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Selective reflection combined with Fabry–Perot effects from two-level atoms confined between two dielectric walls*

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The coefficient of selective reflection at oblique incidence from two-level atoms confined between two dielectric walls is calculated in this paper. It is found to be related to the transient behaviour of atoms after colliding with the wall and the distribution of the field inside the vapour corresponds to L/λ, with L the thickness of the film and λ the incident wavelength. We find that the sub-Doppler structure is manifest both for normal incidence and small angle oblique incidence. It is feasible to detect the real part of selective reflection in several cases that have not been achieved before.

Keywords: selective reflection, confined atomic vapour film, sub-Doppler structure
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1. Introduction

Selective reflection (SR) spectroscopy is known as an excellent tool to detect the resonant properties of the medium in the vicinity of the interface.[1−11] It is notably beneficial to obtain high-resolution spectroscopy in order to study atom–surface interaction,[3,7−11] dilute and high-density gas atomic medium[2−4,10,11] sensitively. The general solution of SR spectroscopy is obtained in a two level system at normal incidence, for very thin vapour cells, and especially the cell thickness is smaller than the optical wavelength or comparable with it, the measured reflection are strongly dependent on the intrinsic Fabry–Perot (FP) nature of the thin cell, sub-Doppler features are obtained both in theory and experiments.[10,11]

It is an approximation to say that the light in experiment is under normal incidence, especially when the vapour film is very thin. Theory for the cases of oblique incidence has been discussed before[1] and can be used in a macroscopic vapour film ($L \to \infty$). However, when the vapour film is very thin, more cases can be treated as the cases in which the field is under oblique incidence; meanwhile the FP effects cannot be eliminated totally though the second window is antireflection coated in experiment.[10] This requires a more general theory to calculate the SR from a very thin atomic vapour film. In this paper, we will discuss some peculiarities of SR from a very thin atomic vapour film combined with FP effects in the situation of oblique incidence, which gives a recipe for studying ultrathin atomic vapour film SR spectroscopy.

2. Basic theory in a two level system

At normal incidence, the problem which we consider is a pure one dimensional problem in preceding work. For oblique incidence (see Fig.1), we will treat the problem as a two-dimensional problem in which the diffraction effects are neglected. An ultrathin atomic layer (medium) is sandwiched between two dielectric windows assumed to be transparent (i.e., no absorptive or scattering losses) whose refractive indices are n_1 for $z < 0$ and n_2 for $z > L$. The atomic medium is excited under oblique incidence by an inci-

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dent field $E_{\rm in}(x,z,t)$ with a small incident angle θ in the first window:

$$E_{\rm in}(x,z,t) = \frac{1}{2}E_{\rm in}\exp[-{\rm i}(\omega t - k_x n_1 x - k_z n_1 z)] + {\rm c.c.} \quad (1)$$

Here, ω is the circular frequency and k_x, k_z are the components of wave vector k respectively. The amplitude of incident field $E_{\rm in}$ is split into a reflected component $E_{\rm r}^{(0)}$ in the dielectric and a refracted component $E_{\rm of}^{(0)}$ inside the vapour at the lower surface, $E_{\rm of}^{(j)}(j=1,2,3,\cdots,\infty)$, $E_{\rm ob}^{(j)}$, $E_{\rm t}^{(j)}(j=0,1,2,3,\cdots,\infty)$ inside the vapour and $E_{\rm r}^{(j)}(j=1,2,3,\cdots,\infty)$ inside the dielectric represent the amplitudes of waves reflected at the lower surface, reflected at the upper surface, refracted at the upper surface and refracted at the lower surface respectively, then we have[12]

$$E_{\rm of}^{(j)}(x,z,t) = \frac{1}{2}E_{\rm of}^{(j)}(x,z)\exp[-{\rm i}(\omega t - k_x x - k_z z)] + {\rm c.c.}, \quad (2)$$

$$E_{\rm ob}^{(j)}(x,z,t) = \frac{1}{2}E_{\rm ob}^{(j)}(x,z)\exp[-{\rm i}(\omega t - k_x x - k_z z] + {\rm c.c.}, \quad (3)$$

$$E_{\rm t}^{(j)}(x,z,t) = \frac{1}{2}E_{\rm t}^{(j)}(x,z)\exp[-{\rm i}(\omega t - k_x n_2 x - k_z n_2 z] + {\rm c.c.}, \quad (4)$$

