# Narrowing of the Bennett Hole In Collisional Plasma

E. V. Podivilov, D. A. Shapiro, and M. G. Stepanov
*Institute of Automation and Electrometry, Novosibirsk, 630090, Russia*
(Received 26 July 1994)

The profile of a Bennett hole, induced by a laser field, in the ionic distribution of a collisional plasma is calculated. The influence of Chandrasekhar's dependence of the coefficients of velocity space transport on the profile is included in the calculation for the first time. It is found that the hole narrows down as the field-detuning frequency increases. The physical cause for this effect is the falling dependence of the Coulomb collision frequency on the ionic velocity. Estimations show that the effect is quite observable under conditions of a high-current gas-discharge plasma.

PACS numbers: 52.20.Hv, 42.62.Fi, 52.80.–s

The nonlinear spectroscopic technique as a plasma diagnostic tool is a subject of considerable recent interest because of its novelty and potential in basic and applied plasma physics. Of particular interest are the spectroscopic effects of Coulomb scattering [1], since it differs radically in appearance from binary collisions in gases [2,3]. In linear absorption spectra the Doppler width usually exceeds collisional broadening, so it masks the effects of the scattering. However, the velocity distribution of particles interacting with a monochromatic field near the resonance is changed significantly by the scattering. If the frequency $\omega$ of the field is near resonant with Bohr's frequency $\omega_{mn} = (E_m - E_n)/\hbar$ between the levels $m$ and $n$ of atoms with velocity $\boldsymbol{v}$ that satisfies the condition $\boldsymbol{k}\boldsymbol{v} = \omega - \omega_{mn} \equiv \Omega$, then the induced transitions $m \leftrightarrow n$ occur ($\boldsymbol{k}$ denotes the wave vector). As a result, a narrow peak or dip (Bennett hole [4]) arises in the velocity distribution of level populations. The steady-state profiles of holes are controlled by both the homogeneous width of the transition $m$-$n$ and the scattering processes with a change in velocity. By measuring the width, shift, and asymmetry of nonequilibrium structures in the distribution with the help of the probe-field technique one can extract information about the scattering.

Previous calculations of Coulomb broadening [1,5] were based on the assumption of constancy of the diffusion coefficient in the velocity space. This assumption is applicable when the detuning frequency $\Omega$ is less than the Doppler width $kv_T$, where $v_T = \sqrt{2T_i/m}$ is the thermal velocity of ions and $m$ is the ion mass. The constant diffusion model was enough to interpret Lamb dip measurement, in an argon laser plasma or one with near-resonance pumping. In contrast, farther away from the resonance, at $\Omega > kv_T$, one must use Chandrasekhar's velocity dependence of the diffusion tensor, which complicates the evaluation. One way around this problem was shown in the linear theory of Dicke narrowing in the ionic spectrum, which was developed for low intensity of the incident wave [6].

In the present Letter we study a first-order nonlinear problem, the influence of collisions in a plasma on the profile of the Bennett hole. The calculation presented below for nonlinear structure in the velocity distribution is performed on the basis of density matrix formalism [3,7]. First, we derive the formula for the distribution, then analyze it under some simple but realistic conditions, and at the end estimate whether the effect obtained is observable in experiment. In particular, we show that the width of the hole decreases with the detuning of the electromagnetic field from the resonance. By observing the effect, one could directly measure the diffusion tensor. It would be helpful to confirm our understanding and offer a scope for new diagnostic methods.

The diffusion coefficient as a function of the velocity was measured for a $Q$-machine plasma at low density by Bowles, McWilliams, and Rynn [8]. The effect of the transient spread of a Bennett hole in the distribution of a tagged ion population in a metastable state was exploited. However, that effect cannot be used for denser plasmas, because the diffusion time becomes smaller and the measurement requires a faster registration technique. In this paper we shall examine the stationary hole shape. Analysis of its width is useful for diagnostics at high as well as low density.

