Collective excitations of a heavy atom

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It is shown that the excitation spectrum of the electron shell of a heavy atom contains not only the single-particle levels but also two dipole-type collective levels of relatively small width. In the first approximation, the latter levels correspond to the oscillations of the atomic core relative to the nucleus. The collective levels are optically active, and the lower of the two has a substantial oscillator strength. The corresponding excitation energies are of the order Z Ry (Z is the atomic number) and correspond to the far-ultraviolet (soft x ray) region.

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1. It is usually considered that the atomic excitation spectrum is single-particle in character: excitations correspond to the transition of one or more electrons to higher energy states with the formation of the corresponding number of holes. It has been established in recent years (see, for example, [1]) that other electrons filling the same subshells are, in fact, involved in these transitions as well. However, this type of collective effect does not affect the standard single-particle classification of excitation levels, which is determined by the quantum numbers of electrons and holes in the corresponding self-consistent field.

The existence of special (truly collective) excitations in the electron shell of a heavy atom, which could be described by specific quantum numbers, remained an open question for a considerable time. In classical language, such collective states correspond to the oscillations of a charged liquid drop which could be used as a model of the atomic shell of a heavy atom. On the other hand, collective states might be the analogs of plasma oscillations of an electron gas in macroscopic systems. Questions of this kind are discussed in [2] where, in addition to the history of the problem, there is a review of many physical aspects of collective states.

The question as to whether the collective states exist at all is not a simple one by far, and the difficulties associated with answering it have been overcome only recently. The theory of the dielectric response of the electric shell of the atom, which provides an adequate description of the attenuation of collective states, was proposed more than ten years ago [3,4] (see also [2]), but the answer to the above question (which reduces to the determination of the ratio of the width of the collective state to its energy) could not be obtained without performing very complicated numerical calculations. Such calculations have now been performed and their results show (see the brief account given in [5]) that the excitation spectrum of a heavy atom does, in fact, contain two dipole-type collective states of relative small width. These states are optically active and are capable of providing an appreciable contribution to processes connected with the excitation of the atomic shell.

2. The description of collective states is most simply formulated in the language of response functions. These include the longitudinal permittivity[1)] ϵ and the polarization operator Π which are defined by

$$\int dx' \varepsilon(x, x', \omega)\delta U(x', \omega) = \delta U_e(x, \omega), \tag{1}$$

$$\delta n(x, \omega) = \delta n_e(x, \omega) + \int dx' \Pi(x, x', \omega)\delta U(x', \omega). \tag{2}$$

In these expressions, δU_e, δU, δn_e, and δn are the Fourier components of the variations of, respectively, the external and total potential and the external and total charge density. The expressions given by (1) and (2) are written for the general case of a spatially inhomogeneous system such as an atom.

Combining (2) with the Poisson equation, we obtain

$$\Delta\delta U(x, \omega) + 4\pi \int dx' \Pi(x, x', \omega)\delta U(x', \omega) = -4\pi\delta n_e(x, \omega).$$

Collective states correspond to the natural oscillations of the system, and are described by the last equation [or Eq. (1)] without the right-hand side:

$$\Delta\delta U(x, \omega) + 4\pi \int dx' \Pi(x, x', \omega)\delta U(x', \omega) = 0. \tag{3}$$

Its eigenvalues $\omega = \Omega + i\Gamma$ give the frequency Ω and the attenuation Γ of the collective states. All this must, of course, be augmented by two boundary conditions. One is

$$\delta U|_{r=0} < \infty. \tag{4}$$

However, the question of the second condition is not at all trivial, and has given rise to difficulties for many of the researches concerned with collective states. At first sight, this second condition might be chosen in the form

$$\delta n|_{r\to\infty} \propto \Delta\delta U|_{r\to\infty} \to 0. \tag{5}$$

It turns out, however, that (3), (4), and (5) together predict that, depending on the chosen expression for Π, the lower level in the spectrum corresponds either to very small or zero energy. The low-lying levels correspond to disturbances δn on the distant periphery of the atom (see, for example, [6]). It is clear that such levels are not collective but single-particle states, i.e., simply the excitations of the outer electrons of the atom. We note in this connection that the energy of the collective states, which can be estimated from the formula for the plasma frequency

$$\Omega = (4\pi n)^{1/2} \sim Z \tag{6}$$

(according to the Thomas-Fermi model, $n \sim Z^2$; here and henceforth, we are using atomic units), substantially exceeds the ionization energies for the outer electrons. It follows that the collective state lies in the continuous spectrum of single-particle excitations.

