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DIFFUSION-BROADENED LINE SHAPE NEAR A TURNING POINT

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An absorption spectrum of the gas with large Doppler width and soft collisions between particles is studied. Particles are assumed having a nonlinear dependence of the resonance frequency on velocity. The shape of the narrow peak in the spectrum resulting from an extreme point of this dependence is calculated analytically for the first time. Without collisions it has the characteristic asymmetric shape. Collisions are shown to broaden the line and change its shape. The profile of probe-field spectrum in a three-level system with the strong field at adjacent transition is obtained. Components of Autler – Townes doublet are found to spread and repel each other because of collisions.

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An investigation of the resonant interaction of a gas of particles and electromagnetic wave is a promising way of studying collision processes in gas [1 – 3]. Of particular interest is the case when collisions can be described as diffusion in the velocity space; for example, particles are ions in plasma or heavy atoms in the buffer gas of light ones. Landau collision term describes well the spectroscopic effects of Coulomb ion-ion scattering [4]. For some reason the frequency of exact resonance between the wave and some particle can depend on the velocity. If this dependence results from Doppler shift then it is linear, the spectral line shape within the linear approximation in field intensity has been calculated [5, 6]. First nonlinear corrections to the absorption spectrum due to saturation were obtained [7], too.

Besides the effects of saturation and nonlinear interference, the field of monochromatic wave splits energy levels of a particle [8]. Consider the interaction of gas with strong and probe waves resonant to adjacent transitions between intrinsic states of the particle. Without the strong field the dependence of the resonance frequency for the probe wave on the velocity is linear due to Doppler shift. If one turns on the field then for each particle there are two resonances between it and the probe wave. Their positions coincide with Rabi frequencies which are nonlinear functions of the velocity. However, the computation of splitting in a system with large Doppler width was done for collisionless case only [9].

Nowadays a challenging task is to gain tunable CW UV coherent radiation. Using stimulated Raman scattering tunable radiation in Na_2, Ne was obtained. The ions have higher energy levels, so there is hope to reach short-wave radiation by Raman up-conversion in Ar^+ [10, 11]. Thus, strong-field effects are interesting for experiment along with soft collisions.

In the present Letter we study the absorption spectra of a gas of particles with both soft collisions and nonlinear *resonance frequency* $\Omega_R(v)$, the frequency of the field at which the exact resonance between it and the particle with velocity v occurs. In Ref.[5, 6, 7] the linear dependence $\Omega_R = kv$ was considered. Here we examine the nonlinear function

$\Omega_R(v)$ arisen from the interaction with a strong monochromatic wave. The extreme point of $\Omega_R(v)$ is of special interest in our consideration. The simplest nonlinearity is quadratic. If particles are concentrated near velocity v_0 then one can interpret this dependence as Taylor expansion of Ω_R near v_0 to order $(v - v_0)^2$. If for some v_0 the linear term in the expansion is equal to zero then it would appear reasonable that the main term in it is quadratic or Ω_R is constant. In brief, the quadratic nonlinearity seems sufficient to describe all new effects associated with nonlinearity.

Let us calculate the spectrum of light absorbed (or emitted) by monokinetic beam of particles with given initial velocity v_0 throughout its whole time evolution, the so-called *beam spectrum* with velocity v_0. After that it is possible to find the absorption spectra for arbitrary velocity distribution. The spectrum is given by the expression

$$I(\Omega) = \frac{1}{\pi}\mathrm{Re}\int_0^\infty \mathrm{d}t\, \Phi(t)\, e^{-\mathrm{i}\Omega t}, \quad \Phi(t) = \left\langle \exp\left(\mathrm{i}\int_0^t \mathrm{d}\tau\, \Omega_R(v(\tau))\right)\right\rangle, \tag{1}$$

where $\Phi(t)$ is the correlation function. The width of the beam spectrum is the inverse time of the dephasing t_D^{-1}. When the dependence of the resonance frequency Ω_R is linear $\Omega_R(v) = kv$, then correlation function is given by [5, 6]

$$\Phi(v_0, t) = e^{\mathrm{i}kv_0t - Dk^2t^3/3}. \tag{2}$$

Roughly, the deviation of velocity from its initial value is of the order of $\Delta v(t) \sim \sqrt{Dt}$, then the phase deviation is

$$\Delta\varphi = \Delta\int_0^t \mathrm{d}\tau\, \Omega_R(\tau) \sim t\cdot k\Delta v \sim \sqrt{Dk^2t^3}.$$

The dephasing happens when the latter reaches π, thus the spectrum width or the inverse dephasing time is of the order of $t_D^{-1} \sim (Dk^2)^{1/3}$. If the particles decay in time then one should add $\mathrm{i}\Gamma$ to Ω_R or multiply $\Phi(t)$ by $e^{-\Gamma t}$, where Γ is the inverse lifetime of the particles.

