

Selective reflection spectroscopy of a resonant vapor at the interface with a metallic layer

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We discuss the properties of selective reflection spectroscopy at the interface between a resonant vapor and a complex dense medium, such as a metallic film layered on a dielectric substrate. We show that in the approximation of a low-density vapor, the signal mixes up the absorptive and dispersive components of the “effective resonant susceptibility” of the vapor, with the mixture amount governed by simple laws of linear optics. Preliminary experiments performed at the interface between Cs vapor (D_2 , 852-nm line) and a silver-coated glass window are reported, that show qualitatively the effect of the atom-surface van der Waals interaction.

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I. INTRODUCTION

Reflection spectroscopy at the interface between a homogeneous, nondispersive, dense material and a resonant medium is an efficient method to probe the resonant properties of the medium in the vicinity of the interface, at distances on the order of an optical wavelength. When the resonant medium is a vapor, the “selective reflection” (SR) spectroscopy (Fig. 1), devised long ago [1], is known to be an excellent tool, exhibiting sub-Doppler resolution [2–4], notably suited for studying interatomic collisions in the high-density regime [5], when the gas medium is opaque to transmission. In the last years, interest in the SR technique has been renewed by the search for local field effects [6], collisional studies between excited states [7], and by its ability to probe atom-surface interaction sensitively [8].

In the ideal case of an interface between a transparent dielectric medium and a dilute vapor of motionless atoms, one derives, via Fresnel formulas that resonant changes in the reflection coefficient relate with the *real* part of the refractive index of the vapor (i.e., vapor dispersion) [9]. Conversely, one may extrapolate from Fresnel formulas that if the dense transparent medium (i.e., with a real index) could be replaced by an ideal metal (i.e., with a purely imaginary index), the reflection signal would yield information on the vapor absorption. However, the strong absorption of metals (infinite for an ideal metal) makes this situation nonpractical, and instead one has to consider the case when the interface includes a metallic film sandwiched between a resonant vapor and a transparent dielectric substrate. This is the topic of the present paper, in which a simple theoretical model is presented, along with a few preliminary experimental results.

Among the motivations for such a study, one may mention the following. (i) Although *dispersive* line shapes have unique advantages in the context of laser frequency stabilization [10], the detection of an absorption line shape can provide a superior spectral resolution, notably in the wings. (ii) There is a special interest in the extension of SR spectroscopy, usually limited to the probing of atomic interaction with a transparent dielectric media, to the study of atomic interaction with a real metal surface. Indeed, this interaction is generally stronger and is more straightforwardly described

in a cavity QED context (iii) It also offers richer possibilities, including the interaction of atoms with surface plasmons.

In this paper, we first establish (Sec. II) the dependence of the resonant SR signal in a general way, as a function of the optical properties of the interface. This is illustrated in Sec. III with a discussion of the respective contributions of the vapor absorptive and dispersive components to the SR signal, as a consequence of the specific properties of the metal film (index and thickness). In Sec. IV we turn to the frequency-modulated (FM) version of SR spectroscopy, in order to establish that the results of Sec. II can also be applied to the sub-Doppler singularities, due to the atomic motion, appearing in SR spectra. In Sec. V we present preliminary experimental results, performed in the FM mode of SR spectroscopy, on a Cs vapor cell with a silver-coated window. Finally (Sec. VI), we include in the model for the SR signal at a metal-vapor interface the atom-surface van der Waals attraction, in order to interpret the experimental line shapes.

II. OPTICAL PROPERTIES OF A STRATIFIED INTERFACE AND SR SPECTROSCOPY

In SR spectroscopy, one measures the reflected intensity at the (plane) interface of a plane wave $E_0(\omega)$ (circular frequency ω) incident from a dense medium to a resonant vapor [see Fig. 1(a)]. The field transmitted in the dilute medium drives a macroscopic atomic polarization, that is a source for the reradiated field, which propagates back in the dense medium along the same direction as the nonresonant reflected field [11]. Hence, the detected reflection coefficient $R(\omega)$ is given by

$$R(\omega) = |E_{\text{NR}} + E_{\text{res}}(\omega)|^2 / |E_0|^2, \quad (1)$$

with E_0 the incident field and $E_{\text{res}}(\omega)$ and E_{NR} the complex amplitudes of the resonant and nonresonant contributions to the reflection, respectively. The SR signal is the resonant change $\Delta R(\omega)$ in the reflection coefficient $R(\omega) = R_0 + \Delta R(\omega)$. The standard hypothesis of a *dilute* medium implies $|E_{\text{res}}(\omega)| \ll |E_{\text{NR}}|$ [12]. This means that the SR signal, given by

