OPTICAL PROPERTIES OF METALS IN THE INFRARED REGION

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Landau's theory of a Fermi liquid is employed to describe the optical properties of metals in the infrared region. It is shown that the results obtained differ substantially from the corresponding results of the ordinary electron theory of metals.

1. In the region of infrared radiation ($\omega/2\pi = 10^{12}$ to 3×10^{14} sec^{-1}), great interest (from the viewpoint of obtaining information on the properties of the conduction electrons of a metal[1]) attaches to the range of frequencies much less than the frequencies of quantum absorption, and at the same time frequencies which are large in comparison with the collision frequency $1/\tau$ (τ is the characteristic time of free flight of the electrons*). For many metals, the frequency of quantum absorption is of the same order of magnitude as the frequency† $\omega_0 = \sqrt{4\pi e^2 N/m} \approx \sqrt{3 \times 10^9 N}$ of plasma oscillations of the conduction electrons, which value appreciably exceeds the upper limit of the infrared region. Therefore we shall consider frequencies which satisfy the inequality

$$\omega_0 \gg \omega \gg 1/\tau. \tag{1}$$

The optics of metals in such a region were studied by a number of authors.[1,3,4] In these researches, as is usually done in conduction theory, the representation of the conduction electrons as a gas of noninteracting particles is employed as one of the principal assumptions. In reality, the interaction between the electrons is by no means small, and they ought to be considered as a degenerate electron liquid. The latter is now possible with the use of the theory of a Fermi liquid formulated by Landau[5] and extended in Ref. 6 to an electron liquid.

An essential difference was discovered in Ref. 2 of the complex dielectric constant of a metal in the infrared region and the corresponding expression for ordinary theory.[1,3] However, only the real part of the complex dielectric constant was considered in that work. Below, we have obtained an expression for the imaginary part of the constant in the case in which such exists (see Ref. 1). In the case of an anomalous skin effect in the infrared region, an expression is obtained below for the surface impedance of the metal. In all these cases, the theory of the Fermi liquid modifies considerably the description of the properties of the metals.

We note that in the region of small frequencies, in which we can make use of the statistical characteristics of the metal, the theory of conductivity, which starts out from assumptions on the electron liquid, does not differ from the usual theory. The same also applies to the prominent anomalous skin effect.[2]

2. The electrons — quasi-particles produced by the electrons of the liquid — are described by the equation

$$\frac{\partial n}{\partial t} + \frac{\partial n}{\partial \mathbf{r}}\frac{\partial \varepsilon}{\partial \mathbf{p}} - \frac{\partial n}{\partial \mathbf{p}}\frac{\partial \varepsilon}{\partial \mathbf{r}} + e\left(\mathbf{E} + \frac{1}{c}\left[\frac{\partial \varepsilon}{\partial \mathbf{p}}\mathbf{H}\right]\right)\frac{\partial n}{\partial \mathbf{p}} = I(n) \tag{2}$$

in the theory of a Fermi liquid.[5,6] Here n is the distribution function, $I(n)$ is the collision integral, and ϵ is the energy of the quasi-particle. In this case,

$$\delta\varepsilon = \int d\mathbf{p}'\Phi(\mathbf{p}, \mathbf{p}')\,\delta n'. \tag{3}$$

The latter relation is useful because we are almost always interested in states which differ slightly from equilibrium, in which

$$n = n_0 + \delta n, \quad |\delta n| \ll n_0 = \frac{2}{(2\pi\hbar)^3}\left[\exp\left\{\frac{\varepsilon_0 - \mu}{kT}\right\} + 1\right]^{-1}; \tag{4}$$

$$\varepsilon = \varepsilon_0 + \delta\varepsilon.$$

In this case the theory of the Fermi liquid gives the following expression for the density of the electric current:

$$\mathbf{j} = e\int d\mathbf{p}\, n\,\frac{\partial \varepsilon}{\partial p} = e\int d\mathbf{p}\left\{\delta n - \delta\varepsilon\,\frac{\partial n_0}{\partial \varepsilon_0}\right\}\frac{\partial \varepsilon_0}{\partial \mathbf{p}}. \tag{5}$$

*At room temperature, τ 0.3 × 10^{-13} sec.