While only the integral of $\Phi(t)e^{-\mathrm{i}\Omega t}$ over time is of interest, one can reduce the problem to simpler one: there is a source of particles with velocity v_0 and there is a steady-state distribution for polarization of particles $\rho(v)$ governed by the equation

$$\left(\mathrm{i}(\Omega - \Omega_R) - D\frac{\mathrm{d}^2}{\mathrm{d}v^2}\right)\rho = \delta(v - v_0), \tag{3}$$

and the beam spectrum given by the expression $I_B(\Omega, v_0) = \frac{1}{\pi}\mathrm{Re}\int \mathrm{d}v\, \rho(v)$.

Let us now consider the case when the dependence of resonance frequency is quadratic $\Omega_R(v) = \omega + kv + av^2/2$ if only near v_0. If $v_0 \simeq -k/a$ then the linear shift of the resonance frequency in change of the velocity vanishes, so the diffusion of phase arises from the quadratic one

$$\Delta\varphi \sim t\cdot a(\Delta v)^2 \sim Dat^2,$$

the spectrum width is of the order of $t_D^{-1} \sim (D|a|)^{1/2}$. This simple estimates are confirmed below by detailed calculations. The point $v = -k/a$, or generally the point where $\mathrm{d}\Omega_R/\mathrm{d}v = 0$, can be called a *turning point* [9, 12], because if you pull the velocity through it the sign of $\mathrm{d}\Omega_R/\mathrm{d}v$ (or the direction of Ω_R variation) changes.

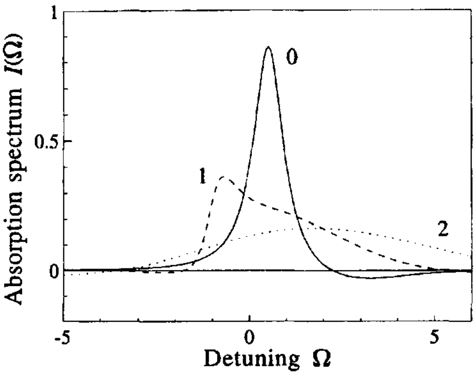

Fig.1. Beam spectra with velocity $v_0 = n$, $n = 0, 1, 2$ (curve n) at $\Omega_R(v) = v^2/2$, $D = 2$

Introduce a new variable $z = \alpha(v + k/a)$, $\alpha = (a/2iD)^{1/4}$, $z_0 = z|_{v=v_0}$ and decompose ρ, $\delta(v - v_0)$ in a series over functions $\psi_n(z)$

$$\psi_n(z) = \alpha \frac{e^{-z^2/2} H_n(z)}{\sqrt{\pi} 2^n n!}, \quad \rho = \sum_{n=0}^{\infty} \rho_n \psi_n(z), \quad \delta(v - v_0) = \sum_{n=0}^{\infty} \psi_n(z)\, e^{-z_0^2/2} H_n(z_0),$$

where $H_n(z)$ is the nth Hermite polynomial. The quantities ρ_n are found immediately since all ψ_n are eigenvectors of the operator in l.h.s. of (3). After integration over v one gets

$$I_B(\Omega, v_0) = \frac{\sqrt{2}}{\pi} \mathrm{Re} \sum_{n=0}^{\infty} \frac{e^{-z_0^2/2} H_{2n}(z_0)}{2^{2n} n! (2\beta n + x)} = \frac{1}{\pi} \mathrm{Re} \int_0^{\infty} \mathrm{d}\tau \, \frac{e^{(\beta/2 - x)\tau}}{\sqrt{\mathrm{ch}\, \beta\tau}} \exp\Big(- \tfrac{1}{2} z_0^2 \mathrm{th}\, \beta\tau \Big). \quad (4)$$

$$\beta = (2Da/\mathrm{i})^{1/2}, \quad x = \beta/2 + \mathrm{i}(\Omega - \omega - k^2/2a).$$

