Ultrathin atomic vapor film transmission spectroscopy: analysis of Dicke narrowing structure

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Transmission sub-Doppler spectroscopy with confined atomic vapor film between two dielectric walls is theoretically studied. Because of atoms flying from wall to wall, where they get de-excited, the atom-field interaction time is anisotropic so that the contribution of slow atoms is enhanced, a sub-Doppler transmission spectroscopy (Dicke narrowing effect) can be obtained when the thickness of the film is much small or comparable with the wavelength even at small angle oblique incidence. It is feasible to get a sub-Doppler structure in a new region ($L < \lambda/4$) in experiments.

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Since the early days of cavity quantum electrodynamics (cavity QED), it has been known that, inside a cavity or waveguide, the spectrum of the electromagnetic fields modes is strongly modified for wavelengths that are comparable with or larger than the physical dimensions[1]. Both in microwave and optical domain, sub-Doppler spectroscopy related to thin atomic vapor film has been reported in several articles[2−11]. In this letter, a more general theory about transmission spectra of atomic thin vapor film (the thickness is comparable with or much less than the wavelength) at oblique incidence is investigated, especially when the ratio of L/λ is less than 1/4.

Figure 1 shows that 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 an incident field $E_{\rm in}$ with a small incident angle θ in the first window

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

where 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 $E_{\rm r}^{(0)}$

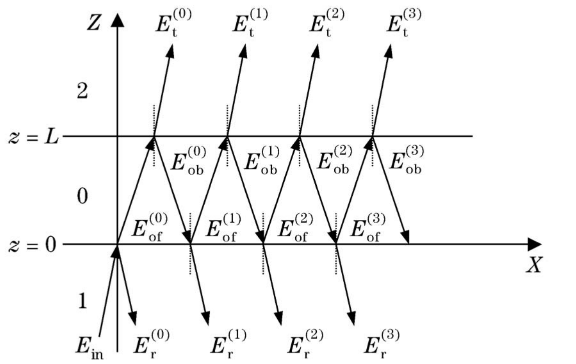

Fig. 1. Ultrathin atomic layer film $(0 - L)$ between two dielectric windows.

in the dielectric and a refracted component $E_{\rm of}^{(0)}$ inside the vapor at the lower surface, $E_{\rm of}^{(j)}$ $(j = 1,2,3,\cdots,\infty)$, $E_{\rm ob}^{(j)}$ and $E_{\rm t}^{(j)}$ $(j = 0,1,2,3,\cdots,\infty)$ represent the amplitudes of waves reflected at the lower surface, of waves reflected at the upper surface, and of waves refracted at the upper surface respectively, then we write

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

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

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

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

In a very dilute medium, assuming that the incident laser beam diameter largely exceeds the film thickness, we neglect the resonant part fields contribution, considering the effects of interference, the field $E_0(x,z,t)$ driving polarization at any position z inside the vapor consists of two parts: the total forward field $E_{\rm of}(x,z,t)$ and the total backward fields $E_{\rm ob}(x,z,t)$. Thus, the amplitude of the total field $E_0(z)$ inside the vapor can be expressed as

$$E_0(z) = E_{\rm of}(z) + E_{\rm ob}(z) = \{1 + r_2\exp[2ik_z(L-z)]\}\frac{t_{10}}{F}E_{\rm in}, \tag{5}$$

with $F = 1 - r_1 r_2\exp(-i\phi)$, $r_1 = \frac{k_z - n_1 k\cos\theta}{k_z + n_1 k\cos\theta}$, $r_2 = \frac{k_z - n_2 k\cos\theta}{k_z + n_2 k\cos\theta}$, $t_{10} = \frac{2n_1 k\cos\theta}{n_1 k\cos\theta + k_z}$ or $r_1 = \frac{k\cos\theta - n_1 k_z}{k\cos\theta + n_1 k_z}$, $r_2 =$

$\frac{k\cos\theta-n_2k_z}{k\cos\theta+n_2k_z}$, $t_{10}=\frac{2n_1k\cos\theta}{n_1k_z+k\cos\theta}$ for polarization normal to or in the plane of incidence respectively. Where, we used the wave boundary conditions at $z=0$ and $z=L$.