The correct boundary condition which would free us from the background of low-lying single-particle excitations can be obtained from the following physical considerations, the validity of which is confirmed by subsequent calculations. The collective excitation is localized in the atomic core of radius $R \sim Z^{-1/3}$ in which there is a sufficiently dense electron gas and where coordinated mo-

tions of a large number of particles are, in fact, possible. Such an excitation has a finite lifetime. Among the more important channels for its decay, we have a) decay into a particle-hole pair in the region of localization of the collective state (analog of the Landau damping), b) radiative decay corresponding to the emission of quanta of electromagnetic radiation, and c) excitation (ionization) of outer electrons by the field due to the oscillating atomic core. In the foregoing discussion, we are essentially concerned with the decay products of the collective state corresponding to this last channel.

Collective-state calculations should, therefore, be focused on excitations localized in the atomic core, and outer electrons should be regarded only as a source of one of the partial widths of its decay. Correspondingly, the condition given by (5) should be replaced by the boundary condition

$$\delta n|_{r=R}\infty\Delta\delta U|_{r=R}=0, \tag{7}$$

where R should be varied within certain limits. The validity of the foregoing scheme will be confirmed by the appearance of a plateau on the functions $\Omega(R)$ and $\Gamma(R)$. The values of Ω and Γ corresponding to this plateau can be looked upon as the energy and attenuation of the collective state. More precisely, the quantity Γ obtained in this way is the partial width corresponding to decay channel (a). The other partial widths should be added to it.

3. The other difficulty encountered in connection with the collective problem is that, in contrast to the homogeneous medium, the ratio Γ/Ω for the atom does not contain small literal parameters. It is therefore only a numerical calculation, performed within the framework of a theoretical scheme capable of describing collective-state decay, that could lead to a small value for this ratio, and thus to the conclusion that the collective state does indeed exist.

All this ensues from the following considerations. It is well known (see, for example, [7]) that, in a homogeneous compressed electron gas [force parameter $\beta = e^2/\hbar v = p_F^{-1} \ll 1$, where $p_F = (3\pi^2 n)^{1/3}$ is the Fermi momentum], the decay of a collective state into one particle-hole pair is strictly forbidden by the energy-momentum conservation laws if the collective state momentum lies below a critical value, whereas decay into two or more pairs is suppressed because β is small. Hence, for long-wave collective states in homogeneous media, $\Gamma/\Omega \sim \beta \ll 1$. In the case of an atom, on the other hand, the situation is quite different. Although here too, the force parameter for a heavy atom is small ($\beta \sim Z^{-2/3} \ll 1$) and, therefore, the decay of the collective state into two or more pairs can be neglected, the channel corresponding to the decay into one pair is always open. This is so because the distribution of electrons in the atom is essentially inhomogeneous. A measure of this inhomogeneity is the length $l \sim Z^{-1/3}$ within which there is an appreciable change in the density. Because of the inhomogeneity, the momentum is a poor quantum number and is no longer conserved during the decay of the collective state. This corresponds to the loss or gain of momentum $\sim l^{-1}$ during scattering by inhomogeneities in the system.

Since momentum is not conserved, the channel representing the decay of the collective state into one pair in the atomic core [channel (a)] is open. The corresponding ratio Γ_a/Ω for the atom is of the order of unity, i.e., not zero. This follows from the fact that the collective state problem contains two characteristic lengths, namely, the distance between the particles $d \sim p_F^{-1} \sim Z^{-2/3}$ and the Debye screening length $r_D \sim (\beta^{1/2} p_F)^{-1} \sim Z^{-1/3}$. On the other hand, this ratio can be expressed in terms of dimensionless parameters containing l in the denominator. These parameters are d/l and r_D/l. The latter is of the order of unity, and this leads to the above result for Γ_a/Ω.

From this and from (6), we find that

$$\Gamma_a \sim \Omega \sim Z. \tag{8}$$

We emphasize that it is precisely the width Γ_a that plays the most important role and is the only one that can be comparable with the energy Ω of the collective state.[2)] To establish the existence of the collective state, it is sufficient to confine our attention to the width given by (8). We shall do this, and henceforth omit the subscript a on Γ. It is also important to note that the numerical coefficient in (8) is anomalously low (see below). Hence, one cannot exclude the possibility that the true collective-state width will be determined by some other decay channel. However, in any case, the total width of the collective state will be relatively small in comparison with its energy.

