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Transmission spectroscopy in a thin vapour layer and its derivative properties*

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A novel method of high-resolution spectroscopy of gases is proposed. The method is based on observing the optical behaviour of thin vapour layer when the particles cross the light beam or collide with the cell walls. The atomic motion and electronic quenching on gas-solid interfaces are explored by numerical analysis. The periodicity of peak values of transmission spectra and its derivative spectra demonstrates a sub-Doppler structure corresponding to the resonant transitions.

Keywords: thin vapour layer, sub-Doppler technique, spectral periodicity, derivative spectra
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1. Introduction

The sub-Doppler technique is widely applied in fields of atomic hyperfine structure, nonlinear process and interface physics. As a sensitive detection method with sub-Doppler structure, the selective reflection spectroscopy is usually used to monitor the interaction between vapour and medium surface.[1−3] In the case of the absorption spectra of thin vapour, its thickness should be in the range of 10–1000μm, for which the atomic flight time from wall to wall is less than the hyperfine energy pumping time, such that a physical mechanism is built up which can select the low-velocity atomic excitation, thereby dispelling the Doppler effect in the direction of laser pumping, and realize the laser cooling in one dimension.[4,5] In recent years, Izmailov[6], Zambon and Nienhuis[7], Vartanyan and Lin[8] made some theoretical studies about the optical symmetry of the thin vapour layer. When a vapour is enclosed in a narrow region between two parallel walls, the behaviour of spectra of two-state atoms will be dominated by the thickness of thin vapour. In the case of dilute gases and weak light beam, the spectrum near resonance is notably affected by the fact that atoms depart from the surface mainly in their ground state, and will have traversed some distance before the induced dipole moments of the vapour atoms reach their steady state. The reflection spectra in a weak field have a striking variation as a function of the thickness with odd and even behaviour around resonance.[7,8] The Bloch and Ducloy group have done many experiments and found that the signals are affected by optical pumping to a non-absorbing state.[4,5]

Harmonic detection is another method which reduces the line width by the mathematical means and gives high-sensitivity and high-resolution spectra.[9,10] Chardonnet *et al.* have obtained a linewidth of 80Hz(HWHM) for the P(39)A_1^3(-) line of O_sO_4 by selecting slow molecules in saturation spectroscopy at low laser fields and low pressures using the harmonic detection.[11]

In this paper, we first study the periodicity of peak values in transmission spectra of thin vapour layer, then discuss the properties of its second harmonic. The narrower absorption lines can be demonstrated through this model.

2. Theoretical model

The dipole polarization of the vapour layer affects the transmission behaviour of the light. We will con-

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sider an atomic gas included between two walls of a dispelling one-dimensional cell with the length l of the order of wavelength λ of the incident light, and the gas is assumed to be sufficiently rarefied, so that one can neglect collisions between atoms (see Fig.1).

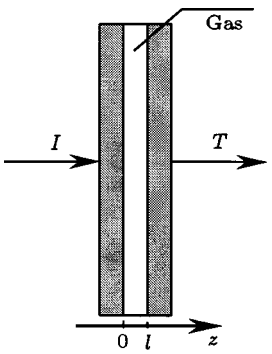


Fig.1. The structure of thin vapour cell.

In particular, such a scheme is fit for alkali metal vapours (the pressure being about 10^{-6}Torr). The transition frequency of two-level atoms between levels $|1\rangle$ and $|2\rangle$ is ω_{21}. The space $z<0$ and $z>l$ is assumed as vacuum. At the resonance region, $\omega \gg \omega_{21}-\omega|$, we express the electric field of the monochromatic light field by

$$E(z,t) = 1/2E(z)\exp(-\mathrm{i}\omega t) + c.c. \tag{1}$$

and the dipole polarization by

$$P(z,t) = 1/2P(z)\exp(-\mathrm{i}\omega t) + c.c. \tag{2}$$

The spatial parts of the emitted field can be obtained by solving Maxwell's equation

