Multiphoton absorption in thin superconducting films

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Multiphoton absorption following specular or diffuse reflection of electrons from the surface of a thin superconducting film is considered. The film is assumed to be sufficiently pure: $\omega_0 \tau > 1$, where ω_0 is the field frequency and τ is the electron mean free time. Only the effect of the wave electric field on the electrons is taken into account, a procedure permissible for thin films. In a first approximation with respect to $(\omega_0 \tau)^{-1}$ multiphoton absorption is found to be possible only in the superconducting state. It is shown that in the case of specular reflection from the boundary the probability of *n*-photon absorption, with n > 1, is primarily determined by the amplitude for the backscattering of the electrons by the impurity. In the case of diffuse reflection the absorption probability is expressed in terms of the transport time between collisions.

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1. It is known that at zero temperature a weak electromagnetic field of frequency $\omega_0 < 2\Delta$ is not absorbed in a superconductor. With increasing field amplitude, however, the absorption probability increases and the condition $\omega_0 > 2\Delta$ is no longer necessary. A consistent theoretical description of multiphoton absorption calls for the calculation of the Green's functions of the superconductor in an electromagnetic field without assuming its amplitude to be small, while the electric and magnetic components of the wave lead, generally speaking, to effects of equal order of magnitude.^[1] The situation simplifies somewhat in thin films of thickness $d \ll \delta$, where δ is the depth of penetration of the field. Under these conditions, only the electric high-frequency field component which is homogeneous over the cross section of the film is significant, ^[1] and the magnetic field of the wave can be disregarded. In this approximation it is possible to obtain the multiphoton absorption probability in sufficiently pure superconducting films by a method similar to that used by Keldysh in the problem of multiphoton ionization of an atom.^[2]

The Gor'kov equations for the time-dependent Green's functions in a pure superconducting film in a homogeneous field with a vector potential $\mathbf{A}(t) = \mathbf{A}_0 \cos \omega_0 t$

$$\begin{cases} i\frac{\partial}{\partial t} + \frac{1}{2m} \left(\nabla_r - \frac{ie}{c} \mathbf{A}(t) \right)^2 + \mu \\ G(x, x') + i\Delta(x)F^+(x, x') = \delta(x - x'), \\ \left\{ i\frac{\partial}{\partial t} - \frac{1}{2m} \left(\nabla_r + \frac{ie}{c} \mathbf{A}(t) \right)^2 - \mu \\ F^+(x, x') - i\Delta^+(x)G(x, x') = 0 \end{cases}$$
(1)

have solutions

$$G.F^{+}(x,x') = G_{0}, \quad F_{0}^{+}\left(\mathbf{r}-\mathbf{r}'-\frac{e}{mc}\int_{t'}^{t}\mathbf{A}(\tau)d\tau, \ t-t'\right),$$
(2)

where $\Delta(x) = \Delta$, $x \equiv (\mathbf{r}, t)$, $G_0(x - x')$, and $F_0(x - x')$ are equilibrium Green's functions satisfying (1) at A = 0. The solutions (2) have a meaning at frequencies $\omega_0 \gg \nu_0$, ^[3] where ν_0 is the frequency of the homogeneous relaxation, which in superconductors is a relatively slow process, ^[4] and as noted by Gor'kov and Éliashberg, ^[3] its influence can be disregarded. It is shown in ^[3] that the condition $\Delta(x) = \Delta$ imposes a limitation on the amplitude of the high-frequency field. As $\nu_0 \rightarrow 0$, however, the correction to the gap tends to zero.

Expression (2) describes the displacement of the entire Fermi sphere under the influence of the electric field. In a coordinate system moving with velocity

$$v = \frac{eA_0}{mc} \cos \omega_0 t$$

the Green's function takes an equilibrium form. The presence of impurities causes the Green's function in the field no longer to be expressed by formulas (2) in terms of equilibrium formulas. In a moving coordinate system the potential $\tilde{u}(\mathbf{r}, t)$ exerted by the impurity atoms on the electrons is explicitly dependent on the time

$$\widetilde{u}(\mathbf{r},t) = u\left(\mathbf{r} - \frac{e}{mc}\int \mathbf{A}(\tau) d\tau\right)$$
$$= \sum_{i,n} \int \frac{d\mathbf{q}}{(2\pi)^3} v(\mathbf{q}) J_n\left(\frac{e\mathbf{A}_0\mathbf{q}}{mc\omega_0}\right) \exp\left\{i\mathbf{q}(\mathbf{r}-\mathbf{R}_i) - in\omega_0t\right\},$$
(3)

where $v(\mathbf{q})$ is the Fourier component of a potential of an individual impurity atom located at the point \mathbf{R}_i (in the immobile coordinate system), and $J_n(x)$ is a Bessel function. The result is a number of nonstationary effects, including multiquantum absorption.