To develop the theory of nonequilibrium structures in the velocity distribution, let us describe the two-level subsystem of a probe ion in an ideal nondegenerate plasma by the spectroscopic density matrix $\rho_{ij}(\boldsymbol{r}, \boldsymbol{v}, t)$. This matrix over internal states $i, j$ is at the same time the Wigner function of position $\boldsymbol{r}$ and velocity $\boldsymbol{v}$. It has been shown [5,6] that elements of the matrix satisfy the quantum kinetic equation with the classical Landau collision term. This equation makes it possible to analyze resonant transitions between levels in the field of the traveling wave $\boldsymbol{E} = \boldsymbol{E}_0 \exp(-i\omega t + i\boldsymbol{k}\boldsymbol{r}) + \text{c.c.}$

Denoting the relaxation constants of level populations as $\Gamma_j$ and that of the coherence as $\Gamma$, we write steady-state equations for off-diagonal $\rho_{mn}(\boldsymbol{r}, \boldsymbol{v}, t) = \rho(\boldsymbol{v}) \exp(-i\Omega t + i\boldsymbol{k}\boldsymbol{r})$ and diagonal $\rho_{jj} \equiv \rho_j$ elements

0031-9007/95/74(20)/3979(4)$06.00  

of the density matrix as

$$(\Gamma - i\Omega + i\boldsymbol{k}\boldsymbol{v})\rho - \nu\hat{V}\rho = iG\Delta N,$$
$$\Gamma_j\rho_j - \nu\hat{V}\rho_j - \lambda_j(\boldsymbol{v}) = \mp 2\,\mathrm{Re}(iG^*\rho), \tag{1}$$

where the upper (lower) sign corresponds to the upper (lower) level. Here $\Delta N \equiv \rho_m - \rho_n$, $G = \boldsymbol{E}_0\boldsymbol{d}_{mn}/2\hbar$, $\boldsymbol{d}_{mn}$ is the matrix element of the dipole moment,

$$\nu = \frac{16\sqrt{\pi}LN_i(Z_a e^2)^2}{3m^2 v_T^3} \tag{2}$$

is the Coulomb collision frequency, $Z_a e$ is the charge of a probe ion, and $L$ is the Coulomb logarithm. We assume that there are only singly charged ions in the plasma; $N_i$ is their concentration. The excitation of ionic levels $j = m, n$ usually occurs from the ground or metastable state, so we are justified in assuming the shape of the excitation function $\lambda_j(\boldsymbol{v})$ to be Maxwellian

$$\lambda_j(\boldsymbol{v}) = Q_j W(\boldsymbol{v}), \qquad W(\boldsymbol{v}) = \frac{1}{v_T^3\pi^{3/2}}\exp\left(-\frac{v^2}{v_T^2}\right), \tag{3}$$

where $Q_j$ is the excitation rate of level $j$.

For a plasma in a state close to thermodynamic equilibrium with ionic temperature $T_i$, the distribution of buffer particles has a Maxwellian shape. Then it is possible to write an explicit expression for the collision operator $\hat{V}$ involving the dynamic friction and the diffusion in velocity space,

$$\hat{V} = \frac{1}{2}\frac{\partial}{\partial\xi_\alpha}\Phi_{\alpha\beta}\left(\frac{\partial}{\partial\xi_\beta} + 2\xi_\beta\right), \tag{4}$$

where $\xi_\alpha = v_\alpha/v_T$ is the $\alpha$ component of the dimensionless velocity and the differential operator $\partial/\partial\xi_\alpha$ acts on the right. The functions $\Phi_l(\xi)$ and $\Phi_{\mathrm{tr}}(\xi)$, occurring in the expression for the diffusion tensor

$$\Phi_{\alpha\beta} = \Phi_l(\xi)\frac{\xi_\alpha\xi_\beta}{\xi^2} + \Phi_{\mathrm{tr}}(\xi)\left(\delta_{\alpha\beta} - \frac{\xi_\alpha\xi_\beta}{\xi^2}\right), \tag{5}$$

can be written as single integrals

$$\Phi_l(\xi) = 3\int_0^1 \lambda^2 e^{-\lambda^2\xi^2}\,d\lambda,$$
$$\Phi_{\mathrm{tr}}(\xi) = \frac{3}{2}\int_0^1 (1 - \lambda^2)e^{-\lambda^2\xi^2}\,d\lambda. \tag{6}$$