†For determination of N, see Refs. 1, 2. For many metals, N is of the order of 0.5 × 10^{-23} cm^2 and, consequently, $\omega_0 \sim 10^{16}$ sec^{-1}

As is shown in the Appendix, the collision integral in the theory of the Fermi liquid is identical with the usual collision integral, but is only written down for the combination $\delta n - \delta\epsilon\, \partial n/\partial\epsilon_0$.* Thus, in Eq. (2), all the quantities have a concrete meaning. Considering the variable field to be a periodic function of time with frequency ω, and taking Eqs. (3) and (4) into account, we can represent Eq. (2) in the following form

$$i\omega\delta n + \left(v_z \frac{\partial}{\partial z} + \frac{e}{c}\,[\mathbf{v}\times\mathbf{H}]\,\frac{\partial}{\partial \mathbf{p}}\right)\left\{\delta n - \delta\varepsilon\,\frac{\partial n_0}{\partial\varepsilon_0}\right\} - I\left(\delta n - \delta\varepsilon\,\frac{\partial n_0}{\partial\varepsilon_0}\right) = -e\,\mathbf{E}\mathbf{v}\,\frac{\partial n_0}{\partial\varepsilon_0}, \tag{6}$$

where $\mathbf{v} = \partial\epsilon/\partial\mathbf{p}$ is the velocity of the electrons in the equilibrium state. Here it is assumed that the metal is bounded by a plane surface, perpendicular to the z axis. Moreover, it is assumed that there is also a constant field $\mathbf{H}_0$.

3. In the infrared region, the skin depth is approximately c/ω_0. This leads to the circumstance that the term containing the coordinate derivative in Eq. (6) is of the order of $(v/c)\omega_0\delta n$. In metals, the velocity of the electrons is about 10^8 cm/sec. For this reason, Eq. (6) can be written for zeroth approximation in the form

$$i\omega\delta n^{(0)} = -e\,\mathbf{E}\mathbf{v}\,\partial n_0/\partial\varepsilon_0. \tag{7}$$

From this we can easily determine the zeroth approximation of the complex conductivity tensor $\sigma_{\alpha\beta}$ ($j_\alpha = \sigma_{\alpha\beta}E_\beta$) and correspondingly the complex dielectric constant tensor $\epsilon'_{\alpha\beta} = \tilde{\epsilon}_{\alpha\beta} - i4\pi \times \sigma_{\alpha\beta}/\omega$, where $\tilde{\epsilon}_{\alpha\beta}$ is that part of the dielectric constant which is not occasioned by the conduction electrons (in the following equations, this quantity is omitted, since its relative contribution is small for $\omega \ll \omega_0$).

It is easy to see that in the approximation employed, $\epsilon'_{\alpha\beta}$ is real and is equal to†

$$\mathrm{Re}\,\varepsilon'_{\alpha\beta} = -\frac{8\pi e^2}{\omega^2(2\pi\hbar)^3}\int\frac{dS}{v}\,v_\alpha V_\beta, \tag{8}$$

where the integration is carried out over the Fermi surface, dS is an element of that surface and

$$V_\alpha = v_\alpha - \int dp'\,\Phi(\mathbf{p},\mathbf{p}')\,\frac{\partial n_0}{\partial\varepsilon'_0}\,v'_\alpha. \tag{9}$$

*We note that for static problems of conduction theory, this reduces to a coinciding of the results of ordinary electron theory of metals with that based on the application of the theory of a Fermi liquid (for further details, see Ref. 7).

†Correspondingly,

$$\sigma^{(0)}_{\alpha\beta} = -\frac{ie^2}{\omega}\,\frac{2}{(2\pi\hbar)^3}\int\frac{dS}{v}\,v_\alpha V_\beta. \tag{8'}$$

It is also convenient to determine the value of the surface impedance of the metal:

$$Z_x = 4\pi E_x(0)/cH_y(0) = -(4\pi i\omega/c^2)\,E_x(0)/E'_x(0) = E_x(0)/J_x, \tag{10}$$

where $E_i(0)$ and $H_i(0)$ are the values of the field at the surface of the metal and $J_x = \int_0^\infty dz\, j_x(z)$.

