signals. After developing a formal approach to FM-selective reflection in presence of surface-induced atomic level shifts and broadening, we analyze the effect of a long-range Van der Waals-London interaction potential and its alteration by the angular spectral broadening, in the case of oblique incidence.

## 2. General theoretical approach.

2.1 E.M. FIELD EQUATIONS AND EFFECTIVE SUSCEPTIBILITY. — As in previous publications [10-11], we consider the interface between a dielectric with a real refractive index  $n$ , and an

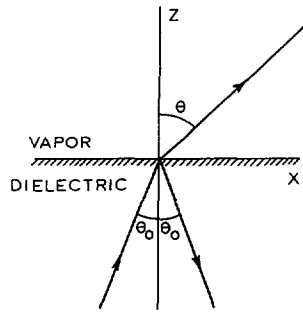


Fig. 1. — Geometry of incident, refracted and reflected waves. The interface is the  $xy$  plane, and the  $xz$  plane is the incidence plane.

atomic vapor. The interface is taken as the  $xy$  plane, and the atomic vapor fills the  $z > 0$  half-space. A plane wave, with wavevector  $\mathbf{k}_0$ , incident on the interface, is partly reflected back into the dielectric, and partly refracted in the vapor (Fig. 1). The refracted field drives a macroscopic electric dipole polarization in the atomic gas, which then re-radiates an e.m. field in the direction of the reflected wave, thereby modifying the reflection coefficient.

We assume all over this study that the resonant reflection change is small, i.e. the vapor refractive index deviates negligibly from unity. This means that the reflection coefficient can be evaluated to first order in the gas density, and that the back reaction of the gas polarization on the field experienced by atoms is negligible. Thus the e.m. field transmitted into the vapor may be assumed to be a plane wave of frequency  $\omega$  and amplitude  $\delta$

$$E(\mathbf{r}, t) = \delta e^{i(\mathbf{k} \cdot \mathbf{r} - \omega t)} + c.c. \tag{1}$$

where the wavevector is given by its vacuum value

$$\mathbf{k} = k(\alpha \hat{x} + \beta \hat{z}) \tag{2}$$

with

$$\alpha = \sin \theta ; \quad \beta = \cos \theta \tag{3}$$

( $\theta$ : refraction angle into vacuum).

With these assumptions, the macroscopic dipole polarization induced in the vapor can be written as

$$P(\mathbf{r}, t) = p(z) e^{i(\mathbf{k} \cdot \mathbf{r} - \omega t)} + c.c. \tag{4}$$

Because of translational invariance along  $x$ , the slowly-varying amplitude  $p$  may depend only on  $z$ . As will be discussed in detail below, the spatially inhomogeneous character of the atomic response could result from either motion-induced spatial dispersion, or surface-induced atomic shifts, or inhomogeneous atomic density due to surface effects.

Solving Maxwell equations for a dipole polarization like (4) shows that the reflected field change induced by the atomic polarization is proportional to [10]

$$\int_0^{+\infty} dz p(z) e^{2i\beta kz} \tag{5}$$

Equation (5) can be interpreted by noting that  $\beta kz$  represents the excess phase experienced by atoms located at distance  $z$  from the surface (compared to  $z = 0$  atoms), and thus

$\exp(2i\beta kz)$  yields the round-trip phase factor appearing in the reflected field reradiated by those atoms. It is thus possible to introduce an effective vapor susceptibility [11-12]

$$\bar{\chi} = -\frac{2ik\beta}{\varepsilon_0 \xi} \int_0^{+\infty} dz p(z) e^{2i\beta kz} \quad (6)$$

( $\varepsilon_0$  is the vacuum permittivity). The corresponding effective refractive index of the vapor,

$$n_v = 1 + \bar{\chi}/2 \quad (7)$$

may be straightforwardly introduced in the Fresnel reflection formulae. Thus the e.m. field reflected into the dielectric can be written as  $\xi_R = r\xi_0$ , where  $\xi_0$  is the incident field and  $r$ , the field reflection coefficient is given by  $r = r_0 + \rho\bar{\chi}$  ( $r_0$  is the dielectric-vacuum reflection coefficient,  $\rho\bar{\chi}$  is the vapor-induced reflectivity change).

For instance, one gets, for light polarization *perpendicular* to the incidence plane [10-12]

$$r^\perp = \frac{nb - \beta}{nb + \beta} - \frac{nb}{\beta} \frac{\bar{\chi}}{(nb + \beta)^2} \quad (8a)$$

and, in the case of a polarization *parallel* to the incidence plane

$$r^\parallel = \frac{n\beta - b}{n\beta + b} - (\beta^2 - \alpha^2) \frac{nb}{\beta} \frac{\bar{\chi}}{(n\beta + b)^2}. \quad (8b)$$

In the above equations,  $n$  is the refractive index of the dielectric;  $a = \sin \theta_0$ ,  $b = \cos \theta_0$  with  $\theta_0$  the incidence angle in the dielectric (see Fig. 1). We always assume that  $|\bar{\chi}| \ll 1$ , and  $\theta_0$  is far enough from the critical angle.

Finally, it should be noted that in the case of a *local* and *homogeneous* response ( $p(z) = p_0$ ), equation (6) reduces to the usual relation  $p_0 = \bar{\chi}\varepsilon_0 \xi$  [13].

**2.2 INDUCED DIPOLE POLARIZATION.** — It therefore remains to evaluate the dipole polarization  $P$  (Eq. (4)), assuming that the atoms move in an external e.m. field,  $E$ , equation (1). One assumes that the field frequency,  $\omega$  is tuned close to an atomic resonance,  $\omega_0$ , so that one can describe the gas as a collection of two-level atomic systems (ground state  $g$ ; excited state  $e$ ), whose density matrix is governed by the usual Bloch equations. The atomic dipole moment is thus proportional to the off-diagonal density matrix element,  $\sigma_{eg}$ , measuring the amount of optical coherence between  $e$  and  $g$ . The macroscopic dipole polarization is determined by the expectation value of atomic dipole moments at position  $\mathbf{r}$ , averaged over the velocity distribution

$$p(z) = N\mu \int_{-\infty}^{+\infty} W(\mathbf{v}) \sigma_{eg}(\mathbf{r}, \mathbf{v}) d^3\mathbf{v} \quad (9)$$

$\mu$  is the e-g electric dipole moment,  $N$  the atom number density and  $W$  the normalized velocity distribution.

