Diffusion-broadened line shape near a turning point

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The absorption spectrum of a gas with a large Doppler width and soft collisions between particles is studied. The particles are assumed to have a nonlinear dependence of the resonance frequency on velocity. The shape of the narrow peak in the spectrum resulting from an extremum of this dependence is calculated analytically for the first time. In the absence of collisions it has a characteristic asymmetric shape. Collisions are shown to broaden the line and change its shape. The profile of the probe-field spectrum is obtained for a three-level system with the strong field at an adjacent transition. The components of the 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 an electromagnetic wave is a promising way of studying collision processes in gases.[1–3] Of particular interest is the case when collisions can be described as diffusion in velocity space; for example, when the particles are ions in a plasma or heavy atoms in a buffer gas of light atoms. The Landau collision term describes well the spectroscopic effects of ion–ion Coulomb scattering.[4] For some reason the frequency of exact resonance between a wave and a particle can depend on the velocity. If this dependence results from the Doppler shift then it is linear; the spectral line shape within the linear approximation in the field intensity has been calculated previously.[5,6] The first nonlinear corrections to the absorption spectrum due to saturation have been obtained,[7] too.

Besides the effects of saturation and nonlinear interference, the field of a monochromatic wave splits the energy levels of a particle.[8] Consider the interaction of a 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 the 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 the Rabi frequencies, which are nonlinear functions of the velocity. However, the computation of the splitting in a system with large Doppler width has been done for the collisionless case only.[9]

Nowadays a challenging task is to get tunable cw UV coherent radiation. Using stimulated Raman scattering, tunable radiation has been obtained in Na_2, Ne. The ions have higher energy levels, so there is hope of reaching short-wavelength radiation by Raman up-conversion in Ar^+ (Refs. 10 and 11). Thus strong-field effects are of interest 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 a nonlinear resonance frequency $\Omega_R(v)$, the frequency of the field at which the exact resonance between it and a particle with velocity v occurs. In Refs. 5–7 the linear dependence $\Omega_R = kv$ was considered. Here we examine a nonlinear function $\Omega_R(v)$ arising from the interaction with a strong monochromatic wave. The extremum of $\Omega_R(v)$ is of special interest in our consideration. The simplest nonlinearity is quadratic. If particles are concentrated near a velocity v_0, then one can interpret this dependence as a Taylor expansion of Ω_R about 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 leading term in it is quadratic or that Ω_R is constant. In brief, the quadratic nonlinearity seems sufficient to describe all the new effects associated with nonlinearity.

Let us calculate the spectrum of light absorbed (or emitted) by a monokinetic beam of particles with a 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 an arbitrary velocity distribution. The spectrum is given by the expression

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

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

$$\Phi(v_0,t) = e^{ikv_0 t - Dk^2t^3/3}. \tag{2}$$

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

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

The dephasing happens when the latter reaches π, and 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 $i\Gamma$ to Ω_R or multiply $\Phi(t)$ by $e^{-\Gamma t}$, where Γ is the inverse lifetime of the particles.

When only the integral of $\Phi(t)e^{-i\Omega t}$ 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 the polarization of the particles, $\rho(v)$, which is governed by the equation

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

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

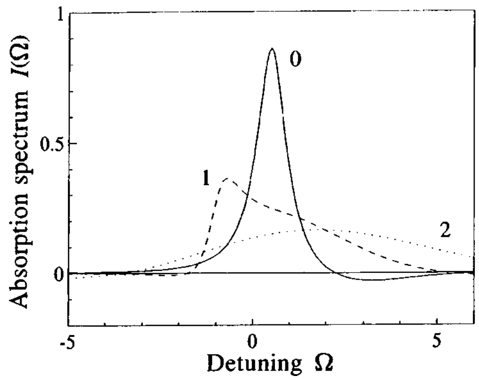

FIG. 1. Beam spectra with velocity $v_0 = n$, $n = 0,1,2$ (curve n) at $\Omega_R(v) = v^2/2$, $D = 2$.