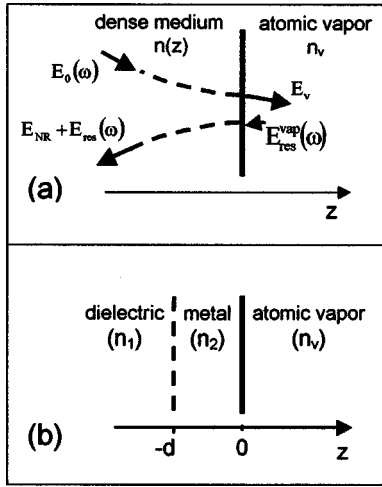


FIG. 1. (a) Principal scheme of selective reflection spectroscopy at a vapor interface; in the generalized approach, the nonresonant medium can exhibit inhomogeneities through an $n(z)$ dependence. (b) Specific case of a sandwiched metallic film.

$$\Delta R(\omega) = 2 \operatorname{Re}[(E_{\text{NR}})^* E_{\text{res}}(\omega)] / |E_0|^2, \quad (2)$$

is the homodyne beat between the nonresonant reflection and the resonantly reradiated field.

Such a description is extremely general. (For the sake of simplicity, in the following we exclude all problems related to the birefringence of the media.) It satisfactorily describes the reflection spectroscopy at a resonant vapor interface with any dense medium, that can be simple (a homogeneous transparent dielectric medium), more complex [an absorbing medium (dielectric or metallic)], or have a longitudinal inhomogeneity (stratified medium or coated window, suitable, e.g., for a plasmon excitation). In this description, E_{NR} and $E_{\text{res}}(\omega)$ are evaluated at the place of detection, i.e., at the incidence face of the structure. If the dense medium is homogeneous E_{NR} is naturally derived from the Fresnel formulas for reflection, and $E_{\text{res}}(\omega)$ is obtained from $E_{\text{res}}^{\text{vap}}(\omega)$, the reflected field on the vapor side, through Fresnel formulas in transmission. Both E_{NR} and $E_{\text{res}}(\omega)$ can be obtained through successive boundary conditions when the interface is more complex. Hence, for a given structure, one essentially has to evaluate the resonant microscopic dipole polarization, which is the source for $E_{\text{res}}^{\text{vap}}(\omega)$.

In the calculation of this microscopic polarization, the *dilute vapor* assumption (implying a low optical density) permits the major simplification that the driving field for the atomic vapor is unaffected by the resonance, and consists solely of the refracted field. Also, the refracted field in the vapor is simply a plane wave (or the evanescent wave induced by a plane wave in the dense medium) whatever the internal complexity of the dense medium may be. Hence, *on the vapor side*, the problem is completely identical to the one encountered at the interface between a vapor and a transparent dielectric material.

In spite of this simplification, a remaining difficulty, which is a major feature of SR spectroscopy, is that, in general, the vapor response to the reflected field cannot be de-

scribed by a local index model: due to the combined effects of the thermal motion and of atom-surface collisions which phase-interrupt the atom-field interaction, the vapor response is *nonlocal* in the sense that the induced polarization is not proportional to the local electromagnetic field, but integrates the whole history of the atom-field interaction [3]. Actually, within the first-order approximation in the vapor density, it has been shown that an “effective susceptibility” [13] can provide a simple description of the resonant change in the reflectivity at the interface. This effective vapor susceptibility χ ($\chi \ll 1$) is derived using the conditions of field continuity at the interface, and is given by

$$\chi = -\frac{2ik\eta}{\varepsilon_0 E_v} \int_0^{+\infty} p(z) \exp(2i\eta kz) dz, \quad (3)$$

with the related effective complex refractive index thus given by

$$n_v = 1 + \chi/2. \quad (4)$$

In Eq. (3), E_v is the driving-field amplitude in the vapor, $p(z)$ the local amplitude of the macroscopic dipole polarization induced by E_v , k is the modulus of the wave vector, and η depends on the incidence angle ($\eta = \cos \theta$, with θ the angle—*real* or *imaginary*—of the field refracted in the vapor, $\eta = 1$ for the normal incidence case). Note that in general, $p(z)$ requires both a velocity integration and a spatial integration over atomic trajectories to account for the transient effects in the atomic response. Also, $p(z)$ can include saturation [14] and the influence of surface effects [15].

