

## AMPLITUDE MODULATION AND NON-ADIABATICITY IN THE STARK BROADENING OF HYDROGEN LINES IN A PLASMA

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The deviations from the quasistatic (Holtsmark) profile of hydrogen spectral lines, due to thermal motion of the charged plasma particles, are calculated. The effects connected with rotation of the vector of the electric microfield, and leading to non-adiabaticity and to amplitude modulation of the light wave radiated by the atom, are considered for the first time. It is shown that amplitude modulation and the non-adiabaticity of the rotation play a decisive role in the deviation from the Holtsmark line profile. For example, for the "binary" wing of the  $L_{\alpha}$  line the correction for the thermal motion with allowance for all the indicated effects exceeds by almost two orders of magnitude the heretofore considered correction due only to phase modulation. The theory is generalized to include the multiple case, which takes into account simultaneous action of all the perturbing particles on the radiating atoms, and the total profile of the line is obtained with allowance for deviations from quasi-static behavior. The results of the calculation are used to analyze and to correct the criteria for quasi-static and adiabatic approximations, on which the theory of the Stark broadening of lines is based. A number of other effects related to those considered is discussed, namely, the role of ions in the broadening of the central Stark component, amplitude modulation at an arbitrary broadening mechanism, etc.

### 1. INTRODUCTION

THE basis for the analysis of the broadening of spectral lines of atoms in a plasma has for a long time been the model of an oscillator with a variable frequency (see, for example<sup>[1]</sup>). In this case allowance for the rapidly-alternating electronic component of the electric microfield was carried out within the framework of the impact approximation, whereas the contribution of the slow ionic component was taken into account in the quasi-static approximation. Further development of the impact approximation<sup>[2]</sup> proceeded mainly along the line of foregoing the oscillator model, whereas the quasi-static approximation was connected as before with this model<sup>1)</sup>. For the broadening of the hydrogen lines, a fundamental role in this model is played by Holtsmark's quasistatic theory<sup>[4]</sup>. Subsequent generalizations of this theory were connected with allowance for effects of the Debye screening and of ion-ion correlations<sup>[5-7]</sup>, the inhomogeneities of the ionic microfield<sup>[8]</sup>, and also the effects of thermal motion of the perturbing ions<sup>[9-11]</sup>. Whereas the first ones of the aforementioned effects can be treated within the framework of the oscillator model, the last effect touches directly on the very foundation of this model. It was precisely this effect which was not taken into account in papers<sup>[9-11]</sup> devoted to the generalization of the Holtsmark theory, connected with the thermal motion of the ions. Therefore the effects due to this motion deserve a new analysis.

The model of oscillator with varying frequency is based on two fundamental assumptions: 1) adiabaticity of the action of the ionic microfield on the atom, and 2) the presence of only phase modulation of the light wave radiated by the atom. These two simplifying as-

sumptions mean the neglect of certain other effects, also due to the thermal motion under investigation.

As to the phase modulation, it is actually the decisive factor in the limit of quasistatic broadening, and this, apparently, is the reason why it was assumed that it remained the same when account was taken of the thermal motion of the ions. Yet an electric field of the ions  $F$  changes in the course of time not only in magnitude (which indeed is the cause of the phase modulation), but also in direction. If the rotation of the vector  $F$  is sufficiently slow, then the dipole moment  $d$  of the atom follows adiabatically this rotation, maintaining all the time a constant projection on the direction of  $F$ . At such a reorientation of the atom, the projection  $d$  on the direction of propagation of the light wave  $k$  changes, and since the square of this projection determines the intensity of the light radiated by the atom, some amplitude modulation of the light should take place. The importance of taking such a "rotation of the quantization axis of the atom" into account in radiation processes was demonstrated in<sup>[12]</sup>, where it was possible to explain on this basis the anomalous behavior of the polarization of radiation from atoms excited by electron impact. In the present paper we shall show that amplitude modulation plays no less an important role than phase modulation in that part of the Stark broadening of the spectral lines which is due to thermal motion of the ions.

Besides amplitude modulation, rotation of the quantization axis is accompanied also by a non-adiabaticity effect. Indeed, rotation of the quantization axis of the atom is equivalent, according to the Larmor theorem<sup>[13]</sup>, to the appearance (in a coordinate system rotating together with the field  $F$ <sup>2)</sup>) of an additional interaction between the atom and some "magnetic field." This

<sup>1)</sup>For a rigorous quantum-mechanical justification of both approximations, see [3].

<sup>2)</sup>We note that the model of the oscillator is connected precisely with this rotating system (see [1]).

interaction leads to a change of the wave function and the energy of the stationary state of the radiating atom, which evidently affects the magnitude of the corresponding matrix elements of the dipole moment and the phase of the radiated wave. We shall show below that these effects also make a considerable contribution to the deviation from the quasistatic profile of the line.

Thus, the question of the role of thermal motion of ions in the broadening of spectral lines can be formulated in its entirety, and the analysis of the effects indicated above can be used to clarify the region of applicability of the classic-adiabatic model of the oscillator and the closely related quasistatic approximation of Holtmark<sup>3)</sup>.

The present paper is devoted to the realization (both in the binary and in the multiple case) of the aforementioned program of systematic consideration of the influence of the thermal motion of the ions on the deviation of the spectral-line profile from its quasistatic limit and the derivation of the corresponding adiabaticity and quasistatic-behavior criteria.

## 2. PHYSICAL PICTURE OF COLLISION OF A RADIATING ATOM WITH AN ION

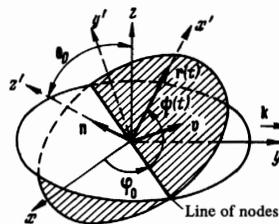
We start with a consideration of the character of the evolution of the dipole moment  $d(t)$ , an evolution responsible for the effect of amplitude modulation, and for simplicity we shall frequently resort to the simplest binary picture of collision (the results, as will be shown below, are not connected with this limitation).