In the following, the incident field is supposed to be weak enough so that atomic response is linear. To first order in the field, the evolution of  $\sigma_{eg}$  is governed by (in the rotating wave approximation) [10]

$$\frac{d}{dt} \sigma_{eg} = - \left[ \frac{\gamma}{2} - i(\omega - \omega_0 - \mathbf{k} \cdot \mathbf{v}) \right] \sigma_{eg} + i\Omega/2 \quad (10)$$

in which  $\mathbf{k} \cdot \mathbf{v} = \alpha k v_x + \beta k v_z$  is the Doppler shift,  $\Omega = 2 \mu \mathcal{E} / \hbar$  is the Rabi frequency, and the total hydrodynamic time derivative is  $\frac{d}{dt} = \frac{\partial}{\partial t} + \mathbf{v} \cdot \nabla$ . Because of long-range atom-surface interactions, the optical transition linewidth,  $\gamma$ , and frequency,  $\omega_0$ , are assumed to depend on atom-surface distance,  $z$ . Translational invariance along the  $x$ - $y$  plane implies that, in the steady-state regime,  $\sigma_{eg}$  depends only on  $z$ , so that equation (10) reduces to

$$v_z \frac{d\sigma_{eg}}{dz} = - \left[ \frac{\gamma}{2} - i(\omega - \omega_0 - \mathbf{k} \cdot \mathbf{v}) \right] \sigma_{eg} + i \frac{\Omega}{2}. \quad (11)$$

The general solution of (11) may be found, if we introduce  $\mathcal{L}_0(z)$  as the integral of  $\frac{1}{2} \gamma - i(\omega - \omega_0)$ :

$$\frac{d}{dz} \mathcal{L}_0 = \frac{1}{2} \gamma(z) + i[\omega_0(z) - \omega]. \quad (12)$$

Thus the general solution of (11) is obtained by a straightforward integration:

$$\sigma_{eg}(z, \mathbf{v}) = i \frac{\Omega}{2 v_z} \int_{z_0}^z dz' e^{[\mathcal{L}(z') - \mathcal{L}(z)]/v_z} \quad (13)$$

with

$$\begin{aligned} \mathcal{L}(z) &= \mathcal{L}_0(z) + iz\mathbf{k} \cdot \mathbf{v} \\ &= \mathcal{L}_0(z) + ikz(\alpha v_x + \beta v_z). \end{aligned} \quad (14)$$

In equation (13), the integration constant,  $z_0$ , represents the atom-surface separation for which the optical coherence vanishes. The solution depends on  $v_z$ .

(i)  $v_z > 0$ : the atom is going away from the surface. It is thus highly likely that the dipole phase is disrupted (or, at least, randomly distributed) on the wall, and that  $\sigma_{eg} = 0$ . This leads us to  $z_0 = 0$ , and

$$\sigma_{eg}(z, v_z > 0) = i \frac{\Omega}{2 v_z} \int_0^z dz' e^{[\mathcal{L}(z') - \mathcal{L}(z)]/v_z} \quad (15)$$

(ii)  $v_z < 0$ . The atoms approach the surface, coming from the  $z = +\infty$  side, where they are not excited. Setting  $z_0 = +\infty$  leads to the solution

$$\sigma_{eg}(z, v_z < 0) = i \frac{\Omega}{2 v_z} \int_{+\infty}^z dz' e^{[\mathcal{L}(z') - \mathcal{L}(z)]/v_z} \quad (16)$$

(iii)  $v_z = 0$ . The solution of (11) is

$$\sigma_{eg}(z, v_z = 0) = i \frac{\Omega}{2} \frac{1}{\frac{\gamma(z)}{2} - i[\omega - \omega_0(z) - \mathbf{k} \cdot \mathbf{v}]} \quad (17)$$

Equation (17) may also be obtained by continuity from (15) or (16), with  $v_z \rightarrow 0$ .

**2.3 GENERAL PROPERTIES OF THE EFFECTIVE SUSCEPTIBILITY.** — The effective susceptibility of the atomic vapor is given by (Eqs. (6), (9))

$$\bar{\chi} = - \frac{2 ik\beta N \mu}{\varepsilon_0 \mathcal{E}} \int_{-\infty}^{+\infty} d^3\mathbf{v} W(\mathbf{v}) \int_0^{+\infty} dz \sigma_{eg}(z, \mathbf{v}) e^{2i\beta k z} \quad (18)$$

with  $\sigma_{eg}$  given by equations (15)-(17).

Let us introduce an effective susceptibility per atom of velocity  $\mathbf{v}$  :

$$\chi(\mathbf{v}) = -\frac{2ik\beta\mu}{\varepsilon_0\hbar} \int_0^{+\infty} dz \sigma_{eg}(z, \mathbf{v}) e^{2i\beta kz} \quad (19)$$

The vapor susceptibility is

$$\bar{\chi} = N \int_{-\infty}^{+\infty} d^3\mathbf{v} W(\mathbf{v}) \chi(\mathbf{v}). \quad (20)$$

A noteworthy property of  $\chi(\mathbf{v})$  is to be an even function of  $v_z$  :

$$\chi(v_x, v_y, v_z) = \chi(v_x, v_y, -v_z). \quad (21)$$

Indeed, for  $v_z > 0$ , one has (Eq. (15))

$$\chi(v_x, v_y, v_z > 0) = \frac{2k\beta\mu^2}{\varepsilon_0\hbar} \frac{1}{v_z} \int_0^{+\infty} dz \int_0^z dz' e^{[\zeta(z') - \zeta(z)]/v_z} e^{2i\beta kz} \quad (22)$$

In (22), the order of integration can be reversed, by using the relationship

$$\int_0^{+\infty} dz \int_0^z dz' = \int_0^{+\infty} dz' \int_{z'}^{+\infty} dz. \quad (23)$$

This yields

$$\chi(v_z > 0) = -\frac{2k\beta\mu^2}{\varepsilon_0\hbar} \frac{1}{v_z} \int_0^{+\infty} dz' \int_{+\infty}^{z'} dz e^{[\zeta(z') - \zeta(z)]/v_z} e^{2i\beta kz} \quad (24)$$

After interchange of  $z$  and  $z'$ , (24) is easily shown to be identical to  $\chi(v_x, v_y, -|v_z|)$ , such as is obtained from (14), (16) and (19). One thus demonstrates the property that  $(+v_z)$  and  $(-v_z)$  atomic velocity groups yield identical contributions to the linear reflection signal. This extends to atoms interacting with the surface, the property previously demonstrated for « free » atoms [4, 10].

Note that for atoms moving along the surface ( $v_z = 0$ ), the effective susceptibility per atom is simply

$$\chi(v_x, v_y, 0) = \frac{2k\beta\mu^2}{\varepsilon_0\hbar} \int_0^{+\infty} dz \frac{e^{2i\beta kz}}{\frac{\gamma}{2} - i(\omega - \omega_0 - \mathbf{k} \cdot \mathbf{v})} \quad (25)$$

**2.4 DISCUSSION.** — Aside from straightforward assumptions like small index changes ( $|\bar{\chi}| \ll 1$ ), or linear response ( $\Omega \ll \gamma$ ), there are a few important hypotheses.

(i) One assumes that the surface-induced frequency shift, and line width change, *depend only on the instantaneous position of the atom*. This implies that retardation effects should be negligible, at least for which concerns the atom's *external* degrees of freedom. Possible time delays are correlated with the atom-surface, light round trip time,  $\tau = 2z/c$ . The corresponding frequency change is  $|\Delta\omega_0| \approx \left| \tau \frac{d\omega_0}{dt} \right| = \left| 2z \frac{v_z}{c} \frac{d\omega_0}{dz} \right|$  leading to the following condition

$$2 \left| \frac{v_z}{c} z \frac{d\omega_0}{dz} \right| \ll |\omega - \omega_0|, \gamma. \quad (26)$$

The above hypothesis does not apply to the atomic *internal* variables (like electron motion), for which retardation effects play a major role as soon as  $z \approx \lambda$  (i.e.,  $\omega_0 \tau \approx 1$ ).