The absorption at any position z inside the vapor is[8]

$$\alpha = 2\mathrm{Re}\left(\frac{|E_0(0)|-|E_0(L)|}{|E_0(0)|}\right) = -\frac{2}{E_0(0)}\int_0^L \mathrm{d}z\mathrm{Re}\left(\frac{\partial E_0(z)}{\partial z}\right). \quad (6)$$

The wave equation about the transmitting field is

$$\nabla^2 E_0 - \frac{1}{c^2}\frac{\partial^2 E_0}{\partial t^2} = \mu_0\frac{\partial^2 P_0}{\partial t^2}, \quad (7)$$

where μ_0 is the magnetic permeability, c is the velocity of the light in vacuum, and the amplitude of the polarization can be written as

$$P_o(x,z,t) = \frac{1}{2}P_{\mathrm{of}}(z)\left\{1+r_2\exp[2ik_z(L-z)]\right\} \times\exp[-i(\omega t-k_x^x-k_zz)]+c.c., \quad (8)$$

with $P_{\mathrm{of}}(z)$ driven by the forward fields amplitudes. If the back reflection of the vapor is not considered, by using slowly varying amplitude approximation, combining Eqs. (2)—(8), one gets the transmitting signal

$$S_{\mathrm{T}}^{\mathrm{lin}} = E_0^*(0)\frac{k}{\varepsilon_0}\cdot\frac{k}{k_z} \times\int_0^L \mathrm{d}z\mathrm{Im}\{1+r_2\exp[2ik_z(L-z)]\}P_{\mathrm{of}}(z), \quad (9)$$

with ε_0 the vacuum permittivity, $E_0^*(0)$ the conjugate value of $E_0(0)$.

In a two-level system with energy levels $|g\rangle$ and $|e\rangle$, assuming that $E_0(z)\approx E_0(0)$ inside the vapor, when the Rabi frequency Ω_{R} (defined as $\Omega_{\mathrm{R}}=\frac{\mu_{\mathrm{eg}}E_0(0)}{\eta}$ with μ_{eg} the dipole transition element) is much less than the dipole relaxation rate γ_{eg}, by solving Bloch equations, note that $t=z/v_z$ for $v_z>0$ and $t=(z-L)/v_z$ for $v_z<0$, the polarization $P_{\mathrm{of}}(z,t)$ can be written as[8]

$$P_{\mathrm{of}}(z,t) = \frac{iN\mu_{\mathrm{eg}}\Omega_{\mathrm{R}}}{\Lambda\varepsilon_0}\int_{-\infty}^{+\infty}\mathrm{d}v_x \times\left\{\int_{-\infty}^{0}\mathrm{d}v_z\left[1-\exp\left(-\Lambda\frac{z-L}{v_z}\right)\right]W(v_x)W(v_z) + \int_0^{\infty}\mathrm{d}v_z\left[1-\exp\left(-\Lambda\frac{z}{v_z}\right)\right]W(v_x)W(v_z)\right\}. \quad (10)$$

In Eq. (10), $\Lambda=\gamma_{\mathrm{eg}}-i[(\omega-\omega_0)-k_xv_x-k_zv_z]$, γ is the homogeneous linewidth, σ is the density matrix element, N is the atomic density, $W(v_x)$ and $W(v_z)$ are Maxwell-Boltzmann velocity distribution functions along x and z respectively, $W(v_i)=\frac{1}{u\sqrt{\pi}}\exp[-(\frac{v_i}{u})^2]$ $(i=x,z)$ with u the probable velocity of atoms in the vapor film, $u=\sqrt{\frac{2k_{\mathrm{B}}T}{m}}$ with k_{B} the Boltzmann constant, m the atomic mass, and T the temperature of the vapor.

