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A NONTHRESHOLD PHOTOCONDUCTIVE EFFECT IN METALS WITH INTERSECTING BANDS

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The electric conductivity due to interaction with the electromagnetic field of electrons located near degeneracy points is calculated for metals with intersecting bands (like graphite). Various cases of the frequency dependence of electric conductivity near threshold are investigated.

THE direct absorption of photons by conduction electrons is known to result from band-to-band transitions. Since the momentum of a photon is negligibly small at infrared or lower frequencies, the energy conservation law in such cases becomes

$$\varepsilon_s(\mathbf{p}) + \hbar\omega = \varepsilon_{s'}(\mathbf{p}) \qquad (s \neq s'); \tag{1}$$

$\epsilon_s(\mathbf{p})$ is the energy of an electron with quasi-momentum $\mathbf{p}$ belonging to the s-th band; $\hbar\omega$ is the energy of a photon with frequency ω. We have, as a rule, $\min[\epsilon_{s'}(\mathbf{p}) - \epsilon_s(\mathbf{p})] \neq 0$, as a result of which absorption begins at some threshold frequency ω_{th} ($\hbar\omega_{th} = \min[\epsilon_{s'}(\mathbf{p}) - \epsilon_s(\mathbf{p})]$).

It is sometimes possible for constant-energy surfaces $\epsilon_s(\mathbf{p}) = \epsilon$ to contain one or more points of degeneracy at which the equation $\epsilon_s(\mathbf{p}) = \epsilon$ can be solved for two different values of the band index s. These are called band-intersection points. From the principle that terms do not cross,[1,2] it follows that only two bands can intersect at a degeneracy point, as a general rule. The existence of band intersection points eliminates the lower limit of absorbable frequencies. In other words, metals with intersecting bands should exhibit a nonthreshold photoconductive effect.[1)]

It is the purpose of the present work to calculate the part of the complex dielectric constant that is associated with band-to-band transitions at relatively low frequencies. However, the frequencies are assumed to be large compared with the collision frequency $\nu = 1/\tau$, where τ is the relaxation time of the electron gas. The condition $\omega\tau \gg 1$ enables us in calculating the effect of interest to entirely neglect relaxation effects in the electron system. In choosing a frequency satisfying the condition $\omega\tau \gg 1$ it must be remembered that as a result of quantum effects the collision frequency is greatly dependent on ω.[3,4] Therefore most of our results will be applicable to the far infrared region.

The interaction Hamiltonian of electrons with an arbitrary dispersion law in an electromagnetic field is written conveniently, introducing the electron velocity operator, as follows:

$$\hat{H}_{int} = -(e/c)\,\hat{\mathbf{v}}\mathbf{A}, \tag{2}$$

where $\mathbf{A}$ is the vector potential of the electromagnetic field, and $c^{-1}\partial\mathbf{A}/\partial t = \mathbf{E}$, where the electric field $\mathbf{E}$ in the metal is taken to be spatially homogeneous.

It will be convenient to include an explicit dependence on the energy difference $\epsilon_s - \epsilon_{s'}$ in the velocity operator $\hat{\mathbf{v}}$; from [1] we have

$$\mathbf{v}_{ss'} = (\partial\varepsilon_s(\mathbf{p})/\partial\mathbf{p})\,\delta_{ss'} + (\varepsilon_s - \varepsilon_{s'})\,\hbar^{-1}\boldsymbol{\Omega}_{ss'}. \tag{3}$$

The operator $\hat{\boldsymbol{\Omega}}$ is associated with band-to-band transitions and appears in the expression for the coordinate operator in the quasi-momentum representation:[1,5] $\hat{\mathbf{r}} = (\hbar/i)\,\partial/\partial\mathbf{p} + \hat{\boldsymbol{\Omega}}$. The matrix elements of the operator $\hat{\boldsymbol{\Omega}}$ do not generally vanish at the degeneracy points [where $\epsilon_s(\mathbf{p}) = \epsilon_{s'}(\mathbf{p})$].

