LETTER TO THE EDITOR

Diffusion-broadened lineshape under a strong field

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Abstract. The Bennett hole is analysed for a medium of two-level particles with weak collisions beyond the scope of the perturbation theory. In the limit of small homogeneous width, the analytical expression for the hole shape is of the form of a Bessel function. The result is compared with data obtained by the variational approximation and numerical calculation. It is shown that the approximation gives the correct width, but an incorrect shape.

A strong monochromatic field resonant with a dipole transition between two excited states of an ion equalizes their population. If the ions execute a thermal motion then only part of them hit the resonance due to the Doppler shift. Bennett holes appear in the velocity distribution of the upper and lower energy level populations [1], with the width of the Bennett hole being determined by different processes. The relaxation of field-induced polarization leads to a homogeneous width determined by the lifetime of the polarization. Effects of saturation under a strong light field give us an additional width proportional to the square root of the intensity of the light. This happens because the amplitude of the Bennett hole on any of the two levels grows, while the intensity of field is increasing for all ion velocities, but as the populations become closer this growth slows down. Moreover, during the lifetime of the excited level the ions interact with the field of other charged particles and change their velocities, then there is a diffusion width determined by the ratio of the transport collision frequency and the rate of the decay to underlying levels [2]. The theory of the diffusion or Coulomb broadening summarized in [4] predicts strong broadening when either of the two states is long-lived; however, the theory was mainly concerned with the case of a weak field and ignored field broadening of the holes due to saturation.

Under the field of a relatively weak travelling electromagnetic wave the Bennett hole shape for a long-lived level is cusp-like [2]. In the case of strong saturation the problem becomes more complicated. Even when one level is short-lived the exact solution has the form of integrals with spheroidal functions, then some approximations are applied for experimental data processing. The two-parametric variational approximation [3] gives the width of the resonance with an error of less than 2%. However, this approximation gives the wrong shape, especially in the limit of a weak field.

The aim of this paper is to calculate the shape of the Bennett hole in a two-level system with strongly distinct level widths when diffusion and field broadening are comparable. The long-lived upper level is typical for continuous ion lasers, the metastable lower state often occurs in the absorbing transition of an anti-Stokes Raman laser—a source of tunable short-wave coherent radiation, so the problem posed above is of interest for experiments [5].

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For definiteness let us consider a two-level system with an upper level m and lower level n, where the lower level is long-lived. To take into account the Coulomb diffusion in the velocity space one should use the density matrix formalism. Let us consider the case when the diffusion width of the Bennett hole on the upper level $w_{mD} = v_T(\nu/2\Gamma_m)^{1/2}$ is small in comparison with the homogeneous width $w_H = \Gamma/k$. Here Γ_j is the relaxation constant of level j; Γ is the relaxation constant of polarization ρ_{mn}, k is the wavenumber of the light field; v_T is thermal velocity of the ions, $v_T = \sqrt{2T/m}$, T, m are the temperature in units of energy and the mass of the ions, respectively; ν is the Coulomb effective transport collision frequency. This case is realistic for the upper level of a Raman ionic laser [5]. Then one can neglect the diffusion operators in the Fokker–Planck equations for the density matrix elements ρ_{mm}, ρ_{mn} [4], and the distribution of the lower state population $\rho_n \equiv \rho_{nn}$ over $v_\parallel$ satisfies the second-order differential equation

$$\left(\Gamma_n + \frac{2\Gamma|G|^2}{\mathcal{T}^2 + (\Omega - kv_\parallel)^2}\right)\rho_n = \frac{\nu v_T^2}{2}\frac{\mathrm{d}^2}{\mathrm{d}v_\parallel^2}\rho_n + q_n + \frac{2\Gamma|G|^2}{\mathcal{T}^2 + (\Omega - kv_\parallel)^2}\frac{q_m}{\Gamma_m}\,. \tag{1}$$