(ii) The atomic motion is treated *classically* (no quantization of center-of-mass motion), and in the approximation of *rectilinear* trajectories. One thus neglects, in a first approach, both the curvature of atomic trajectories due to surface-induced forces, and the motion-induced correlations between external and internal atomic variables (i.e. internal-state-dependent trajectories). This will be discussed in more detail at the end of the paper.

### 3. FM selective reflection for freely-precessing atomic dipoles.

In this section, one assumes that the evolution of atomic dipoles is not affected by the surface, so that  $\gamma$  and  $\omega_0$  do not depend on  $z$ .

Thus (see Eqs. (12)-(14))  $\mathfrak{L}$  is simply given by  $Lz$  with

$$L = \frac{1}{2} \gamma - i(\delta - \mathbf{k} \cdot \mathbf{v}) \quad (27)$$

and

$$\delta = \omega - \omega_0. \quad (28)$$

The general solution (Eq. (13)) becomes

$$\sigma_{\text{eg}}(z, \mathbf{v}) = i \frac{\Omega}{2L} [1 - e^{L(z_0 - z)/v_z}]. \quad (29)$$

For  $v_z > 0$ , one has  $z_0 = 0$  and the solution reflects the transient behavior of ground-state atoms leaving the surface

$$\sigma_{\text{eg}}(z, \mathbf{v}) = i \frac{\Omega}{2L} (1 - e^{-Lz/v_z}). \quad (30)$$

For  $v_z \leq 0$ , one gets the steady-state dipole response ( $z_0 = +\infty$ )

$$\sigma_{\text{eg}}(z, \mathbf{v}) = i \frac{\Omega}{2L} \quad (31)$$

When either (30) or (31) are replaced in equation (19), this leads to a single-atom effective susceptibility which is an even function of  $v_z$ , as expected, and is given by

$$\chi(\mathbf{v}) = i \frac{\mu^2}{\varepsilon_0 \hbar} \left[ \frac{\gamma}{2} - i(\delta - \alpha k v_x + \beta k |v_z|) \right]^{-1} \quad (32)$$

The vapor susceptibility can be written

$$\bar{\chi} = i \frac{N \mu^2}{\varepsilon_0 \hbar} \int_{-\infty}^{+\infty} dv_x \int_{-\infty}^{+\infty} dv_y \int_0^{+\infty} dv_z \frac{W(v_x, v_y, v_z) + W(v_x, v_y, -v_z)}{\frac{\gamma}{2} - i(\delta - \alpha k v_x + \beta k v_z)} \quad (33)$$

This result is equivalent to the ones obtained previously by several authors [4, 8, 10], in the limit of weak field (linear response) and low vapor density. In the case of a Maxwell-Boltzmann velocity distribution,

$$W(\mathbf{v}) = (u \sqrt{\pi})^{-3} e^{-v^2/u^2} \quad (34)$$



( $u$ , thermal velocity), the vapor susceptibility becomes

$$\bar{\chi} = i \frac{2 N \mu^2}{\varepsilon_0 \hbar \pi u^2} \int_{-\infty}^{+\infty} dv_x \int_0^{+\infty} dv_z \frac{e^{-(v_x^2 + v_z^2)/u^2}}{\frac{\gamma}{2} - i(\delta - \alpha k v_x + \beta k v_z)} \quad (35)$$

Note that this type of susceptibility applies to the cases when either the desorbed atoms are thermalized, or there are no desorbed atoms (because of chemical transformations, etc.).

**3.1 GENERAL FORMALISM.** — Frequency-modulation of the incident field is taken into account by replacing in equation (1) the field amplitude by

$$\begin{aligned} \delta &= |\delta| e^{-iM \sin \omega_m t} \\ &= |\delta| \sum_N J_N(M) e^{-iN \omega_m t} \end{aligned} \quad (36)$$

where  $M$  and  $\omega_m$  are the FM modulation index and frequency respectively.  $J_N$  is the Bessel function of integer order  $N$ . Thus the reflected field may be written as

$$\varepsilon_R = \sum_N r(\omega + N \omega_m) \varepsilon_0 J_N(M) e^{-iN \omega_m t} \quad (37)$$

$r(\omega + N \omega_m)$ , the reflection coefficient at frequency  $\omega + N \omega_m$ , is given by :

$$r(\omega + N \omega_m) = r_0 + \rho \bar{\chi}(\omega + N \omega_m). \quad (38)$$

The light reflectivity  $R = |\varepsilon_R/\varepsilon_0|^2$  is thus given by

$$R = r_0^2 + r_0 \rho \sum_{N, N'} [\bar{\chi}(\omega + N \omega_m) + \bar{\chi}^*(\omega + N' \omega_m)] J_N J_{N'} e^{-i(N - N') \omega_m t}. \quad (39)$$

The modulated terms in  $R$  allow one to discriminate the frequency-dependent vapor contribution to the reflectivity. For instance, the AM contribution at frequency  $\omega_m$  is

$$\delta R(\omega_m) = r_0 \rho \sum_N [\bar{\chi}(\omega + N \omega_m) + \bar{\chi}^*(\omega + N \omega_m - \omega_m)] J_N J_{N-1} e^{-i \omega_m t} + \text{c.c.} \quad (40)$$

For weak modulation index, one can limit the summation in equation (40) to the  $J_0 J_{\pm 1}$  terms.

$$\delta R(\omega_m) = r_0 \rho J_0 J_1 [\bar{\chi}(\omega + \omega_m) - \bar{\chi}^*(\omega - \omega_m) + \bar{\chi}^*(\omega) - \bar{\chi}(\omega)] e^{-i \omega_m t} + \text{c.c.} \quad (41)$$

For  $\omega_m$  large compared with the spectral features of  $\bar{\chi}$ , phase-sensitive detection of the AM modulated reflectivity allows one to monitor both real and imaginary parts of  $\bar{\chi}$ . This is one of the well-known advantages of FM spectroscopy [14-16]. Reflection spectroscopy usually does not yield the absorptive response, except for metallic reflection [17].

In equation (41), the in-phase component yields the dispersion part of the atomic response at the vapor-dielectric interface, under the form of a dispersion doublet of opposite sign,

$$\text{Re}[\bar{\chi}(\omega + \omega_m) - \bar{\chi}(\omega - \omega_m)]. \quad (42)$$

On the other hand, the quadrature component gives access to the absorptive features of the atomic response at the interface, under the form of an absorption triplet :

$$\text{Im}[\bar{\chi}(\omega + \omega_m) + \bar{\chi}(\omega - \omega_m) - 2 \bar{\chi}(\omega)]. \quad (43)$$

Note that fluorescence experiments under evanescent wave excitation are also able to provide absorption features. Generally, in the Doppler limit,  $\text{Im } \bar{\chi}$  yields *half a Voigt lineshape* (i.e. its lower frequency part, with a sudden decrease to zero at line center, in a frequency interval of order  $\gamma$ , or  $ku \beta$ ). For a Maxwell-Boltzmann distribution, one has :

$$\text{Im } \bar{\chi} \propto \int_{-\infty}^{+\infty} dv_x \int_0^{+\infty} dv_z \frac{\exp - (v_x^2 + v_z^2)/u^2}{\frac{\gamma^2}{4} + (\delta - \alpha kv_x + \beta kv_z)^2} \tag{44}$$

**3.2 NORMAL INCIDENCE.** — Predicted reflection lineshapes are given in figures 2 and 3, at normal incidence ( $\alpha = 0, \beta = 1$ ) for various modulation frequencies,  $\omega_m$ .