4. We must now determine the polarization operator in (3). According to (7), our analysis is confined to the atomic core, where, except for the unimportant region of small distances $r < Z^{-1}$, we have the quasiclassical condition for the electrons $(lp_F)^{-2} \sim Z^{-2/3} \ll 1$.

Therefore, in the expressions discussed below, we can pass to the quasiclassical limit. In particular, the ground state of the atom can be described by the Thomas-Fermi model. Another source of simplification is the fact that the parameter $\beta \sim Z^{-2/3}$ is small (see Sec. 3). This enables us to work in lower perturbation-theory order in the interaction between the electrons (for the polarization operator), and this corresponds to the usual random-phase approximation for the description of homogeneous systems. The accuracy of the approximations used below is, therefore, determined by the parameter $Z^{-2/3}$.

The final result of the analysis is relatively simple:

$$\Pi(\mathbf{x},\mathbf{x}',\omega)=-\frac{p_F(\mathbf{x})}{\pi^2}\left\{\delta(\mathbf{x}-\mathbf{x}')+i\omega\int_{-\infty}^{0}dt\,e^{-i\omega t}\langle\delta(\mathbf{x}(t)-\mathbf{x}')\rangle\right\} \tag{9}$$

where $p_F(\mathbf{x}) = [2(\mu - U(\mathbf{x}))]^{1/2}$, μ is the Fermi energy, $\mathbf{x}(t)$ is the solution of Newton's equation of motion for a particle in the Thomas-Fermi field

$$\ddot{\mathbf{x}}=-\nabla U \tag{10}$$

subject to the initial conditions

$$\mathbf{x}(0)=\mathbf{x},\quad \dot{\mathbf{x}}(0)=p_F(x)\mathbf{n}, \tag{11}$$

$\mathbf{n}$ is a unit vector in the direction of initial velocity, and $\langle\ldots\rangle$ represent averaging over $\mathbf{n}$.

The expression given by (9) was obtained in [3] (see also [2,4]) by the Green function method. We shall not repeat this derivation here, and will merely outline its principle. The polarization operator corresponds to a closed particle-hole loop, and is given by

$$\Pi(\mathbf{x},\mathbf{x}',\omega)=-2i\int_{-\infty}^{\infty}\frac{d\varepsilon}{2\pi}G(\mathbf{x},\mathbf{x}',\varepsilon+\omega)G(\mathbf{x}',\mathbf{x},\varepsilon),$$

where the Green function is given by

$$G(\mathbf{x},\mathbf{x}',\varepsilon)=i\int_{-\infty}^{0}dt\exp(it(\hat{H}-\mu))\,[\theta(t)-\theta(\varepsilon-\mu)]\,\delta(\mathbf{x}-\mathbf{x}').$$

Substituting this expression into the preceding one, and passing to the quasiclassical limit, we obtain (9), where the classical particle trajectory $\mathbf{x}(t)$ arises from the Heisenberg position operator $\exp(i\hat{H}t)\mathbf{x}\exp(-i\hat{H}t)$ and $\hat{H}$ is the Hamiltonian for the particle in the Thomas-Fermi field.

A simpler derivation, which we shall reproduce in full, is based on the utilization of the classical collisionless-transport equation (see, for example, [8])

$$\left(\frac{\partial}{\partial t}+\mathbf{p}\nabla-\nabla U\nabla_{p}\right)\delta f(\mathbf{x},\mathbf{p},t)=\nabla_{p}f_{0}\,\nabla\delta U. \tag{12}$$

In this expression, $f_0=\theta[p_F^2(\mathbf{x})-p^2]$ is the electron distribution function in the ground state and δf is its variation, where

$$\delta n(\mathbf{x},\omega)-\delta n_e(\mathbf{x},\omega)=2\int d^3p\,\delta f(\mathbf{x},\mathbf{p},\omega). \tag{13}$$