$$E_{\rm r}^{(j)}(x,z,t) = \frac{1}{2}E_{\rm r}^{(j)}(x,z)\exp[-{\rm i}(\omega t - k_x n_1 x - k_z n_1 z] + {\rm c.c.} \quad (5)$$

where there is a phase difference ϕ between $E_{\rm of}^{(j)}$ and $E_{\rm of}^{(j+1)}$, $E_{\rm ob}^{(j)}$ and $E_{\rm ob}^{(j+1)}$, $E_{\rm t}^{(j)}$ and $E_{\rm t}^{(j+1)}$, $E_{\rm r}^{(j)}$ and $E_{\rm r}^{(j+1)}$ respectively, with $\phi = 2k_z L\dfrac{k_z}{k}$ and $j = 0,1,2,3,\cdots,\infty$.

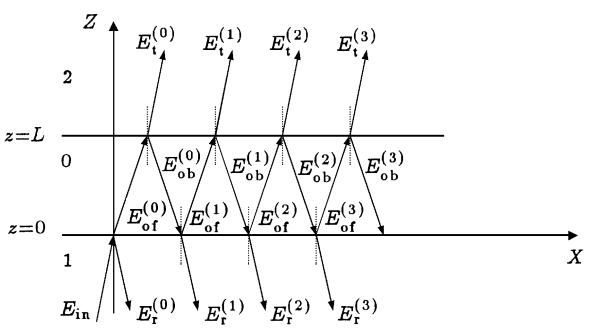


Fig.1. Ultrathin atomic layer film (0-L) between two dielectric windows.

The fields in the gas drive the active atoms, thereby creating a dipole polarization. This polarization emits radiation in the dielectric of the reflected wave, and modifies the reflection coefficient.[1] The purpose of this paper is to calculate the reflection fields consisting of two parts: the total fields $E_{\rm rv}$ from internal reflection when the half space $z>0$ is vacuum and the resonant contribution $E_{\rm rp}$ due to the dipole polarization from the vapour for two cases where the incident field and the polarization are normal to the plane of incidence (S polarization), or in this plane (P polarization).

When the incident field and the dipole polarization are directed normal to the plane of incidence, the same is true for reflected fields and refracted fields, at the $z=0$ boundary

$$E_{\rm ob}^{(j)}(0) + E_{\rm of}^{(j+1)}(0) = E_{\rm rv}^{(j+1)}(0), \quad (6a)$$

$$[E_{\rm ob}^{(j)}(0) - E_{\rm of}^{(j+1)}(0)]\frac{k_z}{k} = n_1 E_{\rm rv}^{(j+1)}(0)\cos\theta, \quad (6b)$$

and for the incident field $E_{\rm in}$, we have

$$E_{\rm in} + E_{\rm rv}^{(0)}(0) = E_{\rm of}^{(0)}(0), \quad (6c)$$

$$n_1[E_{\rm in} - E_{\rm rv}^{(0)}(0)]\cos\theta = E_{\rm of}^{(0)}(0)\cdot\frac{k_z}{k}. \quad (6d)$$

At the $z=L$ boundary

$$E_{\rm of}^{(j)}(L) + E_{\rm ob}^{(j)}(L) = E_{\rm t}^{(j)}, \quad (6e)$$

$$[E_{\rm of}^{(j)}(L) - E_{\rm ob}^{(j)}(L)]\cdot\frac{k_z}{k} = n_2 E_{\rm t}^{(j)}\cos\theta, \quad (6f)$$

where, $j = 0,1,2,3,\cdots,\infty$.

From Eqs.(6a)–(6f), one gets

$$E_{\rm of}^{(0)}(0) = t_{10}E_{\rm in}, \quad (7a)$$

$$E_{\rm rv}^{(0)}(0) = -r_1 E_{\rm in}, \quad (7b)$$

$$E_{\rm ob}^{(j)}(L) = r_2 E_{\rm of}^{(j)}(L), \quad (7c)$$

$$E_{\rm of}^{(j+1)}(0) = r_1 E_{\rm ob}^{(j)}(0), \quad (7d)$$

$$E_{\rm rv}^{(j+1)}(0) = t_{01}E_{\rm ob}^{(j)}(0) \quad (7e)$$

with

$$r_1 = \frac{k_z - n_1 k\cos\theta}{k_z + n_1 k\cos\theta}, \quad r_2 = \frac{k_z - n_2 k\cos\theta}{k_z + n_2 k\cos\theta},$$

$$t_{10} = \frac{2n_1 k\cos\theta}{n_1 k\cos\theta + k_z}, \quad t_{01} = \frac{2k_z}{n_1 k\cos\theta + k_z}.$$

