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MUTUAL ELECTRON CORRELATIONS IN METAL OPTICS

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The absorption of infrared electromagnetic waves in metals is considered on the basis of Landau's Fermi-liquid theory,[1] with the quantum properties of the electromagnetic field taken into account.

THE present paper is devoted to an investigation of the absorption in metals of electromagnetic waves in the infrared region of the spectrum from the point of view of the Fermi-liquid theory developed by Landau.[1] This problem has already been considered by Silin.[2*] However, in his paper the classical transport equation was used to describe the Fermi liquid, whereas in the infrared region of the spectrum the quantum properties of the electromagnetic field (see references 3 to 5) are essential for an evaluation of the volume absorption (due to the collisions of the electrons with one another and with the lattice phonons).

If the system is in a state close to equilibrium, the energy of the quasi-particles is of the form

$$\varepsilon(\mathbf{p}, \mathbf{r}, t) = \varepsilon^{(0)}(\mathbf{p}) + \delta\varepsilon(\mathbf{p}, \mathbf{r}, t) \tag{1}$$

where $\epsilon^{(0)}(\mathbf{p})$ is the dispersion law of the quasi-particles when there is no external field, and

$$\delta\varepsilon(\mathbf{p}, \mathbf{r}, t) = \int d\mathbf{p}' \cdot \Phi(\mathbf{p}, \mathbf{p}')\, \delta f(\mathbf{p}', \mathbf{r}, t).$$

In the last formula $\delta f(\mathbf{p}, \mathbf{r}, t)$ is the non-equilibrium correction to the distribution function,

$$f(\mathbf{p}, \mathbf{r}, t) = f_0(\mathbf{p}) + \delta f(\mathbf{p}, \mathbf{r}, t),$$

$$f_0(\mathbf{p}) = \left[1 + \exp\left(\frac{\varepsilon^{(0)}(\mathbf{p}) - \varepsilon_0}{kT}\right)\right]^{-1};$$

the function $\Phi(\mathbf{p}, \mathbf{p}')$ characterizes the interelectron correlation.

In correspondence with (1) it is natural to write the "electron" Hamiltonian in an external electromagnetic field in the form

$$\hat{H}(1) = \hat{H}^{(0)}(1) + \delta\hat{H}(1),$$

where

$$\hat{H}^{(0)} = \varepsilon^{(0)}\left(-i\hbar\nabla + \frac{e}{c}\mathbf{A}(\mathbf{r}, t)\right),$$

$$\mathbf{A}(\mathbf{r}, t) = \mathbf{A}(\mathbf{r})\, e^{i\omega t} + \mathbf{A}^*(\mathbf{r})\, e^{-i\omega t}.$$

Using the calculations performed in references 3 and 5 we easily obtain a quantum transport equation for the system of such electron quasi-particles interacting with the lattice vibrations and colliding one with another. In the linear approximation this equation is of the form

$$\begin{aligned} i\omega\delta f + \mathbf{v}\frac{\partial}{\partial \mathbf{r}}\left(\delta f - \delta\varepsilon\frac{\partial f_0}{\partial\varepsilon^{(0)}}\right) - e\mathbf{E}\cdot\mathbf{v}\frac{\partial f_0}{\partial\varepsilon^{(0)}} &= (\hat{J}_{ep} + \hat{J}_{ee})\left(\delta f - \frac{e}{i\omega}\mathbf{E}\cdot\mathbf{v}\frac{\partial f_0}{\partial\varepsilon^{(0)}}\right) \\ &\quad - (\hat{K}_{ep} + \hat{K}_{ee})\left(\frac{e}{i\omega}\mathbf{E}\cdot\mathbf{v} - \delta\varepsilon\right). \end{aligned} \tag{2}$$