In the Lagrange picture, the equation given by (12) can be written in the form

$$\frac{d}{dt'}\delta f(\mathbf{x}(t'-t),\mathbf{p}(t'-t),t')=A(\mathbf{x}(t'-t),\mathbf{p}(t'-t),t'),$$

where $\mathbf{p}(t)=\mathbf{x}(t)$, $\mathbf{x}(0)=\mathbf{x}$, $\mathbf{p}(0)=\mathbf{p}$, and A represents the right-hand side of (12). Hence, using the causality principle, we have

$$\delta f(\mathbf{x},\mathbf{p},t)=\int_{-\infty}^{0}dt'A(\mathbf{x}(t'),\mathbf{p}(t'),t+t').$$

The right-hand side of this equation can be written in the form

$$-2\delta(p_F^2(\mathbf{x})-p^2)\int d\mathbf{x}'\int_{-\infty}^{0}dt'\frac{d}{dt'}[\delta(\mathbf{x}(t')-\mathbf{x}')]\,\delta U(\mathbf{x}',t+t'),$$

where we have used the fact that $p_F^2(\mathbf{x}(t))-p^2(t)=p_F^2(\mathbf{x})-\mathbf{p}^2$, $(\mathbf{p}(t)\nabla)F(\mathbf{x}(t))=\dot{F}$. Substitution of these expressions in (13) and comparison with (2) does, in fact, result in (9).

The characteristic feature of (9) is that it contains the classical trajectory of the electron in the self-consistent field. This can also be seen in the well-known expression for the polarization operator (or permittivity) in simpler problems, for example, in the case of an infinite or semi-infinite homogeneous medium, a plasma in a uniform magnetic field, and so on (see [8]). However, in the case of a nonuniform distribution of electrons inside the atom, the form of the classical trajectories turns out to be very much more complicated. We note that, in the case of the Thomas-Fermi field, the application of the scale transformation $\mathbf{x}\to Z^{-1/3}\mathbf{x}$, $t\to Z^{-1}t$ frees $\mathbf{x}(t)$ from explicit dependence on Z. It follows, in particular, that the frequency is present in (3) in the form of the self-similar combination ω/Z. This is in accordance with (8). [9]

5. The expression given by (9) is the kernel of the integral equation (3) and takes the form of a triple integral with respect to t, cos θ, and φ, where the last two arguments define the position of the unit vector $\mathbf{n}$. This difficulty can be avoided by substituting for the δ function in (9) in accordance with the following expression:

$$\delta(\mathbf{x}(t)-\mathbf{x}')=\sum_{n}D_n^{-1}\delta(t-t_n)\,\delta(\cos\theta-\cos\theta_n)\,\delta(\varphi-\varphi_n). \tag{14}$$

Here, t_n, cos θ_n, φ_n are the solutions of the equations $\mathbf{x}(t)=\mathbf{x}'$, $\mathbf{x}(0)=\mathbf{x}$, $\dot{\mathbf{x}}(0)=p_F(\mathbf{x})\mathbf{n}$, which can be interpreted, respectively, as the time of transit between $\mathbf{x}$ and $\mathbf{x}'$ and the angles defining the direction of the initial velocity of the particle on the trajectory connecting these points. There may be many such trajectories; we shall label them by the index n. In particular, in the case of periodic motion, summation was carried out over the number of cycles completed in both directions.

Next, the quantity D_n is the Jacobian

$$D_n=\left|\frac{\partial(x_1(t),x_2(t),x_3(t))}{\partial(t,\cos\theta,\varphi)}\right|$$

at the point t_n, cos θ_n, and φ_n. The derivatives $\partial x_i/\partial t$ in this expression are the velocity components at the point $\mathbf{x}'$.

Substitution of (14) in (9) yields the following final expression for the kernel in (3):

$$\Pi(\mathbf{x},\mathbf{x}',\omega)=-\frac{p_F(\mathbf{x})}{\pi^2}\left[\delta(\mathbf{x}-\mathbf{x}')+\frac{i\omega}{4\pi}\sum_{n}\frac{\exp(it_n\omega)}{D_n}\right],\quad t_n>0. \tag{15}$$

We note that, in explicit language, this expression corresponds to the geometric-optics approximation for electrons. This approximation is unsuitable if the points $\mathbf{x}$ and $\mathbf{x}'$ are joined by a continuum of trajectories, i.e., if they lie on caustics or, in a particular case, coincide with the foci of a beam of trajectories. However, the corresponding singularities are localized in a region of volume λ^3, where $\lambda\sim p_F^{-1}$ is the wavelength of the particle. It is readily shown that their contribution to (3) is determined by the quantity $(\lambda/l)^3\sim Z^{-1}$, and can be neglected.