Here q_j is the excitation rate of level j,

$$G = \boldsymbol{E}\boldsymbol{d}_{mn}/2\hbar \qquad \Omega = \omega - \omega_{mn}$$

where $\boldsymbol{E}$ and ω are the amplitude and the frequency of the field, respectively, $\boldsymbol{d}_{mn}$ is the matrix element of the dipole moment, ω_{mn} is the Bohr frequency of the m–n transition; $\mathcal{T}$ is an auxiliary width $\mathcal{T}^2 = \Gamma^2 + 2\Gamma|G|^2/\Gamma_m$. We also neglect the frictional force which is possible while the concentration of charge carriers in the plasma is moderate. If $\rho_n(v_\parallel)$ has a width w, then for an estimate one can substitute $1/w$ for each derivative $\mathrm{d}/\mathrm{d}v_\parallel$. The diffusion width w_{nD} appears from the condition of compensation of the term $\Gamma_n\rho_n$ and the diffusion term. Then the friction force term $\nu v_\parallel(\mathrm{d}\rho_n/\mathrm{d}v_\parallel)$ has a term of order $(\nu\Gamma_j)^{1/2}v_\parallel\rho_n$ and we can neglect it while $\nu \ll \Gamma_n$ (the velocity change during the lifetime Γ_j^{-1} due to the frictional force is of the order of $v_\parallel\nu/\Gamma_j$ which is small compared with the diffusional change $v_T(\nu/\Gamma_j)^{1/2}$).

When $|\Omega - kv_\parallel| \gg \mathcal{T}$ we can neglect $\mathcal{T}^2$ in the denominators of (1), so its solution has the form

$$\rho_n = \frac{q_n}{\Gamma_n} + \left(\frac{q_m}{\Gamma_m} - \frac{q_n}{\Gamma_n}\right)A\sqrt{\mathrm{i}X}\left(CK_\alpha(X) - S_{-3/2,\alpha}(\mathrm{i}X)\right) \tag{2}$$

where $X = |\Omega - kv_\parallel|/kw_{nD}$, $A = (w_F/w_{nD})^2$ and w_F is the field width of the Bennett hole due to saturation $w_F^2 = 2\Gamma|G|^2/\Gamma_n k^2$, $\alpha = (A + \frac{1}{4})^{1/2}$, $S_{\mu,\nu}(z)$ is Lommel's function [6] and C is the constant defined from joining together (2) and the solution for ρ_n at small X.

Since the upper level is much wider than the lower one, $\Gamma_n \ll \Gamma_m$, in the case of a strong field, $|G|^2 \gg \Gamma\Gamma_n$, we have $w_F \gg \mathcal{T}/k$. If $X \simeq \mathcal{T}$ we can neglect Γ_n in the brackets in the left-hand side of (1). Then ρ_n has the form of a sum of some constant and two hypergeometric functions ${}_2F_1$ whose parameters depend only on α. It gives us a hope that it is possible to join these two asymptotics for $\rho_n(v_\parallel)$. But there is one rougher but simpler way of doing this—under these conditions the width of the Bennett hole is much greater than $\mathcal{T}$, so we can define a constant C from the condition of nonsingular behaviour of (2) at $X = 0$. Then

$$C = \frac{2^{-3/2}}{\pi}\Gamma\left(\frac{-\frac{1}{2} - \alpha}{2}\right)\Gamma\left(\frac{-\frac{1}{2} + \alpha}{2}\right)\cos\left(\pi\frac{\frac{3}{2} + \alpha}{2}\right)\mathrm{e}^{-\mathrm{i}\alpha\pi/2} \qquad \rho_n|_{X=0} = \frac{q_m}{\Gamma_m}\,.$$