In this approach with an “equivalent medium” whose response would be local, the SR spectral features result from the combination of two independent problems: (i) one has to deal with the (nonresonant) optical problem of the reflection at a complex interface, the vapor being treated as a homogeneous medium whose index is n_v (with the added peculiarity that this index depends on the incidence angle); and (ii) one has to calculate the specific resonant response of the vapor, in the same way as in usual SR spectroscopy. In addition, when the atom-surface interaction is neglected, one can show that $\operatorname{Re}(\chi)$ is simply related with the *dispersive* part of the atomic response, and $\operatorname{Im}(\chi)$ with the *absorptive* part of the response. Combining the principles of homodyne detection in SR spectroscopy [Eq. (2)] and the concept of effective refractive index, we now have the required background to determine the SR line shapes at a “dense medium” atomic vapor interface.

III. SR SPECTROSCOPY AT NORMAL INCIDENCE AT THE INTERFACE WITH A COATED WINDOW

In the following, we concentrate on a system in which the dense medium is a transparent dielectric medium (real index n_1) coated with a metallic thin film (thickness d), as shown in Fig. 1(b), and irradiated under *normal incidence*. The metal film, supposed to be isotropic and homogeneous, is described by a scalar complex index

$$n_2 = \alpha + i\beta. \quad (5)$$

The dilute vapor is described by a complex effective index

$$n_v = 1 + \delta n(\omega), \quad (6)$$

with $\delta n(\omega) = \chi/2$ and $|\chi| \ll 1$; see Eq. (4).

Replacing with Fresnel formulas, one easily finds the amplitude reflection coefficient for a two-interface system,

$$r(\omega) = -\frac{(n_v + n_2)(n_2 - n_1) + (n_v - n_2)(n_2 + n_1)e^{-\phi}}{(n_v + n_2)(n_2 + n_1) + (n_v - n_2)(n_2 - n_1)e^{-\phi}}, \quad (7)$$

with

$$\phi = -2in_2kd, \quad (8)$$

an argument that stands for the attenuation and dephasing related to the propagation inside the metal film [in Eq. (8), one has taken $k = \omega/c$]. From the first-order development on the resonant vapor contribution term $\delta n(\omega)$, one derives that the SR signal obeys a general expression

$$\Delta R(\omega) = 2R_0 \text{Re}[F \delta n(\omega)], \quad (9)$$

with F a complex value depending on the optical properties of the coated window (indices, film thickness, or more generally incidence angle), and R_0 is given by $|r(\omega)|^2$ for $n_v = 1$:

$$F = \frac{8n_1n_2^2e^{-\phi}}{(1+n_2)^2(n_2^2-n_1^2) + 2e^{-\phi}(1-n_2^2)(n_1^2+n_2^2) + e^{-2\phi}(1-n_2)^2(n_2^2-n_1^2)}. \quad (10)$$