$$\nabla^2 E - \nabla(\nabla\cdot E) + k^2 E = -4\pi k^2 P(z) \tag{3}$$

with the polarization

$$P(z) = P_0(z)\exp(\mathrm{i}kz) = 2nM\langle\rho_{21}(z,v)\rangle, \tag{4}$$

where $k=\omega/c$, n is the number density of atoms, M is the transition dipole moment, v is the z component of the atomic velocity and $\langle\ \rangle$ is the symbol for average over the velocity distribution, which we can take as Maxwellian.

In fact, we can write the off-diagonal density matrix elements $\rho_{21}(z,v,t)$ as $\rho_{21}(z,v)\exp(-\mathrm{i}\omega t)$, in the steady state, ρ_{21} satisfies the optical Bloch equation

$$v\frac{\partial}{\partial z}\rho_{21} = \frac{i}{2\hbar}ME(z) - (\Lambda + i\Delta + ikv)\rho_{21}, \tag{5}$$

Λ is the homogeneous linewidth (FWHM) and Δ is the resonance detuning. The Bloch Eq. (5) shows that atoms departing from the wall at $z=0$ $(v>0)$ feel the light field with a detuning shift $-kv$, we set the boundary conditions $\rho_{21}(z=0, v>0)=0$, $\rho_{21}(z=l, v<0)=0$ and using the perturbation theory with $E(z)=E_0\exp(ikz)$, the Eq.(5) can be solved as

$$\rho_{21}(z,v>0) = \frac{iME_0}{2\hbar}\frac{\exp(ikz)-\exp\left(-\dfrac{\Lambda+i\Delta}{v}z\right)}{\Lambda+i(\Delta+kv)},$$

$$\rho_{21}(z,v<0) = \frac{iME_0}{2\hbar}\frac{\exp(ikz)-\exp\left[ikl-\dfrac{\Lambda+i\Delta}{v}(z-l)\right]}{\Lambda+i(\Delta+kv)}, \tag{6}$$

where, $v>0$ means the atoms moving to the right and $v<0$ to the left. When we insert (6) in Eq.(4), the polarization $P(z)$ can be obtained. Then the solution is formally given by $E(z) = 2\pi ik\int_0^l P(z')\mathrm{e}^{ik|z-z'|}\mathrm{d}z$ using the Green function.

The different regions have different physical meaning. For $z>l$, the transmitted field is expressed as

$$E_t(z) = 2\pi ik\int_0^l P(z')\mathrm{e}^{ikz'}\mathrm{d}z'\exp(ikz). \tag{7}$$

To calculate the characteristic of the transmitted beam, the intensity is

$$I_t \propto |E_0-E_t|^2 = E_0^2 - 2E_0\mathrm{Re}(E_t) + |E_t|^2. \tag{8}$$

Because E_0 is much larger than E_t, the term $|E_t|^2$ may be neglected in Eq.(8), and the absorption profile is determined by $\mathrm{Re}(E_t)$ or $\mathrm{Re}(E_t/E_0)$. Now we calculate the transmitted field of a thin vapour. It can be found from Eqs.(3)(4)(6)(7), and the result is

$$E_t/E_0 = -\frac{2\sqrt{\pi}nM^2}{\hbar k\nu_T}(G_1+G_2+G_3). \tag{9}$$

Here

$$G_1 = \phi\int_{-\infty}^{\infty}\frac{\exp(-\nu^2)}{\Gamma - i(\Omega-\nu)}\mathrm{d}\nu,$$

$$G_2 = \int_{-\infty}^{0}\frac{\nu\exp(-\nu^2)}{[\Gamma-i(\Omega-\nu)]^2}\mathrm{d}\nu - \int_0^{\infty}\frac{\nu\exp(-\nu^2)}{[\Gamma-i(\Omega-\nu)]^2}\mathrm{d}\nu$$

and

$$G_3 = \int_{-\infty}^{0} -\mathrm{e}^{i\phi}\frac{\nu\exp[-\nu^2+(\Gamma-i\Omega)\phi/\nu]}{[\Gamma-i(\Omega-\nu)]^2}\mathrm{d}\nu + \int_0^{\infty}\mathrm{e}^{-i\phi}\frac{\nu\exp[-\nu^2-(\Gamma-i\Omega)\phi/\nu]}{[\Gamma-i(\Omega-\nu)]^2}\mathrm{d}\nu$$