Inside a very dilute medium, the absorption is very weak; suppose the incident laser beam diameter largely exceeds the film thickness, and consider the effects of interference, the total amplitude of the reflected fields $E_{\rm rv}(z=0)$ from internal reflection for the incident field normal to the incident plane can be written as

$$\begin{aligned} E_{\rm rv}(0) =& E_{\rm in}\left[-r_1 + t_{01}t_{10}r_2\sum_{j=0}^{\infty}(r_1 r_2)^j \exp(-ij\phi)\right] \\ =& \left(-r_1 + \frac{t_{01}t_{10}r_2}{F}\right)E_{\rm in}, \end{aligned} \tag{8}$$

where $F = 1 - r_1 r_2 \exp(-{\rm i}\phi)$.

Equations (7) and (8) are valid too for the incident field, the polarization are in the plane of incidence, in this case we use notation

$$r_1 = \frac{k\cos\theta - n_1 k_z}{k\cos\theta + n_1 k_z}, \quad r_2 = \frac{k\cos\theta - n_2 k_z}{k\cos\theta + n_2 k_z},$$

$$t_{10} = \frac{2n_1 k\cos\theta}{n_1 k_z + k\cos\theta} \quad \text{and} \quad t_{01} = \frac{2k_z}{n_1 k_z + k\cos\theta}.$$

To calculate the resonant contribution $E_{\rm rp}$ due to the dipole polarization, we consider the total field $E_{\rm o}(x,z,t)$ driving polarization at any position z inside the vapour consists of two parts: the total forward fields $E_{\rm of}(x,z,t)$ and the total backward fields $E_{\rm ob}(x,z,t)$. One gets the total amplitude of the forward fields $E_{\rm of}(z)$ and $E_{\rm ob}(z)$ of the backward fields inside the vapour as follows:

$$E_{\rm of}(z) = t_{10}E_{\rm in}\sum_{j=0}^{\infty}(r_1 r_2)^j \exp(-ij\phi) = \frac{t_{10}}{F}E_{\rm in}, \tag{9a}$$

$$\begin{aligned} E_{\rm ob}(z) =& t_{10}r_2 E_{\rm in}\exp[2ik_z(L-z)] \\ & \times \sum_{j=0}^{\infty}(r_1 r_2)^j \exp(-{\rm i}j\phi) \\ =& \frac{t_{10}r_2\exp[2{\rm i}k_z(L-z)]}{F}E_{\rm in}. \end{aligned} \tag{9b}$$

We then obtain the amplitude of the total field inside the vapour as

$$\begin{aligned} E_{\rm o}(z) =& E_{\rm of}(z) + E_{\rm ob}(z) \\ =& \{1 + r_2\exp[2{\rm i}k_z(L-z)]\}\frac{t_{10}}{F}E_{\rm in}. \end{aligned} \tag{10}$$

Thus the polarization inside the vapour can be defined as

$$P_{\rm o}(x,z,t) = P_{\rm of}(x,z,t) + P_{\rm ob}(x,z,t), \tag{11a}$$

where

$$P_{\rm of}(x,z,t) = \frac{1}{2}P_{\rm of}(z)\exp[-{\rm i}(\omega t - k_x x - k_z z)] + {\rm c.c.}, \tag{11b}$$

$$\begin{aligned} P_{\rm ob}(x,z,t) =& \frac{1}{2}P_{\rm ob}(z)\exp[-{\rm i}(\omega t - k_x x - k_z z)] + {\rm c.c.} \\ =& \frac{1}{2}P_{\rm of}(z)r_2\exp[2{\rm i}k_z(L-z)] \\ & \times \exp[-{\rm i}(\omega t - k_x x - k_z z)] + {\rm c.c.}, \end{aligned} \tag{11c}$$

where $P_{\rm of}(z)$ and $P_{\rm ob}(z)$ are driven by the forward fields amplitudes and backward fields amplitudes respectively.