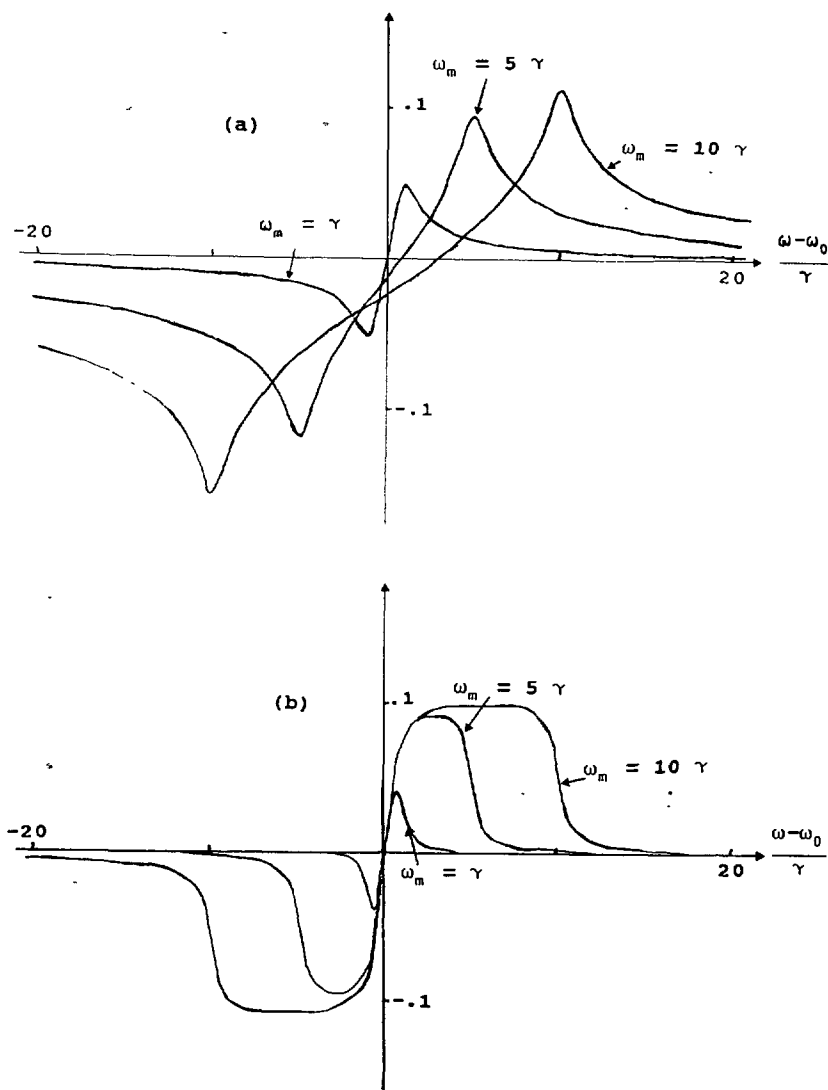


Fig. 2. — FM reflection signal at normal incidence, for various modulation frequencies :  $\omega_m = \gamma, 5 \gamma, 10 \gamma$  ( $ku = 50 \gamma$ ) ; dispersive (a) and absorptive (b) responses.

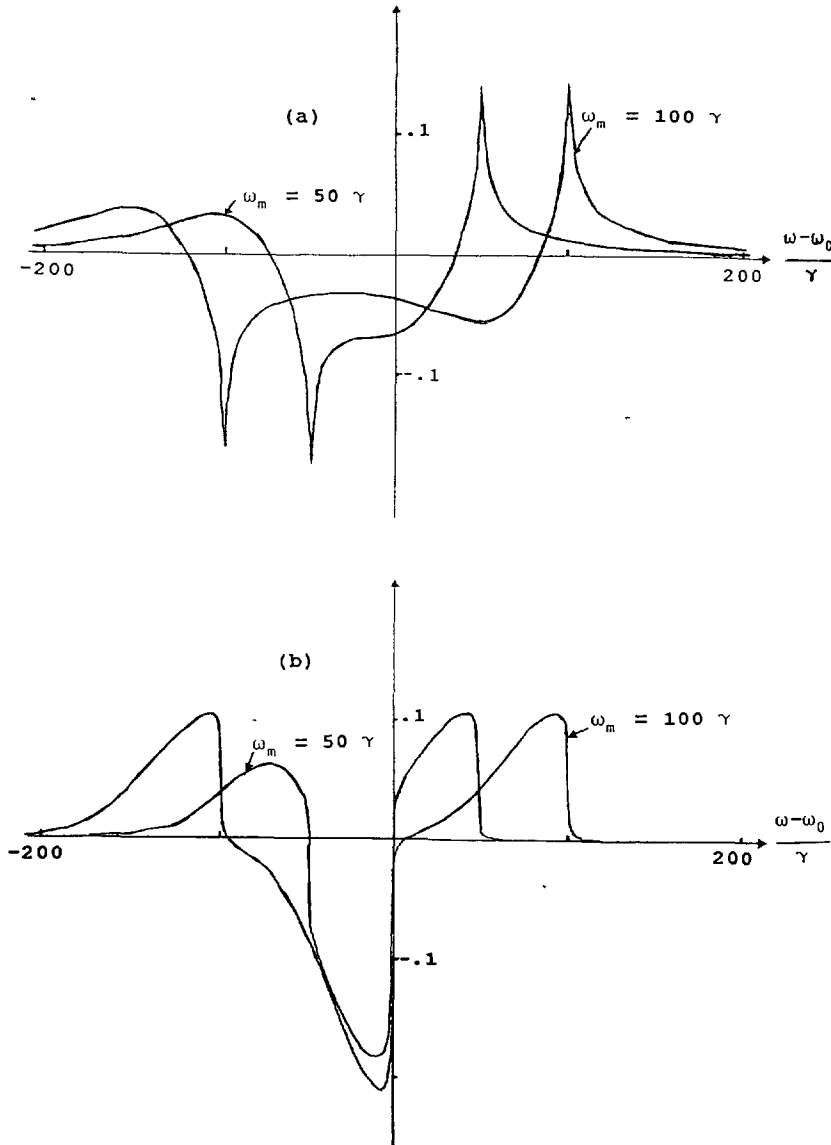


Fig. 3. — FM reflection signal at normal incidence, in the case of very large modulation frequencies :  $\omega_m = 50 \gamma$ ,  $100 \gamma$  ( $ku = 50 \gamma$ ) ; dispersive (a) and absorptive (b) responses.

In the case of a low frequency of modulation ( $\omega_m \leq \gamma$ ), one gets the usual frequency-derivative for the in-phase (*dispersive*) component :

$$\delta R(\omega_m) = 2 r_0 \rho J_0 J_1 \omega_m \left( \frac{d\bar{\chi}}{d\omega} + \frac{d\bar{\chi}^*}{d\omega} \right) \cos \omega_m t . \tag{45}$$

The quadrature component (smaller by a factor  $\omega_m/\gamma$ ) is proportional to the second derivative of the absorption,  $\text{Im } d^2\bar{\chi}/d\omega^2$ .