In the zeroth approximation, it follows from the condition (1) that the electrical charge density must be zero. In this connection, the continuity equation has the form $\operatorname{div}\mathbf{j} = 0$. The latter leads to the fact that $j_z = 0$ in this approximation. From this condition we get [by making use of the expression for the conductivity tensor (8′)]

$$E^{(0)}_z = -\left(\sigma^{(0)}_{zx}E^{(0)}_x + \sigma_{zy}E^{(0)}_y\right)/\sigma^{(0)}_{zz}. \tag{11}$$

Finally, eliminating the magnetic field and the z component of the electric field from Maxwell's equations, we get, in zeroth approximation,

$$E^{(0)''}_\alpha - \frac{4\pi e^2}{c^2}\left(\frac{N}{m}\right)_{\alpha\beta}E^{(0)}_\beta = 0, \quad (\alpha,\beta = x, y), \tag{12}$$

where

$$\left(\frac{N}{m}\right)_{\alpha\beta} = \frac{2}{(2\pi\hbar)^3}\left\{\int\frac{dS}{v}\,v_\alpha V_\beta - \frac{\left(\int v_\alpha V_z dS/v\right)\left(\int v_z V_\beta dS/v\right)}{\int v_z V_z dS/v}\right\}. \tag{13}$$

Directing the x and y axes parallel to the principal axis of the tensor N/m, we get (see Ref. 3)

$$E^{(0)}_x(z) = E_x(0)\,e^{-z/\delta_x}, \quad E^{(0)}_y(z) = E_y(0)\,e^{-z/\delta_y}, \tag{14}$$

where

$$\delta_x = c/\Omega_x,\ \delta_y = c/\Omega_y,\ \Omega_x^2 = 4\pi e^2(N/m)_x,\quad \Omega_y^2 = 4\pi e^2(N/m)_y, \tag{15}$$

and $(N/m)_i$ is the principal value of the tensor (N/m). The solution (14) reduces to the following formulas for the zeroth approximation of the surface impedance

$$Z^{(0)}_x = 4\pi i\omega\delta_x/c^2 = 4\pi i\omega/c\Omega_x;\quad Z^{(0)}_y = 4\pi i\omega\delta_y/c^2 = 4\pi i\omega/c\Omega_y. \tag{16}$$

Corresponding to the fact that in this approximation, ϵ' is real, the expression obtained for the surface impedance is a pure imaginary.

4. The emergence of an imaginary part of the complex dielectric constant, or of a real part (R) of the surface impedance, is brought about by the small terms of Eq. (6), which were neglected in the zeroth approximation of Eq. (7). Let us first consider the case in which the collision integral and the term containing the constant magnetic field dominate the term of Eq. (6) containing the coordinate derivative. Obviously, this takes place under the condition that[7]

$$(v/c)\,\omega_0 \ll 1/\tau \quad \text{or} \quad \omega_H \sim eH_0/mc. \tag{17}$$

Solving Eq. (6) in first approximation (7), we easily find that

$$\sigma^{(1)}_{\alpha\beta} = -\frac{e^2}{\omega^2}\frac{2}{(2\pi\hbar)^3}\int\frac{dS}{v}V_\alpha\left\{J(V_\beta) - \frac{e}{c}\left([\mathbf{v}\times\mathbf{H}_0]\frac{\partial}{\partial\mathbf{p}}\right)V_\beta\right\}. \tag{18}$$

Here it is taken into account that

$$I(V_\beta\partial n_0/\partial\varepsilon_0) = (\partial n_0/\partial\varepsilon_0)\,J(V_\beta).$$

Correspondingly, the imaginary part of ϵ' takes the form

$$\operatorname{Im}\varepsilon'_{\alpha\beta} = -\frac{4\pi}{\omega}\sigma^{(1)}_{\alpha\beta}$$

$$= \frac{8\pi e^2}{\omega^3(2\pi\hbar)^3}\int\frac{dS}{v}V_\alpha\left\{J(V_\beta) - \frac{e}{c}\left([\mathbf{v}\times\mathbf{H}_0]\frac{\partial}{\partial\mathbf{p}}\right)V_\beta\right\}. \tag{19}$$

In the case in which there is a mean time of flight τ, we must consider $J = -1/\tau(\mathbf{p})$ in Eq. (19).