For the vector potential A in the Lorentz gauge, the wave equation is

$$(\nabla^2 + k^2)A = {\rm i}\omega\mu_0 P, \tag{12}$$

where μ_0 is the vacuum permeability. We then obtain the wave emitted by the dipole polarization, and incident on the surface[1]

$$E_{\rm rp}(x,z) = \frac{1}{2\varepsilon_0 k_z}\exp[{\rm i}(k_x x - k_z z)]\int_0^L {\rm d}z_0 \left(k^2 - (k_x\ k_z)\begin{pmatrix} k_x \\ k_z \end{pmatrix}\right) P(z_0). \tag{13}$$

According to Eq.(13) and laws of Fresnel and Snellius, we then obtain the resonant contribution $E_{{\rm rp}\perp}$ and $E_{{\rm rp}//}$ for the polarization normal to the plane of incidence and in this plane respectively as follows[1]

$$E_{{\rm rp}\perp} = \frac{S}{n\cos\theta + k_z/k}, \tag{14a}$$

$$E_{{\rm rp}//} = \frac{2(k_z/k)^2 - 1}{n_1 k_z/k + \cos\theta}S, \tag{14b}$$

where

$$S = {\rm i}\frac{k}{\varepsilon_0}\int_0^L {\rm d}z\exp({\rm i}k_z z)P_{\rm o}(z), \tag{15}$$

ε_0 is the vacuum permittivity.

In a two-level system with a ground state $|g\rangle$ and an excited state $|e\rangle$, when the Rabi frequency Ω (assuming that the absorption is very weak inside the vapour, and $E_{\rm o}(0) \approx E_{\rm o}(z)$, then we define

$\Omega = \dfrac{\mu_{\rm eg}E_{\rm o}(0)}{\hbar}$, where $\mu_{\rm eg}$ is the dipole transition element) is much less than the dipole relaxation rate $\gamma_{\rm eg}$, the polarization $P_{\rm o}(z)$ can be written as

$$\begin{aligned} P_{\rm o}(z) =& N\mu_{\rm eg}\int_{-\infty}^{+\infty}{\rm d}v_x\int_{-\infty}^{+\infty}{\rm d}v_z W(v_x)W(v_z) \\ & \times[\Theta(v_z)\sigma_{\rm eg}(v,v_z>0) \\ & +\Theta(-v_z)\sigma_{\rm eg}(v,v_z<0)], \end{aligned} \tag{16}$$

where $\Theta(v_z)$ and $\Theta(-v_z)$ are the Heaviside functions which ensure the separation of contributions from atoms with $v_z>0$ and $v_z<0$ respectively, N is the atomic density, $W(v_x)$ and $W(v_z)$ are Maxwell–Boltzmann velocity distribution functions along x and z respectively: i.e. $W(v_i)=\dfrac{1}{u\sqrt{\pi}}\exp\left[-\left(\dfrac{v_i}{u}\right)^2\right]$ $(i=x,z)$, where $u=\sqrt{\dfrac{2k_{\rm B}T}{m}}$ is the probable velocity of atoms in the vapour film, $k_{\rm B}$ is the Boltzmann constant, m is the atomic mass and T is the temperature of the vapour, $\sigma_{\rm eg}(v)$ is the off-diagonal (raising) part of the density matrix element. When the intensity of the light is not strong enough to saturate the resonant transition, it satisfies the equation

$$v\nabla\sigma_{\rm eg}+\Lambda\sigma_{\rm eg}={\rm i}\frac{\mu_{\rm eg}}{\hbar}E_{\rm o}(z), \tag{17}$$

where

$$E_{\rm o}(z)=E_{\rm o}(0)\exp({\rm i}k_z z), \tag{18}$$

$\Lambda=\gamma_{\rm eg}-{\rm i}[(\omega-\omega_0)-k_xv_x-k_zv_z]$, γ is the homogeneous linewidth, ω_0 is the transition frequency from $|g\rangle$ to $|e\rangle$.