Here the indices ep and ee denote terms arising respectively due to electron-phonon and electron-electron collisions; $\mathbf{E}(\mathbf{r}) = -(i\omega/c)\mathbf{A}(\mathbf{r})$ is the electrical field strength. $\hat{J}_{ep}$ and $\hat{J}_{ee}$ can be obtained from the corresponding classical collision operators by substituting for $\delta(a)$ (the δ-function that describes the conservation of energy during a collision) the average[3,5] $\frac{1}{2}[\delta(a + \hbar\omega) + \delta(a - \hbar\omega)]$ (we have denoted by a the difference between the energies of the colliding particles before and after the collision). Furthermore,

$$\begin{aligned} \hat{K}_{ep}\delta\varepsilon(\mathbf{p}) = &-\frac{\Delta}{(2\pi\hbar)^3}\frac{\pi}{Mu\hbar}C^2\int\limits_{|\mathbf{q}| \leqslant q_0} d\mathbf{q}\cdot q\,(\delta\varepsilon(\mathbf{p}+\mathbf{q}) - \delta\varepsilon(\mathbf{p}))\frac{1}{2\hbar\omega} \\ &\times\{[\delta(a + \hbar\omega) - \delta(a - \hbar\omega)] \\ &\times[-(\bar{N}_q + 1) f_0(\mathbf{p})(1 - f_0(\mathbf{p}+\mathbf{q})) + \bar{N}_q f_0(\mathbf{p}+\mathbf{q}) \\ &\times(1 - f_0(\mathbf{p}))] + [\delta(b + \hbar\omega) - \delta(b - \hbar\omega)] \\ &\times[-\bar{N}_q f_0(\mathbf{p})(1 - f_0(\mathbf{p}+\mathbf{q})) \\ &+ (\bar{N}_q + 1) f_0(\mathbf{p}+\mathbf{q})(1 - f_0(\mathbf{p}))]\}; \end{aligned}$$

$$a = \varepsilon(\mathbf{p}) - \varepsilon(\mathbf{p}+\mathbf{q}) - h\nu_q, \quad b = \varepsilon(\mathbf{p}) - \varepsilon(\mathbf{p}+\mathbf{q}) + h\nu_q,$$

$$\bar{N}_q = \left[\exp\left(\frac{h\nu_q}{kT}\right) - 1\right]^{-1}$$

For the sake of simplicity we have here assumed that the phonons possess a very simple dispersion law: $h\nu_{\mathbf{q}} = u|\mathbf{q}|$, where $\mathbf{q}$ is the phonon momentum, u the sound velocity in the metal, $q_0 = k\Theta/u$

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[*] The author is deeply grateful to V. P. Silin for making his paper available in advance of publication.

the limiting phonon momentum, Θ the Debye temperature, M the mass of an atom in the lattice, Δ the volume of an elementary cell in the crystal, and C a constant of the same order of magnitude as the average energy of an electron in the metal.

$$\hat{K}_{ee}\,\delta\varepsilon(\mathbf{p}) = -\frac{\pi e^4}{\hbar(2\pi\hbar)^6}\sum_{g}\iiint d\mathbf{p}'\,d\mathbf{p}_1\,d\mathbf{p}_1'$$

$$\times[\nu(|\mathbf{p}-\mathbf{p}_1|)-\nu(|\mathbf{p}-\mathbf{p}_1'|)]^2\,\delta(\mathbf{p}+\mathbf{p}'-\mathbf{p}_1-\mathbf{p}_1'+2\pi\hbar\,\mathbf{g})$$

$$\times(\delta\varepsilon+\delta\varepsilon'-\delta\varepsilon_1-\delta\varepsilon_1')\,\frac{1}{2\hbar\omega}\,[\delta(\varepsilon+\varepsilon'-\varepsilon_1$$

$$-\varepsilon_1'+\hbar\omega)-\delta(\varepsilon+\varepsilon'-\varepsilon_1-\varepsilon_1'-\hbar\omega)]\,\{f_0(\mathbf{p})\,f_0(\mathbf{p}')\,[1-f_0(\mathbf{p}_1)]$$

$$\times[1-f_0(\mathbf{p}_1')]-f_0(\mathbf{p}_1)f_0(\mathbf{p}_1')\,[1-f_0(\mathbf{p})]\,[1-f_0(\mathbf{p}')]\},$$

where the summation extends over all reciprocal lattice vectors **g**.

$$\nu(|\mathbf{p}|) = \int d\mathbf{r}\,e^{i\,\mathbf{p}\cdot\mathbf{r}/\hbar}\varphi(|\mathbf{r}|),$$

$\varphi(|\mathbf{r}|)$ is the interaction potential of two electrons.

