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PLASMA OSCILLATIONS OF THE ELECTRON SHELL OF THE ATOM

D. A. KIRZHNITS and Yu. E. LOZOVIK

P. N. Lebedev Physics Institute, Academy of Sciences, U.S.S.R.

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THE question of plasma (collective) oscillations of the atomic shell, raised more than thirty years ago[1], has been recently getting more attention. The possible manifestations of an atomic plasmon in such processes as loss of energy by a fast particle in a medium[1-2], plasma oscillations and characteristic losses of electrons in a metal[3-4], etc. were already discussed in the literature in their time. Of special interest at present is the participation of the plasmon in various types of atomic reactions brought about by electron-atom and atom-ion collisions or by interactions of electromagnetic radiation with the atom[5-8]. This interest has been stimulated by the appearance of new experimental data on atomic reactions with energy transfer ranging from tens to thousands of electron volts. Besides experiments on electron-atom collisions[9-11], special interest is evinced in new and to a considerable degree unique data on ion-atomic collisions[12-14] obtained by using a coincidence technique to register the states of the two participants in the reaction.*

Plasma oscillations in the atomic shell have the same nature as in the electron fluid of a metal or in a plasma: when the particle density deviates from its equilibrium value, a Coulomb restoring force is produced. In quantum language, the plasmon as an elementary excitation constitutes a definite superposition of ordinary single-particle excitations of the particle-hole type. This superposition encompasses, with one weight or another, all the filled levels of the atom.†

To determine the nature of the atomic plasmon it is necessary to evaluate primarily the following factors:

a) the spectrum of the oscillations—the natural frequencies damping, and the character of the oscillations (radial, dipole, etc.);

b) the characteristics describing the probability of excitation of the plasma oscillations (in particular, the oscillator strengths);

c) the degree and character of participation of the plasmon in atomic reactions.

*A special conference on collective effects in the atom was held in February of this year at the Physicotechnical Institute im. A. F. Ioffe of the Academy of Sciences, where most of these data were reported.

†In spite of the prevalent opinion, the plasmon cannot be regarded as a bound state of a particle and a hole, since the corresponding annihilation diagrams correspond not to attraction but to repulsion. Accordingly, the plasmon energy in a homogeneous medium lies above the energy levels of the interacting particles and holes.

We must point out immediately that these factors have not yet been uniquely evaluated. The experimental data do not lend themselves to an unambiguous interpretation, and the theoretical analysis encounters serious difficulties. The problem is particularly acute when it comes to the damping of plasma oscillations, that is, essentially to the very existence of the atomic plasmon (see[4,15] and Sec. 4 below concerning this question).

In this brief review we describe the present status of the theory of the atomic plasmon. We use the atomic units $e = \hbar = m = 1$.

1. The simplest and historically the earliest method of describing plasma oscillations in a heavy atom is to start with their analogy with the hydrodynamic oscillations of a charged liquid drop. By considering the acoustic approximation to the nonstationary Thomas-Fermi equation, that is, by regarding the deviation from the equilibrium density and the velocity as small quantities, we arrive[1-2] at the following expressions for the natural frequencies ω_n and the oscillator strengths f_n:

$$\omega_n = k_n Z, \qquad f_n = q_n Z, \tag{1}$$

where Z is the number of electrons in the atom, and k_n and q_n are numerical coefficients of the order of unity.

It must be noted that the foregoing estimates are valid when $Z \gg 1$ also outside the framework of the hydrodynamic approach; thus, an estimate for $\omega_n \sim v_0/l$ (v_0—characteristic velocity, l—characteristic length) is obtained directly from the relations $v_0 \sim Z^{2/3}$ and $l \sim Z^{-1/3}$, which follow from the Thomas-Fermi equation.

Formula (1) shows thus that the plasma oscillations correspond to a region intermediate between the optical and the x-ray spectra (see[7] for details).

Relations (1) were used in[1-2] to determine the energy loss of a fast particle in a medium (the Baker-Bloch formula). Although this formula is in good agreement with experiment, we still can not conclude from this that the atomic plasmon really exists, since we are uncertain whether we deal with a real excitation of plasma oscillation or with some averaged effect of single-particle excitations[16].

