Moscow University Physics Bulletin
Vol. 55, No. 2, pp. 8–12, 2000

Vestnik Moskovskogo Universiteta. Fizika
UDC 533.951

INTERPRETATION OF SOME FEATURES OF THE PLASMON SPECTRUM OF SIMPLE METALS

I. M. Aleshin and D. V. Peregudov

The influence of a periodic nonuniform background on the propagation of charge density waves in simple metals is studied. It is shown that the incorporation of spatial nonuniformity in the Vlasov–Poisson equations leads to a plasmon spectrum that is qualitatively consistent with experimental data. Namely, calculations show that charge density waves decay in a nonuniform medium even at a zero quasimomentum value; at some values of the inverse lattice vector, the dispersion curve has a negative slope.

Experimental investigation of the plasmon spectrum in simple metals [1] showed that the model of a "free" electron gas [2] commonly used in calculations needs refinement: firstly, inverse dispersion is observed in the long-wave region of the spectrum of some alkali metals (such as cesium); secondly, the plasmon attenuation remains finite at a zero value of the wave vector (see also [3]). Theoretical studies [4, 5] explain these effects by a weak nonuniformity of the "gas" of valence electrons, produced by the crystal lattice potential. However, the formalism of equilibrium statistical mechanics used by the authors of [4, 5] leads to tedious calculations, which, in our view, conceals the physical mechanism of the phenomenon. Moreover, each of these studies explains only a part of the observed phenomena.

In the present paper, we propose a simple method for calculating the long-wave part of the plasmon spectrum, with the use of the kinetic theory tools [6]. The problem was first stated in this formulation in [7], where it was shown that the spectrum of plasma waves in a crystal has a zonal character.

In our calculations, we supposed that the medium is nonuniform in one direction and, in addition, the equilibrium velocity distribution of electrons was assumed Maxwellian. Thus, the problem at hand is a model one. In actual metals, the electron gas is strongly degenerate since its adequate description requires a kinetic equation for the density matrix [8] and the three-dimensional structure of the crystal lattice has to be taken into account as well. The realization of this program is not a particular problem, but it still requires a significant amount of calculations. However, even in the framework of our model assumptions, it is possible to qualitatively describe both the plasma wave dispersion in metals and the wave attenuation features*.

Consider the simplest model crystal, consisting of two types of particles: positive ions (i) and electrons (e). The ions are assumed to be stationary to form a periodic nonuniform background. We write the ion distribution function in the form

$$f_i = [n + \rho(\mathbf{r})]\delta(\mathbf{p}), \quad \langle\rho(\mathbf{r})\rangle = 0,$$

* When comparing the results with the experimental data, the temperature T (the characteristic parameter of the Maxwell distribution) should simply be replaced with the Fermi energy ε_F (the characteristic parameter of the Fermi distribution).

where $\rho(\mathbf{r})$ is the periodic function; n is the mean concentration of ions; $\mathbf{p}$ is the momentum, $\mathbf{p} = m\mathbf{v}$; $\delta(\mathbf{p})$ is the three-dimensional Dirac delta. The angular brackets here and below denote the average over the lattice period.

The dynamics of the electron component is described by the Vlasov–Poisson equations

$$\partial_t f_e + \mathbf{v}\partial_\mathbf{r} f_e + e\partial_\mathbf{r}\phi\partial_\mathbf{p} f_e = 0,$$

$$\Delta\phi = -4\pi e \int d^3p\,(Zf_i - f_e),$$

where e is the elementary charge, f_e is the electron distribution function, ϕ is the self-consistent potential, Z is the ion charge. In the electron distribution function, we isolate three terms: F_0 describing the uniform background, an equilibrium periodic nonuniform additive f_0, and a nonequilibrium perturbation f_1,

$$f_e = F_0(\mathbf{p}) + f_0(\mathbf{p},\mathbf{r}) + f_1(\mathbf{p},\mathbf{r},t).$$