It is justified to assume that atoms get de-excited at a collision with the wall, and are in the ground states at the instant that they leave the wall after a collision. Thus, we have the boundary condition

$$\sigma_{\rm eg}(z=0,v_z>0)=\sigma_{\rm eg}(z=L,v_z<0)=0. \tag{19}$$

Combined Eqs.(17–19), one finds

$$\sigma_{\rm eg}(z,v_z>0)={\rm i}\frac{\mu_{\rm eg}E_0(0)}{\hbar}\frac{1}{\Lambda+{\rm i}k_zv_z}\left(\exp({\rm i}k_zz)-\exp\left(-\Lambda\frac{z}{v_z}\right)\right), \tag{20a}$$

$$\sigma_{\rm eg}(z,v_z<0)={\rm i}\frac{\mu_{\rm eg}E_0(0)}{\hbar}\frac{1}{\Lambda+{\rm i}k_zv_z}\left(\exp({\rm i}k_zz)-\exp\left({\rm i}k_zL-\Lambda\frac{z-L}{v_z}\right)\right). \tag{20b}$$

Substituting Eqs.(20a–20b) to Eq.(16), then to Eq.(15), and changing the integration orders of v and z, one obtains

$$S=-N\mu_{\rm eg}^2E_{\rm o}(0)\frac{k}{\varepsilon_0}\int_{-\infty}^{\infty}{\rm d}v_x\int_{-\infty}^{\infty}{\rm d}v_z\frac{1}{\Lambda+{\rm i}k_zv_z}W(v_x)W(v_z)[\Theta(v_z)F_1+\Theta(-v_z)F_2], \tag{21a}$$

where

$$F_1=\frac{1}{2{\rm i}k_z}[\exp(2{\rm i}k_zL)-1]+\frac{v_z}{{\rm i}k_zv_z-\Lambda}\left[1-\exp\left({\rm i}k_zL-\Lambda\frac{L}{v_z}\right)\right], \tag{21b}$$

$$F_2=\frac{1}{2{\rm i}k_z}[\exp(2{\rm i}k_zL)-1]+\frac{v_z}{{\rm i}k_zv_z-\Lambda}\left[\exp\left({\rm i}k_zL+\Lambda\frac{L}{v_z}\right)-\exp(2{\rm i}k_zL)\right]. \tag{21c}$$

For convenience, we introduce a quantity T which governs the SR (see, e.g., Refs.[1–6])

$$T=S/E_{\rm o}(0)=-\frac{N\mu_{\rm eg}^2k}{\varepsilon_0}\int_{-\infty}^{\infty}{\rm d}v_x\int_{-\infty}^{\infty}{\rm d}v_z\frac{1}{\Lambda+{\rm i}k_zv_z}W(v_x)W(v_z)[\Theta(v_z)F_1+\Theta(-v_z)F_2]. \tag{22}$$

Thus, we obtain the reflection coefficients $r_\perp$ and $r_{//}$ from Eqs.(8), (14a–14b), (10) and (22) for a polarization normal to the plane of incidence and in this plane respectively:

$$r_\perp=\left(-r_1+\frac{t_{01}t_{10}r_2}{F}\right)+\frac{t_{10}[1+r_2\exp(-2{\rm i}k_zL)]}{F(n\cos\theta+k_z/k)}T, \tag{23a}$$

$$r_{//}=\left(-r_1+\frac{t_{01}t_{10}r_2}{F}\right)+\frac{t_{10}[1+r_2\exp(-2{\rm i}k_zL)][2(k_z/k)^2-1]}{F(nk_z/k+\cos\theta)}T. \tag{23b}$$

The first terms on the right-hand sides of Eqs.(23a–23b) represent the nonselective reflection contribution and the second terms the selective reflection contribution. It is notable that SR is strongly modified by the FP nature, the modulation factors $\exp(-2ik_zL)$ and $\exp(-ik_zL)$ indicate that the line shape and amplitudes have some periodic peculiarities,[6,10,11] correspond to the thickness of the vapour film. In Section 3, we will discuss these peculiarities which have not been achieved before in different thickness region.

3. Numerical results and conclusion

To study the line profile of SR, we will solely consider the real part of the second terms $\mathrm{Re}(E_{\mathrm{rp}\perp})/E_{\mathrm{in}}$ and $\mathrm{Re}(E_{\mathrm{rp}//})/E_{\mathrm{in}}$ on the right-hand in Eqs.(23a–23b).[1,6] When the incident and the selected polarization are linear, then numerical results (intensity in arbitrary units) are shown in Fig.2 and Fig.3 at normal incidence and at oblique incidence ($\theta = 10°$) respectively for (a) $L/\lambda = 1/16$, (b) $L/\lambda = 1/8$, (c) $L/\lambda = 1/4$, (d) $L/\lambda = 1/2$, with $S_{\mathrm{SR}}(i)$ the selective reflection signal, where i=S for polarization normal to the plane of incidence (solid lines) and i=P for polarization in this plane (hollow circles). The selected values are $ku = 50\gamma_{\mathrm{eg}}$ and $n = n_1 = n_2 = 1.82$.

