Spectroscopic Effects Nonlinear in Atomic Density Caused by the Free Motion of Atoms in a Gas

V.I. Yudin[1,2,3,*], A.V. Taichenachev[1,2], M.Yu. Basalaev[1,2,3], O.N. Prudnikov[1,2], S.N. Bagayev[1,2]
[1]Novosibirsk State University, ul. Pirogova 1, Novosibirsk, 630090, Russia
[2]Institute of Laser Physics SB RAS, pr. Akademika Lavrent'eva 13/3, Novosibirsk, 630090, Russia
[3]Novosibirsk State Technical University, pr. Karla Marksa 20, Novosibirsk, 630073, Russia
[*] viyudin@mail.ru

Summary—We have found new effects nonlinear in the atomic density caused by the free motion of atoms, considering the self-consistent solution of the Maxwell-Bloch equations in the mean-field approximation and for one-atomic density matrix in a gas of two-level atoms. These effects distort the Doppler lineshape (shift, asymmetry, broadening), but are not associated with an atom-atom interaction. In particular, in the case of dominating Doppler broadening of the linewidth (in respect with collisional broadening), founded atomic-motion-induced effects significantly exceed the well-known influence of the dipole-dipole interatomic interaction (e.g., Lorentz-Lorenz shift) by more than one order of magnitude.

Keywords—optical atomic clocks, collisional shift

According to the prevailing views, one of the main physical reasons for the nonlinear in atomic density effects in a gas is associated with the interatomic dipole-dipole interaction. With regard to laser spectroscopy of resonant gas media and high-precision atomic clocks, collective effects are of importance because of interatomic dipole-dipole interaction, which distort the resonance lineshape (shift, broadening, asymmetry) [1,2]. If we consider an ensemble of two-level atoms with an unperturbed frequency ω_0 for a closed optical transition $|g\rangle \to |e\rangle$ (see Fig.1), then, the scale of the dipole-dipole interaction is determined by the value of Lorentz-Lorenz shift $\Delta_{LL} = -\pi n k_0^{-3}\gamma_0$, where n is the atomic density (number of atoms per unit volume), $k_0 = \omega_0/c$ is the wave number (c is the speed of light in vacuum), γ_0 is the spontaneous decay rate of the upper level (see Fig.1). In particular, for an ensemble of atoms confined within a plane of thickness l, we have the total redshift induced by dipole-dipole interaction [1]:

$$\Delta_{dd} = \Delta_{LL} - \frac{3}{4}\Delta_{LL}\left(1 - \frac{\sin 2k_0 l}{2k_0 l}\right), \tag{1}$$

where the second term is the collective Lamb shift. For a thick slab ($k_0 l \gg 1$), the redshift is

$$\Delta_{dd} = \frac{1}{4}\Delta_{LL} \approx -0.8 n k_0^{-3}\gamma_0 \ . \tag{2}$$

In contrast to above, we describe new spectroscopic effects nonlinear in the atomic density n, which follow from the self-consistent solution of the Maxwell-Bloch equations for atomic gases and lead to distortion of the spectroscopic signal (shift, asymmetry, broadening). These effects are due to the free motion of atoms and can significantly exceed the effects of the dipole-dipole interatomic interaction. In particular, the frequency shift of the Doppler lineshape for a monochromatic running wave is more than one order of magnitude larger than the estimate (2) and has an opposite sign. We emphasize that our results are obtained in the standard mean-field approximation and without taking into account the atom-atom interaction. The presented atomic-motion-induced effects have not previously been discussed in the scientific literature, as far as we know.

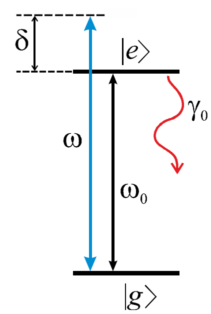

Fig.1. Two-level system.