All the features of the SR spectroscopy at the metallic-coated window can now be extrapolated from the F value. As expected, for a thick metal layer [i.e., in the limit $\exp(-\phi) \rightarrow 0$] the sensitivity to the vapor resonance, given by $\Delta R(\omega)/R_0$, decreases due to the strong metal attenuation. One can even note that for an *ideal metal* layer ($\alpha = 0$ and in the limit $\beta \gg 1$), the behavior would remain purely dispersive [$F \approx -8n_1/(n_1^2 + \beta^2)e^{-\phi}$ is real], while Fresnel formulas for a single metal-vapor interface would yield an absorption behavior (if propagation in a metallic half-space would be possible): indeed, the dielectric-metal interface introduces an extra phase factor in the reflection. For a very thin metal layer (i.e., $\phi \rightarrow 0$), the initial dispersive behavior mixes with an absorptive component. The growth of this absorptive component with the film thickness may be very rapid (like $\beta\phi$) if the metal is nearly ideal. The absorption-to-dispersion behavior for intermediate thicknesses depends on the interface used. Thus it is worthwhile to evaluate the respective contribution of $\text{Im}(\chi)$ and $\text{Re}(\chi)$ in the SR signal for various situations of practical interest. In Fig. 2, these contributions of absorption and dispersion [$\text{Im}(F)$ and $\text{Re}(F)$, respectively] have been plotted as functions of the thickness of the metal layer in two typical cases of interfaces. As a rule, for a *good metal* ($\beta \gg 1 \gg \alpha$), one observes [Fig. 2(a)] that the imaginary (absorptive) contribution starts to grow linearly with the metal thickness, as stated earlier, and that the ratio $\text{Im}(F)/\text{Re}(F)$ reaches a limit. This limit, whose value compares with unity, can be reached asymptotically or can be attained for a finite thickness [$kd \sim 0.04$ in the case of Fig. 2(a)]. In all cases, the absorptive contribution can become more or less comparable with the real (dispersive) one, but it never becomes dominant. Note that this regime, in which the metal film thickness is sufficient to profoundly alter the SR features, can be effectively reached in SR spectroscopy, yielding a non-negligible signal, because there is still light transmitted through the metallic film.

Bad metals (with $\beta, \alpha \gg 1$), which are far enough from the ideal metal behavior, can appear specifically appealing for SR spectroscopy, because the absorptive contribution can become strongly dominant. Indeed, and as exemplified in Fig. 2(b) for a typical case of a “bad metal,” one finds a pronounced *decrease* in the nonresonant reflectivity R_0 for a specific layer thickness, associated with a vanishing contribution of the dispersion [i.e., $\text{Re}(F) \rightarrow 0$; note, in any case, that the minimal value of R_0 is always nonzero, but for $n_1 = 1$]. Hence this favorable property of imperfect metals may be used in practical applications, when only the absorptive part of the vapor response is desirable. It essentially requires a sufficient accuracy in the thin-film fabrication, most probably a monitoring of the film deposition at the atomic level, because of the low values required for kd . Note that once this minimum of R_0 has been reached, an increase in the layer thickness leads to a revival of the dispersive contribution (with a sign change) that remains comparable with the absorption on a large range, while the overall signal decreases [16].

IV. SUB-DOPPLER FEATURES AND FM MODE OF SR SPECTROSCOPY

As mentioned above (Sec. 2), in the absence of long-range atom-surface interaction, $\text{Re}(\chi)$ and $\text{Im}(\chi)$ relate, respectively, to the dispersive and absorptive parts of the vapor response. However, for a Doppler-broadened vapor, the corresponding line shapes do not exhibit the usual symmetries [antisymmetry for $\text{Re}(\chi)$ and symmetry for $\text{Im}(\chi)$], because of a singularity in the velocity integration. Indeed, arriving atoms are in a steady state, and departing atoms experience a transient regime of interaction with the light field. This singularity is responsible for the well-known sub-Doppler features of SR spectroscopy, making the SR line shapes relatively complex. In the FM mode of the SR technique, when