In this result, we have defined the dimensionless parameters $\Gamma = \Lambda/k\nu_T$, $\Omega = \Delta/k\nu_T$, $\phi = kl$, $\nu = v/\nu_T$, where ν_T is the most probable thermal velocity of atoms.

3. Numerical results and discussion

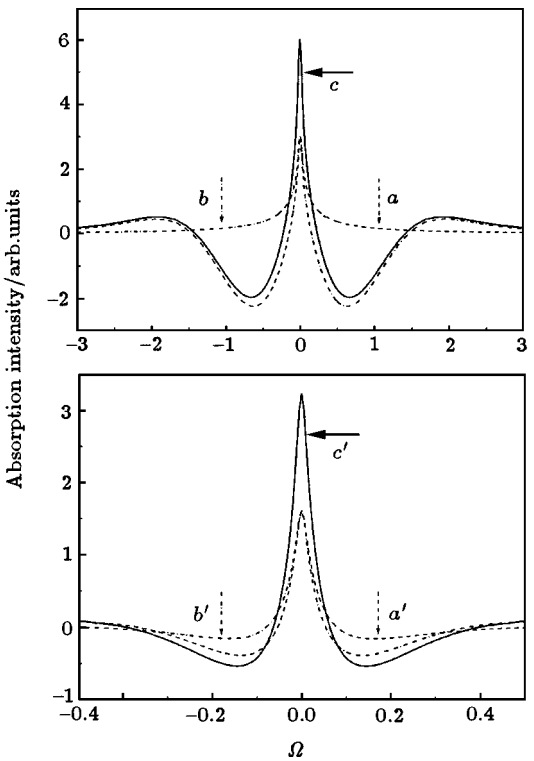


Fig.2. The contribution to the profile of transition spectrum depends on the atoms moving in different direction ($\phi = 3\pi$). a and a' are integral contribution with the velocities from $-\infty \to 0$ in Re(G_2) and Re(G_3), respectively. b and b' are integral contribution with the velocities from $0 \to +\infty$ in Re(G_2) and Re(G_3), respectively. c and c' are the contribution for all velocities in Re(G_2) and Re(G_3), respectively. The thickness of thin vapour cell is $\phi = 3\pi$.

Here we present the results of numerical calculations and discuss the relevant physics. In our computation we take Γ=0.01, which is fit for alkali-metal atomic gases under normal experimental conditions. From Eq.(9), the first term G_1 is integrated over the whole range of velocity distribution, the Re(G_1) term provides the general steady-state Doppler-broadened absorption with the transient effects expressed by the Re(G_2) and Re(G_3) terms, the maximum peak Re($G_{1\max}$) increases linearly with ϕ (or l). Actually, the atomic free path is larger than the thickness of vapour, and atoms flying nearly parallel to the wall yield a stronger signal which appears insensitive to the Doppler shift. Re(G_2) gives the response to the pumping field, and Re(G_3) reflects the finiteness of the interaction time ϕ/ν, which is limited by the cell thickness, the overall absorption at line centre ($\omega_{21} = \omega_0$) becomes stronger when ϕ is a half-integer. In Fig.2., we note that atoms with negative and positive velocity have different effects to the transition (compare a with b and a' with b'), at the same time, we find the sub-Doppler structure will arise, and this property is quite prominent (see c and c').