Substitute Eq. (10) into Eq. (9), change integrated orders of z and v_z, we obtain the transmitting signal in the vapor film at $z=L$

$$S_{\mathrm{T}}^{\mathrm{lin}} = C\times\mathrm{Re}\left[\int_{-\infty}^{+\infty}W(v_x)\mathrm{d}v_x\left(\int_{-\infty}^{+\infty}W(v_z)\mathrm{d}v_zT_{\mathrm{F}} + \int_{-\infty}^{0}W(v_z)\mathrm{d}v_zT_{\mathrm{B1}} + \int_0^{+\infty}W(v_z)\mathrm{d}v_zT_{\mathrm{B2}}\right)\right], \quad (11)$$

with

$$T_{\mathrm{F}} = -\frac{kL}{\Lambda}+\frac{k|v_z|}{\Lambda^2}-\frac{k|v_z|}{\Lambda^2}\exp(-\frac{\Lambda L}{|v_z|}),$$

$$T_{\mathrm{B1}} = r_2\cdot\frac{k}{\Lambda}\left\{\frac{1}{2ik_z}[\exp(2ik_zL)-1] + \frac{v_z}{\Lambda+2ik_zv_z}\{1-\exp[(\frac{\Lambda}{v_z}+2ik_z)L]\}\right\},$$

$$T_{\mathrm{B2}} = r_2\cdot\frac{k}{\Lambda}\left\{\frac{1}{2ik_z}[\exp(2ik_zL)-1] + \frac{v_z}{\Lambda+2ik_zv_z}\exp(-\frac{\Lambda}{v_z}L)\{1-\exp[(\frac{\Lambda}{v_z}+2ik_z)L]\}\right\},$$

and

$$C = \frac{t_{10}^2\{1+r_2^2+r_2[\exp(-2ik_zL)+\exp(2ik_zL)]\}E_{\mathrm{in}}^2}{F^2} \times\frac{1}{\varepsilon_0\sqrt{1-n_1^2\sin^2\theta}}.$$

Equation (11) is the main result of the transmitting signal, it relates to the Fabry-Perot (F-P) intrinsic nature by a factor of F, and there is a damping resonant modulation with a period of π/k_z versus the thickness of the film, it has been shown in Refs. [9] and [10]. T_{F} corresponds to the forward fields contribution, T_{B1} and T_{B2} to the backward fields contribution for $v_z<0$ and $v_z>0$ respectively.

Theoretical line shapes ($ku=50\gamma_{\mathrm{eg}}$, $n=n_1=n_2=1.82$) according to Eq. (11) at normal incidence (Fig. 2) and at oblique incidence ($\theta=5°$) (Fig. 3) in a two level system are shown for $L/\lambda=1/16$, 1/8, 1/4, 1/2, 3/4, and 1, respectively. $S_{\mathrm{TF}}(i)$, $S_{\mathrm{TB}}(i)$, and $S_{\mathrm{T}}(i)$ represent the forward fields contribution, backward fields contribution and transmitting signal respectively, $i=s$ for polarization normal to the plane of incidence and $i=p$ for polarization in this plane.

From Eq. (11), Figs. 2 and 3, one can find that:

i) At normal incidence and at oblique incidence with a small incident angle, the line shapes of transmitting signal for s-polarization are similar to the shapes for p-polarization, this is due to the similar behavior of atoms excited by the total fields which have the similar distribution inside the vapor film for s-polarization and p-polarization.