The discussed hydrodynamic approach has many shortcomings. First, it is impossible to solve within its framework the question of the damping without introducing special dissipative terms. This in itself already indicates the need for enrolling additional microscopic information. In addition, even when applied to a homogeneous medium, a correct result is obtained for ω_n only in the long-wave limit[4]. Yet in an atom this limit is unattainable because of the boundedness of the system (the wavelength of the plasmon cannot exceed the dimensions of the atom).

2. A consistent microscopic description of plasma oscillations is best expressed in the language of the dielectric constant. To abbreviate the notation, we assume the longitudinal dielectric constant of the atom* $\epsilon(\omega, \mathbf{x}, \mathbf{x}')$ to be a matrix relative to $\mathbf{x}$ and $\mathbf{x}'$ and denote it by the symbol $\hat{\epsilon}(\omega)$.

Denoting by $\epsilon(\omega)$ the eigenvalue of the matrix $\hat{\epsilon}(\omega)$, we can determine the natural frequencies ω_n and the damping γ_n of the plasma waves from the equation $\epsilon(\omega_n + i\gamma_n) = 0$. In other words,

$$\int d\mathbf{x}'\varepsilon(\omega_n + i\gamma_n, \mathbf{x}, \mathbf{x}')\Phi(\mathbf{x}') = 0, \tag{2}$$

where $\Phi(\mathbf{x})$ is the eigenfunction of the matrix $\hat{\epsilon}(\omega)$, subject to the required boundary conditions.† In more compact form, the equation for the determination of ω_n and γ_n can be written as the condition for the vanishing of the resolvent of the kernel $\epsilon(\omega, \mathbf{x}, \mathbf{x}')$:

$$\exp(-\mathrm{Sp}\ln\hat{\varepsilon}(\omega_n + i\gamma_n)) = 0. \tag{3}$$

With the aid of the matrix $\hat{\epsilon}(\omega)$ we can express practically all the pertinent characteristics of the system[17-18]. In particular, the spectral density of the oscillator strengths is

$$g(\omega) = -\frac{\omega}{2\pi^2}\,\mathrm{Im}\int dx\,dx'\hat{\varepsilon}^{-1}(\omega). \tag{4}$$

This quantity satisfies the usual sum rule

$$\int_0^\infty g(\omega)\,d\omega = Z.$$

To determine the dielectric constant of the atom itself, many workers employ the so called quasihomogeneous approach. It consists in substituting in the expression for ϵ for a homogeneous medium, which depends on the values of the density ρ as a parameter, the quasilocal value $\rho(\mathbf{x})$ at the given point of space in place of ρ.

In order for this approach to be valid it is necessary that $\rho(\mathbf{x})$ vary sufficiently smoothly. If we introduce the characteristic inhomogeneity length l, over which $\rho(\mathbf{x})$ changes noticeably, then it is necessary that l be much larger than all the characteristics of the system (with dimensions of length), entering in the expression for ϵ. These include the mean distance between particles $d \sim \rho^{-1/3}$ and a quantity which in the homogeneous case has the meaning of the Debye screening radius, $r_D \sim \rho^{-1/6}$. In an atom with $Z \gg 1$ we have $\rho \sim Z^2$, $l \sim Z^{-1/3}$, and $r_D \sim Z^{-1/3}$. Thus, although $l \gg d$, we always have $l \sim r_D$. Therefore the quasihomogeneous approach to the problem of calculating ϵ for an atom is not valid in principle[6,15].* This circumstance makes worthless the quantitative deductions of most papers devoted to the calculation of the spectrum of the atomic plasmon.†