The velocity distribution of populations $\rho_j$ of both levels $j = m, n$ needs to be found. To simplify the analysis, we assume the electromagnetic field to be weak $|G|^2 \ll \Gamma\Gamma_j$ and will neglect the effects of strong saturation. The right sides of Eqs. (1) can therefore be regarded as a perturbation. The formal solution of (1) in terms of operator exponents is

$$\rho_j^{(0)} = \frac{Q_j}{\Gamma_j}W(\boldsymbol{v}), \qquad \rho^{(0)} = 0; \qquad \Delta N(\boldsymbol{v}) = \rho_m^0(\boldsymbol{v}) - \rho_n^{(0)}(\boldsymbol{v}) = N_{mn}W(\boldsymbol{v});$$
$$\rho^{(1)}(\boldsymbol{v}) = -iG\int_0^\infty dt\,\exp(-\hat{A}t + \nu\hat{V}t)\Delta N(\boldsymbol{v}), \qquad \hat{A} = \Gamma - i\Omega + i\boldsymbol{k}\boldsymbol{v}, \tag{7}$$
$$\rho_j^{(1)}(\boldsymbol{v}) = \mp\int_0^\infty dt\,\exp(-\Gamma_j t + \nu\hat{V}t)2\,\mathrm{Re}[iG^*\rho^{(1)}(\boldsymbol{v})]. \tag{8}$$

It remains to write out the explicit solution and analyze its limiting cases. For this purpose we must calculate the commutators of operators $\hat{A}$ and $\hat{V}$ and get, in the general case, the infinite series. Fortunately, the relation $[[[\hat{V},\hat{A}],\hat{A}],\hat{A}] = 0$ allows us to break the series within the first order in $\nu$:

$$\rho_j^{(1)}(\boldsymbol{v}) = \mp 2|G|^2\Delta N(\boldsymbol{v})\,\mathrm{Re}\int_0^\infty dt\int_0^\infty d\tau\,\exp[-\Gamma_j\tau - (\Gamma - i\Omega + ikv_z)t]$$
$$\times\exp\left[\frac{\nu}{2}[-\Phi_{zz}(\boldsymbol{\xi})k^2v_T^2(t^2\tau + t^3/3) + 2ikv_z\Phi_l(\xi)(2t\tau + t^2)] + O(\nu^2)\right], \tag{9}$$

where the axis $z$ is chosen along the wave vector $\boldsymbol{k}$. The function

$$\Phi_{zz}(\boldsymbol{\xi}) = [\Phi_l(\xi) - \Phi_{\mathrm{tr}}(\xi)](\xi_z^2/\xi^2) + \Phi_{\mathrm{tr}}(\xi) \tag{10}$$

is the $zz$ component of the diffusion tensor. This function depends on two variables $\xi \equiv |\boldsymbol{\xi}|$ and $\xi_z$. The integrand in (9), obtained by the expansion, is inappropriate at long times $t, \tau$. Consequently, for application of this expansion it is necessary to have the main contribution to integral (9) in the domain where the quadratic terms in $\nu$ remain small. Since $|\partial\Phi_{\alpha\beta}/\partial\xi_\gamma| < |\Phi_{\alpha\beta}|$, estimation of those terms gives the following applicability conditions of the expansion (9):

$$\alpha = \frac{\nu}{\Gamma_j} \ll 1, \qquad \beta = \frac{\Gamma}{kv_T} \ll 1. \tag{11}$$

The latter inequality means the Doppler limit; the former coincides with the condition of smallness of the diffusion broadening compared to the Doppler width. The collision frequency in a high-current gas-discharge plasma is about $\nu \sim 10^5$–$10^7\ \mathrm{s}^{-1}$ (see [1]). It is by a few orders of magnitude less than the Doppler width $kv_T \sim 10^{10}\ \mathrm{s}^{-1}$ and, as a rule, less than the relaxation rates $\Gamma \sim 10^8$–$10^9\ \mathrm{s}^{-1}$ and $\Gamma_j \sim 10^6 - 10^9\ \mathrm{s}^{-1}$. That is why we may use of small parameters $\alpha, \beta$ and it is enough to keep only terms linear in $\nu$.