Consider the one-dimensional task of propagation along the z axis of a plane monochromatic wave with a real electric field $E(t,z)$ in a gas medium of free-moving two-level atoms (see Fig.1). The atom-field interaction is described by the operator of electro-dipole interaction. Our analysis will be carried out within the framework of a self-consistent solution of the Maxwell-Bloch equations system, which includes the wave equation for the field (in the CGS system):

$$\left(\frac{\partial^2}{\partial z^2} - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)E(t,z) = \frac{4\pi}{c^2}\frac{\partial^2}{\partial t^2}P(t,z)\,, \tag{3}$$

where $P(t,z)$ is the mean-field polarization of the medium. The atomic gas is described by the one-atomic density matrix $\hat{\rho}(v)$ (v is the velocity of the atom). In the linear approximation in the field E (i.e., in the small saturation limit), we have the following Bloch equation for non-diagonal matrix element $\rho_{eg}(v)$:

$$\left(\frac{\partial}{\partial t} + v\frac{\partial}{\partial z} + \gamma_{eg} + i\omega_0\right)\rho_{eg}(v) = i d_{eg} E(t,z) f(v)/\hbar\,, \tag{4}$$

where $f(v)$ describes the Maxwellian velocity distribution of atoms:

$$f(v) = e^{-(v/\overline{v})^2}/(\overline{v}\sqrt{\pi}), \quad \overline{v} = \sqrt{2k_B T/m} \tag{5}$$

where k_B is the Boltzmann constant, T is the temperature of the gas, m is the mass of the atom. The polarization of the medium P in our one-dimensional task is defined as:

$$P(t,z) = n\langle D\rangle_v = n\int_{-\infty}^{+\infty} D(v)dv, \quad D(v) = d_{ge}\rho_{eg}(v) + c.c. \quad (6)$$

where $\langle D\rangle_v$ is the velocity-averaged dipole moment of the atom. Thus, the equations (3)-(6) constitute the Maxwell-Bloch equations system in our case.

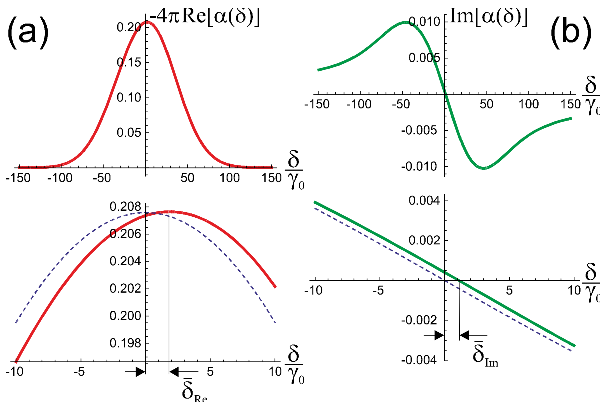

Fig.2. Spectroscopic dependence $\alpha(\delta)$ as a solution of the Eq.(9)

In the case of a resonant monochromatic wave (with the frequency ω traveling in the positive direction along the z axis, the equations for the density matrix (4) usually use the expression for the field in vacuum $E(t,z) = E_0 \exp\{-i(\omega t - kz)\} + c.c.$ (where $k = \omega/c = 2\pi/\lambda$ is the wave number in vacuum, λ is the wavelength in vacuum), which leads to the standard expression for the Doppler lineshape. However, it is more correct to define the function $E(t,z)$ as a self-consistent solution of the equations system (3)-(6), which we will seek in the form:

$$E(t,z) = E_0 e^{-i\omega t + (i+\alpha)kz} + c.c. \quad (7)$$

where α is an unknown complex number. Substituting this expression into Eq.(4), we find in the rotating wave approximation

$$\rho_{eg}(v) = \frac{i d_{eg} f(v)/\hbar}{\gamma_{eg} + i\delta + (i+\alpha)kv} E_0 e^{-i\omega t + (i+\alpha)kz}, \quad (8)$$

where $\delta = (\omega - \omega_0)$ is the frequency detuning. Then, using Eqs.(6)-(7), we obtain from Eq.(3) the equation with respect to the unknown α:

$$\alpha^2 + 2i\alpha = -i3\pi n k_0^{-3}\gamma_0 \int_{-\infty}^{+\infty} \frac{f(v)dv}{\gamma_{eg} - i\delta + (i+\alpha)kv}, \quad (9)$$

where we have used the well-known expression $\gamma_0 = 4k_0^3|d_{eg}|^2/(3\hbar)$ for the spontaneous decay rate of the upper level $|e\rangle$ (see Fig.1). Thus, we will use Eq.(9) to determine the frequency dependence $\alpha(\delta)$. In this case, the correct (physical) solution must satisfy the condition $\mathrm{Re}[\alpha(\delta)] < 0$, which corresponds to the attenuation of the wave during propagation along the positive direction of the z axis.