The situation is somewhat more complicated in the case in which

$$\omega_0(v/c) \gg 1/\tau, \quad \omega_H. \tag{20}$$

In such a case we say that an anomalous skin effect occurs. By virtue of the fact that the correction term of Eq. (6) now contains a space derivative, it is necessary, for solution of the kinetic equation, to make use of the boundary conditions of the function δn. As such a condition, we assume that as $z \to \infty$, δn goes to zero, while at the boundary of the metal, we assume that the so-called diffuse reflection exists, and $\delta n - \delta\epsilon \times \partial n_0/\partial\epsilon = 0$ for $v_z \geq 0$. Such a condition corresponds to the usual[8] in the sense that it is written for a quantity which determines the current density, just as is done in the ordinary theory of δn. Furthermore, in the computation of the correction of the first approximation to the current density, $\delta j_\alpha = j^{(1)}_\alpha - j^{(0)}_\alpha$, we assume that we can use the zeroth approximation for the electric field entering into δj_α.

$$E^{(0)}_\alpha(z) = E^{(0)}_\alpha \exp(-z/\delta_\alpha).$$

As a result, we get

$$\delta J_\alpha = \int_0^\infty \delta j_\alpha dz = \sum_\beta E_\beta(0)\frac{e^2}{\omega^2}\frac{2}{(2\pi\hbar)^3}\int_{v_z\geq 0}\frac{dS}{v}V_\alpha v_z V_\beta. \tag{21}$$

Here it is taken into consideration that the Fermi surface possesses a center of symmetry and, in particular, $\Phi(-\mathbf{p}, -\mathbf{p}') = \Phi(\mathbf{p}, \mathbf{p}')$.

Assuming that the radiation incident on the metal is polarized along the x axis, and that in this case, by Eqs. (14) and (11),

$$E^{(0)}_x(z) = E_x(0)\,e^{-z/\delta_x}, \quad E^{(0)}_y = 0,$$
$$E^{(0)}_z(z) = -E_x(0)\,(\sigma^{(0)}_{zx}/\sigma^{(0)}_{zz})\,e^{-z/\delta_x}, \tag{22}$$

we find the following expression for the real part of the impedance:

$$R_x = \frac{4\pi}{c^2}\left\{\int_{v_z\geq 0}\frac{dS}{v}v_z V_x\left(V_x - V_z\frac{\int dS'' v''_z V''_x/v''}{\int dS'' v''_z V''_z/v''}\right)\right\}$$
$$\times\left\{\int\frac{dS}{v}v_x\left(V_x - V_z\frac{\int dS' v'_z V'_x/v'}{\int dS' v'_z V'_z/v'}\right)\right\}^{-1}. \tag{23}$$

We now determine the "oblique" terms of the surface impedance:[3]

$$Z_{xy} = E_x(0)/J_{xy} \approx R_{xy}; \qquad E_y(0)\,J_{yx} = Z_{yx} \approx R_{yx}. \tag{24}$$

Here $J_{\alpha\beta}$ is the α-component of the current which arises in the metal for incidence of radiation polarized along the β-axis. Employing (21) under the condition (22), we get

$$R_{xy} = \frac{\omega^2}{e^2}\frac{(2\pi\hbar)^3}{2}\left\{\int_{v_z\geq 0}\frac{dS}{v}v_z V_y\left(V_x - V_z\frac{\int v''_z V''_x dS''/v''}{\int v''_z V''_z dS''/v''}\right)\right\}^{-1}. \tag{25}$$

The inequalities (17) and (20) do not always hold. Therefore the case is of interest in which the skin effect cannot be considered normal, and in which at the same time one cannot consider it anomalous. The solution of such a problem is actually contained in the results worked out above.