One can easily verify that in the classical limiting case ($\hbar\omega/kT \to 0$)

$$\hat{K}_{\mathbf{ep}}\delta\varepsilon = -\hat{J}^{(\mathbf{cl})}_{\mathbf{ep}}\delta\varepsilon\,\partial f_0/\partial\varepsilon^{(0)},\quad \hat{K}_{ee}\,\delta\varepsilon = -\hat{J}^{(\mathbf{cl})}_{ee}\delta\varepsilon\,\partial f_0/\partial\varepsilon^{(0)}.$$

The dependence of the field and the electron distribution on the coordinates does not affect the volume absorption, i.e., (2) we can drop terms containing spatial derivatives in Eq. 2, which can then be easily solved by successive approximations (since the terms on the right hand side of the transport equation are small in the infrared region of the spectrum),

$$\delta f^{(I)}(\mathbf{p}) = \frac{e}{i\omega}\,(\mathbf{E}\cdot\mathbf{v})\,\frac{\partial f_0}{\partial\varepsilon^{(0)}},\quad \delta f^{(II)} = \frac{e}{\omega^2}(\hat{K}_{\mathbf{ep}}+\hat{K}_{ee})\,\mathbf{E}\cdot\mathbf{V}(\mathbf{p}),$$

$$\mathbf{V}(\mathbf{p}) = \mathbf{v}(\mathbf{p}) + \int\frac{dS_{\mathbf{p}'}}{v(\mathbf{p}')}\,\Phi(\mathbf{p},\mathbf{p}')\,\mathbf{v}(\mathbf{p}'), \tag{3}$$

where the integration extends over the surface $\epsilon(\mathbf{p}) = \epsilon_0$ in momentum space.

The electrical current density is of the form[2]

$$\mathbf{j} = -\frac{2e}{(2\pi\hbar)^3}\int d\mathbf{p}\cdot\mathbf{v}(\mathbf{p})\,f(\mathbf{p})$$

$$\cong -\frac{2e}{(2\pi\hbar)^3}\int d\mathbf{p}\cdot\mathbf{v}(\mathbf{p})\left[\delta f(\mathbf{p}) - \delta\varepsilon\,\frac{\partial f_0}{\partial\varepsilon^{(0)}}\right].$$

If we substitute the function of the first approximation into this equation we are led to the following dielectric-constant tensor,[2]

$$\varepsilon_{\alpha\beta} = -(4\pi e^2/\omega^2)\,(N/m)_{\alpha\beta},\quad(\alpha,\beta = x, y),$$

where

$$(N/m)_{\alpha\beta} = a_{\alpha\beta} + a_{\alpha z}\,a_{z\beta}/a_{zz},\quad a_{ik} = \frac{2}{(2\pi\hbar)^3}\int\frac{dS_{\mathbf{p}}}{v(\mathbf{p})}v_i(\mathbf{p})\,V_k(\mathbf{p}).$$

We have chosen here a Cartesian coordinate system with a z axis directed out of the metal perpendicularly to its boundary.

The function $\delta f^{(II)}$ gives the conduction current, which can in a natural way be written in the form

$$j_i = [\sigma^{(\mathbf{ep})}_{ik}(\omega) + \sigma^{(ee)}_{ik}(\omega)]\,E_k.$$