In order to calculate the complex dielectric constant we define the mean current as $\mathbf{j} = \mathrm{Sp}\,\{\hat{\rho}\hat{\mathbf{j}}\}$. Here $\hat{\mathbf{j}}$ is the second-quantized current density operator:

$$\hat{\mathbf{j}} = \frac{e}{V}\sum_{\mathbf{p}ss'} a^{+}_{s\mathbf{p}}\mathbf{v}_{ss'}\,a_{s'\mathbf{p}}, \tag{4}$$

[1)] The intersection of bands is relatively rare among metals. An example is found in graphite, photon absorption in which will be discussed below. Band intersection should not be confused with band overlapping, in which case $\min \epsilon_s < \max \epsilon_{s'}(\mathbf{p})$ and $\min \epsilon_{s'}(\mathbf{p}) < \min \epsilon_s(\mathbf{p})$. In the case of band overlapping (without intersection) nonthreshold absorption is impossible since the coinciding energy values in the different bands correspond to different values of the quasi-momentum.

where V is the volume of the crystal, a^+_{sp} and a_{sp} are the electron creation and annihilation operators in a state of band s with quasi-momentum **p**, and $\hat{\rho}$ is a statistical operator satisfying

$$i\hbar\dot{\hat{\rho}} = [\hat{H}, \hat{\rho}], \quad \hat{H} = \hat{H}_0 + \hat{H}_t, \tag{5}$$

where $\hat{H}_0$ is the Hamiltonian of the electron system:

$$\hat{H}_0 = \sum_{s\mathbf{p}} \varepsilon_s(\mathbf{p})\, a^+_{s\mathbf{p}} a_{s\mathbf{p}}, \tag{6}$$

and $\hat{H}_t$ is the interaction operator (2), which has the second-quantized form

$$\hat{H}_t = -\frac{e}{c} \sum_{ss'\mathbf{p}} a^+_{s\mathbf{p}} a_{s'\mathbf{p}} \mathbf{v}_{ss'} \mathbf{A}(t). \tag{7}$$

Using the definition $j_i = \sigma_{ik}E_k$ of the electric conductivity tensor, we obtain by the standard procedure

$$\sigma_{ik} = -\frac{ie^2}{(2\pi\hbar)^3\,\omega} \sum_{\substack{ss' \\ \gamma\to 0}} \int d\tau_{\mathbf{p}} \left\{ \frac{v^i_{ss'} v^k_{s's}}{\varepsilon_s(\mathbf{p}) - \varepsilon_{s'}(\mathbf{p}) - \hbar\omega - i\gamma} + \frac{v^i_{ss'} v^k_{s's}}{\varepsilon_s(\mathbf{p}) - \varepsilon_{s'}(\mathbf{p}) + \hbar\omega + i\gamma} \right\} (n_{s\mathbf{p}} - n_{s'\mathbf{p}}), \tag{8}$$

where n_{sp} is the Fermi equilibrium function

$$n_{s\mathbf{p}} = \left(\exp \frac{\varepsilon_s(\mathbf{p}) - \zeta}{T} + 1 \right)^{-1}, \tag{9}$$

and ζ is the chemical potential of the electron gas [$\epsilon_s(\mathbf{p}) = \zeta$ is the Fermi surface].

It will be recalled that we are calculating only that part of σ_{ik} that is associated with band-to-band transitions. In order to calculate the total electric conductivity tensor we must take into account the coordinate dependence of the electromagnetic field potentials.[4] We now write (8) in the case of an electron transition from the first to the second band:

$$\sigma_{ik} = -\frac{2ie^2}{(2\pi\hbar)^3\,\omega} \int_{\gamma\to 0} d\tau_{\mathbf{p}} \left\{ \frac{v^i_{12} v^k_{21}}{\varepsilon_2 - \varepsilon_1 - \hbar\omega - i\gamma} + \frac{v^i_{21} v^k_{12}}{\varepsilon_2 - \varepsilon_1 + \hbar\omega + i\gamma} \right\} (n_1 - n_2), \tag{10}$$