By virtue of the quasineutrality condition

$$\int d^3p\,F_0(\mathbf{p}) = Zn, \quad \left\langle \int d^3p\,f_0(\mathbf{p},\mathbf{r}) \right\rangle = 0,$$

$$\int d^3r \int d^3p\,f_1(\mathbf{p},\mathbf{r},t) = 0.$$

We represent the potential ϕ as the sum of the equilibrium part and the nonequilibrium perturbation $\phi = \phi_0(\mathbf{r}) + \phi_1(\mathbf{r},t)$, where

$$\Delta\phi_0(\mathbf{r}) = -4\pi e\left[Z\rho(\mathbf{r}) - \int d^3p\,f_0(\mathbf{p},\mathbf{r})\right],$$

$$\Delta\phi_1(\mathbf{r},t) = 4\pi e \int d^3p\,f_1(\mathbf{p},\mathbf{r},t).$$

We also set $\langle\phi_0(\mathbf{r})\rangle = 0$, which can always be ensured by an appropriate choice of the potential origin.

The equilibrium distribution function is determined from the stationary Vlasov equation ($\partial f_0/\partial t = 0$). We are interested in the case where F_0 is a Maxwell distribution. Then

$$f_0(\mathbf{p},\mathbf{r}) = \left[C\exp\left(\frac{e}{T}\phi_0(\mathbf{r})\right) - 1\right]F_0(\mathbf{p}).$$

The constant C is determined from the condition $\left\langle\int d^3p\,f_0(\mathbf{p},\mathbf{r})\right\rangle = 0$, and $C = \left\langle\exp\left(\frac{e}{T}\phi_0(\mathbf{r})\right)\right\rangle^{-1}$.

In what follows, we assume that the function ϕ_0 is available from experimental data and equal to the metal's pseudopotential [9].

The equation for the perturbations has the form

$$\partial_t f_1 + \mathbf{v}\partial_\mathbf{r} f_1 + e\partial_\mathbf{r}\phi_0\,\partial_\mathbf{p} f_1 + e\partial_\mathbf{r}\phi_1\,\partial_\mathbf{p}(F_0 + f_0) + e\partial_\mathbf{r}\phi_1\,\partial_\mathbf{p} f_1 = 0. \tag{1}$$

The last, nonlinear term will be henceforth omitted. This can be done under the condition $2e\phi_1 k^2/m\omega^2 \ll 1$, where ω and k are the frequency and the wave vector of the perturbation.

The linearized equation is an equation with periodic coefficients, whose solutions have the form of Bloch waves:

$$f_1(\mathbf{p},\mathbf{r},t) = \mathrm{e}^{-i\omega t + i\mathbf{k}\mathbf{r}}\,h_0(\mathbf{p}) + \sum_{n\neq 0} h_n(\mathbf{p})\,\mathrm{e}^{-i\omega t + i\mathbf{k}_n\mathbf{r}} = F + \delta f,$$

$$\phi_1(\mathbf{r},t) = \mathrm{e}^{-i\omega t + i\mathbf{k}\mathbf{r}}\,\chi_0 + \sum_{n\neq 0}\chi_n\,\mathrm{e}^{-i\omega t + i\mathbf{k}_n\mathbf{r}} = \Phi + \delta\phi.$$

In order to simplify the notation, we use the compound subscript $n = \{n_1, n_2, n_3\}$ and the inverse lattice vector $\mathbf{b} = \{\mathbf{b}_1, \mathbf{b}_2, \mathbf{b}_3\}$, the notation $\mathbf{b}n$ means $\mathbf{b}_1 n_1 + \mathbf{b}_2 n_2 + \mathbf{b}_3 n_3$.

In the last formulas, F and Φ denote the zero harmonics of the distribution function and the potential, and all terms with $n \neq 0$ are denoted as δf and $\delta\phi$. From the physical standpoint, this means decomposition of the perturbation into components that are slowly ($n = 0$) and rapidly ($n \neq 0$) varying in space. This decomposition makes sense provided $k \ll b$.