Taking an integral over $\tau$ we obtain the correction to the distribution function as a single integral over $t$,

$$\rho_j^{(1)}(\boldsymbol{v}) = \mp 2|G|^2 \Delta N(\boldsymbol{v}) \,\mathrm{Re} \int_0^\infty \frac{\exp[-\hat{A}t - \Phi_{zz}(\boldsymbol{\xi})\nu k^2 v_T^2 t^3/6 + i\nu k v_z \Phi_l(\xi) t^2]}{\Gamma_j + \Phi_{zz}(\boldsymbol{\xi})\nu k^2 v_T^2 t^2/2 - 2i\nu k v_z \Phi_l(\xi) t}\, dt\,. \tag{12}$$

The integrand in (12) regarded as a function of $t$ at $\Omega = k v_z$ is maximal at $t = 0$ and decreases with $t$. The shape of the distribution depends on a few parameters. Considering conditions (11) and the inequality $\Gamma_j \leq \Gamma$, we can distinguish two limiting cases of parameters,

$$\nu k^2 v_T^2 \ll \Gamma^2 \Gamma_j \qquad (\alpha \ll \beta^2)\,, \tag{13}$$

$$\Gamma^2 \Gamma_j \ll \nu k^2 v_T^2 \qquad (\alpha \gg \beta^2)\,. \tag{14}$$

For the domain (13) integral (12) is accumulated at $t \leq T_1 = 1/\Gamma$. The diffusion and friction cause the Bennett hole to change its width and position somewhat. For the limit (14) integral (12) is gathered at $t \leq T_2 = (\alpha/2)^{-1/2}/k v_T$. Diffusion and friction effects control the width and shift of the hole. The width is defined by the maximum among the homogeneous width $\Delta v_H = \Gamma/k$ and diffusion width $\Delta v_D = v_T(\nu/\Gamma_j)^{1/2}$. Thus the scattering has stronger impact on the shape of the hole, while the relaxation constants of selected levels satisfy inequality (14).

Let us find the width as a function of detuning in this limit $\epsilon(\boldsymbol{\xi}) = 2\beta^2/\alpha\Phi_{zz}(\boldsymbol{\xi}) \ll 1$. It is convenient to write the expression for the nonlinear correction to the Maxwellian distribution as a convolution of two functions

$$\rho_j^{(1)}(\boldsymbol{v}) = W(\boldsymbol{v}) \int_{-\infty}^{\infty} f(x - x') g_j(x')\, dx', \qquad x = k v_z - \Omega\,, \tag{15}$$

where

$$g_j(x) = \mp \frac{\pi |G|^2 N_{mn}}{\Gamma \Gamma_j} \frac{\epsilon}{\sqrt{\mu}} \exp\left(-\frac{|x|\sqrt{\mu} + x\sigma}{\Gamma}\right), \tag{16}$$

$$\sigma(\boldsymbol{\xi}) = \frac{2\Gamma \xi_z \Phi_l(\xi)}{k v_T \Phi_{zz}(\boldsymbol{\xi})}\,, \qquad \mu(\boldsymbol{\xi}) = \epsilon(\boldsymbol{\xi}) + \sigma^2 \simeq \epsilon(\boldsymbol{\xi})\,,$$

$$f(x) = \mathrm{Re} \int_0^\infty \frac{dt}{\pi} \exp[-(\Gamma + ix)t - \Phi_{zz}(\boldsymbol{\xi})\nu k^2 v_T^2 t^3/6 + i\nu k v_z \Phi_l(\xi) t^2]\,. \tag{17}$$

In the limit $\epsilon \ll 1$ the width of the function $f(x)$ is much less than that of $g_j(x)$. Accordingly, the central part of the distribution is given by expression (16), $\rho_j^{(1)} \simeq g_j(x)$ at $|x| < \Gamma/\sqrt{\epsilon}$. Meanwhile the function $g_j(x)$ decreases exponentially with $|x|$, so the asymptotic of the distribution $\rho_j^{(1)}(v_z)$ at $|x| \gg \Gamma/\sqrt{\epsilon}$ is given by a narrow but slowly decreasing function $f(x)$ and has a Lorentzian shape.