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

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

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

We 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 on the left-hand side of (3). After integration over v one gets

$$I_B(\Omega, v_0) = \frac{\sqrt{2}}{\pi} \mathrm{Re} \sum_{n=0}^{\infty} \frac{e^{-z_0^2/2} H_{2n}(z_0)}{2^{2n} n! (2\beta n + x)} = \frac{1}{\pi} \mathrm{Re} \int_0^{\infty} d\tau \frac{e^{(\beta/2 - x)\tau}}{\sqrt{\cosh \beta\tau}} \exp\left(-\frac{1}{2} z_0^2 \tanh \beta\tau \right). \tag{4}$$

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

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

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

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

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

where $z=e^{\pm\pi i/4}(\Gamma+i\Omega)/\sqrt{8D|a|}$, $\Gamma(z)$ 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: a width Γ due to decay, and a 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 $\gamma(v_R)/(d\Omega_R/dv)(v_R)$, 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 $N_R\propto(\gamma(v_*)/(d^2\Omega_R/dv^2)(v_*))^{1/2}$, i.e., the field resonantly interacts with a maximal number of particles when $\Omega\simeq\Omega_R(v_*)$. The spectrum wings are asymmetric:

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

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

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

To calculate the spectrum $I(\Omega)$ one can replace the beam spectrum $I_B(\Omega,v_0)$ by $\delta(\Omega-\Omega_R(v_0))$ if the linear shift $\Omega'(v)=(d\Omega_R/dv)(v)$ does not change substantially within the domain $v-v_0\sim\gamma(v_0)/\Omega'(v_0)$. In a more general problem one should also require that the integrated intensity of the beam spectrum change insignificantly 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 that the two waves are 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

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

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

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

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

where $2\eta=\Omega-kv$, $2\Gamma_\pm=\Gamma_{13}\pm\Gamma_{23}$. One may think of this as there being 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 these two branches of hyperbola (8) independently and apply the theory developed above. Only two elements of the density matrix are mixed by the strong field. Then there are no cubic and higher shifts in the equation for ρ_{32}. One can get this equation by operating with $(\Omega_\mu-\Omega_{1B}+iDd^2/dv^2)$ on (7).

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

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

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

By analogy with (5) the spectrum is given by

$$I_\mu(\Omega_\mu)\propto\sum_{j=1}^{2}(-1)^j\,\mathrm{Re}\int dv\int_0^\infty dt\,\frac{\exp(i(\Omega_R^{(j)}-\Omega)t)}{\sqrt{\cosh\tau_j}}$$
$$\times\frac{(\Omega_R^{(j)}-\Omega_{1B})(\rho_2-\rho_3)+G^*\rho_{21}}{\Omega_R^{(1)}-\Omega_R^{(2)}}\exp(h_j(\Gamma_{j1}/\Gamma_2)^3(\tau_j-\tanh\tau_j)), \tag{10}$$

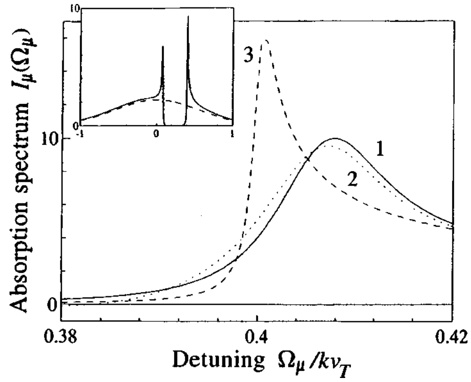

FIG. 2. Absorption spectra $I_\mu(\Omega_\mu)$ in arbitrary units. $(Dk^2)^{1/3}=6.3\times10^{-2}kv_T$: numerical calculations (curve *1*) and approximation formula (10) (curve *2*); and $D=0$ (curve *3*). $\Gamma_{ij}=10^{-3}kv_T$, $\Omega=0.3kv_T$, $|G|=0.2kv_T$, $k_\mu=0.8k$. The population of levels *1* and *2* coincide. The dashed curve in the inset corresponds to $G=0$.

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

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

The diffusion width Γ_2 of each narrow peak in the spectrum is found to be much less than the 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 a plasma (Ar^+, 488 and 514.5 nm, V-scheme with common short-lived level 2, $\Gamma_{23}=250$ MHz) was observed in Ref. 12. If we take the diffusion coefficient measured under similar conditions,[13] then $\Gamma_1=170$ MHz and $\Gamma_2=20$ 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 diffusional 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 extremum 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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