In fact, as follows from Eq. (6), the excitations which lead to the correction of the first approximation are additive. In this regard, the correction of first approximation for the current density and surface impedance will arise additively from the term of Eq. (6) which contains the spatial derivatives (and which is most significant in the anomalous skin effect), from the collision integral and from the term containing the constant magnetic field (which is most important in the region of the normal skin effect). In the general case, therefore, the usual impedance of the metal can be represented in the form

$$Z_x \approx Z^{(0)}_x + R^{(a)}_x + R^{(H)}_x; \quad Z_{xy} \approx R^{(a)}_{xy} + R^{(H)}_{xy}, \tag{26}$$

with the accuracy considered by us, where $Z_x^{(0)}$, $R_x^{(a)}$, $R_{xy}^{(a)}$ are defined respectively by Eqs. (16), (23), and (25), while $R_x^{(H)}$ and $R_{xy}^{(H)}$ can easily be obtained from Eqs. (8), (11), and (19).

5. The difference of Eqs. (8), (13), (23), and (25) from the corresponding relations of the research of Kaganov and Slezov[9] is due to the function Φ. It is natural to attempt to make clear what evidence can be obtained on this function from experiments in the infrared region. However, we can point out at once that the results which we have obtained above are always complicated by the anisotropies of the metal. Therefore, without a knowledge of the Fermi surface, it is difficult to obtain information on the function Φ from experiments in the infrared region. Evidently, for the determination of effects which distinguish the conduction electrons from the Fermi gas, we must first determine the form of the Fermi surface with the aid of experiments in the radio-frequency range for the case of a sharply defined anomalous skin effect.[9]

The results obtained above are greatly simplified when the metal can be regarded as isotropic.[1] The latter is quite natural for polycrystalline samples. We shall consider this case in more detail below. Under such an assumption, we have, in place of Eq. (8),

$$\operatorname{Re}\varepsilon' = -\frac{4\pi e^2 N}{m\omega^2}, \quad N = \frac{8\pi p_0^2 v_0 m}{3(2\pi\hbar)^3}\{1 + p_0^2 \textstyle\int d\Omega \cos\chi F(\cos\chi)\};$$
$$F = [2/(2\pi\hbar)^3]\,\Phi(\mathbf{p}, \mathbf{p}')/v_0, \tag{8''}$$

where p_0 and v_0 are the momentum and velocity of the electron on the Fermi surface, m = mass of the free electron. Such an expression for the real part of the complex dielectric constant was obtained in Ref. 2. The non-diagonal elements of the tensor of the real part of the complex dielectric constant tensor in the approximation under consideration are equal to zero. In the absence of a magnetic field, the imaginary part ϵ', which is defined by Eq. (19), is also diagonal and is equal to

$$\operatorname{Im}\varepsilon' = \frac{8\pi e^2 p_0^2}{3\omega^3 (2\pi\hbar)^3}\int \frac{d\Omega}{v}\,\mathbf{V} J(\mathbf{V}). \tag{19'}$$

We note that formulas which contain the collision integral are valid even in the quantum case,[10] in which the frequency of the light is comparable to, or becomes larger than $kT/\hbar$. In this case, only the value of the collision integral changes.

In the isotropic case, only the nondiagonal terms arise, as a result of the constant magnetic field. Directing H_0 along the z axis, we have

$$\operatorname{Im}\varepsilon'_{xy} = -\operatorname{Im}\varepsilon'_{yx}$$
$$= -\frac{4\pi}{\omega}\,\frac{8\pi e^2 p_0^2 v_0}{3(2\pi\hbar)^3\omega^2}\,\frac{e v_0 H_0}{p_0}\left\{1 + p_0^2\int d\Omega \cos\chi F\right\}^2. \tag{19''}$$

The presence of a magnetic field has no effect on the other components.