3. The exact microscopic expression for the dielectric constant is

$$\hat{\varepsilon}(\omega) = \delta(\mathbf{x}-\mathbf{x}') + \int dx''\frac{\Pi(\omega, \mathbf{x}, \mathbf{x}'')}{|\mathbf{x}''-\mathbf{x}'|},$$

where Π is the polarization operator (a quantity describing the closed particle-hole loop)[6]. This expression can be represented in the form of a Kramers-Kronig dispersion integral[17]

$$\hat{\varepsilon}(\omega) = \delta(\mathbf{x}-\mathbf{x}') - \frac{1}{2\pi}\int_{-\infty}^{+\infty}\frac{d\omega' F(\omega', \mathbf{x}, \mathbf{x}')}{\omega^2 - \omega'^2 + i\delta}. \tag{5}$$

The calculation of $\hat{\epsilon}$ for atoms with relatively small values of Z is an exceedingly difficult problem. The situation becomes much simpler when $Z \gg 1$, a case to which we confine ourselves. More accurately speaking, we neglect terms having a relative order $Z^{-2/3}$.

We recall first that the condition $Z \gg 1$ corresponds to the inequality $r_D \gg d$, as a result of which the particle-pair interaction energy turns out to be small compared with the kinetic energy. This makes it possible to replace Π by its lower-order perturbation-theory expression (the perturbing term is chosen to be the difference between the exact and self-consistent interaction)[6]:

$$\Pi(\omega, \mathbf{x}, \mathbf{x}') = \frac{i}{\pi}\int G_0(\varepsilon+\omega, \mathbf{x}, \mathbf{x}')\,G_0(\varepsilon, \mathbf{x}', \mathbf{x})\,d\varepsilon,$$

where

$$G_0(\varepsilon, \mathbf{x}, \mathbf{x}') = \sum_\nu \frac{\chi_\nu^*(\mathbf{x}')\chi_\nu(\mathbf{x})}{\varepsilon - \varepsilon_\nu + i\delta\,\mathrm{sign}(\varepsilon_\nu - \varepsilon_F)}$$

is the Green's function in the Hartree-Fock approxi-

*This quantity relates the longitudinal components of the induction and of the electric field:

$$D_\omega(\mathbf{x}) = \int d\mathbf{x}'\varepsilon(\omega, \mathbf{x}, \mathbf{x}')E_\omega(\mathbf{x}').$$

†In the spatially-homogeneous problem $\Phi(\mathbf{x}) = \exp(i\mathbf{k}\cdot\mathbf{x})$. Introducing the Fourier transform $\epsilon(\omega, \mathbf{k})$, of the dielectric constant in terms of the coordinate difference, we arrive at the well known equation.

$$\varepsilon(\omega_n + i\gamma_n, \mathbf{k}) = 0$$

(see, for example, [17]).

*At the same time, the quantities that can be expediently determined by means of the cruder self-consistent-field approximation can be calculated with the aid of the quasihomogeneous approach, for the parameter r_D does not enter at all in this approximation. This pertains, for example, to the electron density, a quasihomogeneous description of which is given by the Thomas-Fermi equation.

†We note in addition that the exponential (3) has been erronepusly replaced in [7] 0– the expression $[1 + \mathrm{Sp}\ln\epsilon(\omega)]^{-1}$; this leads, in particular, to an incorrect spectrum in the homogeneous-system limit.

mation, χ_ν and ϵ_ν are the eigenfunction and the energy in this approximation, and ϵ_F is the Fermi energy.

Using further the smallness of the parameter $Z^{-2/3}$, we can disregard the exchange terms and proceed to a quasiclassical description. Consistent utilization of the condition $Z \gg 1$ gives the following final expression for F, which is valid in that region of the atom where most electrons are situated[15]*:

$$F(\omega, \mathbf{x}, \mathbf{x}') = \frac{\omega^2 p_0(\mathbf{x})}{\pi^2} \overline{\left(\frac{1}{|\mathbf{x}(\tau) - \mathbf{x}'|}\right)_\omega} \tag{6}$$