For an arbitrary velocity distribution exhibiting *broad* variations (« Doppler limit »), the  $v_z$ -integral in equation (33) (with  $\alpha = 0$ ) may be performed analytically by replacing

$W(\pm v_z)$  by  $W(0_{\pm})$ , and using the fact that all the  $\omega$  and  $v_z$  dependence is then enclosed in  $(\delta + kv_z)$ . Thus the relationship

$$\frac{d}{dv_z} = k \frac{d}{d\omega} \tag{46}$$

leads to

$$\frac{d\bar{\chi}_0}{d\omega} = -i \frac{N\mu^2}{\epsilon_0 \hbar k} \frac{W(v_z = 0_+) + W(v_z = 0_-)}{\frac{\gamma}{2} - i\delta} \tag{47}$$

[ $\bar{\chi}_0$  stands for  $\theta = 0$ ].

Doppler-free dispersion lineshapes indeed appear in the low-frequency predictions of figure 2. This  $v_z = 0$  velocity selection in FM selective reflection has been first noticed by Akul'shin *et al.* [5]. Note that the quadrature component is antisymmetric up to  $\omega_m \leq 0.2 ku$  (Fig. 2). It thus provides an error signal, for optical frequency stabilization, extending over a range  $\mp 0.2 ku$  with a steep slope around  $\omega = \omega_0$  governed by the homogeneous linewidth  $\gamma$ .

**3.3 ANGULAR BROADENING.** — In the following, only low modulation frequencies are considered (Eq. (45)). Thus the FM-SR lineshape is given by  $\text{Re} \frac{d\bar{\chi}}{d\omega}$ , with  $\bar{\chi}$  given by equation (33) or (35). The Doppler shift along the  $x$  direction, which appears for oblique incidence ( $\theta \neq 0$ ), is responsible for an angular broadening of the dispersion lineshape. Also, in this case, if we assume a slowly-varying amplitude for the velocity distribution (Doppler width large compared with the natural linewidth) and *near-normal incidence* ( $\alpha \ll 1$ ,  $\beta = 1$ ), we make use of equation (46) to get a generalized form of equation (47) :

$$\frac{d\bar{\chi}}{d\omega} = -i \frac{N\mu^2}{\epsilon_0 \hbar ku \sqrt{\pi}} \int_{-\infty}^{+\infty} dv_x \frac{W_+(v_x) + W_-(v_x)}{\frac{\gamma}{2} - i(\delta - \alpha kv_x)} \tag{48}$$

in which  $W_{\pm}(v_x) = u \sqrt{\pi} W(v_x, v_z \rightarrow 0_{\pm})$  [18]. For Maxwellian laws,  $W_{\pm}(v_x) = (u \sqrt{\pi})^{-1} e^{-v_x^2/u^2}$ . Equation (48) predicts a Doppler broadening which originates in the velocity distribution along the surface (Fig. 4). This broadening, experimentally observed in Cs [17, 19], is given by  $1.83 \alpha ku$  (for  $\gamma \ll \alpha ku$ ).

**4. FM selective reflection in presence of long-range surface-atom interactions.**

In this section, we analyse the general solution for the effective susceptibility with  $\omega_0$  and  $\gamma z$ -dependent (Eqs. (15)-(18)). Using parity properties of  $\chi(\mathbf{v})$ , one can write  $\chi$  as

$$\bar{\chi} = N \int_{-\infty}^{+\infty} dv_x \int_{-\infty}^{+\infty} dv_y \int_0^{+\infty} dv_z [W(v_x, v_y, v_z) + W(v_x, v_y, -v_z)] \chi(v_x, v_y, v_z) \tag{49}$$

with  $\chi(\mathbf{v})$  given by equation (22). We assume low modulation frequencies in order that equation (45) applies, and that the AM reflection signal is proportional to  $\text{Re} (d\bar{\chi}/d\omega)$ .

**4.1 FORMAL THEORY AT NORMAL INCIDENCE.** — At normal incidence  $\chi_{\theta=0}(\mathbf{v})$  depends on  $v_z$  only, so that  $v_x$  and  $v_y$  averages are easily performed. This leads to the following expression (cf. Eqs. (12), (14), (22)) :

$$\bar{\chi}_0 = \frac{2 N k \mu^2}{\epsilon_0 \hbar} \int_0^{+\infty} \frac{dv_z}{v_z} [W(v_z) + W(-v_z)] \int_0^{+\infty} dz \int_0^z dz' e^{ik(z+z')} e^{[\epsilon_0(z') - \epsilon_0(z)]/v_z} \tag{50}$$

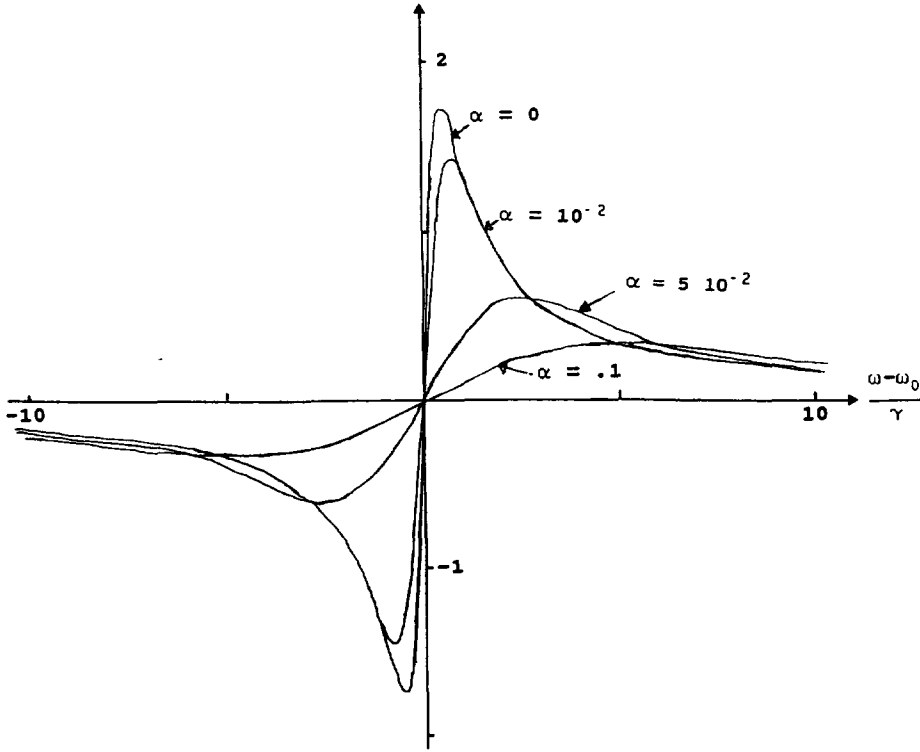


Fig. 4. — FM-SR lineshapes at near-normal incidence, for low modulation frequencies;  $\alpha = 0, 10^{-2}, 5 \times 10^{-2}, 0.1$  ( $ku = 50 \gamma$ ).