Lines in Fig.2 represent the cases without spatial dispersion where as Fig.3 the cases with spatial dispersion. It is notable that, in the case of S polarization with spatial dispersion, SR is a little bit stronger than it is in the case of P polarization, while line shapes are approximately the same for the case without spatial dispersion. It corresponds to the behaviour of atoms induced by the different or similar distribution of the total field inside the vapour. For convenience, we consider line shapes in three groups: the first group with $L = 2m\lambda/4$ (m is integer), the second group with $L = (2m+1)\lambda/4$ and the third group with $L < \lambda/4$. The first and the third group exhibit dispersion-like shapes and the second group mixes with absorption-dispersion-like shapes. Because the path length contribution, the absorption is weaker inside the vapour at normal incidence than it is at oblique incidence.

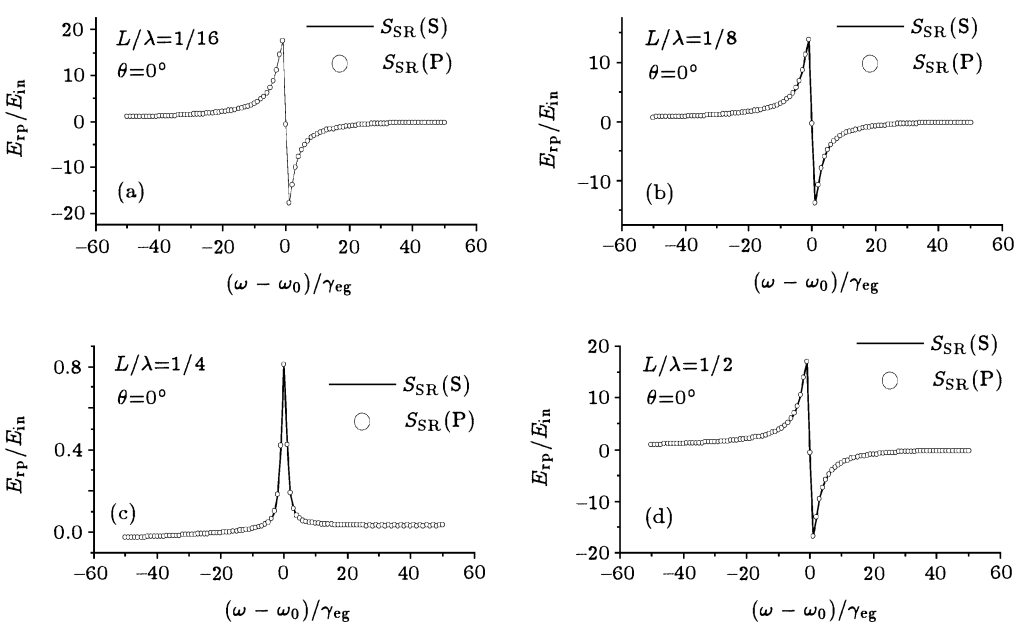


Fig.2. SR at normal incidence in a two level system for (a) $L/\lambda = 1/16$, (b) $L/\lambda = 1/8$, (c) $L/\lambda = 1/4$, (d) $L/\lambda = 1/2$.

In all cases shown in Fig.2 and Fig.3, the sub-Doppler structure is manifest when FP effects are considered, this is different from the transmission situation in which line shapes do not exhibit sub-Doppler structure in some region of L/λ even at small angle oblique incidence (e.g., $\theta = 5°$).[12]. The main reason is that the reflection is mainly sensitive to the thin layer near the interface, and atom-wall collision can give rise to sub-Doppler structure due to the transient behaviour of atoms leaving the wall just after a collision, say, atoms travelling against the incident direction are not in the steady state, they remain in the

ground state right after leaving the back surface, and there is simply not enough time for them to adopt the external fields. Thus atoms moving in opposite direction with the same speed contribute the same amount to the signal just as in the case of a thick film.[6] One notices also that the absorption dip in the reflection is broadened by spatial dispersion, one contribution is from an additional Lorentzian broadening which may be viewed as transit-time broadening induced by spatial dispersion effects, and another is from the component of the wave vector along the surface which is larger than that in the propagation case, and enhances Doppler broadening.[1]