In conclusion of this section, let us consider the imaginary part of the polarization operator (which determines the width Γ of the collective state) in the case of periodic motion. This case is important because the motion of an electron in an external field which can be adequately represented by the Thomas-Fermi field is, in fact, periodic (see next section). The Jacobian is then independent of n, and t_n is equal to t_0+nT for motion from $\mathbf{x}$ to $\mathbf{x}'$ in one direction, and to $(n+1)T-t_0$ for motion in the opposite direction in the closed cycle, where T is the period of the motion, $n=0-\infty$. Taking the imaginary part of (15), and using the causal circuit rule $\omega\to\omega+i\delta$, we obtain

$$\operatorname{Im}\Pi(\mathbf{x},\mathbf{x}',\omega)\propto\operatorname{Im}\operatorname{ctg}\frac{\omega T}{2}\propto\sum_{n}\delta\left(\omega-\frac{2\pi n}{T}\right). \tag{16}$$

The physical significance of this result is that the attenuation of the collective state is connected with a resonance between the frequency ω and the quasiclassical particle excitation frequency $2\pi n/T$. Clearly, this is the analog of Landau damping in the case of inhomogeneous but quasiclassical systems.

6. To analyze the collective state in a neutral heavy atom, we shall use the approximate Tietz formula [10]

$$U=Z/[r(1+\xi)^2],\quad \xi=r/\alpha,\quad \alpha=(9/2Z)^{1/6}. \tag{17}$$

The trajectories for $\mu=0$ and $M\neq 0$ in this field are found to be closed self-intersecting curves (Fig. 1). In terms of the polar coordinates in the plane of motion, the equation describing this curve is

$$\xi+\xi^{-1}=\Delta+1+(\Delta-1)\cos\varphi, \tag{18}$$

where $\Delta=Z\alpha/M^2-1$ and the law of motion is

$$\xi=\Delta-(\Delta^2-1)^{1/2}\cos\eta, \tag{19}$$

$$tM=(\Delta+1)(3\Delta+1)\eta/2-(2\Delta+1)(\Delta^2-1)^{1/2}\sin\eta+(\Delta^2-1)\sin(2\eta)/4.$$

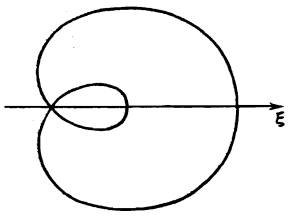

FIG. 1. Trajectory in the Tietz field for $\mu = 0$ and $m \neq 0$.

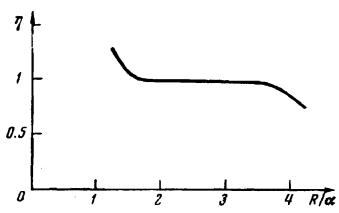

FIG. 2.

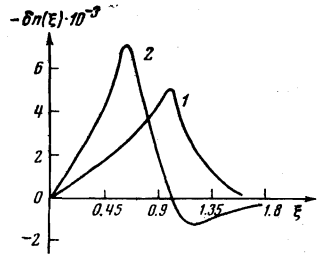

FIG. 3

FIG. 2. Plot of η as a function of R/α, where $\eta = 3 \times 2^{-3/2}\Omega Z^{-1}$Ry.

The period of the motion is given by[3)]

$$T=\pi(\Delta+1)(3\Delta-1)\alpha^2/M. \qquad (20)$$

Returning now to (15), we note, to begin with, that the sum in this expression, evaluated in accordance with the remarks at the end of Sec. 5, has the form

$$\sum_n \exp(i\omega t_n)=\frac{i\cos[\omega(t_0-T/2)]}{\sin(\omega T/2)}$$

[the expression given by (16) follows from this formula]. The quantity t_0 is equal to the smallest value of the difference $t(\eta')-t(\eta)$, where $\xi = \xi(\eta)$, $\xi' = \xi(\eta')$ [see (19)]. It will be convenient to introduce the angle Φ between the radius vectors directed toward the points **x** and **x'**. It is clear that $\Phi = \varphi' - \varphi$, where φ and φ' are found from (18) by substituting ξ and ξ' into the left-hand side. Evaluation of the Jacobian in (15) yields

$$D=\xi(\xi')^{1/2}\frac{[2Q(\xi)Q(\xi')]^{1/2}}{M(\xi+1)^2(\xi'+1)^2}\frac{\partial\cos\Phi}{\partial M},$$

where $Q(\xi) = 2\xi - M^2(\xi + 1)^2$.