where $\epsilon_2 > \epsilon_1$ is assumed. From this formula we derive diagonal values of the real and imaginary parts of the tensor σ_{ik}:

$$\operatorname{Re}\sigma_\alpha = \frac{2\pi e^2}{(2\pi\hbar)^3\omega} \int |v^\alpha_{12}|^2 (n_1 - n_2)\, \delta(\varepsilon_2 - \varepsilon_1 - \hbar\omega)\, d\tau_{\mathbf{p}} \tag{11}$$

$$\operatorname{Im}\sigma_\alpha = -\frac{4e^2}{(2\pi\hbar)^3\omega} \int \frac{(\varepsilon_2 - \varepsilon_1)\,|v^\alpha_{12}|^2 (n_1 - n_2)\, d\tau_{\mathbf{p}}}{(\varepsilon_2 - \varepsilon_1)^2 - (\hbar\omega)^2}, \tag{12}$$

where the integrals in (12) must be understood to mean

$$\int f(\mathbf{p})\, d\tau_{\mathbf{p}} = \int d\varepsilon \oint_{\varepsilon(\mathbf{p}) = \varepsilon} f(\mathbf{p}) \frac{dS}{|\nabla_{\mathbf{p}}\varepsilon|}. \tag{13}$$

Equation (11), in which the character of the dispersion law is not specified, does not restrict us to an investigation of only nonthreshold absorption, which we discuss here. The equation is also valid for the general case of band-to-band transitions, and we can use it to arrive at the frequency dependence of Re σ_α (i.e., of the absorption coefficient) near the absorption threshold. We shall first consider the most probable low-temperature case (or rather, the relatively high frequency case $\hbar\omega \gg T$). The temperature can here be set equal to zero, in which case the integral in (11) becomes

$$I = \int_{(\varepsilon_F - \hbar\omega < \varepsilon_1 < \varepsilon_F)} |v^\alpha_{12}|^2\, \delta(\varepsilon_2 - \varepsilon_1 - \hbar\omega)\, d\tau_{\mathbf{p}}, \tag{14}$$

where ϵ_F is the Fermi energy. The integration region is shown schematically in Fig. 1 by the cross-hatched area.

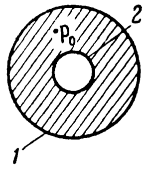

FIG. 1. Integration region in (14). 1 – the surface $\epsilon_1(\mathbf{p}) = \epsilon_F$; 2 – the surface $\epsilon_1(\mathbf{p}) = \epsilon_F - \hbar\omega$.

Two cases are possible, as follows.

In the first case the point $\mathbf{p}_0$ at which the function $\epsilon_2(\mathbf{p}) - \epsilon_1(\mathbf{p})$ reaches its minimum is located in the region of **p**-space over which Eq. (14) is integrated. We then have

$$\varepsilon_2(\mathbf{p}) - \varepsilon_1(\mathbf{p}) = \hbar\omega_{\mathrm{th}} + \tfrac{1}{2}[\partial^2(\varepsilon_2 - \varepsilon_1)/\partial p_i\, \partial p_k]_{\mathbf{p}=\mathbf{p}_0} (p_i - p_{i0})(p_k - p_{k0}),$$

$$\hbar\omega_{\mathrm{th}} = \min(\varepsilon_2 - \varepsilon_1) \equiv [\varepsilon_2 - \varepsilon_1]_{\mathbf{p}=\mathbf{p}_0}. \tag{14a}$$

The principal values of the tensor $\partial^2(\epsilon_2 - \epsilon_1)/\partial p_i \partial p_k$ will be denoted by $1/M_1$, $1/M_2$, and $1/M_3$. Equation (14) then yields

$$I = \pi\, |v^\alpha_{12}(\mathbf{p}_0)|^2 \sqrt{32\, M_1 M_2 M_3 \hbar\, (\omega - \omega_{\mathrm{th}})} \qquad (\omega \gtrsim \omega_{\mathrm{th}}) \tag{15}$$

or

$$\operatorname{Re}\sigma_\alpha \approx \frac{e^2\sqrt{2}}{\pi\hbar^{5/2}\omega_{\mathrm{th}}} |v^\alpha_{12}(\mathbf{p}_0)|^2 (M_1 M_2 M_3)^{1/3} (\omega - \omega_{\mathrm{th}})^{1/2} \quad (\omega \gtrsim \omega_{\mathrm{th}}). \tag{16}$$

Thus in this case the derivative of electric conductivity (and therefore of the absorption coefficient) with respect to frequency possesses a square-root singularity (see Fig. 2).