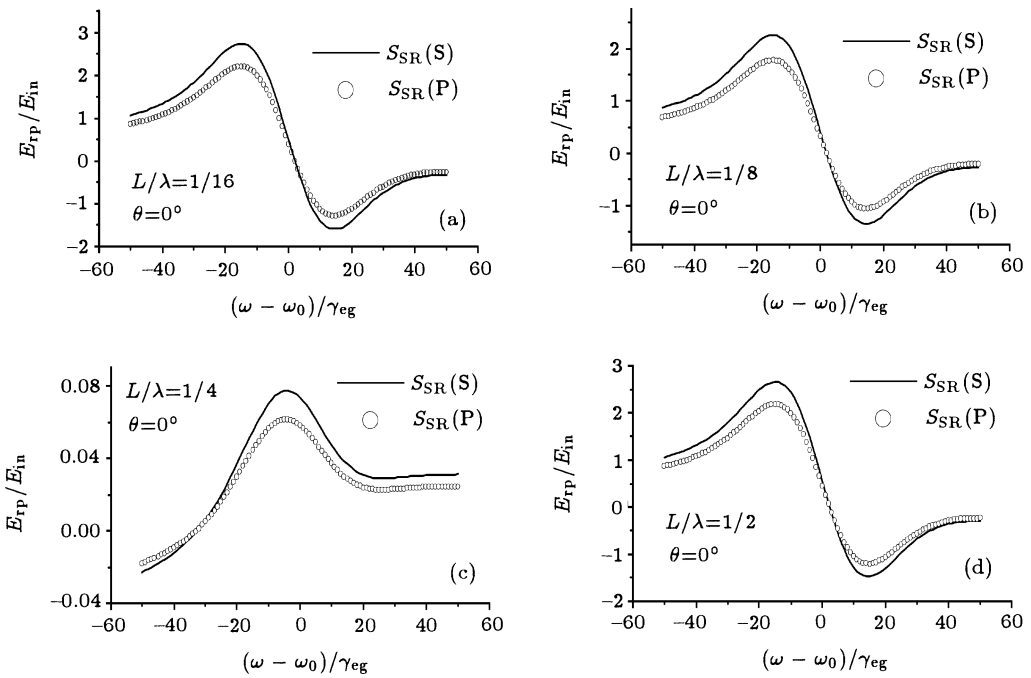


Fig.3. SR line shapes at oblique incidence ($\theta = 10°$) in a two level system for (a) $L/\lambda = 1/16$, (b) $L/\lambda = 1/8$, (c) $L/\lambda = 1/4$, (d) $L/\lambda = 1/2$.

One interesting point as shown in Fig.2 and Fig.3 is that, in a wide range of L/λ, the sharp narrow line shapes, from resonant atoms with slow velocity and the effects of wave selection of FP, are thought to be impossible to get in experiments in preceding work which is possible to be achieved in many cases.

In conclusion, we calculated the coefficient of SR from atoms confined between two dielectric walls for two cases of S polarization and P polarization, it corresponds to the transient behaviour of atoms after colliding with the wall and the distribution of the field inside the vapour depends on L/λ. The results are expressed in Eqs.(23a) and (23b) for S polarization and P polarization respectively. Numerical calculations are presented in Fig.2 and Fig.3 in several cases. We would like to point out that, in previous discussions, the theory is not suitable for interpreting the experiments in a very thin vapour film spectroscopy when the laser is under oblique incidence. In our calculation, the vapour inside the film is supposed to be very dilute, and the absorption inside the vapour is very weak, thus the field distribution which governs the polarization inside the film is obtained by interference theory. With different boundary conditions of density matrix elements at $z = 0$ for $v_z > 0$ and $z = L$ for $v_z < 0$ used in Ref.[6], we then get a general theory combined with FP effects which has not been obtained before in a very thin vapour film for the case of oblique incidence, and this gives a recipe for studying ultrathin atomic vapour film SR spectroscopy. We find that the sub-Doppler structure is manifest both for normal incidence and small angle oblique incidence. Indeed, in experiment, the SR spectroscopy from ultrathin atomic vapour film does not exactly satisfy the condition of normal incidence, it can be found from our calculation that, though with a robust control of incident angle, the sub-Doppler SR spectroscopy can still be achieved in several cases.

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