Let the incoming wave be polarized along one of the principal directions of the symmetrical tensor $(N/m)_{\alpha\beta}$. We shall take the x and y axes along the principal directions of this tensor. The corresponding volume absorption has then the form

$$A^{(\alpha)} = \frac{8\pi\omega^2}{c^3}\,\delta_\alpha^3\left[\sigma_{\alpha\alpha} - 2\,\frac{a_{\alpha z}}{a_{zz}}\,\sigma_{\alpha z} + \left(\frac{a_{\alpha z}}{a_{zz}}\right)^2\sigma_{zz}\right],$$

where

$$\delta_\alpha^{-2} = (4\pi e^2/c^2)\,(N/m)_{\alpha\alpha}.$$

If the metal surface is oriented in such a way that all three axes x, y, and z can be directed along principal axes of the a_{ik} tensor then

$$A^{(\alpha)} = (8\pi\omega^2/c^3)\,\delta_\alpha^3\sigma_{\alpha\alpha}.$$

The phonon-conductivity tensor $\sigma^{(\mathrm{ep})}_{ik}$ depends in general in rather a complicated fashion on T and ω. If $\hbar\omega \gg kT$, which is practically always satisfied in the near infrared region of the spectrum, we have*

$$\sigma^{(\mathbf{ep})}_{ik}(\omega) = \frac{e^2}{\omega^2}\int_0^{q_0} dq\,q^3\coth\left(\frac{h\nu_q}{2kT}\right)B_{ik}(q),$$

$$B_{ik}(q) = \frac{\Delta}{(2\pi\hbar)^6}\,\frac{2\pi}{Mu\hbar}\,C^2$$

$$\times\int d\Omega_{\mathbf{q}}\int\frac{dS_{\mathbf{p}}}{v(\mathbf{p})}\,V_i(\mathbf{p})\,[\mathbf{V}(\mathbf{p}+\mathbf{q})-\mathbf{V}(\mathbf{p})]_k\,\delta(\varepsilon(\mathbf{p}+\mathbf{q})-\varepsilon_0),$$

where $d\Omega_{\mathbf{q}}$ is an element of solid angle in momentum space. In the low temperature region ($T \ll \Theta$) it follows from this formula that

$$\sigma^{(\mathbf{ep})}_{ik}(\omega)\,\frac{\omega^2}{e^2} \approx \int_0^{q_0} dq\cdot q^3 B_{ik}(q) + \left(\frac{T}{\Theta}\right)^5 48q_0^5B'_{ik}(0) + \ldots.$$

At high temperatures ($T \gg \Theta$) we have

$$\sigma^{(\mathbf{ep})}_{ik}(\omega)\,\frac{\omega^2}{e^2} \approx \frac{T}{\Theta}\,2q_0\int_0^{q_0} dq\,q^2B_{ik}(q)$$

$$+\frac{\Theta}{T}\,\frac{1}{6q_0}\int_0^{q_0} dq\cdot q^4B_{ik}(q) + \ldots\,.$$

Taking the correlation between electrons into account does therefore in both limiting cases not lead to a qualitative change in the temperature dependence of the conductivity and only influences the numerical values of the coefficients of the corresponding powers of T/Θ.

A simple calculation shows (cf. reference 5) that

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*Here and henceforth we have assumed that the quantities $\hbar\omega$, kT, and kΘ are small compared to the Fermi energy ε_0.

$$\sigma_{ik}^{(ee)}(\omega) = (e/\omega)^2\,[(kT)^2 + (\hbar\omega/2\pi)^2]\,b_{ik}\,,$$

where b_{ik} are the constant coefficients

$$b_{ik} = \frac{4e^4\pi^3}{3\hbar\,(2\pi\hbar)^9} \sum_{\mathbf{g}} \iiiint \frac{dS_{\mathbf{p}}}{v(\mathbf{p})} \frac{dS_{\mathbf{p}'}}{v(\mathbf{p}')} \frac{dS_{\mathbf{p}_1}}{v(\mathbf{p}_1)} \frac{dS_{\mathbf{p}_1'}}{v(\mathbf{p}_1')}$$

$$\times\,[\nu(|\mathbf{p}-\mathbf{p}_1|) - \nu(|\mathbf{p}-\mathbf{p}'_1|)]^2$$

$$\times\,V_i(\mathbf{p})\,[\mathbf{V}(\mathbf{p}) + \mathbf{V}(\mathbf{p}') + \mathbf{V}(\mathbf{p}_1) - \mathbf{V}(\mathbf{p}_1')]_k$$

$$\times\,\delta(\mathbf{p} + \mathbf{p}' - \mathbf{p}_1 - \mathbf{p}_1' + 2\pi\hbar\mathbf{g}).$$