The most interesting feature of the distribution obtained is the narrowing of its central part as the detuning $\Omega$ increases, due to the falling velocity dependence of the collision frequency. One can estimate the half-width by substituting the extreme value of the longitudinal velocity $v_z = \Omega/k$ into all components of the diffusion tensor,

$$\Delta x \simeq \frac{\Gamma}{\sqrt{\mu}} \simeq k v_T \sqrt{\frac{\nu \Phi_{zz}(\xi, \Omega/k v_T)}{\Gamma_j}}\,.$$

This quantity is proportional to the square root of the $zz$ component of the diffusion tensor. It decreases with detuning $\Omega$. The effect is absent in the model of constant collision frequency [9,10].

To describe this effect quantitatively we calculate the $\Omega$ dependence of $x^2$ averaged over the distribution (15). Keeping only the terms linear in parameters $\alpha$ and $\beta$ we obtain

$$F_j(\Omega) = \int d^3 v\, x^2 \rho_j^{(1)}(\boldsymbol{v}) \simeq \mp \frac{2|G|^2 N_{mn}}{\Gamma_j} \left\{ \Gamma + \frac{\nu k^2 v_T^2}{\Gamma_j} \mathrm{Re} \int_0^\infty dt\, \overline{\Phi}_{zz}(t) \exp[-(\Gamma - i\Omega)t]\,, \right\} \tag{18}$$

$$\overline{\Phi}_{zz}(t) = \int d^3 v\, W(\boldsymbol{v}) \Phi_{zz}(\boldsymbol{\xi}) \exp(-i\xi_z t) = \int_0^{1/\sqrt{2}} 6y^2 dy\, \exp\left[-(1 - y^2)\left(\frac{t k v_T}{2}\right)^2\right].$$

The first term on the right-hand side of (18) is independent of $\Omega$ and corresponds to the contribution of the Lorentzian wings of the distribution. The second term describes the collisional broadening and decreases with $|\Omega|$.

Under the approximations the line shape is Voigt's contour. The dependence of the variance $D = \langle [x - \langle x \rangle]^2 \rangle$ on detuning $\Omega$ consists of two terms: the contribution of the wings, which increases with detuning, and the decreasing contribution of the diffusion broadening. At small detuning $|\Omega| \ll kv_T$ we have the explicit expression

$$D \simeq \frac{\Gamma k v_T}{\sqrt{\pi}}\left[1 + \left(\frac{\Omega}{kv_T}\right)^2\right] + \frac{\nu(kv_T)^2}{2\Gamma_j}\left[\frac{3\pi}{2} - 3 - \left(\frac{\Omega}{kv_T}\right)^2\left(15 - \frac{9\pi}{2}\right)\right].$$

Summarizing the results, we conclude that the Bennett hole narrows as detuning increases. The physical reason for the effect is that the width of the hole is not affected by the homogeneous width $\Delta v_H$, but depends on the change in velocity due to diffusion $\Delta v_D = v_T\sqrt{\nu\tau_j}$, while $\Delta v_D \gg \Delta v_H$. The excited state lifetime $\tau_j = \Gamma_j^{-1}$ determines the stationary width of the hole. While the detuning $\Omega$ of the field remains inside the Doppler contour, the field interacts only with a group of slow ions $|v_z| \ll v_T$. When we turn the field off the resonance, the light interacts with faster excited ions. The collision frequency of those ions with ions in the ground state $\nu$ is smaller, therefore the width decreases. It would be more appropriate to call this effect a decrease of the broadening instead of a narrowing.

To observe the narrowing experimentally, probe-field spectroscopy can be used. One should measure the near-resonant absorption or gain of a weak probe wave as a function of its detuning on the same or an adjacent transition. Another possibility is recording the spectrum of spontaneous emission in the presence of a strong resonant continuous field. Let us estimate two alternative effects independent of the detuning, namely, the Stark broadening and the change velocity due to the interaction with electrons. The Stark effect is quadratic in the Ar II spectrum. Its value (Ref. [1], p. 139) is about 100 MHz, which is small compared with both the homogeneous width $\Gamma$ and the diffusion width $kv_T\sqrt{\nu/\Gamma_j}$. The velocity change owing to scattering on electrons is as small as the ratio of the ion and electron thermal velocities $v_{Ti}v_{Te} \sim 10^{-2}$ ([1], p. 155). The ion-ion collision frequency decreases by no more than a factor of 2–3 at the detuning about $kv_T$; therefore neither alternative broadening mechanism can compensate the effect in question. The measured diffusion broadening in a low-temperature argon plasma at zero detuning is by a factor of 3–4 (see Ref. [1], p. 199). At the detuning $\Omega \sim kv_T$ the narrowing factor expected is more than 10%, so this effect seems observable (see Fig. 1).