The periodic functions f_0 and ϕ_0 can be represented in a similar form (we recall that $\psi_0 = 0$ by construction):

$$f_0(\mathbf{p}, \mathbf{r}) = \sum_n g_n(\mathbf{p})\, e^{ibnr} = F_0 + \delta f_0, \quad \phi_0(\mathbf{r}) = \sum_n \psi_n\, e^{ibnr}.$$

When these expressions are substituted into (1), the rapidly and slowly varying terms should be cancelled separately. Isolating the rapidly varying terms, we have

$$\begin{gathered}(\partial_t + \mathbf{v}\partial_\mathbf{r})\,\delta f + e\partial_\mathbf{r}\delta\phi\,\partial_\mathbf{p}F_0 + e\partial_\mathbf{r}\Phi\,\partial_\mathbf{p}\delta f_0 + e\partial_\mathbf{r}\phi_0\,\partial_\mathbf{p}F = 0, \\ \Delta\delta\phi = 4\pi e\int \delta f\, d^3p.\end{gathered} \tag{2}$$

To obtain equations for the slowly varying terms, we average (1) over the lattice period:

$$\begin{gathered}(\partial_t + \mathbf{v}\partial_\mathbf{r})\,F + e\partial_\mathbf{r}\Phi\,\partial_\mathbf{p}F_0 + e\,\langle\partial_\mathbf{r}\delta\phi\,\partial_\mathbf{p}\delta f_0\rangle + e\,\langle\partial_\mathbf{r}\phi_0\,\partial_\mathbf{p}\delta f\rangle = 0, \\ \Delta\Phi = 4\pi e\int F\, d^3p.\end{gathered} \tag{3}$$

Here we have a direct analogy with the method proposed by P. L. Kapitsa for solving the mechanical problem of the motion of a pendulum with an oscillating suspension point [10]. This analogy was earlier used by one of the authors of [11] to solve the equations of hydrodynamic of cold plasma (investigating the nonlinear dynamics of plasma waves in a periodic nonuniform medium).

Equation (2) is valid at all values of the potential ψ_0 but it is difficult to solve exactly. Therefore, to construct a perturbation theory, we draw on the fact that the parameter ϕ_0 is small in simple metals. In what follows, we restrict ourselves to accounting for the first nonvanishing corrections (quadratic in ϕ_0^2).

In order to obtain the function $\omega(\mathbf{k})$ explicitly, we make further simplifying assumptions. Let us consider a one-dimensional problem and assume that the potential ϕ_0 has only one (first) harmonic. It is convenient to introduce characteristic parameters: the Langmuir frequency, the Debye radius, and the thermal velocity: $\omega_p = \left(4\pi e^2 n/m\right)^{1/2}$ (m is the electron mass), $r_D = \left(T/4\pi e^2 n\right)^{1/2}$, $v_T = (T/m)^{1/2}$, and to pass to dimensionless frequency, wave vector, and velocity. In addition, we introduce the dimensionless parameter $\xi = |e\psi_1/T|$.