Let us consider the collision of an excited hydrogen atom with a charged particle. We assume that the trajectory of this particle is linear:  $\mathbf{r}(t) = \mathbf{r}_0 + \mathbf{v}t$ , and "does not penetrate" into the atom:  $|\mathbf{r}(t)| \gg n^2 a_0$ . In the stationary (laboratory) frame, the vector of the electric field  $\mathbf{F} = e\mathbf{r}(t)/|\mathbf{r}(t)|^3$  produced by the passing charged particle changes in magnitude and, in addition, turns through  $180^\circ$  in the plane of the vectors  $\mathbf{r}_0$  and  $\mathbf{v}$  (the collision plane). Let us changeover to a coordinate system in which one of the axes (for example, the  $x$  axis) is directed at each instant of time along the field  $\mathbf{F}(t)$ . In this rotating coordinate system, the electrostatic interaction of the atom  $V(t) = -\mathbf{d} \cdot \mathbf{F}$  is determined by only one projection of the dipole moment  $d_x$ , and therefore cannot cause transitions between states with different magnetic quantum numbers  $m$ .

However, besides the electrostatic field we have here also a "magnetic" interaction due to the rotation<sup>15)</sup>. For the particular case of rotation through an angle  $\varphi$  about the axis  $z$ , it is given by (the field is directed along the  $x$ -axis!):

$$V_{\text{magn}} = \hbar J_x \partial \psi / \partial t = \hbar J_x \dot{\psi}(t), \quad (1)$$

<sup>3)</sup>We note in this connection that the first indications about the need for taking into account the effects of the rotation of the field  $\mathbf{F}$  are contained in the papers of Spitzer [14], where these effects were considered within the framework of the impact approximation and the hypothesis was advanced that the effects of non-adiabaticity and reorientation of the atom may cancel out. Within the framework of the quasistatic approach, these questions were investigated by Wimmel [11], where, in particular, an attempt was made to establish the relation between the adiabatic and quasistatic approaches. However, both Spitzer and Wimmel confined themselves to semiquantitative estimates of the discussed effects, without calculating their influence on the line contour, which is the only way one can judge their true role.



Stationary ( $xyz$ ) and rotating ( $x'y'z'$ ) coordinate systems. The origins of both systems coincide with the center of the radiating atom; the position of the perturbing ion is given by the vector  $\mathbf{r}(t)$  and its velocity is  $-\mathbf{v}$ . The Euler angles  $\varphi_0$ ,  $\theta_0$ , and  $\psi(t)$  and the vector  $\mathbf{n} = \mathbf{r} \times \mathbf{v} / |\mathbf{r} \times \mathbf{v}|$  determine the collision plane;  $\mathbf{k}$  is the wave vector of the radiated wave.

where  $J$  is the operator of the orbital angular momentum of the atom.

Obviously, this interaction can cause transitions between sublevels with different  $m$  (Stark sublevels), but at sufficiently slow (adiabatic) passages of the ions, the characteristic value of this interaction is small compared with the electrostatic interaction, so that it can be accounted for by perturbation theory (see Sec. 3). In the zeroth approximation, neglecting the magnetic interaction, so that it can be accounted for by perturbation theory (see Sec. 3). In the zeroth approximation, neglecting the magnetic interaction, we can regard the magnetic quantum number  $m$  as an integral of the motion.

For "nonpenetrating" trajectories of the perturbing charged particles, the principal quantum number  $n$  of the atom will also be conserved, so that only transitions between states with fixed  $n$  can take place. If we direct one of the axes of the rotating coordinate system along the field  $\mathbf{F}$ , then the electrostatic interaction, as already noted, will not have nonzero nondiagonal matrix elements for the transitions between the Stark sublevels. This means that at such adiabatic collisions the excited hydrogen atom precesses about the direction of the vector of the electric field of the passing particle in such a way that the projections of the angular momentum  $J$  and of the electric dipole moment  $\mathbf{d}$  on this direction are conserved.

The radiation processes are determined, as is well known, by the evolution of the vector of the dipole moment of the atom. Since in practice the criterion for the adiabaticity of rotation is satisfied for all the ions of the plasma, the evolution of the vector  $\mathbf{d}(t)$  will consist of a rotation of its precession axis so as to follow the electric microfield of the ions  $\mathbf{F}(t)$  (see the figure). Observation of the shape of the spectral line is carried out in the laboratory system, relative to which the direction of the microfield can have at the initial instant of time an arbitrary orientation. On the other hand, the examination of the interaction of the atom with microfield  $\mathbf{F}$  is best carried out in a coordinate system whose symmetry axis rotates in the collision plane together with the vector  $\mathbf{F}(t)$ . The transition from the initial laboratory system to these rotating quantization axes can be realized in two stages: at first the laboratory system is rotated through the Euler angles  $\varphi_0$ ,  $\theta_0$ , and  $\psi_0$ , which determine the positions of the quantization axis at the initial instant of time, after which rotation is carried out through an angle  $\psi_1(t)$  about the vector  $\mathbf{n} = \mathbf{r}_0 \times \mathbf{v} / |\mathbf{r}_0 \times \mathbf{v}|$ . The direction of this vector, which

determines the plane of collision for our choice of the coordinate system, coincides with the axis  $z'$  (see the figure). Accordingly, the rotation operator, which transforms the wave functions from the laboratory system to the rotating coordinate system, can be written in the form<sup>[15]</sup>

$$R(t) = R_1(0, 0, \psi_1(t))R_0(\varphi_0, \theta_0, \psi_0) = e^{iJ_z \varphi_0} e^{iJ_y \theta_0} e^{iJ_z \psi_0}. \quad (2)$$

Here  $\varphi = \varphi_0$ ,  $\theta = \theta_0$ ,  $\psi = \psi_0 + \psi_1(t)$

The evolution of the operator of the dipole moment of the atom can now be represented in the form

$$\mathbf{d}(t) = R_0^{-1} R_1^{-1} \mathbf{d} R_1 R_0 = \hat{A}(t) \mathbf{d}', \quad (3)$$

where  $A(t)$  is the matrix of the rotation of the vector through the Euler angles  $\varphi$ ,  $\theta$ , and  $\psi$ <sup>[16,17]</sup>, and  $\mathbf{d}'$  is the dipole-moment vector expressed in terms of the projections on the quantization axis of the atom.