We shall not reproduce the final equation obtained by substituting the expressions obtained in this section in (15) and then in (3). This final expression is rather unwieldy but, nevertheless, manageable. It is amenable to the separation of the angular variables through the consideration of multipole collective states $\delta U \sim Y_{lm}(\theta, \varphi)$. We shall confine our attention to dipole collective states ($l = 1$) which are particularly interesting from the point of view of photoatomic reactions. We note, in particular, that these collective states have the lowest excitation energy because radial oscillations ($l = 0$) are connected with a change in the density of the electron gas which has a high elasticity.

The procedure is to search for the collective state with relatively small width. Accordingly, we neglect the imaginary part of Π in (3) and, by solving the resulting equation, we obtain the excitation energy Ω and the perturbation δU of the potential. The corresponding perturbation in the density, δn, is found from the Poisson equation. The width of the collective state is calculated from the expression

$$\Gamma=-\frac{\langle \operatorname{Im}\Pi\rangle}{\langle \partial \operatorname{Re}\Pi/\partial\Omega\rangle},\quad \langle\ldots\rangle\equiv\int dx\,dx'\,\delta U(x)\delta U(x')(\ldots).$$

Only those solutions are retained for which Γ, found in this way, is small in comparison with Ω.

7. The numerical solution of the eigenvalue equation (3) subject to the boundary conditions given by (4) and (7) was performed on the BESM-6 computer (at the Computational Center Siberian Branch, USSR Academy of Sciences) by the supplemented vector method described in [11]. The $\Omega(R)$ and $\Gamma(R)$ curves were, in fact, found to have well-defined plateaus. Figure 2 shows the $\Omega(R)$ curve for one of the collective states. These calculations have demonstrated the existence of two dipole-type collective states with relatively small widths. Their energies and widths (electron-volts) are:

$$\Omega_1=13.74Z,\quad \Gamma_1=3\cdot10^{-3}Z,$$
$$\Omega_2=36.04Z,\quad \Gamma_2\cong10^{-4}Z. \qquad (21)$$

Figure 3 shows the behavior of the radial part of the density perturbation $\delta n(r)$ for both collective states. It is clear that, for the lower collective state (curve 1), this perturbation reduces in the first approximation to a shift of the shell as a whole relative to the nucleus. In point of fact, there is an attendant deformation of the shell which, however, remains fixed near the nucleus and on the periphery of the atom. For the upper collective state (curve 2), there is an additional node in the density perturbation.

We shall not consider possible experimental manifestations of the collective states and refer the reader to the review given in [2]. We merely note that, in addition to the peaks in the photoabsorption cross section, the collective states may lead to certain features in reactions involving the participation of heavy atoms, which are typical of the Bohr picture.

We are indebted to S. L. Mandel'shtam and E. L. Feĭnberg for their interest in this work and for useful discussions.

[1)]We note that, in the case of an inhomogeneous system, the permittivity which related δn to δn_e is not the same as the quantity in (1).

[2)]Detailed analysis of the partial widths corresponding to decay channels (b) and (c) will be given in a separate publication; here, we confine our attention to semiclassical estimates. The radiation width turns out to be of the order of $(Z/137)^3$, and is indeed small in comparison with Ω. The width connected with the excitation of the outer electrons, on the other hand, is given by an expression whose value decreases with increasing Z, and turns out to be smaller still. The point is that the characteristics of the outer electrons and the field δU which excites them are both independent of Z. Only the excitation energy depends on Z [see (6)], and the cross section for the corresponding process decreases with increasing Z.

[3)]The period is of the same order of magnitude as the characteristic time of the problem, namely, the period Ω^{-1} of the collective oscillation. This means that the real trajectory has nothing in common with the trajectory of rectilinear and uniform motion [for the same initial conditions (1'1)] which corresponds to the approximation frequently used in the theory of collective states. This approximation is based on the utilization of expressions valid for homogeneous media in which the density n is replaced by n(x).

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