In the second case $\mathbf{p}_0$ lies outside the integration region of (14). This means that at the frequency threshold ω'_{th} the surface bounding the integration region is tangent to the surface $\epsilon_2(\mathbf{p})$

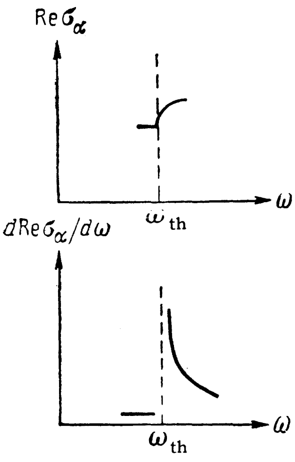

FIG. 2. Frequency dependency of $\mathrm{Re}\,\sigma_\alpha$ near threshold (for the case represented in Fig. 1).

$-\epsilon_1(\mathbf{p}) = \hbar\omega$ (Fig. 3). In the case represented by Fig. 3a we have

$$\mathrm{Re}\,\sigma_\alpha = \frac{e^2 \mid v_{12}^\alpha(\mathbf{p}_1)\mid^2 R_G R_{th}(\omega - \omega'_{th})}{2\pi\hbar^2\omega'_{th}(R_G + R_{th})\,(v_2 - v_1)^2} \quad (\omega \gtrless \omega'_{th}); \qquad (17)$$

R_G and R_{th} are the corresponding radii of curvature (R_F for the Fermi surface, R_G for the surface $\epsilon_1(\mathbf{p}) = \epsilon_F + \hbar\omega'_{th}$, and R_{th} for the surface $\epsilon_2 - \epsilon_1 = \hbar\omega'_{th}$). All values are taken at the point $\mathbf{p}_1$.

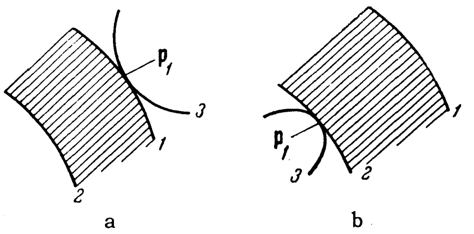

FIG. 3. Contact of the surface $\epsilon_2(\mathbf{p}) - \epsilon_1(\mathbf{p}) = \hbar\omega$ with the surface bounding the integration region in (14). 1 – the surface $\epsilon_1(\mathbf{p}) = \epsilon_F$; 2 – $\epsilon_1(\mathbf{p}) = \epsilon_F - \hbar\omega$; 3 – $\epsilon_2 - \epsilon_1 = \hbar\omega$. The cross-hatched area is a portion of the region represented in Fig. 1.

In the case represented in Fig. 3b we have

$$\mathrm{Re}\,\sigma_\alpha = \frac{e^2 \mid v_{12}^\alpha(\mathbf{p}_1)\mid^2 R_F R_{th}(\omega - \omega'_{th})}{2\pi\hbar^2\omega'_{th}(R_F + R_{th})\mid v_1 - v_2\mid}\left\{\frac{1}{v_1} + \frac{1}{\mid v_1 - v_2\mid}\right\} \quad (\omega \gtrless \omega'_{th}). \qquad (18)$$

Thus in the second case the derivative of $\mathrm{Re}\,\sigma_\alpha$, and therefore of the absorption coefficient, exhibits a finite jump (Fig. 4).

At finite but low temperatures the electric conductivity in the first case depends very slightly on temperature. In the second case, for $\omega_{th} < \omega < \omega'_{th}$ the absorption coefficient is exponentially dependent on temperature.