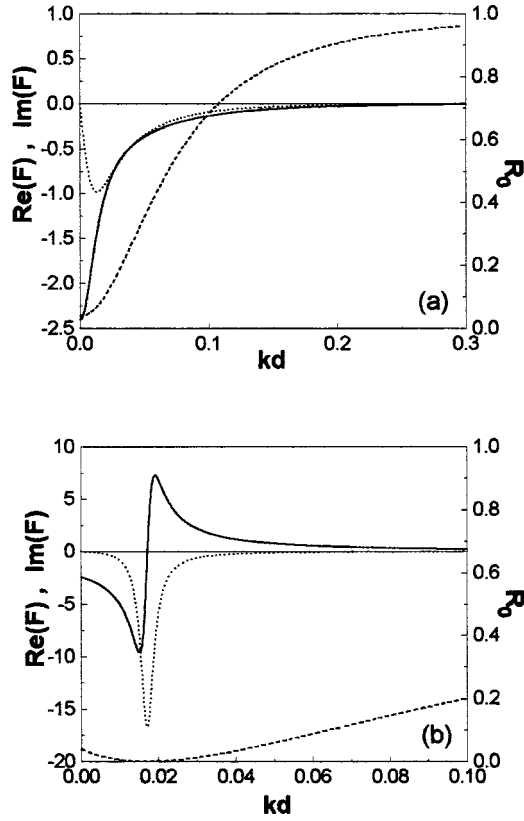


FIG. 2. $\text{Re}(F)$ and $\text{Im}(F)$ (solid and dotted lines, respectively; the vertical scale is on the left), along with the value of R_0 (dashed line; the vertical scale is on the right) as a function of the metal layer thickness (in $\lambda/2\pi$ units). Calculations are for (a) $n_1 = 1.5$ and $n_2 = 0.1 + i5.85$ (the typical case of Ag in the near IR range [19]), (b) $n_1 = 1.5$ and $n_2 = 3.69 + i3.94$ (the typical case of Fe in the near IR range [19]). The (a) and (b) curves are significant examples of a “good metal” film and a “bad metal” film, respectively, and no significant dependence has been found on the n_1 value. Note that in (b), the resonant behavior of the F value helps to discriminate between absorptive and dispersive responses, but does not correspond to a resonance in the reflection behavior, as it is due to the nearly vanishing factor R_0 .

the relevant signal is $d[\Delta R(\omega)]/d\omega$ instead of $\Delta R(\omega)$, it is known that SR spectroscopy [4,8] exhibits simpler features, notably yielding pure Doppler-free line shapes when performed at an interface with a transparent medium. In this section, we show that these results can be extended to the case of a complex interface.

Let us recall that in a two-level atom model [15], the effective susceptibility of the vapor satisfies:

$$\chi \propto N \int_0^{+\infty} dv_z \frac{W(v_z) + W(-v_z)}{(\delta + kv_z) + i(\gamma/2)}, \quad (11)$$

with N the atomic density, $W(v_z)$ the (normalized) distribution of thermal velocity along the normal to the interface, δ the frequency detuning relatively to the atomic resonance, and γ the optical width of the transition. In Eq. (11), the velocity integration is only performed on two half-Maxwellians, because of the different behavior of arriving

and departing atoms. Hence $\text{Re}(\chi)$ comprises a symmetric logarithmic singularity at line center (i.e., the singularity diverges logarithmically in the infinite Doppler width approximation) in addition to the Doppler-broadened dispersion. This is why the (FM) SR signal at a dielectric-vapor interface, governed by $d[\text{Re}(\chi)]/d\omega$, yields a pure Doppler-free dispersive Lorentzian line shape [4], with a suppressed Doppler background. In the same way, $\text{Im}(\chi)$ is not a Doppler-broadened absorption, but is actually *half a Voigt profile*, located on the red side of the transition, decreasing to zero on the resonance as abruptly as γ . As usual, frequency derivation enhances the narrow contribution, so that $d[\text{Im}(\chi)]/d\omega$ becomes a Doppler-free absorption-like line shape, symmetric with respect to the frequency detuning. Note that, until now, observation of this nonlocal absorptive response related to $\text{Im}(\chi)$ or to $d[\text{Im}(\chi)]/d\omega$ was predicted [15] only in the special context of high-frequency FM reflection spectroscopy [17].

When applying the results of Sec. III, one sees that the (FM) line shape for SR spectroscopy at the interface between a complex dense medium and a vapor is predicted to be Doppler free, but without any definite symmetry. As turning to the FM mode does not alter the absorptive-to-dispersive mixture indicated by the value of the optical parameter F , the previous discussion (in Sec. III) on the possible symmetries of the SR line shape fully applies. In particular, a purely absorptive Doppler-free line shape is expected only in the special cases mentioned above, i.e., a “bad metal” layer of the adequate thickness, while at the interface with a good metal, the Doppler-free spectrum should not exhibit any particular symmetry.

V. EXPERIMENTAL OBSERVATIONS

The present paper was triggered by a preliminary observation of SR spectra in several low-pressure Cs-vapor cells, in which one of the glass windows was coated with a thin metallic film on the internal side. The coating was obtained by a controlled deposition of evaporated Ag. The metal film thickness was on the order of 10–25 nm in order to remain in the regime of partial metallic reflection (50–90%). The experiments were performed on the D_2 resonance transition (852 nm) with a laser diode. A small modulation of the injection current was applied to generate a frequency modulation with a weak amplitude, and the (FM) SR spectra were recorded through a subsequent lock-in detection.