ii) The sub-Doppler feature is manifest for $L=\lambda/16$,

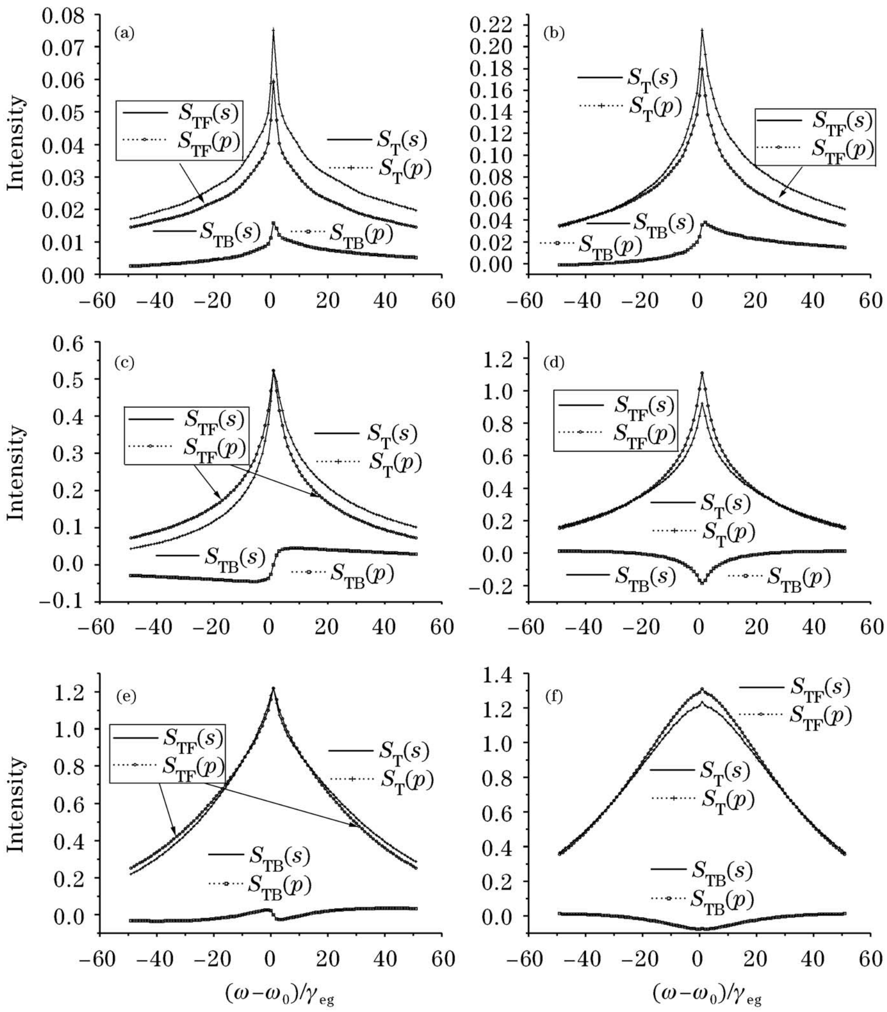

Fig. 2. Theoretical line shapes at normal incidence ($\theta = 0°$) in a two level system for (a) $L/\lambda = 1/16$, (b) $L/\lambda = 1/8$, (c) $L/\lambda = 1/4$, (d) $L/\lambda = 1/2$, (e) $L/\lambda = 3/4$, and (f) $L/\lambda = 1$.

$\lambda/8$, $\lambda/4$, $\lambda/2$, $3\lambda/4$ at normal incidence and for $L = \lambda/4$, $\lambda/2$ at oblique incidence, we should note that the Dicke-type line-width is narrower than that in the case of $L = \lambda/2$ predicted before as the narrowest Dicke-type structure. It is possible to obtain a narrowest Dicke-type structure (Figs. 2(a) and (b)) when the thickness of the cell is less than $\lambda/4$ at normal incidence, the main reason is that the contribution of slow atoms undergoes a long interaction time with the external fields[4−10]. The usual steady-state Doppler structure is dominant in a macroscopic film, and the sub-Doppler features are associated with the transient effects when the film thickness is much small compared with the wavelength. Atoms flying nearly parallel to the wall yield a stronger contribution to the signal while their resonance appears insensitively to the Doppler effects. On the other hand, atoms with a velocity along or near along normal direction of the surface in the film, the first is that they have smaller velocity, the second is that after colliding with the wall, they have no time to absorb photons from the external fields again, thus this group atoms contribute to the linewidth less. Because of the transverse Doppler effect, the narrow structure gets weaker and suffers an additional broadening, then vanishes when $L = \lambda/16$, $\lambda/8$, $3\lambda/4$ in the case of oblique incidence, that is, the Doppler broadening washes out the Dicke narrowing effect.

iii) It is comparable between a symmetric film (the refractive indices are the same for two windows) and a film with second window ideal antireflection coating. Indeed, we considered the backward fields contribution (and the F-P effects) in the case of symmetric film, the transmission signal S_T is not only an absorption properties (signal S_{TF}), it includes a dispersive contribution related to the mixing between absorption and selective reflection line shapes. To compare the transmission peaks and structures, it is convenient to consider 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 two groups are discussed in Refs. [7]—[10]. For the third group, because of the path length contribution, the absorption is weaker than that in the first two groups, in this case, atoms traveling against the incident direction are not in the steady state, they remain in the ground state right after leaving the back surface, 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.