It follows from the foregoing that the evolution of  $\mathbf{d}(t)$ , described above, leads to a change in the amplitude of the light wave for a given Stark state of the atom (i.e., with fixed  $m$ ) (effect of amplitude modulation). On the other hand, the interaction of the atom with the "magnetic field" leads obviously to a change of the Stark state itself (the non-adiabaticity effect). We shall construct below a perturbation theory that takes both these effects into account.

### 3. CORRELATION FUNCTIONS THAT TAKE THE ROTATION OF THE PERTURBING MICROFIELD INTO ACCOUNT

Let us find the expression for the distribution of the intensity in the line (line profile), which would take into account, besides the ordinary phase modulation, amplitude modulation and the non-adiabaticity effect. We confine our analysis to the "near static"<sup>4)</sup> and "near-adiabatic" regions. It is therefore meaningful to speak of the profile  $I(\omega)$  of a certain Stark component produced when the atom goes over from the initial state  $i$  to the final state  $f$ ; for simplicity, we assume the latter to be unperturbed. The profile of the component in question is given by<sup>[11,11]</sup>

$$I_{ij}(\omega) = \frac{1}{\pi} \operatorname{Re} \int_0^{\infty} d\tau e^{i(\omega - \omega_0)\tau} \Phi_{ij}(\tau), \quad (4)$$

where  $\omega_0 \equiv (E_i^{(0)} - E_f^{(0)})/\hbar$  is the unperturbed frequency of the transition, and  $\Phi_{ij}(\tau)$  is the correlation function:

$$\Phi_{ij}(\tau) = \langle \langle \chi_i'(0) | \mathbf{d} | \chi_f'(\tau) \rangle \langle \chi_f'(\tau) | \mathbf{d} | \chi_i'(\tau) \rangle \rangle_{\text{cp}}. \quad (5)$$

Here  $\chi_i'$  and  $\chi_f'$  are the wave functions of the initial and final states in the laboratory frame, and the symbol  $\{\dots\}_{\text{av}}$  denotes averaging over the ensemble.

We now transform the right-hand side of (5) into a rotating coordinate system. The connection between the wave function  $\chi_{i,f}$  in the rotating system and the function  $\chi'_{i,f}$  is given by the relation

$$\chi_{i,f} = R(t) \chi'_{i,f}, \quad (6)$$

where  $R(t)$  is the above-described operator of rotation through the Euler angles  $\varphi_0$ ,  $\theta_0$ ,  $\psi(t)$ . Carrying out the

<sup>4)</sup>We shall use this term to designate the region in which the deviations from the quasistatic (Holtsmark) limit, due to thermal motion, begin to play a noticeable role; it is probably better to use simply "static" limit in place of the "quasistatic" limit.

corresponding transformations in (5), we obtain

$$\Phi_{ij}(\tau) = \sum_{m,n} \left\{ \sum_k A_{km}(0) A_{kn}(\tau) \langle \chi_i(0) | d_m' | \chi_i(0) \rangle \langle \chi_f(\tau) | d_n' | \chi_f(\tau) \rangle \right\}_{\text{cp}}. \quad (7)$$

We average in (7) over the Euler angles  $\varphi_0$ ,  $\theta_0$  and  $\psi_0$ . Using the explicit expressions for the matrices  $A(0)$  and  $A(\tau)$ , we obtain

$$\left\{ \sum_k A_{km}(0) A_{kn}(\tau) \right\}_{\text{av}(\varphi_0, \theta_0, \psi_0)} = 1/3 [1 + 2 \cos \psi_1(\tau)] \delta_{mn}, \quad (8)$$

whence

$$\Phi_{ij}(\tau) = 1/3 [1 + 2 \cos \psi_1(\tau)] \langle \chi_i(0) | \mathbf{d}' | \chi_i(0) \rangle \langle \chi_f(\tau) | \mathbf{d}' | \chi_f(\tau) \rangle_{\text{cp}}. \quad (9)$$

We now proceed to find the wave functions  $\chi(t)$  in the rotating coordinate system. The Schrödinger equation, in accordance with the figure and relations (2) and (6), is

$$i\hbar \partial \chi / \partial t = H(t) \chi \equiv [H_0 + d_x F(t) + \hbar J_z \dot{\psi}_1(t)] \chi, \quad (10)$$

where  $H_0$  is the unperturbed Hamiltonian of the atom.

It is seen from (10) that in a rotating system there is both an electrostatic perturbation ( $d_x F$ ) and a "magnetic" perturbation ( $\hbar J_z \dot{\psi}_1$ ). Since the unperturbed state is degenerate, we can choose as the zeroth approximation the wave functions that diagonalize either the electrostatic or the "magnetic" perturbation. In the near-static region under consideration, it is natural to choose as the zeroth approximation the Stark wave functions that diagonalize the Hamiltonian  $H_0 + d_x F(t)$ , regarding the "magnetic" interaction as a perturbation.

We determine first the complete system of "adiabatic" states  $u_k(t)$ :

$$H(t) u_k(t) = E_k(t) u_k(t). \quad (11)$$

We seek the solution in the form of a series

$$\chi_i(t) = \sum_k a_{ik}(t) u_k(t) \exp\left(-i \int_0^t \omega_k dt'\right); \quad (12)$$

$$\omega_k(t) \equiv E_k(t) / \hbar.$$

Substituting (12) in (10), we arrive at the system<sup>[18]</sup>

$$a_{ik} = \sum_n \frac{a_{in}}{\hbar \omega_{kn}} \exp\left(i \int_0^t \omega_{kn} dt'\right) \left(\frac{\partial H}{\partial t}\right)_{kn}; \quad (13)$$

$$\omega_{kn} \equiv (E_k - E_n) / \hbar.$$

We shall seek the solution of the system (13) by perturbation theory. It must be borne in mind here that perturbation theory is applied also to Eqs. (11), the solutions of which give, obviously, a contribution to the corresponding orders:  $a_{ik} \approx a_{ik}^{(0)} + a_{ik}^{(1)} + a_{ik}^{(2)}$ , and also determine the changes in the phases (energies) of the wave functions. We choose as the zeroth approximation in (11), as already indicated, the Stark wave functions. Then the solution of Eqs. (11) is obviously a perturbation-theory series whose only difference from the series of the ordinary stationary perturbation theory is the dependence of the corresponding frequencies on the time as a parameter:

$$\omega_k(t) \approx \omega_k^{(0)}(t) + \omega_k^{(2)}(t) = \omega_k^{(0)}(t) + \dot{\psi}^2(t) \sum_m \frac{|(J_z)_{km}|^2}{\omega_{km}^{(0)}(t)}; \quad (14)$$