Typical SR spectra are presented in Figs. 3 and 4, in the case when such a coating yields a reflection coefficient $\sim 90\%$ [Fig. 3(b)] or $\sim 50\%$ (Fig. 4) along with a reference spectrum obtained at a simple vapor-dielectric interface [Fig. 3(a)], in comparable conditions (window substrate, Cs pressure, etc). The essential features described here are independent of the experimental conditions. Within the instrumental width of the diode laser (~ 30 MHz, as the diode is kept free running), all spectra appear to be Doppler-free (the Doppler width is ~ 200 MHz). The line shape of the reference spectrum is nearly dispersionlike, in spite of an observable residual asymmetry known to originate in the van der Waals atom-surface attraction [8,18]. For the partially reflecting

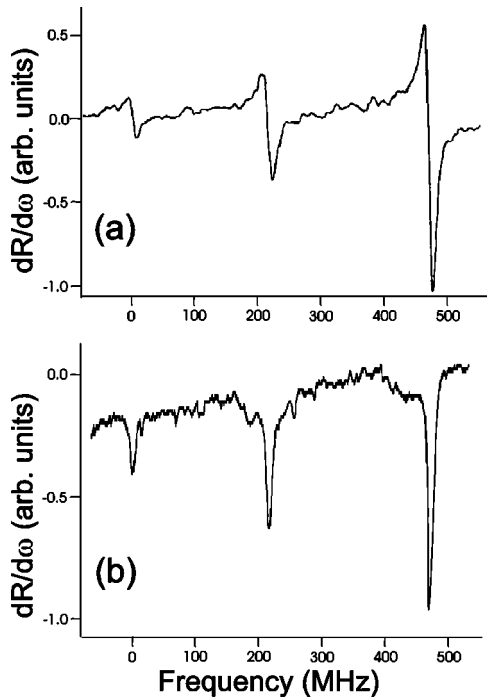


FIG. 3. Experimental comparison between (FM) SR spectroscopy at (a) a glass/Cs interface and (b) a glass/Ag/Cs-vapor interface, with an off-resonance reflection of 90%. The laser frequency is tuned across the Cs D_2 line ($F=4 \rightarrow F'=\{3,4,5\}$). The resolution is limited by the laser emission linewidth ~ 30 MHz.

($\sim 50\%$) window, no special symmetry of the line shape can be recognized. Conversely, it is striking that for the thicker coating ($R_0 \sim 90\%$), a nearly *absorptionlike spectrum* is obtained. This behavior, with nearly symmetric line shapes, is very similar to the one observed on a Cs glass cell, whose window had been accidentally coated with a metallic Cs film (due to a temporary temperature inversion between the cold point and the window), and whose reflectivity, although not perfectly homogeneous on the whole interface, was also $R_0 \sim 90\%$.

A more detailed experimental analysis has been restricted, because of the diode laser frequency resolution, and because, with our preliminary technology, the lifetime of the coatings

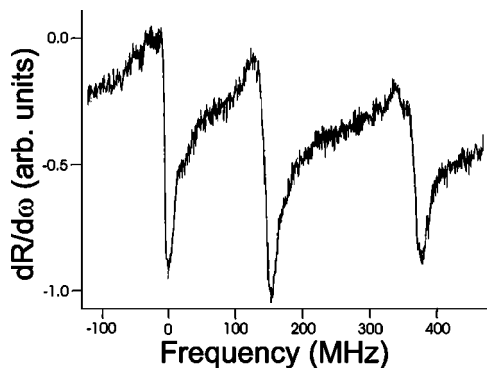


FIG. 4. FM selective reflection spectrum at a glass/Ag/Cs-vapor interface, with an off-resonance reflection of 50%. The laser is tuned across the Cs D_2 line ($F=3 \rightarrow F'=\{2,3,4\}$).