The variation of absorption with the thickness ϕ (or l) when the detuning Ω is zero is shown in Fig.3. Re ($G_{3\max}$) has a damping attenuation with increasing ϕ, its vibrational period being 2π. Re($G_{2\max}$) has a constant value and Re($G_{1\max}$) linearly increases. So we cannot see the hyperfine structure when the cell is thick.

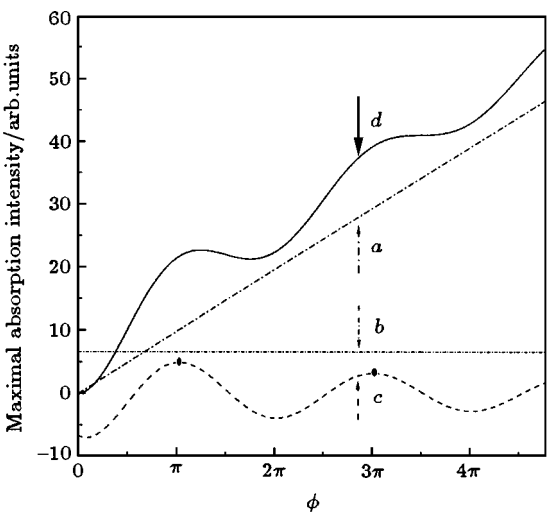


Fig.3. The variation of absorption intensity at resonance (Ω=0) versus the vapour thickness $\left(\phi = 2\pi\frac{l}{\lambda}\right)$. a: Re ($G_{1\max}$) b: Re ($G_{2\max}$) c: Re ($G_{3\max}$) d: Re ($G_{1\max}$+$G_{2\max}$+$G_{3\max}$).

Because of the special line profile, Lorentz structure with Doppler background will be much narrowed by using the harmonic detection. We can contrast the direction absorption signal with its second-order-derivative signal (see Fig.4). If we assume that the Doppler linewidth is 500MHz, then its second-order-derivative signal will be narrowed down to $\Delta\nu$ ≈3MHz. From this figure, we find that this method can dispel the Doppler background.

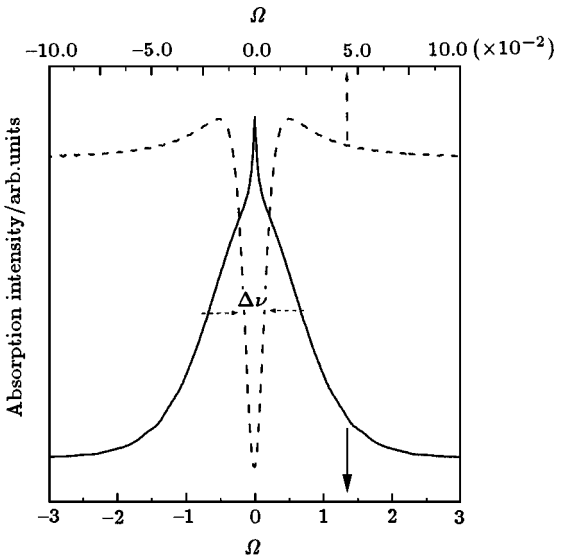


Fig.4. The transmission spectrum (solid line) and its second derivative spectrum (dashed line) for $\phi = 3\pi$.

4. Conclusion

We have discussed a method for obtaining the sub-Doppler spectra, which can realize the one dimension slow atomic detection. The vapour layer is so thin that the optical properties are strongly affected by its thickness. We studied the periodicity of the variation of transmission spectra with layer thickness and its derivative spectroscopy, and found a vibrational period of peak values of 2π (corresponding to $\Delta l = \lambda$) and obtained the second-order-derivative signal with $\Delta\nu \approx$3MHz. Such a method can be attractive to build up Doppler-frequency standards that require ultra-low incident power, or to attain Doppler-free molecular lines that are difficult to saturate.[6]

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