We shall now consider the high-temperature case ($\hbar\omega \ll T$), where

$$n_1 - n_2 \approx -\frac{\partial n}{\partial\varepsilon}\hbar\omega = \frac{\hbar\omega}{T}e^{(\varepsilon_1-\zeta)/T}\left(e^{(\varepsilon_1-\zeta)/T} + 1\right)^{-2}$$

(ζ is the chemical potential) and

$$\mathrm{Re}\,\sigma_\alpha \approx \frac{e^2}{(2\pi\hbar)^2 T}\int \mid v_{12}^\alpha(p_0)\mid^2 \frac{e^{(\varepsilon_1-\zeta)/T}\,\delta(\varepsilon_2 - \varepsilon_1 - \hbar\omega)}{(\exp(\varepsilon_1 - \zeta)/T + 1)^2}\,d\tau_{\mathbf{p}}.$$

Proceeding as with the derivation of (16), we obtain

$$\mathrm{Re}\,\sigma_\alpha \approx \frac{\sqrt{2}\,e^2}{\pi\hbar^{3/2}}\,\frac{\mid v_{12}^\alpha(\mathbf{p}_0)\mid^2}{T}\sqrt{M_1 M_2 M_3\,(\omega - \omega_{th})}\,\frac{e^{(\varepsilon_1(\mathbf{p}_0)-\zeta)/T}}{\left(e^{(\varepsilon_1(\mathbf{p}_0)-\zeta)/T} + 1\right)^2}. \qquad (19)$$

If $\mid\epsilon_1(\mathbf{p}_0) - \zeta\mid \ll 1$, then $\mathrm{Re}\,\sigma_\alpha \sim 1/T$. If $\mid\epsilon_1(\mathbf{p}_0) - \zeta\mid \gg 1$, then $\mathrm{Re}\,\sigma_\alpha$ and the absorption coefficient are exponentially small quantities ($\mathrm{Re}\,\sigma_\alpha \sim e^{-\mid\epsilon_1(\mathbf{p}_0) - \zeta\mid/T}$).

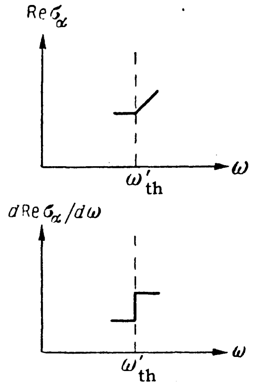

FIG. 4

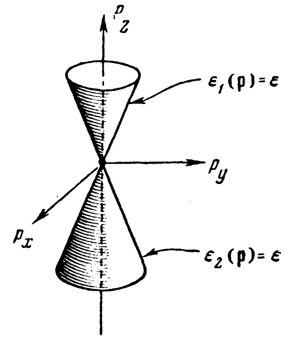

FIG. 5

FIG. 4. Frequency dependence of $\mathrm{Re}\,\sigma_\alpha$ near threshold (for the case represented in Fig. 3).

FIG. 5. The Fermi surface near a point of degeneracy.

We have thus far tacitly assumed that $\mathbf{v}_{12}(\mathbf{p}_0)$ and $\mathbf{v}_{12}(\mathbf{p}_1)$ are different from zero; this is evidently the most frequent case. However, if $\mathbf{p}_0$ is a high symmetry point it can be shown that $\mathbf{v}_{12}(\mathbf{p}_0) = 0$ (a forbidden transition). Expanding $\mathbf{v}_{12}(\mathbf{p})$ in powers of $\mathbf{p} - \mathbf{p}_0$, it is easily shown that in this case $\mathrm{Re}\,\sigma_\alpha = (\omega - \omega_{th})^{3/2}$. Although the vanishing of $\mathbf{v}_{12}(\mathbf{p}_1)$ is extremely unlikely, we note that if $\mathbf{v}_{12}(\mathbf{p}_1) = 0$, then $\mathrm{Re}\,\sigma_\alpha \sim (\omega - \omega_{th})^2$.[2]