In  $\bar{\chi}_0$ , frequency  $\omega$  appears in  $\epsilon_0$  only, and  $\frac{d\epsilon_0}{d\omega} = -iz$ . Thus  $\frac{d\bar{\chi}_0}{d\omega}$  contains the following integral over  $v_z$ :

$$i(z - z') \int_0^{+\infty} \frac{dv_z}{v_z^2} [W(v_z) + W(-v_z)] e^{[\epsilon_0(z') - \epsilon_0(z)]/v_z} \tag{51}$$

In the Doppler limit (Doppler width  $\gg \gamma, |\omega - \omega_0|$ ),  $W(\pm v_z)$  is slowly varying around the  $v_z = 0$  singularity, where the remaining part of the integrand is very large and yields the dominant part of the integral. By taking  $W(\pm v_z)$  out of the integral, the integration of equation (51) is easily carried out and yields:

$$i[W(v_z = 0_+) + W(v_z = 0_-)] \frac{z - z'}{\epsilon_0(z) - \epsilon_0(z')}. \tag{52}$$

This leads us to the final expression:

$$\frac{d\bar{\chi}_0}{d\omega} = i \frac{Nk\mu^2}{\epsilon_0 \hbar} [W(0_+) + W(0_-)] \int_0^{+\infty} dz \int_0^{+\infty} dz' e^{ik(z+z')} \frac{z - z'}{\epsilon_0(z) - \epsilon_0(z')}. \tag{53}$$

In the above formula, the integral over  $z'$  has been extended from 0 to  $+\infty$ , by reversing the integration order (cf. Eq. (23)), and using the  $z/z'$  symmetry of the integrand. Expression (53)

is the key expression for predicting FM selective reflection spectra in presence of atom-surface long-range interactions. In the absence of such interactions,  $\epsilon_0(z) = \left(\frac{1}{2}\gamma - i\delta\right)z$ , and equation (53) reduces to equation (47) (cf. Ref. [13]).

*Note that*

(i) The  $v_z = 0$  singularity is basic in obtaining equation (53). As discussed before (Sect. 3.2), this reflects the dominant contribution to FM-SR spectra, of *atoms moving nearly parallel to the dielectric surface*. For those atoms, the atom-light interaction is not interrupted by wall collisions (within the atomic dipole lifetime).

(ii) The double-space integral over  $z$  and  $z'$  reflects the *non-local* character of the atomic response, originating in the *transient behaviour* of atoms moving in an external inhomogeneous potential. In this sense, it is quite revealing to compare equation (53) with the equivalent prediction for *stationary* atoms, which can be obtained from equations (17)-(18) (with  $\mathbf{v} = 0$ ), and contains a single space average :

$$\chi_{st} = \frac{2Nk\mu^2}{\hbar\epsilon_0} \int_0^{+\infty} dz \frac{e^{2ikz}}{\frac{1}{2}\gamma(z) - i[\omega - \omega_0(z)]} . \quad (54)$$

4.2 SR SPECTRUM FOR VAN DER WAALS INTERACTION FORCES. — For a Van der Waals (VW) London non-resonant potential [20], we take  $\gamma$  constant and

$$\omega_0(z) = \omega_0 - C/z^3 \quad (55)$$

( $C$ , strength of the interaction potential).

This gives

$$\epsilon_0(z) = L_0 z + iC/2 z^2 \quad (56)$$

with

$$L_0 = \frac{1}{2}\gamma - i\delta . \quad (57)$$

In appendix, we show how the susceptibility may be written as (cf. Eq. (A6)) :

$$\frac{d\bar{\chi}_0}{d\omega} = - [W(0_+) + W(0_-)] \frac{iNk\mu^2}{\epsilon_0 \hbar L_0} \left( 1 - \int_0^{+\infty} ds \frac{8i\nu F(q) e^{is}}{s^2 L_0} \right) \quad (58)$$

with  $F(q)$  given by equations (A7)-(A8), and  $\nu = Ck^3$ . The predicted SR lineshapes are shown in figures 5 (a, b, c), for various values of the dimensionless parameter

$$A = 2Ck^3/\gamma . \quad (59)$$

$A$  measures the VW frequency shift at a distance  $\lambda/2\pi$  from the surface, in units of the dipole decay rate,  $\gamma/2$ . In appendix, the lineshape behavior is discussed for small values of  $A$ . For large values of  $A$ , the dispersion lineshape is red-shifted, and gradually distorted, denoting the increasing mixture of absorption and dispersion atomic response, induced by VW forces. This distortion fully transforms the lineshape into an absorptive one ( $A \approx 6$ ), and eventually reverses the sign of the dispersion for  $A > 10$ . Such a peculiar behaviour has been observed on the second resonance line of Cesium ( $6S_{1/2} - 7P_{3/2}$ ;  $\lambda = 456$  nm) [7]. The first Cs resonance line ( $6S - 6P$ ) yields weakly distorted dispersion lineshapes ( $A \approx 0.2$ ) [6].