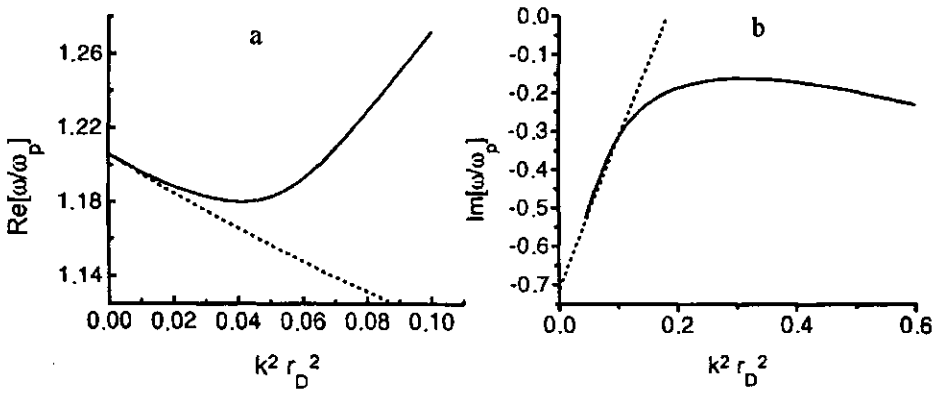

Fig. 1

(a) Frequency and (b) damping factor of plasmons as functions of quasimomentum squared. Solid curves — long-wave approximation, dots — numerical calculation.

The frequency correction is expressed in terms of three reference integrals:

$$J_1(z) = \int\limits_{-\infty}^{\infty} \frac{1}{z-v}\partial_v F(v)\,dv,$$

$$J_2(z_1,z_2) = \int\limits_{-\infty}^{\infty} \frac{1}{z_1-v}\partial_v \frac{1}{z_2-v}\partial_v F(v)\,dv,$$

$$J_3(z_1,z_2) = \int\limits_{-\infty}^{\infty} \frac{1}{z_1-v}\partial_v \frac{1}{z_2-v}\partial_v \frac{1}{z_1-v}\partial_v F(v)\,dv,$$

where $F(v) = \mathrm{e}^{-v^2/2}/\sqrt{\pi}$, and the circumvention of the poles depends on the sign of z [12]. The integrals J_2 and J_3 are reduced to J_1 by means of the formulas

$$J_2(z_1,z_2) = \partial_{z_1}\frac{J_1(z_1)-J_1(z_2)}{z_2-z_1},$$

$$J_3(z_1,z_2) = -\partial_{z_1}\left(\frac{1}{3}\partial_{z_1}+\frac{1}{2}\partial_{z_2}\right)J_2(z_1,z_2).$$

The integral J_1, with due account for a pole circumvention rule corresponding to the Landau rule [12], is expressed in terms of the error integral [13]:

$$J_1(z) = \begin{cases} 1+iz\sqrt{\dfrac{\pi}{2}}\,\mathrm{e}^{-z^2/2}\operatorname{erfc}(-iz/\sqrt{2}), & z>0, \\ 1-iz\sqrt{\dfrac{\pi}{2}}\,\mathrm{e}^{-z^2/2}\operatorname{erfc}(iz/\sqrt{2}), & z<0. \end{cases}$$

The dispersion relation, in the zero approximation, has the form

$$\epsilon(\omega^{(0)},\mathbf{k}) = 1+\frac{1}{k^2}J_1\left(\frac{\omega^{(0)}}{k}\right) = 0.$$

All subsequent calculations were carried out for a one-dimensional crystal with a sinusoidal coordinate dependence of the background potential ϕ_0. In this case, the summation over the inverse lattice is reduced to a sum of two terms with $n = \{0,0,\sigma\}$, $\sigma = \pm 1$. The second-order frequency correction is

$$\omega^{(2)} = \xi^2 \sum_{\sigma=\pm 1}\left[\frac{M_{\sigma 0}^2}{k_\sigma^2\varepsilon_\sigma}+\frac{b\sigma}{k_\sigma}J_2\left(\frac{\omega^{(0)}}{k},\frac{\omega^{(0)}}{k_\sigma}\right)+\frac{b^2\sigma^2}{kk_\sigma}J_3\left(\frac{\omega^{(0)}}{k},\frac{\omega^{(0)}}{k_\sigma}\right)\right].$$