We now come to the case of energy band inter-

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[2] The threshold absorption equations (16) - (19) derived here are analogous to those for semiconductors.[8] It must be noted, however, that the metal equations are more accurate than the semiconductor equations, since metals have no excitons, while in semiconductors the laws $\Gamma \sim (\omega - \omega_{th})^{1/2}$ and $(\omega - \omega_{th})^{3/2}$ hold true only for $\hbar(\omega - \omega_{th}) >> \epsilon_{exc}$ where ϵ_{exc} is the exciton ionization energy.[9] (We are indebted to E. I. Rashba for this observation.)

section. Near a self-intersection point of a constant-energy surface the general dispersion law can be written as

$$\varepsilon_{1,2} = A(p_z) \pm B(p_z, \mathbf{n}_\perp)\, p_\perp, \qquad (20)$$

where $p_\perp = \sqrt{p_x^2 + p_y^2}$, $\mathbf{n}_\perp = \mathbf{p}_\perp / p_\perp$, and the axes are located as in Fig. 5.

In analyzing the frequency dependence of the conductivity it must be taken into account that according to (4) we have

$$\mathbf{v}_{12} = [(\varepsilon_2 - \varepsilon_1)/\hbar]\, \Omega_{12}.$$

Therefore (13) can be written as

$$\operatorname{Re} \sigma_\alpha = \frac{2\pi e^2 \omega}{(2\pi\hbar)^3} \int |\Omega_{12}^\alpha|^2 \, [n(\varepsilon_1) - n(\varepsilon_1 + \hbar\omega)]\, \delta(\varepsilon_2 - \varepsilon_1 - \hbar\omega)\, d\tau_p.$$

We first consider the case of very low temperatures ($\hbar\omega \gg T$). Integrating, subject to the condition $\hbar\omega \ll \epsilon_F$, we obtain

$$\operatorname{Re} \sigma_\alpha \approx \frac{e^2 \omega^3}{8\pi\hbar} \sum \overline{\frac{|\Omega_{12}^\alpha|^2}{B^2 v_z}}, \qquad (21)$$

where the bar denotes averaging over angles in the (p_x, p_y) plane. All quantities included in (21) are taken for $p_\perp = 0$ and $\epsilon_1 = \epsilon_F$. The summation is carried out over all cone points. If we assume $\Omega_{12} \sim a$, $v_z \sim B \sim v$, we obtain in order of magnitude

$$\operatorname{Re} \sigma_\alpha \approx e^2 a^2 \omega^3 / 8\pi\hbar v^3. \qquad (22)$$

It is easily verified that in the opposite extreme case ($\hbar\omega \ll T$) the foregoing derived equation is valid to within terms of the order $\hbar\omega/T$, i.e., $\operatorname{Re} \sigma_\alpha \sim \omega^3$, with only slight temperature dependence in relatively broad frequency and temperature ranges.

We shall now discuss the case of graphite in greater detail.[6] The Fermi surface of graphite contains eight degeneracy points, two of which are located on the axis of symmetry. Near these points (points p_s in Fig. 6) the surface possesses axial symmetry. Knowledge of the quantities appearing in the dispersion law of graphite electrons and holes enables one to estimate the parameters in (21). According to [6] we have $B \sim v_z \sim 10^6$ cm/sec. This makes it possible in principle to evaluate a nondiagonal element of the coordinate operator from a comparison with experimental data.³⁾

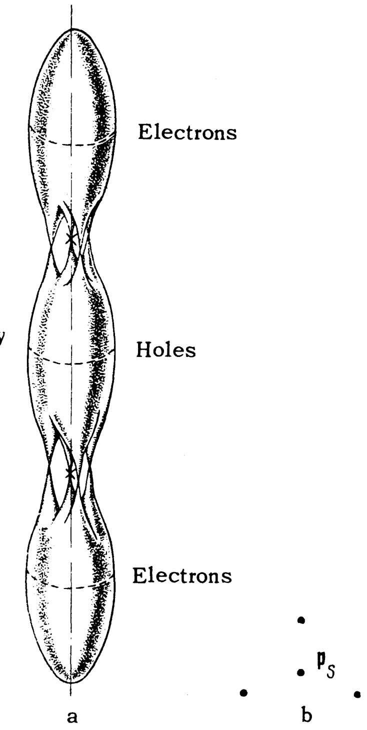

FIG. 6. The Fermi surface of graphite. a – overall view; b – arrangement of degeneracy points in the plane p_z = const. Degeneracy points located on the axis (as the point p_s) are denoted by crosses.