Finally, for an isotropic metal, Eq. (25) vanishes, while (23) takes on the form

$$R_x = \frac{3\pi}{4}\frac{v_0}{c^2}\left\{1 + p_0^2\int d\Omega \cos\chi F(\cos\chi)\right\}. \tag{23'}$$

As was shown in Ref. 2, the relation (8), along with data on the electronic heat capacity of the metal and the results of experiments in the radio-frequency region, in the case of a sharply defined anomalous skin effect, permits us to establish the value of $p_0^2 \int d\Omega \cos\chi\, F(\cos\chi)$. In other words, the possibility is shown of determining the first coefficient in the expansion of $F(\cos\chi)$ in Legendre polynomials. Equation (23) points up the possibility of a similar estimate of this coefficient. Of course, it should be observed that all such estimates are essentially connected with the possibility of using the isotropic model of the metal, and this in each case must be done with caution.[2] We note that in the collision with the surface, the part (2) of the electrons can be scattered specularly. In this case, the expression obtained above for $R^{(a)}$ must be changed by a factor of $(1 - q)$.

In conclusion, I am pleased to express my thanks to L. D. Landau for useful discussions of the problem, explained in the Appendix.

APPENDIX

In order to clarify the form of the collision integral in the theory of a Fermi liquid for the non-equilibrium state, we consider as an example collisions with impurities. In this case, the collision integral has the form

$$I(n) = \int W(n; \mathbf{p}, \mathbf{p}')\{n(\mathbf{p}')[1 - n(\mathbf{p})]$$
$$- n(\mathbf{p})[1 - n(\mathbf{p}')]\}\,\delta\{\varepsilon(\mathbf{p}) - \varepsilon(\mathbf{p}')\}\,d\mathbf{p}'.$$

Taking (4) into account, we can represent $I(n)$ in the form of a sum $I_1 + I_2$, where

$$I_1 = \int W(n_0; \mathbf{p}, \mathbf{p}')\{\delta n(\mathbf{p}) - \delta n(\mathbf{p}')\}\,\delta\{\varepsilon_0(\mathbf{p}) - \varepsilon_0(\mathbf{p}')\}\,d\mathbf{p}',$$

$$I_2 = \int W(n_0; \mathbf{p}, \mathbf{p}')\{n_0(\mathbf{p}') - n_0(\mathbf{p})\}\,\delta\{\varepsilon_0(\mathbf{p})$$
$$+ \delta\varepsilon(\mathbf{p}) - \varepsilon_0(\mathbf{p}') - \delta\varepsilon(\mathbf{p}')\}\,d\mathbf{p}'.$$

Here, I_1 is a quantity which coincides with the collision integral of ordinary theory.

Further, it is convenient to transform I_2 somewhat. In this case, we take it into consideration that

$$n_0(\varepsilon_0) \approx n_0(\varepsilon_0 + \delta\varepsilon) - \delta\varepsilon \partial n_0 / \partial\varepsilon_0.$$

Thanks to the fact that in the law of conservation of energy, $\epsilon = \epsilon_0 + \delta\epsilon$, terms containing only $n_0(\epsilon)$ fall out, and there are left terms proportional to $\delta\epsilon$. For the remaining small terms, the difference between ϵ and ϵ_0 in the conservation law is no longer significant. Therefore,

$$I(n_0 + \delta n) = \int W(n_0; \mathbf{p}, \mathbf{p}') \left\{ \left(\delta n' - \delta\varepsilon' \frac{\partial n_0}{\partial \varepsilon_0'} \right) - \left(\delta n - \delta\varepsilon \frac{\partial n_0}{\partial \varepsilon_0} \right) \right\} \delta(\varepsilon_0 - \varepsilon_0') d\mathbf{p}'.$$

Thus it can be shown that the collision integral for states which differ only slightly from the equilibrium can be obtained by the substitution in the conservation law of the equilibrium value of the energy, and the replacement of n by $n_0 + \delta n - \delta\epsilon\, \partial n_0/\partial\epsilon_0$.

From these considerations, it is obvious that this result does not depend on the particular form of the collision integral selected by us, and also holds for collisions of electrons with phonons, electrons with electrons, etc. In all cases, the difference from the ordinary theory of a Fermi gas reduces to the fact that we have $\delta n - \delta\epsilon \times \partial n_0/\partial\epsilon_0$ in place of δn in the collision integral.

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