We have taken into consideration here the fact that  $\omega_k^{(1)} \sim (J_z)_{kk} = 0$ ;

$$u_k \approx u_k^{(0)} + u_k^{(1)} + u_k^{(2)} = u_k^{(0)} + \dot{\psi}(t) \sum_m \frac{|(J_z)_{km}|^2}{\omega_{km}^{(0)}(t)} u_m^{(0)} \quad (15)$$

$$+ \psi^2(t) \sum'_m \left\{ \sum'_n \frac{(J_z)_{mn}(J_z)_{nk}}{\omega_{km}^{(0)}\omega_{kn}^{(0)}} u_m^{(0)} - \frac{1}{2} \frac{|(J_z)_{mk}|^2}{\omega_{km}^{(0)2}} u_k^{(0)} \right\}$$

(The indices  $k$  and  $m$  denote matrix elements taken over the unperturbed (Stark) wave functions).

Further, according to the described solution procedure, we have

$$\left( \frac{\partial H}{\partial t} \right)_{kn} \approx \left( \frac{\partial H_2}{\partial t} \right)_{kn}^{(1)} + \left( \frac{\partial H}{\partial t} \right)_{kn}^{(2)} = (d_x)_{kn} \dot{F}(t) + (J_z)_{kn} \dot{\psi}(t) + \langle u_k^{(1)} | d_x | u_n^{(0)} \rangle \dot{F}(t) + \langle u_k^{(0)} | d_x | u_n^{(1)} \rangle \dot{F}(t). \quad (16)$$

It is easy to verify that the term  $(\partial H/\partial t)^{(2)}$  vanishes upon averaging over the angle between  $\mathbf{r}_0$  and  $\mathbf{v}$ , and therefore it makes no contribution to  $a^{(2)}$  when the corresponding orders are equated in (13). The terms containing  $(\partial H/\partial t)_{kn}^{(1)}$  likewise make no contribution in either the first ( $a^{(1)}$ ) or the second ( $a^{(2)}$ ) order of perturbation theory, since the quantities  $(\partial H/\partial t)_{kn}^{(1)}$  are proportional to  $\delta_{kn}$ , and the expressions for  $a^{(1)}$  and  $a^{(2)}$  contain only the non-diagonal matrix elements. Thus,  $a^{(1)} = a^{(2)} = 0$ , and accurate to terms of second order of smallness inclusive we have  $a_{kn} = a_{kn}^{(0)} = \delta_{kn}$ .

As a result, the wave function  $\chi_i(t)$ , accurate to terms of second order, is given by

$$\chi_i(t) \approx (u_i^{(0)} + u_i^{(1)} + u_i^{(2)}) \exp \left( -i \int_0^t [\omega_i^{(0)}(t') + \omega_i^{(2)}(t')] dt' \right). \quad (17)$$

Substituting (17) in (9), we get

$$\Phi_{i\tau} \approx \left\{ \frac{1 + 2 \cos \psi_i(\tau)}{3} \exp \left( -i \int_0^\tau [\omega_i^{(0)}(t) + \omega_i^{(2)}(t)] dt \right) \times \langle u_i^{(0)} + u_i^{(1)}(0) + u_i^{(2)}(0) | d' | \chi_i \rangle \langle \chi_i | d' | u_i^{(0)} + u_i^{(1)}(\tau) + u_i^{(2)}(\tau) \rangle \right\}_{av}. \quad (18)$$

The calculation that follows is connected with the use of the "slowness" of the perturbation, i.e., with the expansion of the obtained correlation function in a series in powers of  $\tau$ . We shall first carry out this expansion and the corresponding averaging in the binary case. Certain results not connected with the binary approximation will then be used in the consideration of multiple interaction.

#### 4. PROFILE OF LINE WING

We consider first the influence of the amplitude modulation, neglecting the effects of non-adiabaticity, i.e., assuming in (18)  $\omega_i^{(2)} = 0$  and  $u_i^{(1)} = u_i^{(2)} = 0$ . Then, carrying out in (18) the expansions

$$\psi_1(\tau) \approx \psi_1(0)\tau, \quad \omega_i^{(0)}(t) \approx \omega_i^{(0)}(0) + \dot{\omega}_i^{(0)}(0)t + \ddot{\omega}_i^{(0)}(0)t^2/2$$

etc., we obtain an expression for the adiabatic correlation function  $\Phi_{i\tau}^{ad}(\tau)$ ; this expression takes into account the effects of phase and amplitude modulation:

$$\Phi_{i\tau}^{ad}(\tau) \approx \Phi_{i\tau}^{(0)ad}(\tau) + \Phi_{i\tau}^{(2)ad}(\tau) = |d_{i1}|^2 \{ \exp(-i\omega_i^{(0)}(0)\tau - \exp(-i\dot{\omega}_i^{(0)}(0)\tau) [1/3 \dot{\omega}_i^{(0)2}(0)\tau^3 + 1/6 \ddot{\omega}_i^{(0)}(0)\tau^3 + 1/3 \dot{\psi}_1^2(0)\tau^2] \}_{av}. \quad (19)$$

We have taken here into account the fact that the terms that are linear in  $\dot{\omega}_i^{(0)}(0)$  and  $\dot{\psi}_1(0)$ , and also the crossing terms  $\dot{\omega}_i^{(0)}(0)\dot{\psi}_1(0)$ , give zero after averaging over the angle<sup>5)</sup>.

<sup>5)</sup>This is valid also in the multiple case (see Sec. 5).

The first term in (19) gives, with allowance for (4), the usual quasistatic profile of the component.