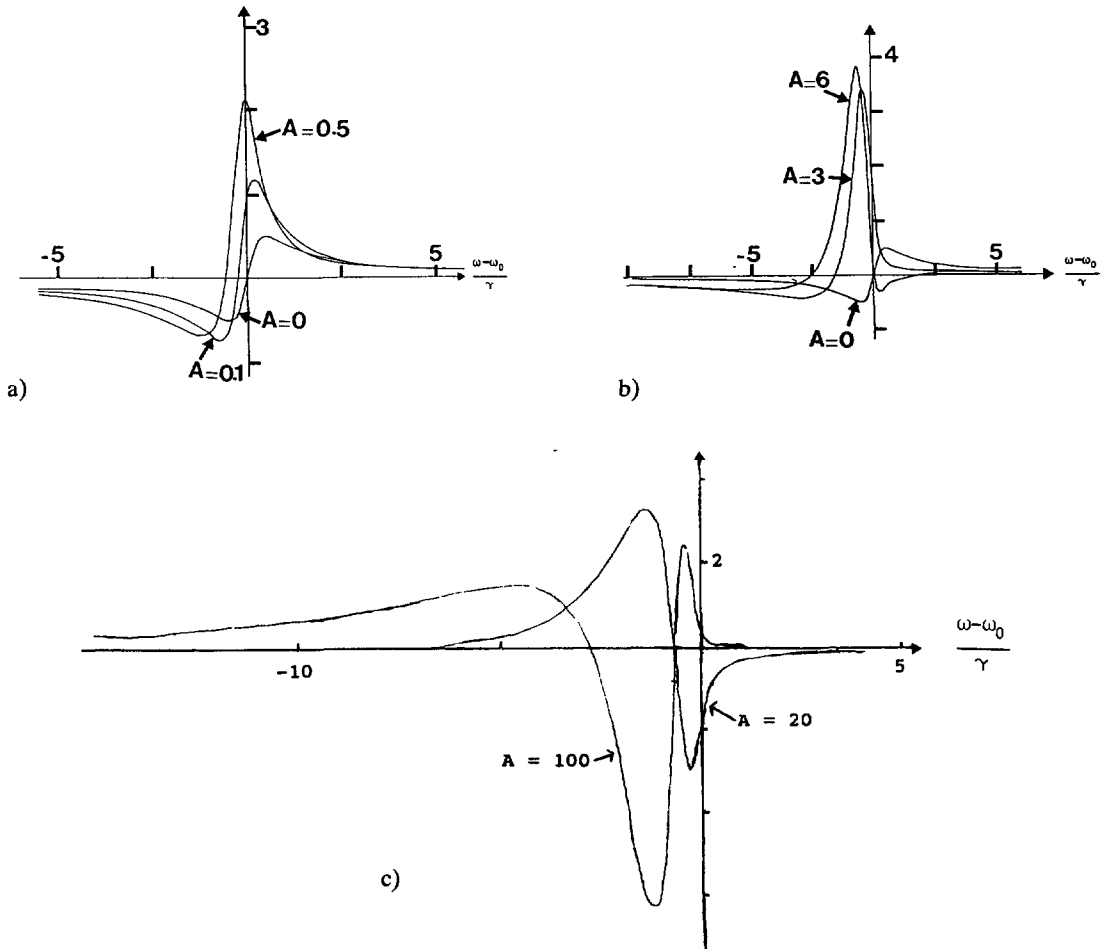


Fig. 5. — FM-SR lineshapes for various strengths of the Van der Waals attraction at normal incidence : (a)  $A = 0, 0.1, 0.5$ ; (b)  $A = 0, 3, 6$ ; (c)  $A = 20, 100$ .

**4.3 OBLIQUE INCIDENCE.** — We have seen in section 3.3 that the deviations of beam incidence from the normal to the interface are responsible for an angular broadening of SR spectra. Here, we address this situation when atom-surface long-range interactions are present. The starting equation is now equation (49) with  $\chi(\mathbf{v})$  given by equation (22). In  $\chi$ , non-vanishing  $\alpha$  introduces an additional  $v_x$ -dependence via  $\mathcal{L}(z)$  (Eq. (14)). After integration over  $v_y$ , and with the assumption of a thermal velocity distribution,  $\bar{\chi}$  can be written under the general form

$$\bar{\chi} = \frac{4 N k \beta \mu^2}{\varepsilon_0 \hbar \pi u^2} \int_{-\infty}^{+\infty} dv_x \int_0^{+\infty} \frac{dv_z}{v_z} e^{-(v_x^2 + v_z^2)/u^2} \times \int_0^{+\infty} dz \int_0^z dz' e^{i\beta k(z+z')} e^{[\mathcal{L}_0(z') - \mathcal{L}_0(z) + i\alpha k v_x(z' - z)]/v_z} \quad (60)$$

For arbitrary values of  $\alpha = \sin \theta$ , one has to perform numerically all the integrations appearing in  $d\bar{\chi}/d\omega$ . However, for  $\alpha \ll 1$ , the Doppler shift,  $\alpha k v_x$ , remains small compared to

the full Doppler linewidth,  $ku$ . Thus, integration over  $v_z$  may be carried out along the very lines detailed in section 4.1, i.e. setting  $e^{-v_z^2/u^2} \approx 1$ , and replacing in equation (51)  $\mathcal{L}_0$  by  $\mathcal{L}_0 + i\alpha kv_x z$ . The final result is

$$\frac{d\bar{\chi}}{d\omega} = i \frac{2 N k \beta \mu^2}{\varepsilon_0 \hbar \pi u^2} \int_{-\infty}^{+\infty} dv_x e^{-v_x^2/u^2} \int_0^{+\infty} dz \int_0^{+\infty} dz' \frac{(z - z') e^{i\beta k(z+z')}}{\mathcal{L}_0(z) - \mathcal{L}_0(z') + i\alpha kv_x(z - z')}. \quad (61)$$

The SR lineshape at  $\theta \neq 0$  is thus the convolution of the normal-incidence atomic response — shifted by the Doppler effect along the  $x$  direction — with the  $v_x$  velocity distribution :

$$\frac{d\bar{\chi}}{d\omega} = \int_{-\infty}^{+\infty} dv_x \frac{e^{-v_x^2/u^2}}{u \sqrt{\pi}} \frac{d\bar{\chi}_0}{d\omega} (\omega - \alpha kv_x). \quad (62)$$

Examples of SR lineshape angular behavior are shown in figures 6 (a, b) in the case of a Van der Waals atom-surface interaction potential: Note that, when the Doppler broadening gets very large compared to the VW shift ( $\alpha ku \gg \nu$ ), the lineshape recovers its dispersive behaviour, which is still red-shifted.

### 5. Conclusion.

In this article, we have presented a general theoretical approach to frequency-modulated selective reflection from an interface between a low-density atomic vapor and a dielectric window. We have shown how Doppler-free linear reflection spectra can be monitored due to the non-local character of the atomic dipole response, which enhances the contribution of atoms moving parallel to the interface.

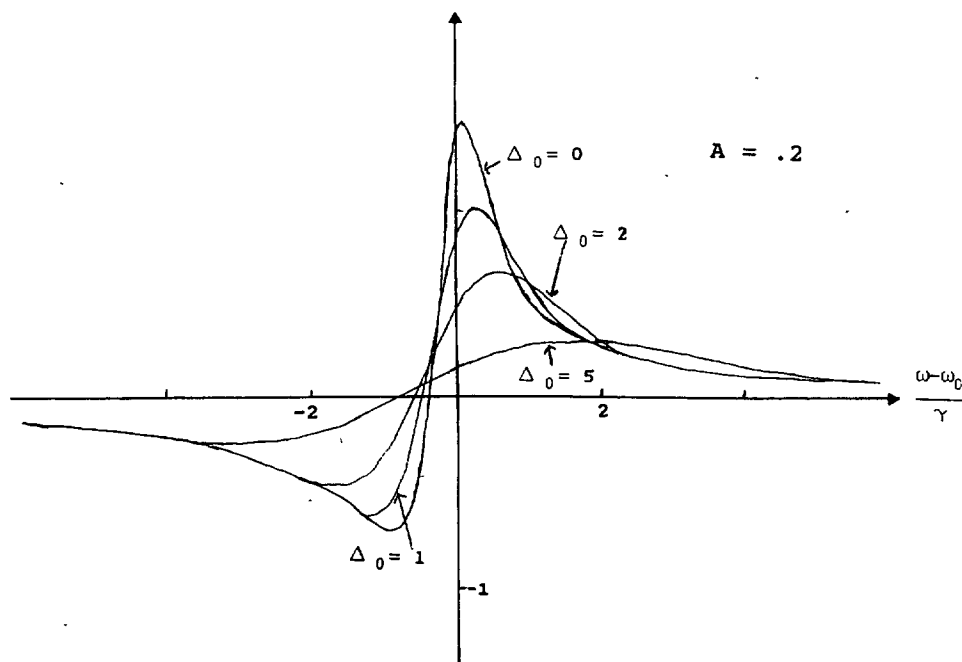
Both the high frequency-resolution (natural-width-limited), and the fact that only atoms located within a wavelength-deep layer contribute to the reflection signal, make the lineshape highly sensitive to atom-surface interactions. We have shown how to incorporate these long-range interactions in the formal theory, and analyzed their influence.

To perform this theoretical study, we have assumed that the atomic trajectories are not affected by the atom-surface interaction forces. This assumption, if valid for heavy atoms (like Cs [6, 7]) and weakly polarizable energy levels, may get unrealistic for lighter atoms and highly excited states. Indeed, bending of atomic trajectories, e.g., by attractive forces, may introduce an additional distortion in the spectral lineshapes, and increase the red lineshift.