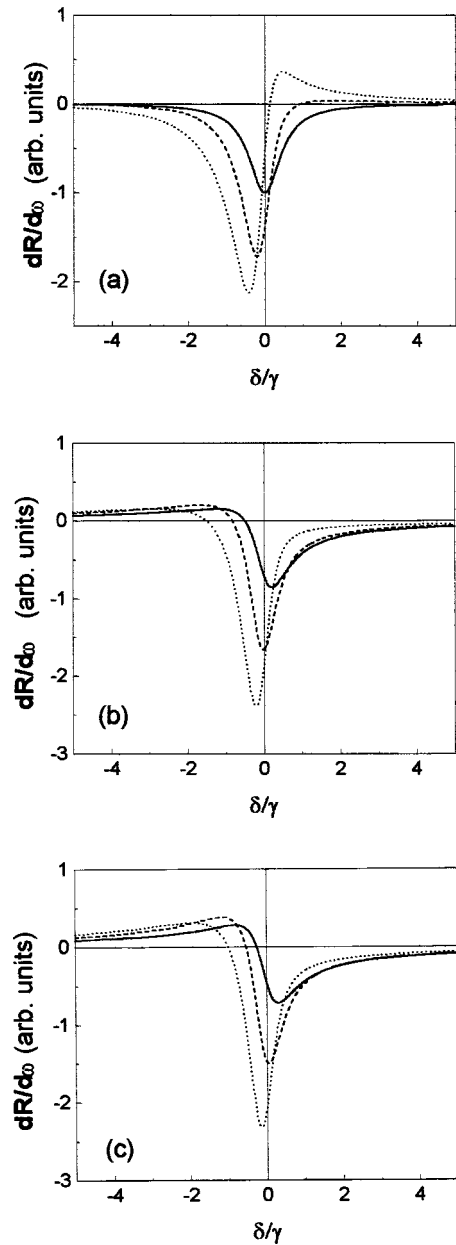


FIG. 5. Theoretical (FM) SR line shapes when the van der Waals long-range interaction is included. The dimensionless parameter $A (A=2C_3k^3/\gamma)$ characterizes the strength of the van der Waals redshift (in previous experiments on the Cs D_2 line at a dielectric interface, one found $A \leq 0.2$). $A=0$ solid (line) 0.1 (dashed line), and 0.5 (dotted line). The spectra in (a), (b), and (c) correspond to interfaces such that (a) $\text{Im}(F)/\text{Re}(F) \rightarrow \infty$ (e.g., a metal film with suppressed dispersion), (b) $\text{Im}(F)/\text{Re}(F)=1$, and (c) $\text{Im}(F)/\text{Re}(F)=0.5$, respectively. The $A=0.1$ and 0.5 line-shape amplitudes are scaled relative to the normalized $A=0$ spectrum.

becomes very limited in the presence of a heated vapor. However, it is clear that these experimental findings (for $R_0 \sim 90\%$) are not well described by the model of Sec. IV when applied to a “good metal.” Indeed, bulk Ag is a good metal in the near-infrared range (e.g., $n_2=0.1+5.85i$ according to Ref. [19]), and the film is not so thin that one should expect major deviations from bulk Ag properties [20].

VI. INFLUENCE OF THE ATOM-SURFACE INTERACTION

As already mentioned, when describing the reference spectrum (the glass-vapor interface), the long-range atom-surface interaction (van der Waals attraction) is known to distort the SR line shapes [8], through a spatially dependent shift of the atomic resonance. In the case of a simple interface between a transparent dielectric window and a vapor, the (FM) SR line shapes have already been calculated with an atom-surface interaction taken into account, and an extensive numeric evaluation of $d[\text{Re}(\chi)]/d\omega$ has been given in the case of a nonretarded van der Waals atom-surface interaction, that shifts the atomic resonance with a z^{-3} dependence [15].