The terms connected with the change of the phase (proportional to  $\tau^4$  and  $\tau^3$ ), lead after averaging in the binary scheme (see (22) below) to the well known correction to the spectrum  $I_{i1}^{(2)P}(\omega)$ <sup>[9,10]</sup>, which takes into account only the phase modulation:

$$I_{i1}^{(2)P}(\omega) = -|d_{i1}|^2 \frac{5\pi}{32} \frac{N\alpha^{1/2}v_0^2}{(\omega - \omega_0)^{3/2}}, \quad (20)$$

where  $N$  is the density,  $v_0$  is the most probable (Maxwellian) ion velocity, and  $\alpha$  is the Stark constant of the component in question.

The correction to the spectrum  $I_{i1}^{(2)A}(\omega)$ , due to the amplitude modulation, can be easily obtained by noting that in the binary case

$$\dot{\psi}_i(0) = \frac{|[\mathbf{r}_0\mathbf{v}]|}{r_0^2} = \frac{v}{r_0} \sin(\widehat{\mathbf{r}_0\mathbf{v}}). \quad (21)^*$$

Putting further  $\omega_i^{(0)}(t) = (\alpha/e)F(t) = \alpha/r^2(t)$ , using (4), and averaging over  $\mathbf{r}_0$ , we obtain

$$I_{i1}^{(2)A}(\omega) = |d_{i1}|^2 N v_0^2 \int_0^\infty 4\pi r_0^2 dr_0 \left\{ \frac{1}{3r_0^2} \delta'' \left( \omega - \omega_0 - \frac{\alpha}{r_0^2} \right) \right\}. \quad (22)$$

This gives ultimately

$$I_{i1}^{(2)A}(\omega) = |d_{i1}|^2 \frac{5\pi}{2} \frac{N\alpha^{1/2}v_0^2}{(\omega - \omega_0)^{3/2}}. \quad (23)$$

From a comparison of (20) and (23) we see that the correction  $I_{i1}^{(2)A}(\omega)$  has a sign opposite to the correction  $I_{i1}^{(2)P}(\omega)$ , and exceeds it in absolute magnitude by 16 times.

We now take into account the non-adiabaticity effect. A direct calculation of the sums in (14), (15), and (18) is in the general case quite unwieldy. At the same time, the characteristic features of the effect of non-adiabaticity can be revealed by means of a concrete example of the line  $L_\alpha$ , to the analysis of which we now turn. In this case the sums in (18) can be easily calculated (we shall henceforth have in mind the lateral Stark component with positive frequency shift  $\omega_i^{(0)} = (\alpha/e)F$ , which is denoted by the index 1, the lower state being denoted by the index 0):

$$\sum'_m \frac{|(J_z)_{1m}|^2}{\omega_{1m}^{(0)}(0)} = \frac{1}{2(\alpha/e)F(0)}, \quad \sum'_m (d)_{m0} \frac{(J_z)_{m1}}{\omega_{1m}^{(0)}} \sum'_n (d)_{0n} \frac{(J_z)_{n1}}{\omega_{1n}^{(0)}} = \frac{|d_{10}|^2}{[(\alpha/e)F(0)]^2}. \quad (24)$$

In addition, as can be readily verified,  $\langle \chi_0 | d | u_1^{(2)} \rangle = 0$ . We then obtain for the correlation function  $\Phi_{10}^{(2)nonid}(\tau)$ , corresponding to the non-adiabaticity effect,

$$\Phi_{10}^{(2)nonid}(\tau) = |d_{10}|^2 \left\{ \exp(-i\omega_1^{(0)}(0)\tau) \times \left[ -\frac{i\tau}{2(\alpha/e)F(0)} + \frac{1}{[(\alpha/e)F(0)]^2} \right] \right\}_{av}. \quad (25)$$

The first term in (25) is due to the change of the energy, and the second to the change of the wave function of the Stark state<sup>6)</sup>. Carrying out in (25) averagings similar to

<sup>6)</sup>We note that the presence of this term in (25) is determined by the value of  $\chi_1(0)$ , which we have taken to equal  $u_1^{(0)}(0)$  (formula (17)), i.e., we assumed that at the initial instant of time the atom is not in a definite Stark state  $u_1^{(0)}$ . Such a choice of the initial conditions is dictated by the fact that we implicitly assume the density matrix  $\rho_{iik}$  to be diagonal ( $\sim \delta_{iik}$ ). This has been justified, as a matter of fact, for the here-employed eigenstates of the atom in thermal equilibrium with the medium.

\* $[\mathbf{r}_0\mathbf{v}] \equiv \mathbf{r}_0 \times \mathbf{v}$ .

(22), we obtain for the spectral correction for the non-adiabaticity  $I_{10}^{(2)\text{nonid}}(\omega)$  the expression

$$I_{10}^{(2)\text{nonid}}(\omega) = |d_{10}|^2 \pi N \alpha^{1/2} v_0^2 / (\omega - \omega_0)^{1/2}. \quad (26)$$

It is seen from (26) and (23) that  $I_{10}^{(2)\text{nonid}}(\omega)$  has the same sign and order of magnitude as  $I_{10}^{(2)A}(\omega)$ . It follows therefore, incidentally, that cancellation of the effects of non-adiabaticity and reorientation of the atom, of the type proposed by Spitzer<sup>[14]</sup>, actually does not take place.

It is clear from the foregoing that it is precisely the effects of microfield rotation considered here (amplitude modulation and non-adiabaticity) which determine the bigger part of the total deviation from the quasi-static profile. For the wing of the  $L_\alpha$  line, the ratio of the contribution of the effects of rotation  $I_{10}^{(2)A}(\omega) + I_{10}^{(2)\text{nonid}}(\omega)$  to the previously known contribution of the effect of phase modulation  $I_{10}^{(2)P}(\omega)$  amounts, according to (20), (23) and (26) to  $-44.8$ .

## 5. COMPLETE NEAR-STATIC LINE PROFILE

The use of the binary approximation is justified only for the wings of the spectral line, where the main effect of broadening is due to the nearest perturbing ion. In considering the total line profile, it is necessary to take into account the multiple character of the electric microfield  $F(t)$  produced by the plasma ions. The multiple character of the perturbation excludes the possibility of using the parameters  $r_0$  and  $v$  of one (nearest) ion in order to fix the collision plane and determine the rotation angle  $\psi_1(\tau)$  of the quantization axis of the atom. We shall therefore first discuss the method of determining, in the multiple case, the plane of collision and the parameters over which the averaging will be carried out in (5).