Thus the spectrum is given by the expression (1) with correlation function

$$\Phi(v_0, t) = \frac{e^{\mathrm{i}\Omega_R(v_0)t}}{\sqrt{\mathrm{ch}\, \tau}} \exp\left(\frac{h}{4} \frac{D(k + av_0)^2}{\Gamma_2^3} (\tau - \mathrm{th}\, \tau) \right), \quad (5)$$

where $h = 1 - \mathrm{i}\, \mathrm{sign}\, a$, $\tau = h\Gamma_2 t$, $\Gamma_2 = \sqrt{D|a|}$. The beam spectrum examples are plotted in Fig.1. When $\Gamma_2 \ll \Gamma_1$ one can expand $\mathrm{th}\, \tau$ to order τ^3 and replace $\mathrm{ch}\, \tau$ by 1. The resultant expression coincides with (2). If $\Omega_R(v)$ is the 2nd degree polynomial then (5) is the exact solution for correlation function (1), otherwise it is valid if characteristic velocity scale of Ω_R is much greater than $(D/|a|)^{1/4}$.

Let us consider the spectrum of particles that are uniformly distributed over velocity. For $\Omega_R(v) = av^2/2$ we have $\Gamma_1 = (Da^2v^2)^{1/3}$, $\Gamma_2 = (D|a|)^{1/2}$ and the spectrum is given by

$$I(\Omega) = \frac{1}{\pi} \mathrm{Re} \int \mathrm{d}v \int_0^{\infty} \mathrm{d}t \, \Phi(v, t)\, e^{-\mathrm{i}\Omega t} = \frac{1}{(2D|a|^3)^{1/4}} \mathrm{Re}\, e^{\pm 3\pi \mathrm{i}/8} \frac{\Gamma(z + \frac{1}{4})}{\Gamma(z + \frac{3}{4})}, \quad (6)$$

where $z = e^{\pm\pi \mathrm{i}/4}(\Gamma + \mathrm{i}\Omega)/\sqrt{8D|a|}$, $\Gamma(z)$ is the gamma function, and the upper/lower sign corresponds to positive/negative value of a. $I(\Omega)$ is an asymmetric peak, which has two characteristic widths: width Γ due to decay and diffusion width $(D|a|)^{1/2}$. If Ω is far from $\Omega_R(v_*)$ where v_* is the turning point, then the number of particles N_R

resonantly interacting with the field is proportional to $\gamma(v_R)/(\mathrm{d}\Omega_R/\mathrm{d}v)(v_R)$, where v_R is the resonance frequency, $\Omega \simeq \Omega_R(v_R)$; and $\gamma(v)$ is the width of the beam spectrum with velocity v. Near a turning point $v_R \simeq v_*$ we have $N_R \propto (\gamma(v_*)/(\mathrm{d}^2\Omega_R/\mathrm{d}v^2)(v_*))^{1/2}$, i.e., the field resonantly interacts with maximal number of particles when $\Omega \simeq \Omega_R(v_*)$. The spectrum wings are assymmetric:

$$I(\Omega) \simeq \begin{cases} \sqrt{\dfrac{2}{a\Omega}}, & a\Omega > 0, \\ \dfrac{1}{\sqrt{2|a\Omega|}}\left(\dfrac{\Gamma}{|\Omega|} + \dfrac{D|a|}{4\Omega^2}\right), & a\Omega < 0. \end{cases}$$

When the diffusion is inessential $\Gamma \gg (D|a|)^{1/2}$, we have

$$I(\Omega) = \mathrm{Re}\left[\frac{2}{a(\Omega - \mathrm{i}\Gamma)}\right]^{1/2} = \frac{\sqrt{|a|\sqrt{\Omega^2+\Gamma^2}+a\Omega}}{|a|\sqrt{\Omega^2+\Gamma^2}}.$$

To calculate the spectrum $I(\Omega)$ one can substitute the beam spectrum $I_B(\Omega, v_0)$ by $\delta(\Omega - \Omega_R(v_0))$ if the linear shift $\Omega'(v) = (\mathrm{d}\Omega_R/\mathrm{d}v)(v)$ does not essentially change within the domain $v - v_0 \sim \gamma(v_0)/\Omega'(v_0)$. In more general problem one should demand also insignificant change of the integral intensity of the beam spectrum with velocity v inside this domain. Here we have $\Omega'(v) = av$, $\gamma(v) \sim \Gamma_1$, so the condition of invariance of Ω' looks like $\Gamma_1/\Omega' \ll v$, i.e., $v \gg (D/|a|)^{1/4}$. The widths of this domain and function $\rho(v)$ at $v_0 = 0$ are of the same order. The shift of the resonance frequency in this domain is of the order of Γ_2.