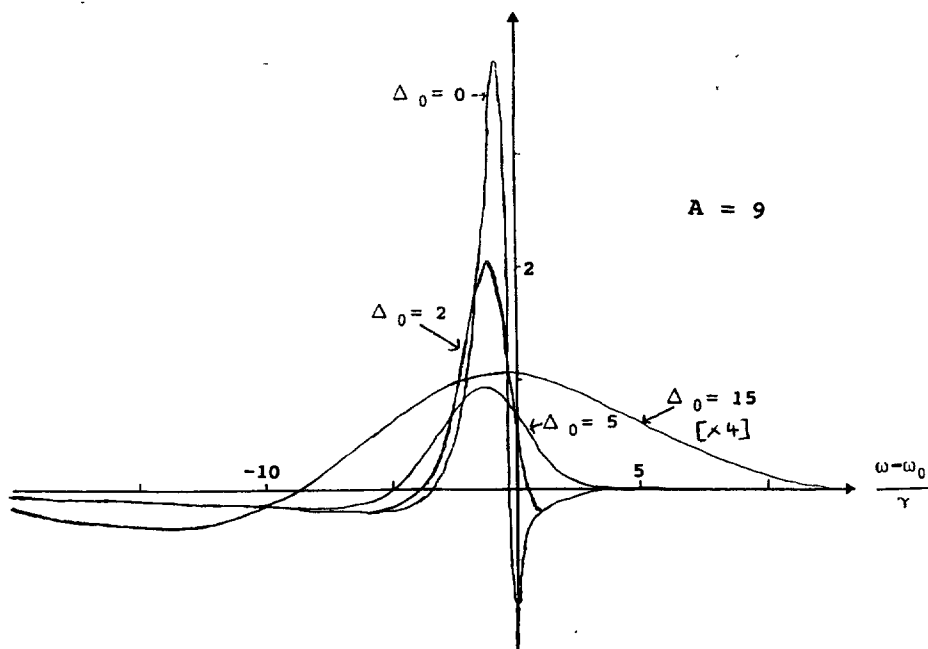
Another cause of spectral modifications relies in the fact that the interaction forces may differ strongly for the ground and excited states. This creates a quantum-mechanical correlation between internal and external degrees of freedom (Stern-Gerlach-type measurement process). The separation of atomic trajectories, in this kind of long-range atom-wall collisions, can be responsible for a reduction of the dipole lifetime.

The proper way of dealing with all these features, is to quantize the atomic motion, as well as the atom internal energy, and analyze the atomic wave packet motion for each internal state. This may lead one to introduce vibrational energy states of the long-range « molecular » complex, formed by the atom — in a given electronic state — interacting with its image in the wall. Such a full quantum-mechanical approach should allow one to predict whether these eventual molecular states, connected with equilibrium position of excited atoms moving parallel to the surface, might be monitored in FM-SR spectra. In the same way, symmetry-forbidden atomic transitions could get allowed in selective reflection because of symmetry breaking induced by atom-wall interaction potential. A detailed study of such processes will need one to account for the angular and parity properties of the interaction potential.





a)



b)

Fig. 6. — FM-SR lineshapes for oblique incidence ( $\Delta_0 = 2 k u \alpha / \gamma$ ): (a)  $A = 0.2$ ,  $\Delta_0 = 0, 1, 2, 5$ ; (b)  $A = 9$ ,  $\Delta_0 = 0, 2, 5, 15$ . The  $\Delta_0 = 15$  signal lineshape is enlarged by a factor 4.

Finally, this analysis could be extended in two directions :

(i) *nonlinear* atomic response, for intense incident e.m. fields (power broadening [8], dynamic Stark Splitting, saturation of the refractive index, etc...),

(ii) *optically dense* vapors, for which the refractive index change is no longer negligible. This will give access to density shifts, as well as pressure effects (collisional broadening and shifts) [21].

#### Acknowledgment.

The authors wish to thank Daniel Bloch for numerous stimulating discussions.

#### Appendix A : Van der Waals lineshape.

The FM-SR lineshape, at normal incidence, is given by the real part of (cf. Eq. (53))

$$\frac{d\bar{\chi}_0}{d\omega} = \frac{iNk\mu^2}{\varepsilon_0 \hbar} [W(0_+) + W(0_-)] I \quad (\text{A1})$$

with

$$I = \int_0^{+\infty} dz \int_0^{+\infty} dz' e^{ik(z+z')} \frac{z-z'}{\varepsilon_0(z) - \varepsilon_0(z')} . \quad (\text{A2})$$

By changing the integration variables ( $X = z + z'$  ;  $Y = z - z'$ ), one gets

$$I = \int_0^{+\infty} dX e^{ikX} \int_0^X \frac{Y dY}{\varepsilon_0\left(\frac{X+Y}{2}\right) - \varepsilon_0\left(\frac{X-Y}{2}\right)} . \quad (\text{A3})$$

For a VW interaction potential (Eqs. (55)-(57)), this yields

$$I = \int_0^{+\infty} dX e^{ikX} \int_0^X \frac{dY}{L_0 - 8iC \frac{X}{(X^2 - Y^2)^2}} . \quad (\text{A4})$$

A new variable transformation ( $s = kX$ ,  $t = kY$ ) leads, after some algebra, to

$$I = -\frac{1}{L_0} \left[ 1 - \int_0^{+\infty} ds e^{is} \int_0^s \frac{8i\nu s dt}{(s^2 - t^2)^2 L_0 - 8i\nu s} \right] \quad (\text{A5})$$

$\nu = Ck^3$  represents the Van der Waals frequency shift at atom-wall distance  $z_0 = k^{-1} = \lambda/2\pi$ . Note that the integral over  $t$  can be carried out analytically :

$$I = -\frac{1}{L_0} \left[ 1 - \int_0^{+\infty} ds \frac{8i\nu F(q) e^{is}}{s^2 L_0} \right] \quad (\text{A6})$$

with

$$q = 2 \frac{1+i}{s^{3/2}} \sqrt{\frac{\nu}{L_0}} \quad (\text{A7})$$

and

$$F(q) = \frac{1}{2q} \left( \frac{1}{\sqrt{1-q}} \tanh^{-1} \left( \frac{1}{\sqrt{1-q}} \right) - \frac{1}{\sqrt{1+q}} \tanh^{-1} \left( \frac{1}{\sqrt{1+q}} \right) \right) . \quad (\text{A8})$$

For  $A = 2 \nu/\gamma \ll 1$  (weak VW interaction),  $F(q)$  may be approximated by  $F(q) \approx i\pi/4 q$ , and

$$I \approx -\frac{1}{L_0} \left( 1 + \pi^{3/2} \sqrt{\frac{\nu}{2L_0}} \right). \quad (\text{A9})$$

In that interaction range, the SR lineshape is not yet red-shifted, but narrows ! Its linewidth is given by  $\gamma(1 - 1.9 A^{1/2})$ . Correlatively, the dispersion amplitude increases proportionally to  $1 + 4.3 A^{1/2}$ . For larger values of  $A$ , the red shift appears, first approximately given by  $-\gamma A^{0.6}$ , and the lineshape starts to broaden (see Fig. 5a).

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