As will be shown below, the effect of rotation of the field vector  $F(t)$  is determined, in the near-static region, by only the first derivative  $\dot{F}(0)$  at the initial instant of time. Therefore it is possible here, too, by way of a natural generalization of the binary case, to define with the aid of the two vectors  $F(0)$  and  $\dot{F}(0)$  the collision plane. Just as in the binary case, specification of the collision plane determines uniquely the Euler angles  $\varphi_0$ ,  $\theta_0$ , and  $\psi_0$ , through which it is necessary to rotate the laboratory frame in order to orient it along the quantization-axis system. One of the axes of the latter system is directed, as above, along the vector  $F$ , and the other along the vector  $n = F(0) \times \dot{F}(0) / |F(0) \times \dot{F}(0)|$ . With such a choice of the axis, the angle of rotation  $\psi_1$  can be approximately represented in the form  $\psi_1(\tau) \approx \psi_1(0)\tau$ , where

$$\dot{\psi}_1(0) = \frac{|[F(0)\dot{F}(0)]|}{F^2(0)} = \frac{|\dot{F}_\perp(0)|}{|F(0)|}. \quad (27)$$

The vector  $\dot{F}_\perp(0)$  is the component of the vector  $\dot{F}(0)$  perpendicular to  $F(0)$ . The component of  $\dot{F}(0)$  parallel to  $F(0)$  will be denoted by  $\dot{F}_\parallel$ .

When averaging in (5) it is necessary to average not only over the three Euler angles  $\varphi_0$ ,  $\theta_0$ , and  $\psi_0$  but also over the angle between  $F(0)$  and  $\dot{F}(0)$ , as well as over the two moduli  $|F|$  and  $|\dot{F}|$ <sup>7)</sup>. We shall henceforth write

<sup>7)</sup>The listed six averaging parameters correspond, obviously, to the six independent components of the vectors  $F(0)$  and  $\dot{F}(0)$ .

simply  $F$  and  $\dot{F}$ , if the values of these vectors are taken at the initial instant of time.

In light of the foregoing it is easy to see that the expressions (19) and (25) for the correlation function are actually valid also in the multiple case, and the symbol  $\{\dots\}_{\text{av}}$  should now be taken to mean the above-described averaging over the ensemble of all the perturbing ions, while the frequency shift, the angular velocity of rotation  $\dot{\psi}_1$ , and their derivatives are to be taken to mean the corresponding multiple analogs of (27) and

$$\dot{F} = (F\dot{F})/F = |\dot{F}_\parallel|, \quad \ddot{F} = (F\ddot{F})/F - \dot{F}_\parallel^2/F. \quad (28)$$

It is easy to verify, in addition, that after averaging over the angles between  $F$  and  $\dot{F}$  and  $\ddot{F}$ , the corresponding mean values in (19) and (25) will contain only  $\{\dot{F}_\parallel^2\}_{\text{av}}$  and  $\{\dot{F}_\perp^2\}_{\text{av}}$ . The calculation of these mean values is best broken up into two stages (see<sup>[11]</sup>): first we average the quantities  $F_\perp^2$  and  $\dot{F}_\parallel^2$  at a fixed value of the modulus of the ion field  $F$ , and then integrate over all the  $F$  with the Holtmark distribution function  $W_{\mathcal{H}}(F)$ . It is then possible to use during the first stage of averaging the results of Chandrasekhar and von Neumann<sup>[19]</sup> (see also<sup>[11]</sup>):

$$\{\dot{F}_\parallel^2\}_{\text{av}(r)} = 4s/(\omega r F_0)^2 \beta^{1/2} G(\beta) / \mathcal{H}(\beta), \quad (29)$$

$$\{\dot{F}_\perp^2\}_{\text{av}(r)} = 4s/(\omega r F_0)^2 \beta^{1/2} [G(\beta) - I(\beta)] / \mathcal{H}(\beta), \quad (30)$$

where  $\omega r = \lambda^{1/2} v_0 N^{1/3}$  is the characteristic scale of the frequency at which the ion field changes<sup>[11]</sup>,  $F_0 = \lambda e N^{2/3}$  is the "normal" Holtmark field ( $\lambda = 2.603$ ),  $\beta = F/F_0$ ,  $\mathcal{H}(\beta)$  is the Holtmark function<sup>[19]</sup>, and  $G(\beta)$  and  $I(\beta)$  are functions introduced by Chandrasekhar and von Neumann<sup>[19,10]</sup>:

$$G(\beta) = \frac{2}{\pi} \int_0^\infty \exp\left\{-\left(\frac{y}{\beta}\right)^{3/2}\right\} y^{-1/2} \sin y \, dy,$$

$$I(\beta) = \frac{2}{\pi} \int_0^\infty \exp\left\{-\left(\frac{y}{\beta}\right)^{3/2}\right\} y^{-1/2} (\sin y - y \cos y) \, dy. \quad (31)$$

We change over in (4) and (18) to the dimensionless variables  $z = \Delta\omega_0\tau$ ,  $\omega' = (\omega - \omega_0)/\Delta\omega_0$  ( $\Delta\omega_0 \equiv (\alpha/e)F_0$ ). We then obtain for the correlation function  $\Phi_{10}(z)$  the expression

$$\Phi_{10}(z) = |d_{10}|^2 \left\{ e^{-iz} \left[ 1 - \frac{45}{8} \frac{1}{\lambda h^{3/2}} \left( \frac{z^4 \beta^{1/2}}{8} \frac{I(\beta)}{\mathcal{H}(\beta)} + \frac{iz^3}{6} \frac{G(\beta) - I(\beta)}{\beta^{1/2} \mathcal{H}(\beta)} \right) \right. \right. \\ \left. \left. + \frac{z^2}{3} \frac{G(\beta) - I(\beta)}{\beta^{1/2} \mathcal{H}(\beta)} + iz \frac{G(\beta) - I(\beta)}{\beta^{1/2} \mathcal{H}(\beta)} + \frac{G(\beta) - I(\beta)}{\beta^{1/2} \mathcal{H}(\beta)} \right] \right\}. \quad (32)$$

We have introduced here the characteristic dimensionless parameter of the problem  $h^{1/3}\alpha/v_0$ , which is the ratio of the Stark shift  $\alpha N^{2/3}$  to the frequency at which the ion field changes  $N^{1/3}v_0$ . The effects of phase modulation, amplitude modulation, and non-adiabaticity are described respectively by the first two, the third, and the last two terms in the round brackets of (32). It is seen from (32), in particular, that all three effects have the same order in smallness in the parameter  $h^{-1/3}$ .