Now we will calculate the probe-wave spectrum in the presence of strong field at the adjacent transition. The strong and probe fields are resonant to transitions between states $|2\rangle$ and $|1\rangle$, $|3\rangle$, respectively. We assume both waves copropagating and denote the projection of the velocity by $v = \mathbf{kv}/k$, where $\mathbf{k}$ is the wave vector of the strong field. Denoting the detunings from the resonance of the strong and the probe waves as Ω and Ω_μ, we write kinetic equations for off-diagonal elements of density matrix as

$$\begin{gathered} \rho_{13}(\mathbf{r},\mathbf{v},t) = \rho_{13}(\mathbf{v})\exp(\mathrm{i}(\mathbf{k}_\mu - \mathbf{k})\mathbf{r} - \mathrm{i}(\Omega_\mu - \Omega)t), \\ \rho_{23}(\mathbf{r},\mathbf{v},t) = \rho_{23}(\mathbf{v})\exp(\mathrm{i}\mathbf{k}_\mu\mathbf{r} - \mathrm{i}\Omega_\mu t), \\ \left(\Omega_\mu - \Omega_{1B} + \mathrm{i}D\frac{\mathrm{d}^2}{\mathrm{d}v^2}\right)\rho_{31} = G\rho_{32} - G_\mu^*\rho_{21}, \\ \left(\Omega_\mu - \Omega_{2B} + \mathrm{i}D\frac{\mathrm{d}^2}{\mathrm{d}v^2}\right)\rho_{32} = G^*\rho_{31} - G_\mu^*(\rho_2 - \rho_3), \\ \Omega_{1B} = \Omega + (k_\mu - k)v + \mathrm{i}\Gamma_{13}, \quad \Omega_{2B} = k_\mu v + \mathrm{i}\Gamma_{23}, \end{gathered} \tag{7}$$

where Γ_{13}, Γ_{23} are the relaxation constants of coherence between $|2\rangle$ and $|1\rangle$, $|3\rangle$; $G = \mathbf{E}\mathbf{d}_{21}/2\hbar$, $G_\mu = \mathbf{E}_\mu\mathbf{d}_{23}/2\hbar$, $\mathbf{d}_{ij}$ is the matrix element of the dipole moment, and $\mathbf{E}$, $\mathbf{E}_\mu$ are the amplitudes of the strong and the probe waves, respectively. To find ρ_{31}, ρ_{32} one should know ρ_2, ρ_3 and ρ_{21}.

The beam spectrum with velocity v has two resonances at Rabi frequencies [8]

$$\Omega_R^{(1,2)}(v) = k_\mu v + \eta + \mathrm{i}\Gamma_+ \pm \sqrt{(\eta + \mathrm{i}\Gamma_-)^2 + |G|^2}, \tag{8}$$

where $2\eta = \Omega - kv$, $2\Gamma_\pm = \Gamma_{13} \pm \Gamma_{23}$. One can think that there are two types of particles with different dependence of their resonance frequency on velocity. Strictly speaking the diffusion causes transitions between these types. But if $|G| \gg (Dk^2 9)^{1/3}$ then one can treat this two branches of hyperbole (8) independently and apply the theory developed above. Only two elements of the density matrix are mixed by the strong field. Then there is no cubic and higher shifts in the equation for ρ_{32}. One can get this equation by the action of operator $(\Omega_\mu - \Omega_{1B} + \mathrm{i}D\,\mathrm{d}^2/\mathrm{d}v^2)$ on (7).

If $k_\mu < k$ then there is one turning point in each of this two frequency branches posed at

$$v_{1,2} = \frac{\Omega}{k} \mp \frac{(2k_\mu - k)|G|}{k\sqrt{k_\mu(k-k_\mu)}}, \quad \Omega_R^{(1,2)}(v_{1,2}) = k_\mu\frac{\Omega}{k} \pm \frac{2|G|}{k}\sqrt{k_\mu(k-k_\mu)},$$

$$\frac{\mathrm{d}\Omega_R^{(1,2)}}{\mathrm{d}v}(v_{1,2}) = 0, \quad a = \frac{\mathrm{d}^2\Omega_R^{(1,2)}}{\mathrm{d}v^2}(v_{1,2}) = \pm\frac{2(k_\mu(k-k_\mu))^{3/2}}{k|G|}. \tag{9}$$

Here for simplicity we have neglected the decay Γ_{13}, Γ_{23}. Expressions for coordinates of turning points (9) coincide with the result for collisionless case [9]. The curvature of the frequency branch $a \propto 1/|G|$, $N_R \propto |G|^{1/2}$, then the absorption grows with $|G|$.