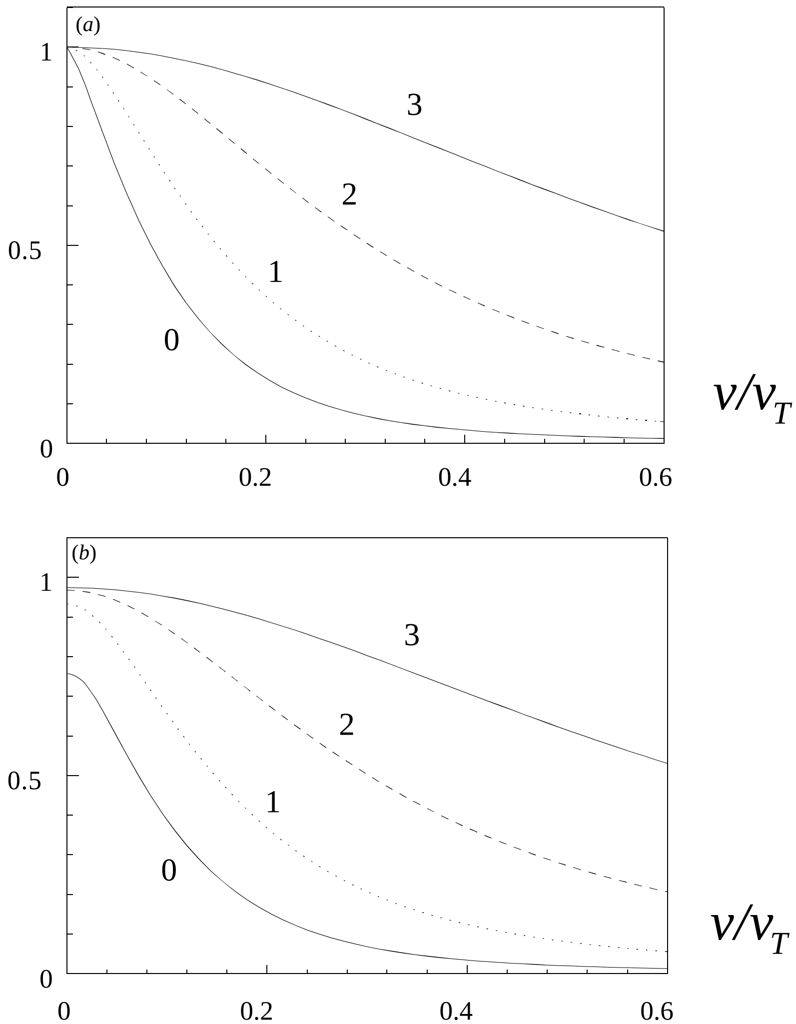

Figure 1. Bennett hole $(\rho_n - q_n/\Gamma_n)/(q_m/\Gamma_m - q_n/\Gamma_n)$: ($a$) solution (2) and ($b$) the numerical calculation. Curves $n = 0, 1, 2, 3$ correspond to $|G| = 10^{n/3-2} k v_T$, $\Gamma_n = 10^{-3} k v_T$, $\Gamma_m = 4 \times 10^{-2} k v_T$, $\Gamma = (\Gamma_n + \Gamma_m)/2$, $\nu = 10^{-5} k v_T$.

Solution (2) contains the imaginary unit i, nevertheless, it is real and involves Lorentzian and cusp-like contours as limiting cases:

$$\rho_n = \frac{q_n}{\Gamma_n} + \left(\frac{q_m}{\Gamma_m} - \frac{q_n}{\Gamma_n}\right) \begin{cases} \dfrac{1}{1 + X^2/A} & w_{nD} \ll w_F \\ e^{-X} & w_{nD} \gg w_F \, . \end{cases}$$

Figure 1(a) represents solution (2) and numerical solutions of four connected diffusion equations for the whole density matrix, which were found by a 4×4 matrix sweep method from the one-dimensional boundary problem (figure 1(b)). The values of the density matrix at the boundary $v_\parallel = \pm 4 v_T$ were chosen according to analytical Lorentzian asymptotics. In figure 1(a) one can see a transition from a cusp-like contour to a Lorentzian (from curve 0 to curve 3). Curves 1, 2, 3 in (a) and (b) almost coincide, the difference between (a) and

(b) in the vicinity of the hole centre $|\Omega - kv_{\|}| \simeq \mathcal{T}$ under weak field conditions (curve 0) can be explained by a non-zero homogeneous width and the influence of collisions while the ion is on the short-lived level.

The shape of solution (2) as a function of X depends only on the parameter $A = (w_F/w_{nD})^2$, so the half-width v_L at the proportion $L, 0 < L < 1$ of the total height is a homogeneous function of w_{nD} and w_F with the degree being equal to 1. It means that if we multiply w_{nD} and w_F by a number $y > 0$ then the width v_L will also be multiplied by y. Then different level curves of v_L with the same L on the (w_F^2, w_{nD}^2) plane are homothetic with point $w_F = 0, w_{nD} = 0$ as the centre of the extension. For example, the half-width at half-maximum

$$v_{1/2} = \begin{cases} w_F\left(1 + \dfrac{1}{4}\left(\dfrac{w_{nD}}{w_F}\right)^2\right) & w_{nD} \ll w_F \\ w_{nD}\log 2 & w_{nD} \gg w_F \end{cases}$$