Here $p_0(\mathbf{x}) = [3\pi^2 \rho(\mathbf{x})]^{1/3}$ is the limiting momentum defined by the Thomas-Fermi equation $\Delta p_0^2 = 8p_0^3/3\pi - 2Z\delta(\mathbf{x})$. The quantity $\mathbf{x}(\tau)$ is the classical coordinate of the particle moving in a self-consistent field, that is, the solution of the equation $\ddot{\mathbf{x}} = -\nabla p_0^2/2$ with initial conditions $\mathbf{x}(0) = \mathbf{x}$ and $\dot{\mathbf{x}}(0) = p_0(\mathbf{x})\mathbf{n}$. The superior bar denotes averaging over the directions of the unit vector $\mathbf{n}$, and the symbol $(\ldots)_\omega$ is the Fourier component with respect to τ. If the motion of the particle is periodic with period T, then the quantity $(\ldots)_\omega$ is replaced by the corresponding component of the Fourier series multiplied by the factor $[1 - \exp(-i\omega T)]^{-1}$. The poles of this expression correspond to quasiclassical single particle excitation energies.

The quasihomogeneous approach discussed above corresponds to replacing the real quantity $\mathbf{x}(\tau)$ by the trajectory of free motion with the same initial conditions, that is, $\mathbf{x} + p_0(\mathbf{x})\mathbf{n}\tau$. In this language, the inapplicability of the quasihomogeneous approach is manifest in the fact that after a characteristic time $\sim 1/\omega_n$ the true motion of the particle deviates noticeably from the uniform straight-line motion.†

We present by way of illustration the results of the calculation of the trajectory in the field of the atom. Choosing for $p_0^2(\mathbf{x})$ the expression[19]

$$p_0^2(\mathbf{x}) = \frac{2Z}{r}(1+\xi)^{-2},$$

$$\xi = \frac{r}{\alpha}, \quad \alpha = \left(\frac{9\pi}{16}\right)^{1/3} Z^{-1/3}$$

and introducing

$$\Delta = \frac{Z\alpha}{M^2} - 1$$

(M is the moment of the particle), we have in polar coordinates in the plane of motion

$$\xi + \frac{1}{\xi} = \Delta + 1 + (\Delta - 1)\cos\varphi.$$

This is a closed self-intersecting trajectory, corresponding to a period

$$T = \frac{\pi(\Delta+1)(3\Delta-1)\alpha^2}{M}.$$

The latter is precisely of the order of $1/\omega_n$.

We see from the foregoing relations the complicated computations involved in the problem of describing an atomic plasmon. This problem is still far from solved.

4. As already indicated, the most acute problem for the atomic plasmon is its damping. We recall that in the case of homogeneous medium the plasmon damping connected with its decay into single-particle excitations is small so long the plasmon momentum k does not exceed a certain value k_{cr}, which amounts to $(0.1-0.3)\ p_0$ in the density range of interest to us.[20,5] When $k < k_{cr}$ the energy and momentum conservation laws prevent the plasmon decay into one particle-hole pair, and decay into two or more pairs is suppressed to the same degree as the Debye radius exceeds the mean distance between particles.

Going over into the atomic plasmon, let us point out several factors that make its damping larger than for a homogeneous medium. First, as already noted, the wavelength of the plasmon in the atom is bounded from above and the effective value k_{eff} is accordingly bounded from below. This raises the question whether the condition $k_{eff} < k_{cr}$ is satisfied for the atomic plasmon, that is, whether the channel for plasmon decay into a particle-hole pair always remains open. This aspect of the problem was investigated in[5] using a very simple model with rectangular charge distribution. It was found that most possible types of oscillations indeed do not satisfy this condition, the only exception being the dipole type* oscillations with the lowest excitation energy.

The situation becomes much more aggravated by inhomogeneity in the particle distribution. Since scattering by the inhomogeneities leads to a momentum transfer of the order of $1/l$ (see Sec. 2), even the conservation laws may turn out to be satisfied in the decay of a plasmon into a particle-hole pair if the condition $k_{eff} < k_{cr}$ is fulfilled. In other words, since k is no longer a good quantum number, the plasmon spends part of the time in a state with $k > k_{cr}$. This results in a damping which is essentially connected with the inhomogeneity, and which is apparently the principal damping for the atomic plasmon.

The ratio of this damping to the plasmon frequency should be expressed in terms of dimensionless com-

*For several quite accidental reasons, this expression can be obtained also from the classical kinetic equation (for details see [15]).