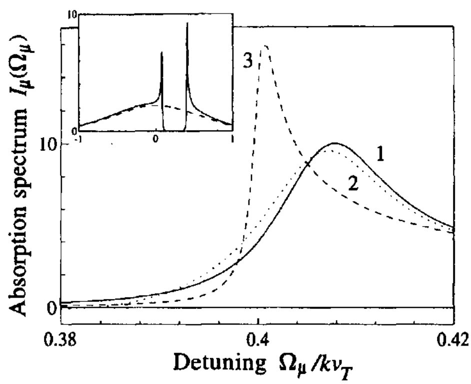

Fig.2. Absorption spectra $I_\mu(\Omega_\mu)$ in arbitrary units. $(Dk^2)^{1/3} = 6.3\cdot 10^{-2}kv_T$: numerical calculations (curve 1) and approximation formula (10) (curve 2); and $D = 0$ (curve 3). $\Gamma_{ij} = 10^{-3}kv_T$, $\Omega = 0.3kv_T$, $|G| = 0.2kv_T$, $k_\mu = 0.8k$. The population of levels 1 and 2 coincide. The dashed curve in the inset corresponds to $G = 0$

By analogy with (5) the spectrum is given by

$$I_\mu(\Omega_\mu) \propto \sum_{j=1}^{2}(-1)^j \mathrm{Re}\int \mathrm{d}v \int_0^\infty \mathrm{d}t \frac{\exp\left(\mathrm{i}(\Omega_R^{(j)} - \Omega)t\right)}{\sqrt{\mathrm{ch}\,\tau_j}} \times$$

$$\times \frac{(\Omega_R^{(j)} - \Omega_{1B})(\rho_2 - \rho_3) + G^*\rho_{21}}{\Omega_R^{(1)} - \Omega_R^{(2)}} \exp\left(h_j\left(\Gamma_{j1}/\Gamma_2\right)^3\left(\tau_j - \mathrm{th}\,\tau_j\right)\right), \tag{10}$$

$$\Gamma_{j1} = \left(D\frac{\mathrm{d}\Omega_R^{(j)}}{\mathrm{d}v}\right)^{1/3}, \quad h_{1,2} = 1 \mp \mathrm{i}\,\mathrm{sign}(k - k_\mu), \quad \Gamma_2 = \left(\frac{2Dk_\mu|k-k_\mu|}{\Omega_R^{(1)} - \Omega_R^{(2)}}\right)^{1/2}, \quad \tau_j = h_j\Gamma_2 t.$$

The probe-wave spectrum is illustrated in Fig.2. In the inset there are two narrow peaks that come from the return points. Curve 1 is derived from numerical solution of coupled diffusion equations for the whole 3×3 density matrix with taking into account the friction force and collisional mixing of the frequency branches.

The diffusion width Γ_2 of each narrow peak in the spectrum is found to be much less then width $\Gamma_1 = (Dk_\mu^2)^{1/3}$ resulting from the linear shift. Their ratio is $\Gamma_2/\Gamma_1 = = (2\Gamma_1\kappa/|G|)^{1/2}$, where $\kappa = (k/k_\mu - 1)^{3/2}k_\mu/k$. Such peaks in plasma ($Ar^+$, 488 and 514.5 nm, V-scheme with common short-lived level 2, Γ_{23} = 250 MHz) was observed in [12]. If we take the diffusion coefficient measured under similar conditions [13], then Γ_1 = 170 MHz, Γ_2 = 20 MHz. However, the width of the peaks was about 200 MHz. The diffusion width of the peak arises from the nonlinear shift near a turning point; otherwise, the peak would be appreciably wider.

Thus, the absorption spectrum of the gas of particles whose velocity evolves in a diffusion way is obtained. The dependence of the resonance frequency on the velocity of the particle can deviate from linear. The universal shape of the narrow peak in the spectrum (6), which comes from the extreme point of the velocity dependence of the resonance frequency, is found. The collisional width of the peak is proportional to the square root of the diffusion coefficient.

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