The auxiliary variable $M_{\sigma 0}$ is

$$M_{\sigma 0} = \frac{k}{k_\sigma}J_1\left(\frac{\omega^{(0)}}{k_\sigma}\right)+\frac{\sigma b}{k_\sigma}J_2\left(\frac{\omega^{(0)}}{k_\sigma},\frac{\omega^{(0)}}{k}\right) = M_{0\sigma},$$

$$\varepsilon_\sigma \equiv \varepsilon\left(\omega^{(0)},k_\sigma\right).$$

In the long-wave region of the spectrum, simpler and more explicit formulas can be obtained by expanding the expression for $\omega^{(2)}$ into a series with respect to k,

$$\omega^{(2)} = \xi^2 R(b)+\frac{3}{2}k^2(1+\xi^2 S(b))+\ldots, \quad R(b) = \frac{(b^2+b^4)(1-J_1(1/b))}{b^2+J_1(1/b)}.$$

The cumbersome expression for $S(b)$ in terms of the reference integral J_1 is omitted here.

The frequency real and imaginary parts are shown in Fig. 1 as functions of quasimomentum. In metals, the value of b is usually on the order of the Debye radius [2] (in our dimensionless variables, on the order of unity). It turns out that for such values of b there hold the inequalities $\mathrm{Re}\, R > 0$, $\mathrm{Im}\, R < 0$, $\mathrm{Re}\, S < 0$, $\mathrm{Im}\, S > 0$. The inequalities for R mean that in the nonuniform background model the plasmon spectrum width is finite even at zero quasimomentum which is observed in the experiments [1]. The inequalities for S mean that, at sufficiently large ξ in the region of low wave vector values, inverse dispersion will be observed which is also confirmed experimentally.

The behavior of the real and imaginary frequency parts at large k can be investigated numerically. The calculations show that the above formulas for the long-wave approximation are valid for $k < 0.2$. For greater k, the subsequent terms of the expansion are important, which qualitatively change the curve shapes, bringing back "normal" dispersion and attenuation.

This approach provides for a simple and graphical interpretation of the long-wave plasmon attenuation. If the measuring device "resolution" is much higher than the size of small-scale nonuniformities, then the rapidly oscillating space-time components of the measured quantities are "averaged off" during experimental observation. The presence of a periodic potential leads to the energy overflow from the "slow" field component to the "fast" one. Therefore the finite width of the plasmon spectrum (in coordinate representation this corresponds to plasma wave attenuation) is explained by the growth of the small-scale part of the perturbation.

REFERENCES

1. A. vom Felde, J. Sprosser-Prou, and J. Fink, *Phys. Rev.*, vol. B40, p. 10181, 1989.
2. D. Pines, *Elementary Excitations in Solids*, New York, 1963.
3. K.J. Krane, *J. Phys. F.*, vol. 8, p. 2133, 1978.
4. K. Sturm, *Z. f. Phys. B.*, vol. 25, p. 247, 1976; K. Sturm, Solid State Commun., vol. 82, p. 295, 1992.
5. A. Fleszar, R. Stumpf, and A. G. Eguiluz, *Phys. Rev.*, vol. B55, p. 2068, 1997.
6. A.A. Vlasov, *Zh. Eksp. Teor. Fiz.*, vol. 8, p. 291, 1938.
7. P.S. Zyryanov, *Zh. Eksp. Teor. Fiz.*, vol. 25, p. 441, 1953.
8. Yu.L. Klimontovich and V.P. Silin, *Zh. Eksp. Teor. Fiz.*, vol. 23, p. 151, 1952.
9. W. Harrison, *Solid State Theory*, McGraw-Hill, New York, 1970.
10. P.L. Kapitsa, *Zh. Eksp. Teor. Fiz.*, vol. 21, p. 588, 1951.
11. I.M. Aleshin, *Vestn. Mosk. Univ. Fiz. Astron.*, no. 4, p. 11, 1997.
12. L.D. Landau, *Zh. Eksp. Teor. Fiz.*, vol. 16, p. 574, 1946.
13. M. Abramowitz and I. Stegun, eds., *Handbook of Special Functions* (in Russian), p. 119, Nauka, Moscow, 1979.

9 April 1999

Department of Physics of the Earth