†We note that (5) and (6) are obtained from the exact expression by neglecting the parameters d/r_D and d/l, but retaining all the powers of the parameter r_D/l. Yet in the quasihomogeneous approach the latter is also neglected.

*Oscillations corresponding to the shift of the shell as a whole relative to the nucleus are also dipole. This very simple model of plasma oscillations was considered long ago by E. L. Feĭnberg, and yields for ω_n estimates that are similar to (1). The possibility of satisfying in this case the adiabaticity of the oscillations and the smallness of the damping has, however, a low probability.

binations that contain l in the denominator: d/l, r_D/l, etc. The parameter r_D/l, according to the estimates of Sec. 2, is of the order of unity. Therefore there are no small parameters in the atom capable of making the ratio of the plasmon damping to its frequency small. The possible occurrence of small numerical coefficients is, of course, not excluded in principle, but the solution of this problem calls for the performance of a complete numerical calculation.*

So far we have been dealing essentially with plasmon decay processes that can occur in an atomic core. There is also a definite plasmon decay mechanism connected with the outer shells. It is sufficient to note that the plasmon energy in a heavy atom greatly exceeds the first ionization potentials. Therefore the plasmon level always lies in the continuous spectrum. If there is an effective mechanism for transferring the excitation energy to the outer shells then, provided the momentum and parity conservation laws are satisfied, the plasmon can always decay into a pair with production of a hole in the outer shell. From the point of view of the analysis that follows, the greatest interest attaches to the case when the corresponding particle falls in the continuous spectrum. The partial width of the plasmon level corresponding to this case will be denoted by γ_n^{ion}. When all the foregoing conditions are satisfied, the contribution of γ_n^{ion} to the total width γ_n may be appreciable.

5. If nevertheless the damping of the atomic plasmon remains not too large, then the presence of such a collective level in the atomic excitation spectrum can radically change the course of the atomic reactions, and the energy transfer may be of the order of the resonant energy ω_n. Under these conditions the Bohr mechanism may compete with the direct reaction mechanism, at least in principle[6].

We shall speak for concreteness of the collision between an electron and an atom. When a resonant electron falls into the atom, a virtual plasma oscillation is excited in the latter. The incident electron transfers its energy to a large number of electrons of the atom and "sticks" in the shell, which goes over into a strongly excited "heated" state (the corresponding effective temperature is of the order of 10^5 deg). This results in a relatively long-lived intermediate state—the "compound atom." The excitation energy can subsequently be concentrated on the outer electrons of the atom, and thus cause ionization ("evaporation" of the outer electrons)†; other secondary processes competing with ionization are, of course, also possible.

The most characteristic feature of a reaction proceeding via a compound atom is the absence of correlation between the states of the reaction products and the states of the colliding particles. The reaction proceeds, as it were, in two independent stages: formation of the intermediate system, and its decay with emission of the end products. The cross section for the reaction can accordingly be represented by a product of two factors:

$$\sigma^{if}(\omega) = \sigma_0^i(\omega)\, w^f(\omega), \tag{7}$$

the first of which represents the cross section for the formation of the compound atom, and the second the probability of its decay in a given channel f. The cross section σ^{if} is given by the well known Breit-Wigner formula; near resonance the cross section σ_0 is close to geometric:

$$\sigma_0 \leqslant \frac{1}{\omega_n} \sim \frac{1}{Z}.$$

$w^f = \gamma_n^f/\gamma_n$, where γ_n^f is the partial width corresponding to the f channel. In particular, for the ionization cross section we have $w^{ion} = \gamma_n^{ion}/\gamma_n$ (see end of Sec. 4).

We emphasize that if the mechanism considered is actually realized in electron-atom collisions, then the traditional juxtaposition of this process with the collision between slow nucleons and a nucleus is unjustified. In order to make the analogy with nuclear reactions, where strong interaction between particles is important, even more complete we point out that the interaction of the electrons in the atom in the resonance region is also strong. In order not to complicate the problem, let us consider a homogeneous medium. It is well known that when allowance is made for the interaction between the particles, the effective potential of the interaction is obtained by replacing the Coulomb potential $4\pi/k^2$ by the quantity $4\pi/k^2\epsilon(\omega, \mathbf{k})$. This quantity may turn out to be appreciable near resonance where $\epsilon(\omega, \mathbf{k}) = 0$ (see Sec. 2).