As follows from the basic equation (9), the $\alpha(\delta)$ has a complex nonlinear dependence on the $nk_0^{-3}\gamma_0$. Indeed, Fig.2(a) shows the dependence of the absorption coefficient $\mathrm{Re}[\alpha(\delta)]$, where we see (Fig.2(a), lower graph) a positive shift $\bar{\delta}_{\mathrm{Re}}$ for the top of the Doppler absorption lineshape. Fig.2(b) shows a correction to the dispersion law $\mathrm{Im}[\alpha(\delta)]$ (also obtained from solving of Eq.(9)), which has the shift $\bar{\delta}_{\mathrm{Im}}$. Our calculations show that under the conditions $\gamma_{eg} \ll k\bar{v}$ and $nk_0^{-3} < 0.5$ these shifts are well described by the formulas:

$$\bar{\delta}_{\mathrm{Re}} \approx 19nk_0^{-3}\gamma_0, \quad \bar{\delta}_{\mathrm{Im}} \approx 11nk_0^{-3}\gamma_0. \quad (10)$$

Comparing (10) with the estimate (2), we can assert that the atomic-motion-induce effects can be more than one order of magnitude greater than an influence of the dipole-dipole interaction. Moreover, shifts (10) have a positive sign (blueshifts), unlike redshift (2).

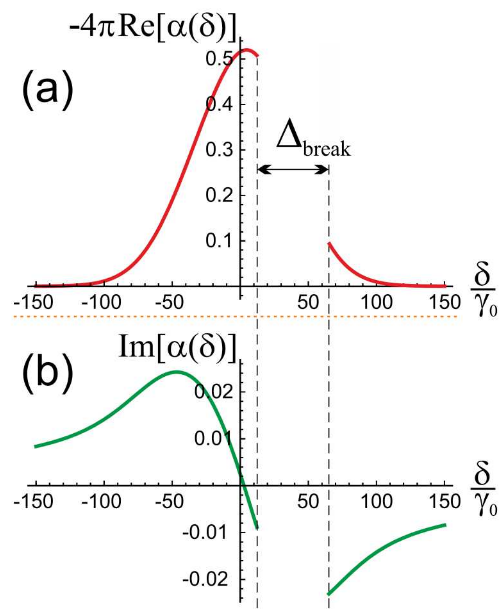

Fig.3 Spectroscopic dependence $\alpha(\delta)$ in the case where there is a frequency interval Δ_{break}, within which there is no solution of Eq.(9).

In addition to the above, we have found an extremely unexpected result, where there exists no solution to Eq. (9) within a frequency interval _break starting from some value nk_0^{-3} [i.e., only the trivial solution E=0 takes place for the Maxwell-Bloch equations (3)-(6)]. This is clearly seen in Figs. 3(a) and (b), as a discontinuity in the functional dependence $\alpha(\delta)$. Numerical analysis shows that when $\gamma_{eg} \ll k\bar{v}$ such a frequency interval Δ_{break} exists under the condition

$$nk_0^{-3} > 0.27\gamma_{eg}/\gamma_0. \quad (11)$$

We call this problem the atomic-motion conditioned catastrophe for Maxwell-Bloch equations (AMCMBE-catastrophe). Alternatively, the problem may be related to the mean-field approximation, which we call the atomic-motion conditioned catastrophe of the mean-field approximation (AMCMF-catastrophe). In any case, this mathematical problem concerns the basic principles of the theoretical description of light propagation in a gas and requires further research.

We have developed a theory of the Doppler-broadened lineshape in an atomic gas, basing on a self-consistent solution of the Maxwell-Bloch equations in the mean-field approximation and one-atomic density matrix. The presence of nonlinear effects in the atomic density caused by the free motion of atoms (which affect the lineshape shift, asymmetry, and broadening) was found. It is shown that in the regime of dominating Doppler broadening and temperatures T>300 K, these effects can exceed by more than one order of magnitude the influence of interatomic dipole-dipole interaction. Thus, in a resonant gas medium, the physical picture of spectroscopic effects nonlinear in atomic density seems as complicated mixing of both the collective effects, caused by the interatomic interaction, and effects due to an atomic motion. Obtained results are important for laser spectroscopy, atomic clocks and fundamental physics. In particular, the shift of the clock resonance depending on the atomic density n determines the inaccuracy and long-term instability (due to temperature variations) of the vapor-cell atomic clock, since the value of n depends on the temperature T.

In addition, in certain area of parameters, we have found that there exists a frequency interval Δ_{break} with only the trivial self-consistent solution to the Maxwell-Bloch equations (i.e., only E=0). This problem (called by us AMCMBE- or AMCMF-catastrophe) concerns the basic theoretical description of light propagation in a gas and, therefore, requires further research.

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