In this way, the absorption due to interelectronic collisions changes only in absolute magnitude. However, this fact can apparently be observed experimentally, if we compare the results obtained in the near infrared region of the spectrum with the results of measurements of the static electrical conductivity of the metal. In the latter case the measurements must be performed in the very low temperature range ($T \lesssim 10°K$) so that collisions with the phonons can be neglected. At such temperatures the resistivity of even very pure specimens is mainly caused by impurities. Using this fact, we can easily show that in our case the static electrical conductivity is

$$\sigma_{ik} \approx e^2 a_{ik}^{(0)}/\nu^{(en)} - e^2\,(kT)^2 b_{ik}^{(0)}\,/\,(\nu^{(en)})^2,$$

where the quantities $a_{ik}^{(0)}$ and $b_{ik}^{(0)}$ are the same respectively as a_{ik} and b_{ik} for $\mathbf{V}(\mathbf{p}) = \mathbf{v}(\mathbf{p})$; for the sake of simplicity we have assumed that frequency $\nu^{(en)}$ of collisions with the impurities is isotropic.

If the interelectronic correlation is essential, the quantities $a_{ik}^{(0)}$ and $b_{ik}^{(0)}$ are, generally speaking, different from a_{ik} and b_{ik}. The quantities a_{ik} and b_{ik} can be determined from measurements in the near-infrared region of the spectrum (1 to 10 μ): a_{ik} from the polarization of the wave, reflected from the metal surface, and b_{ik} from the frequency dependence of the absorption.

In the case of an isotropic Fermi surface, we can easily show that

$$\mathbf{V}(\mathbf{p}) = \mathbf{v}(\mathbf{p})\,(1 + \gamma(p)),\quad \gamma(p) = \int \frac{dS_{\mathbf{p}'}}{v(\mathbf{p}')}\,\Phi(\mathbf{p},\mathbf{p}')\cos\chi,$$

where χ is the angle between the vectors $\mathbf{p}$ and $\mathbf{p}'$. For an isotropic metal accounting for the mutual electron correlation thus reduces to multiplying the earlier expression for the volume absorption by a factor $[1 + \gamma(p_0)]^2$

$$A = A_0\,(1 + \gamma(p_0))^2,$$

where

$$A_0 = (2\delta/c)\,(\nu^{(ep)} + \nu^{(ee)}),\quad \delta = (4\pi Ne^2/mc^2)^{-1/2},$$

$\nu^{(ep)}$ and $\nu^{(ee)}$ are the respective effective collision frequencies. According to reference 4 we have in the near-infrared region of the spectrum

$$\nu^{(ep)}(T,\omega) = 2\nu_0^{(ep)}\,(\Theta)\left(\frac{T}{\Theta}\right)^5 \int_0^{\Theta/T} dx\cdot x^4 \coth\frac{x}{2}\,,$$

where $\nu_0^{(ep)}(T)$ is the classical high-temperature collision frequency (which is proportional to T).

The interelectronic collision frequency is of the form[5]

$$\nu^{(ee)}(T,\omega) = \nu_0^{(ee)}(T)\,[1 + (\hbar\omega/2\pi kT)^2],$$

where $\nu_0^{(ee)}(T)$ is the corresponding classical collision frequency which is well known to be proportional to T^2.

As was noted in reference 5, $\nu_0^{(ee)}(T) \neq 0$ only when Umklapp processes are possible. This statement is justified also in the case of an arbitrary quadratic dispersion law $\epsilon(\mathbf{p}) = \frac{1}{2}\alpha_{ik}p_i p_k$. It is of interest that when there are correlations between the electrons the absorption is, generally speaking, different from zero also when there are no Umpklapp processes in the case of an anisotropic quadratic dispersion law. Indeed, from the fact that $\mathbf{v}(\mathbf{p})$ is a linear function of the components of the vector $\mathbf{p}$ ($v_i = \alpha_{ik}p_k$), it does not follow that the vector $\mathbf{V}(\mathbf{p})$ must possess the same property (see reference 3).

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