We note that excitation of the plasma oscillation in the shell can be effected not only by an electron incident from the outside, but also by an electron falling into the shell, so to speak, "from the inside." We have in mind a β electron emitted from the nucleus during radioactive decay. The corresponding excitation probability is given by the following estimating formula indicated by E. L. Feĭnberg:

$$W \approx 0.1 Z^{2/3} E_\beta^{-1},$$

where E_β is the kinetic energy of the β electron.

Let us also dwell briefly on the absorption of electromagnetic radiation by the atom. The corresponding cross section

$$\sigma(\omega) = 2\pi^2 g(\omega)$$

(see (4)) has obviously a resonant character near ω_n.

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*Certain indirect evidence in favor of appreciable damping is the presence of a series of broad lines in the spectrum of the characteristic electron losses in a metal; these lines are related to oscillations that are induced in the electron shells of the ions[9].

† Similar notions, although phenomenological to a certain degree, were developed earlier for ionization occurring when atoms collide with atoms [21, 22] (see also [23, 24]).

Using (1) we can readily obtain an estimate for the integral of $\sigma(\omega)$ over the resonance region

$$\int \sigma(\omega)\, d\omega = l_n Z,$$

where l_n is a numerical factor of the order of unity.

6. In conclusion let us consider briefly the available experimental data on atomic reactions in the energy region of interest to us.

For electron-atom collisions using a monoenergetic electron beam, there are data on the ionization cross sections of noble gases over a wide range of energies from threshold to 20 keV[9-11].

The most remarkable feature of the ionization cross sections of a given ion multiplicity k is the appreciable growth in the y fraction of the yield of ions of given k with increasing Z. Thus, the ratio of the maximum cross sections with k = 4 and k = 1 is ~0.001 for argon, ~0.01 for krypton, and ~0.1 for xenon. Inasmuch as the properties of the atomic outer shells from which the electrons are emitted change very slowly with Z, the foregoing data offer undisputed evidence of the collective nature of the ionization process, that is, of the important role of the internal shells of the atom.

However, the available experimental data are still insufficient to disclose the nature of this collective process. It is not clear, in particular, whether the plasma level* comes into play here or whether a direct reaction occurs, connected for example with the Auger effect.

The most extensive and most detailed set of data was obtained in experiments on the collisions between ions and atoms of noble gases[12-14].

We note first that the relative probability of deviation of the total charge of the produced ions from the corresponding maximum value is given by a universal Gaussian curve, which fits the points corresponding to different states of the incident particles[12]. This fact does not contradict the Bohr picture (see (7)).

However, other characteristic features observed in experiment are not readily interpreted in the spirit of the Bohr picture. This pertains, in particular, to the mechanism based on plasma excitations.†

It must be emphasized that in general there is still a certain disagreement between the data obtained by different authors. Nor is it clear whether the observed lines of the characteristic losses pertain to individual atoms or to a "quasimolecule" produced during the first stage of the process[13,14]. The interpretation of these experiments is already the subject of many specialized papers[8,25,26], but the proposed schemes can hardly be regarded as satisfactory from the point of view of the entire available experimental material.

It seems to us that in order to explain these experiments and to disclose the role of the atomic plasmon it is necessary to perform additional experiments. Especially important for the solution of the problem of the atomic plasmon would be an investigation of the photoatomic reactions with heavy atoms at energies of the order of several keV.

*We note that in this case the ionization cross sections should have a resonance at an energy ω_n. Resonances of this kind, perhaps not very reliable, were observed at $\omega \approx 1$ keV, for the cross sections of Xe^{3+} and Xe^{6+} in[10].

†It is impossible to agree with the notion introduced in[8], of plasma oscillations of individual shells of the atom, even for the sole reason that the excitations of the internal shells are not plasma but single-particle excitations (see[7] on this subject).

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