can be approximated as

$$v_{1/2} \simeq \left(w_F^2 + w_{nD}^2/2\right)^{1/2} . \tag{3}$$

It should be noticed that the simplicity of expression (3) comes from the following approximate equality $\sqrt{2}\log 2 \simeq 1$ (if $w_F = 0$ because of (3) we have $v_{1/2} = w_{nD}/\sqrt{2} \simeq w_{nD}\log 2$). In [3] the Lorentzian profile with uncertain height and width is explored as a probe function for the variational approximation. The width of the approximating Lorentzian satisfies the fourth-degree algebraic equation

$$v_{1/2}^4 - (w_H^2 + w_F^2 + w_{nD}^2/2)v_{1/2}^2 - 2w_H w_{nD}^2 v_{1/2} - 3w_H^2 w_{nD}^2/2 = 0 . \tag{4}$$

Its solution under the condition $w_{nD}, w_F \gg w_H = \Gamma/k$ is in agreement with (3), i.e. the half-width at half-maximum of the Lorentzian obtained from the variational method almost coincides with the same parameter as the solution of (2). The solution of (4) gives level curves as segments of straight lines on the (w_F^2, w_{nD}^2) plane which become parallel (and thus, homothetic) when $w_{nD}, w_F \gg w_H$.

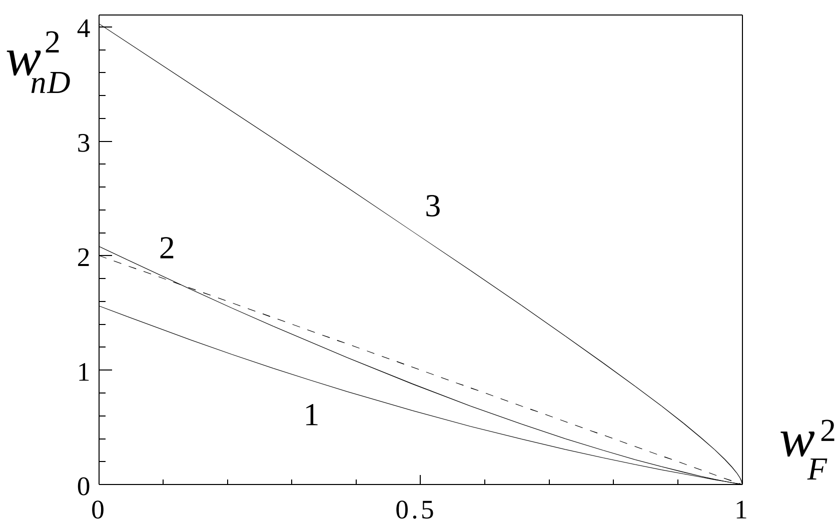

Figure 2. Level curves of $v_{n/4}, n = 1, 2, 3$ of solution (2) corresponding to $v_{n/4} = (4/n - 1)^{1/2}$. The broken curve is approximation (3) for curve 2.

The values of $v_{n/4}, n = 1, 2, 3$ in figure 2 are chosen in such a way that curves 1, 2, 3 would coincide if the shape of the Bennett hole were Lorentzian. The distance between curves is the measure of the shape deviation from Lorentzian. The shape differs from Lorentzian more strongly at the centre of the hole (curve 3 deviates from curve 2 more than curve 1) and in the case of small field broadening in comparison with that of diffusion broadening. When the field broadening is high, $w_F \gg w_{nD}$, curves converge to each other because the field masks the diffusion broadening.

Let us introduce a probe electromagnetic wave which propagates in the opposite direction to the strong one and interacts resonantly with the same transition. If we neglect the population on the short-lived level then the shape of the nonlinear resonance in the spectrum of the probe field is similar to the shape (2) of the Bennett hole, one should substitute only $-\Omega_\mu/k$ for $v_\|$, where Ω_μ is the probe field detuning. The same Bennett hole happens under a standing wave field with zero detuning from resonance, one should substitute only $2|G|^2$ for $|G|^2$, $\sqrt{2}w_F$ for w_F, where the amplitude of one of the two travelling waves is used for the definition of G.

Thus the shape of the Bennett hole on the long-lived level broadened by Coulomb diffusion and the shape of the nonlinear resonance in the probe field spectrum are obtained. The expressions are valid while the homogeneous hole width w_H is small compared with the diffusion w_{nD} and field w_F widths with an arbitrary ratio of the two widths. The expression for the shape contains the smooth transition between a cusp-like exponential and a Lorentzian shape. The width is in agreement with the variational approximation [3].

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