As already stated, the low-frequency limit of (21) is given by $\omega\tau \gg 1$. At high frequencies the limitation is associated with the expansion (20). For graphite this decomposition can be used only if the energy quantum $\hbar\omega$ is considerably smaller than $H_{33} \sim \gamma_1(\gamma_3/\gamma_0)^2$ (given in the notation of [6]). Computations yield $\gamma_1(\gamma_3/\gamma_0)^2 \sim 0.03$ eV, i.e., in addition to the given condition $\hbar\omega \ll \epsilon_F$ we also require the inequality $\omega \ll 4 \times 10^{13}$ sec^{-1}. Neglect of terms $\sim H_{33}$ in the dispersion law leads to a merging of four cone points into one point. The intersection of bands is then accompanied by contact (Fig. 7) and the constant-energy surface possesses axial symmetry.

The dispersion law can be written as

$$\varepsilon_{1,2} = A'(p_z) \pm B'(p_z)\, p_\perp^2. \qquad (23)$$

A calculation analogous to the foregoing, using (13) for $\operatorname{Re} \sigma_\alpha$, yielded

$$\operatorname{Re}\sigma_\alpha \approx \pi e^2 \omega^2 (2\pi\hbar)^{-2} (|\Omega_{12}^\alpha|^2 / B' v_z)_0 \quad (\varepsilon_F \gg \hbar\omega \gg H_{33}). \qquad (24)$$

We recall that the equations derived here for

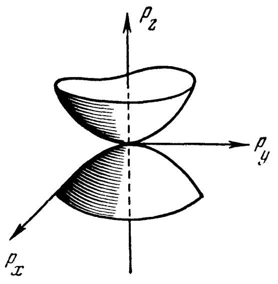

FIG. 7. Self-intersection with contact.

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³⁾We have here evaluated only B and v_z. The parameters of the electronic structure of graphite are known so completely that B and v_z can be calculated very accurately.

Re σ_α describe only the part of the electric conductivity that is associated with band-to-band transitions. A comparison of the equations derived here with the usual equations[5] shows that band-to-band transitions play a fundamental part at relatively high frequencies. The experiments of Boyle and Nozières[7] show that band-to-band transitions can account for the observed absorption pattern. However, the results given in [7] do not account for the analytic dependence of the absorption coefficient.

An additional circumstance must be noted in conclusion. According to (14), Im σ_α is negative at low frequencies and is positive at very high frequencies, the asymptotic frequency dependence being given by

$$\operatorname{Im} \sigma_\alpha \approx \frac{4e^2}{(2\pi\hbar)^3 (\hbar\omega)^2 \omega} \int (\varepsilon_2 - \varepsilon_1) \mid v_{12}^{\alpha} \mid^2 (n_1 - n_2) \, d\tau_p. \qquad (25)$$

Since ϵ_2 and ϵ_1 are close to each other, the sign reversal of Im σ_α must occur at relatively low frequencies and can in principle lead to a sign reversal of the dielectric constant ϵ of a metal ($\epsilon_\alpha = -4\pi \operatorname{Im} \sigma_\alpha / \omega$) in a certain frequency region (or at least to a great change in the value of $|\epsilon_\alpha|$). For $\omega\tau \gg 1$ the reflectivity of a metal is known to be associated with the fact that under these conditions Re $\epsilon < 0$ with $|\epsilon| \gg 1$. The sign reversal of Re ϵ must be accompanied by a considerable reduction of the reflection coefficient.

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