In the case of a more complex interface, an evaluation of $\text{Im}(\chi)$ and $d[\text{Im}(\chi)]/d\omega$ is required along with the evaluation of $\text{Re}(\chi)$. According to the general considerations above, one may even expect, for those special windows whose coating suppresses the dispersion, to observe $\text{Im}(\chi)$ or $d[\text{Im}(\chi)]/d\omega$ directly. Figure 5(a) provides the line shapes of $d[\text{Im}(\chi)]/d\omega$ in a range of perturbative van der Waals attraction (i.e., a van der Waals shift smaller than the optical width at $\lambda/2\pi$) typical of an alkali resonance line [8], and Figs. 5(b) and 5(c) exemplify the expected SR line shapes for various other interfaces. A remarkable point is that the asymmetric behavior induced by the presence of the metal film combines with the asymmetry in the van der Waals-distorted resonance. This induces the possibility of accidentally yielding a nearly symmetric, absorptionlike, Doppler-free reflection line shape, as predicted for a relatively wide range of parameters (film refractive index n_2 , strength of the van der Waals interaction, etc). Note that these nearly symmetric line shapes are predicted even when the $\text{Im}(\chi)$ contribution is not at all dominant relative to $\text{Re}(\chi)$. These general predictions seem sufficient to interpret, in a qualitative manner, the experimental results obtained with highly reflecting metal films (Sec. V). A more quantitative analysis is limited by the inhomogeneities of the metallic film coating (possibly leading to an averaging of various line shapes), and by the possible depen-

dence of the effective strength of the van der Waals interaction with the coating thickness.

VII. CONCLUSION

In conclusion, we have shown that for SR spectroscopy at a vapor interface, replacing the usual transparent dielectric window with some kind of a coated window permits one to explore a variety of line shapes, due to the addition of a variable contribution of the imaginary part $\text{Im}(\chi)$ of the “effective susceptibility” of the vapor. In our preliminary experiments at the interface with a “good metal” film, we have obtained nearly symmetric line shapes, that are unexpected in the absence of a surface interaction. This shows that the atom-surface interaction, usually responsible for perturbations on the line shapes, has dramatic effects on the line shapes, and imposes stronger modifications than when the window is made of a pure dielectric medium. One may infer that the sensitivity to the atom-surface interaction is even larger. In the experiments reported here, the oversimplified technology to coat the window, and the low spectral resolution, have prevented us from quantitatively estimating the strength of the van der Waals attraction, and to compare it with the predictions for a *metallic* surface. In future developments, it seems technologically conceivable to prepare hard-coated windows which would yield a nearly null dispersive response of the vapor. More generally, the FM version of SR spectroscopy should allow one to measure the atom-surface van der Waals interaction for an atom in the vicinity of surfaces of a particular interest, for example metal (thin film), structured layer waveguide, etc. The accuracy should be as good as the one currently obtained at the interface with a transparent medium, once a single off-resonant measurement on the window has provided the $\text{Re}(F)/\text{Im}(F)$ value.

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 - [11] Note that inside the vapor, this additional reflected field exhibits fast variations of its envelope. Indeed, in the direction of the reflected field, the coherence length of the atomic dipole polarization is shorter than an optical wavelength.

- [12] Occasionally, one may have $E_{NR}=0$ for specially designed systems. Here we do not consider this problem of SR spectroscopy with an “antireflecting window.” The special case of a Brewster incidence was addressed in A. M. Akul’shin, V. L. Velichanskii, A. I. Zherdev, A. S. Zibrov, V. I. Malakhova, V. V. Nikitin, V. A. Sautenkov, and H. G. Kharisov, *Kvant. Elektron. (Moscow)* **16**, 631 (1989) [*Sov. J. Quantum Electron.* **19**, 416 (1989)].
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- [14] G. Nienhuis, F. Schuller, and M. Ducloy, *Phys. Rev. A* **38**, 5197 (1988).
- [15] M. Ducloy and M. Fichet, *J. Phys. II* **1**, 1429 (1991).
- [16] One actually calculates several thicknesses for which $\text{Re}(F)$ = 0, but the attenuation of the signal is considerable, due to the large absorption over propagation in the metal film.
- [17] A polarization-sensitive detection may also provide a contribution of the absorptive response (see Ref. [14]). In any case, such a scheme requires a nonzero incidence angle, hence implying a broadening of the sub-Doppler contribution.
- [18] In Fig. 3(a), the distortion looks to be a particularly weak effect, because of a line shape broadening due to the instrumental resolution.
- [19] *American Institute of Physics Handbook*, 3rd ed. (McGraw-Hill, New York, 1972).
- [20] For a very thin film, the usual bulk values for n_2 are not necessarily reliable [see, e.g., F. A. Pudonin, R. Villagomez, and O. Keller, *Opt. Commun.* **170**, 181 (1999)], and a direct measurement of the film properties may be required.