To obtain the dimensionless profile of the Stark component  $I_h(\omega')$ , defined by the relation  $I(\omega)d\omega = |d_{10}|^2 I_h(\omega')d\omega'$ , it remains to calculate the mean value of (32) with the aid of the Holtmark function  $\mathcal{H}(\beta) = F_0 W_{\mathcal{H}}(F)$ , and then carry out a Fourier transformation in accordance with (4). The resulting profile of the component  $1 \rightarrow 0$  (which determines, in this case the

profile of the entire line when  $\omega' > 0$ ) then assumes the following final form:

$$I_h(\omega') = \mathcal{H}(\omega') + h^{-1/2}\Pi(\omega'), \quad (33)$$

$$\Pi(x) = \frac{15}{16\lambda} \left\{ -\frac{3}{4} \frac{d^4}{dx^4} [x^{1/2}I(x)] + \frac{d^3}{dx^3} \left[ \frac{G(x) - I(x)}{x^{1/2}} \right] \right. \\ \left. + 2 \frac{d^2}{dx^2} \left[ \frac{G(x) - I(x)}{x^{1/2}} \right] - 3 \frac{d}{dx} \left[ \frac{G(x) - I(x)}{x^{1/2}} \right] + 6 \left[ \frac{G(x) - I(x)}{x^{1/2}} \right] \right\}. \quad (34)$$

In (34), just as above, the first two terms correspond to phase modulation, the third to amplitude modulation, and the last two to non-adiabaticity.

Let us consider the contributions of the phase modulation and of both rotation effects to the total correction for the quasistatic line contour, due to the thermal motion of the ions, for different regions of  $\omega'$ . To this end we shall use the expansions of the functions  $G(x)$  and  $I(x)$ :

$$G(x) \approx \begin{cases} \sqrt{2/\pi} & x \gg 1 \\ (4/3\pi)x^{3/2} & x \ll 1 \end{cases}; \quad I(x) \approx \begin{cases} \frac{1}{2}\sqrt{2/\pi} & x \gg 1 \\ (4/9\pi)x^{3/2} & x \ll 1 \end{cases}. \quad (35)$$

On the wing of the line ( $\omega' \gg 1$ ) the distribution of the intensity (34) coincides, as it should, with the results obtained from the binary analysis, see (20), (23), and (26).

In the central part of the line ( $\omega' \gg 1$ ) we have

$$\Pi(\omega') \approx 8/\pi\lambda\omega'^2. \quad (36)$$

Such a behavior of the "thermal" correction is due entirely to the contribution of the terms connected with the non-adiabaticity. On the other hand, the terms in (34) connected with the phase and amplitude modulations, for  $\omega' \ll 1$ , as can be readily verified, tend to a constant value and consequently are relatively small.

## 6. DISCUSSION

1. Our analysis shows that the change of the direction of the electric-microfield vector of the ions turns out, in spite of the usual opinion (explicitly expressed or tacitly implied)<sup>[9-11]</sup>, is not only not secondary, but on the contrary, is the decisive one among the effects of thermal motion of the perturbing ions. The result (33) makes it possible to reformulate the quasistatic criterion:

$$h^{-1/2}\Pi(\omega') \ll \mathcal{H}(\omega'). \quad (37)$$

On the line wing ( $\omega' \gg 1$ ) this criterion turns out, as already noted in Sec. 4, to be numerically more stringent (to be sure, by almost 45 times!) than the well known Holstein criterion<sup>[9]</sup> (see also<sup>[10]</sup>).

In the general case ( $\omega \sim 1$ ) the corrections considered here, being connected with derivatives of different order of the Chandrasekhar-von Neumann functions  $G(\omega')$  and  $I(\omega')$ , should generally speaking have also different functional dependence.

In the central part of the line ( $\omega' \ll 1$ ) the criterion (37) differs from that obtained earlier<sup>[10]</sup> qualitatively (by a factor  $h^{1/6} \gg 1$ , also on the more stringent side):

$$\omega' \gg h^{-1/6} \quad \text{or} \quad \omega - \omega_0 \gg \sqrt{aNv_0}, \quad (38)$$

Here, as indicated, this more stringent character of the criterion is due to the non-adiabaticity effects.

For practical estimates of the region of applicability

of the quasi-static approximation it is convenient to establish that value of  $h = h^*$  ( $h = N(\alpha/v_0)^3$ ), at which the total "thermal" correction becomes comparable with the contribution of the zeroth (quasistatic) approximation. According to Sec. 4, for the  $L_\alpha$  line wing there occurs an appreciable increase (by a factor  $(45)^{3/2} \approx 300$ ) of  $h^*$  compared with its value when account is taken of only the phase modulation<sup>[10]</sup>. Thus, for example, for  $\omega = \omega_0 = (3-4)\Delta\omega_0$  we have  $h^* \sim 1$ , whereas according to<sup>[10]</sup> we would obtain  $h^* \sim 0.003$ . It is difficult to obtain a convenient criterion for the central part of the line, since here  $h^*$  depends strongly on  $\omega - \omega_0$ . However, we can indicate by way of an example that when  $\omega - \omega_0 \lesssim \Delta\omega_0$  the value of  $h^*$  also amounts to several units (according to<sup>[10]</sup> we have here  $h^* \sim 0.004$ ). Thus, the quasistatic approximation becomes violated "integrally" already at  $h^* \sim 1$ , and not at  $h^* \ll 1$  as in the case when only the phase modulation is taken into consideration, so that the region of applicability of this approximation  $h \gg h^*$ , becomes much narrower.