In conclusion, we find that a sub-Doppler structure can

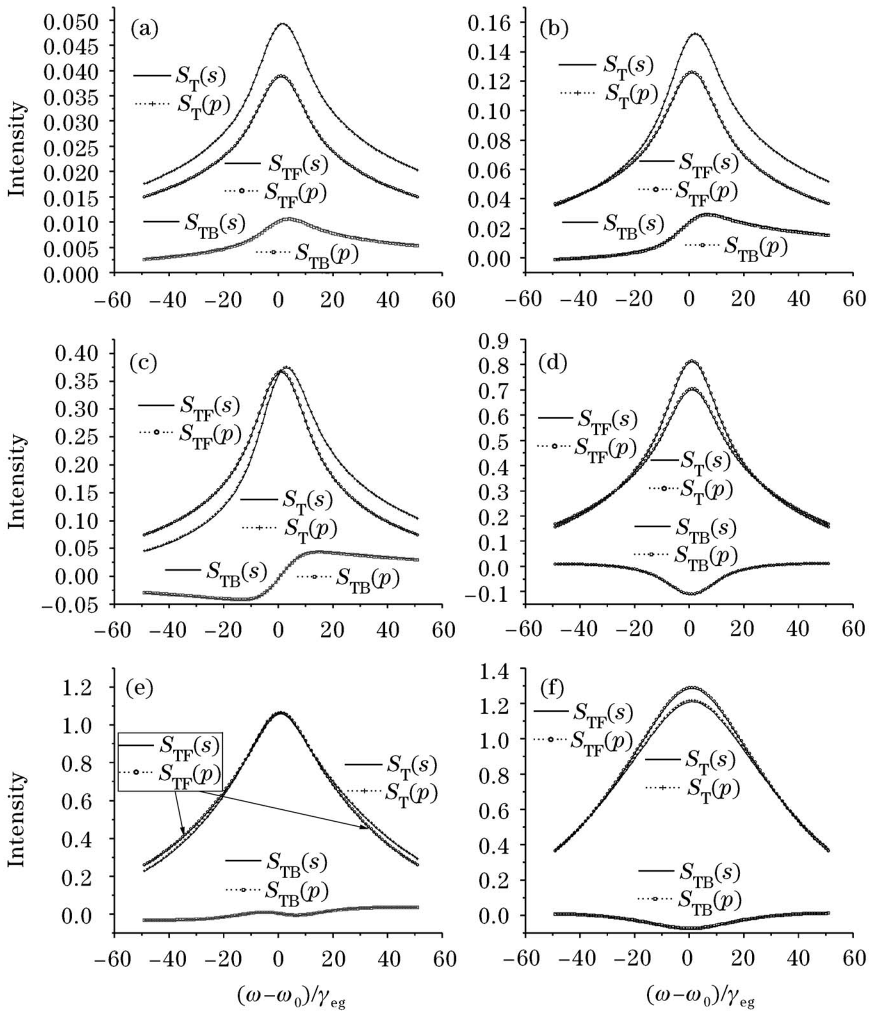

Fig. 3. Theoretical line shapes at oblique incidence ($\theta = 5°$) in a two level system for (a) $L/\lambda = 1/16$, (b) $L/\lambda = 1/8$, (c) $L/\lambda = 1/4$, (d) $L/\lambda = 1/2$, (e) $L/\lambda = 3/4$, and (f) $L/\lambda = 1$.

be obtained in both cases of normal incidence and small angle oblique incidence, especially when the thickness of the film is much less than the wavelength for normal incidence or comparable with wavelength for normal incidence and oblique incidence. It is feasible to get a sub-Doppler structure in a new region ($L < \lambda/4$) in experiments. Because of very small atomic vapor thickness, the signal is very weak, a sensitive spectral analysis, and low noise detection system are required, atom-wall interaction also should be considered.

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