We thank S. A. Babin and S. G. Rautian for stimulating discussions, E. G. Shapiro for excellent assistance in computation, and A. V. Shafarenko for valuable comments. The research described in this publication was made possible in part by Grant No. RCN000 from the International Science Foundation.

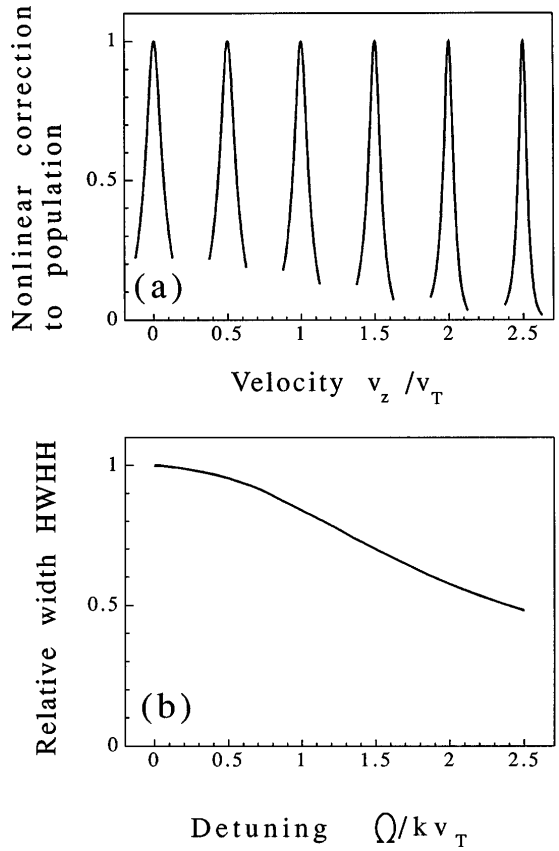


FIG. 1. (a) Distribution $\rho_j^{(1)}(v_z)$ of $j$-level population in ion velocity $v_z$ within the first order of perturbation theory in the field intensity. Different curves correspond to distinct values of detuning $\Omega$ (from left to right $\Omega = 0, 0.5, 1, 1.5, 2, 2.5$). Parameters are assumed to be $\Gamma = 10^{-2}kv_T$, $\Gamma_j = 10^{-3}kv_T$, and $\nu = 10^{-5}kv_T$. (b) Half width at half maximum of contour $\rho_j^{(1)}(v_z)$ as a function of detuning $\Omega$ normalized to unit magnitude.

[1] S. A. Babin and D. A. Shapiro, Phys. Rep. **241**, 119 (1994).
[2] P. R. Berman, Adv. At. Mol. Phys. **13**, 57 (1977).
[3] S. G. Rautian and A. M. Shalagin, *Kinetic Problems of Nonlinear Spectroscopy* (Elsevier, Amsterdam, 1991).
[4] W. R. Bennett, Jr., Phys. Rev. **126**, 580 (1962); Appl. Opt. Suppl. **1**, 24 (1962).
[5] G. I. Smirnov and D. A. Shapiro, Zh. Eksp. Teor. Fiz. **76**, 2084 (1979) [Sov. Phys. JETP **49**, 1054 (1979)].
[6] E. V. Podivilov, A. I. Chernykh, and D. A. Shapiro, Zh. Eksp. Teor. Fiz. **105**, 1214 (1994) [Sov. Phys. JETP **78**, 653 (1994)].
[7] S. Stenholm, Phys. Rep. **43**, 151 (1978).
[8] J. Bowles, R. McWilliams, and N. Rynn, Phys. Rev. Lett. **68**, 1144 (1992).
[9] S. G. Rautian, Zh. Eksp. Teor. Fiz. **51**, 1176 (1966) [Sov. Phys. JETP **24**, 788 (1967)].
[10] L. A. Gel'medova and D. A. Shapiro, J. Mod. Opt. **38**, 573 (1991).