2. Our analysis was based on a systematic allowance for the "adiabatic" non-static character of the effects (phase and amplitude modulations) as well as for the non-adiabaticity effects. Therefore the criterion (37) is simultaneously also the adiabaticity criterion. We see hence that on the line wing the effects of non-adiabaticity and of the "adiabatic" non-static character turn out to be of the same order of magnitude, so that satisfaction of the quasistatic condition implies adiabaticity, and vice versa. In contrast, at the center of the line, the decisive effects are those of non-adiabaticity.

The foregoing consideration enables us also to analyze the tentative criteria obtained by Wimmel<sup>[11]</sup> on the basis of interesting physical considerations<sup>8)</sup>. The effect of amplitude modulation is recognized in<sup>[11]</sup> to be secondary compared with the effect of phase modulation, which, as we have seen, does not correspond to reality. The adiabaticity criterion formulated in<sup>[11]</sup> is in the main correct, but unfortunately during the course of its comparison with the quasistatic criterion, owing to the transition from a spectral criterion to an integral one, an unjustified conclusion was drawn that the adiabatic approximation is applicable all the way to the center of the line.

3. The foregoing analysis pertained to the case of broadening due to the linear Stark effect. It can be shown<sup>[22]</sup> that the results of Sec. 4 for amplitude modulation<sup>9)</sup> admit of a generalization to the case of a general interaction law  $V(t) = C_k/r^k(t)$ . The formula obtained thereby is a generalization of the Holstein formula<sup>[9]</sup>, which takes only phase modulation into account. The ratio of the contribution of the effect of amplitude modulation to the corresponding contribution of the effect of phase modulation is  $-16/(k-1)$ ; for  $k=2$  this result goes over into that obtained in Sec. 4. It follows therefore that for any arbitrary broadening mechanism amplitude modulation plays no less important a role than phase modulation also in the deviations from the quasistatic line profile.

<sup>8)</sup> The question of the corrections that take into account only phase modulation is exhaustively treated in [20, 21], in agreement with [10].

<sup>9)</sup> Allowance for the non-adiabaticity effect is difficult in this case, owing to the difficulty in calculating the sums contained in (15).

4. The phenomenon of amplitude modulation should lead also to a broadening of the central ( $C_2 \equiv \alpha = 0$ ) Stark components of the line, for which there is no phase modulation connected with the Stark splitting. Usually the broadening of the central components is associated only with the impact action of electrons on the atom, without considering the contribution of the ions. Yet the thermal motion of the ions, which leads to the effect of amplitude modulation, should impose a definite limitation on the applicability of such an approach. This limitation can be found with the aid of the results of Sec. 3, by putting in them  $\alpha \equiv 0$ . Simple considerations<sup>[22]</sup> lead to the following condition:

$$\rho_{\text{eff}} \gamma / v_0 \gg 1, \quad (39)$$

where  $\gamma$  is the impact electronic width of the central component;  $\rho_{\text{eff}}$  is the effective impact parameter of the ion; in our case  $\rho_{\text{eff}} \sim N^{-1/3}$ . A condition of this type is violated for the  $L_\alpha$  line in a plasma with temperature  $T_i = T_e \sim 2$  eV already at an electron concentration  $N_e \lesssim 6 \times 10^{16} \text{ cm}^{-3}$ <sup>[8]</sup>, thus pointing to the importance of the aforementioned effect.

5. A criterion of the type (39) has the clear meaning of the condition for the smallness of the "lifetime" of the atom  $1/\gamma$  compared with the characteristic time of variation of the ionic field  $\rho_{\text{eff}}/v_0$ , and was already introduced in the literature on this basis<sup>[23,24,2]</sup> as a criterion for the quasistatic behavior of the ions upon their "convolution" with electrons, and in all cases it was assumed, from intuitive considerations, that  $\rho_{\text{eff}} \sim N^{-1/3}$ . Yet in the general case  $C_2 \equiv \alpha \neq 0$  (non-central components), the problem includes besides  $N^{-1/3}$  one more characteristic length, namely the Weisskopf radius  $\alpha/v_0$ , so that it is not at all evident beforehand which of these lengths (or their combination) plays in each concrete case the role of  $\rho_{\text{eff}}$ ; the latter can be found only from a direct calculation of the contribution of the corresponding broadening mechanism. Thus, according to<sup>[25]</sup>, for pure phase modulation  $\rho_{\text{eff}} \sim \alpha/v_0$ . It is easy to verify that allowance for the amplitude modulation leads only to some numerical decrease (by approximately a factor of four) of this value of  $\rho_{\text{eff}}$ . As to effects of non-adiabaticity, only one of them—the change in the energy of the Stark state, connected with the explicit time variation of the phase, is amenable to analysis in terms of the criterion (39)<sup>10</sup>. The corresponding estimate based on calculating the corrections for thermal motion (see<sup>[25,26]</sup>) gives a value  $\rho_{\text{eff}} \sim N^{-1/3} \ll \alpha/v_0$ , so that the non-adiabaticity effect leads, as in Sec. 1, to a more stringent "adiabatic" criterion of quasistatic behavior.

It is appropriate, in developing the results of<sup>[25,26]</sup>, to summarize the role of criteria of the type (39) connected with the damping in a number of quasistatic criteria. First, they always pertain only to the innermost part of the line ( $\omega - \omega_0 \ll \gamma$ ), whereas in the remaining part of the line ( $\omega - \omega_0 \gg \gamma$ ) a criterion of the "modulation" type (37) is valid and corresponds

formally to the limit  $\gamma = 0$ . Second, the value of  $\rho_{\text{eff}}$  is determined in the general case not only by the characteristics of the microfield (which would lead always to  $\rho_{\text{eff}} \sim N^{-1/3}$ ), but also by the character of its action on the atom ( $\alpha \neq 0$ ). The sometimes realized very simple case  $\rho_{\text{eff}} \sim N^{-1/3}$  is connected either with the fact that  $\alpha = 0$  (central component, amplitude modulation), or with the fact that  $\alpha$  drops out from the result because of some cancellation of the factors (non-central component, non-adiabaticity effect).

6. In conclusion we emphasize that one of the main qualitative conclusions of our analysis is as follows: unlike the quasistatic profile of the line itself, deviations from it as a result of thermal motion of the perturbing particles are due mainly to effects principally outside the framework of the classical adiabatic model of an oscillator with variable frequency.

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