It is easily seen that expression (2) corresponds to the absence of any absorption whatever. To find the Green's functions with allowance for (3) we can use a diagram technique. We assume the impurity density to be small, with $\omega_0 \tau \gg 1$, where τ is the electron mean free time. This enables us to take into account the impurities only in the first nonvanishing approximation when determining the absorption probability.

We consider thin films, so that a major role is played by the conditions of reflection from the boundary. The character of the reflection from the surface is not universal for all metals, particularly for indium films it can be close to specular.^[5] We obtain below the probability of multiquantum absorption both for completely specular and for absolutely diffuse reflection of the electrons from the surface, and show that *n*-photon absorption (n > 1) is possible in the first approximation in $(\omega_0 \tau)^{-1}$ only in the superconducting state. In a normal

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metal, in the same order in $\omega_0 \tau$, there is only onephoton absorption. In specular reflection, the probability of multiphoton absorption at $n \gg 1$ is determined mainly by the amplitude of the backscattering of the electrons by the impurities, whereas in diffuse reflection everything is expressed only in terms of the transport time $\tau_{\rm tr}$ between the collisions.

2. In specular reflection, the problem of absorption in a film reduces to the problem for an infinite sample. At zero temperatures, the diagrams for the Green's functions have graphically the same form as the theory of superconducting alloys, ^[6] but it must be borne in mind that the frequencies in the impurity vertices, in accordance with (3), are no longer conserved. An analytic expression for the correction quadratic in $v(\mathbf{q})$ to the Green's function is given by

$$\delta G(\mathbf{p},\omega,\omega') = N \sum_{\substack{n_1,n_2\\n_1,n_2}} \int \frac{d\mathbf{p}'}{(2\pi)^3} v_{n_1}(\mathbf{p}-\mathbf{p}') v_{n_2}(\mathbf{p}'-\mathbf{p})$$

$$\times \{G_v(\mathbf{p},\omega) G_v(\mathbf{p}',\omega-n_1,\omega_v) G_u(\mathbf{p},\omega-n\omega_v) + G_v(\mathbf{p},\omega)$$

$$\times F(\mathbf{p}',\omega-n_1,\omega_v) F_v(\mathbf{p},\omega-n\omega_v) + F(\mathbf{p},\omega) F^+(\mathbf{p}',\omega-n_1,\omega_v) G_v(\mathbf{p},\omega-n\omega_v)$$

$$+ F_v(\mathbf{p},\omega) G_v(\mathbf{p}',-\omega+n_1,\omega_v) G_v(\mathbf{p},-\omega+n\omega_v)\} \delta(\omega-\omega'-n\omega_v); \quad (4)$$

$$G_v(\mathbf{p},\omega) = \frac{\omega + \zeta_p}{\omega^2 - (\varepsilon_v - i\delta)^2}, \quad F_v(\mathbf{p},\omega) = F_v^+(\mathbf{p},\omega) = \frac{i\Delta}{\omega^2 - (\varepsilon_v - i\delta)^2}, \quad (5)$$

where G_0 and F_0 are the equilibrium functions, $\xi_p = p^2/2m - \mu$, $\mu = p_0^2/2m$, $\varepsilon_p^2 = \zeta_p^2 + \Delta^2$, $n = n_1 + n_2$, N is the number of impurity atoms, $\beta = e/mc\omega_0$, and

$$v_n(\mathbf{p}-\mathbf{p}') = v(\mathbf{p}-\mathbf{p}')J_n[\beta \mathbf{A}_0 \times (\mathbf{p}-\mathbf{p}')] .$$
(6)

We assume that the scattering of the electrons is not accompanied by spin flips. Integrating in (4) with respect to ω and ω' , we get

$$\lim_{t' \to t+0} \delta G(\mathbf{p}, t, t') = -\frac{iN}{2} \sum_{n} \exp(-in\omega_0 t) v_{n_1}(\mathbf{p} - \mathbf{p}')$$

$$\times v_{n_1}(\mathbf{p}' - \mathbf{p}) \{ f(-\varepsilon_p) \varepsilon_p^{-1} D_{n_1}(\varepsilon_p, \varepsilon_{p'}) D_n(\varepsilon_p, \varepsilon_{p'})$$

$$+ f(-\varepsilon_p + n\omega_0) \varepsilon_p^{-1} D_{-n_1}(\varepsilon_p, \varepsilon_p) D_{-n_2}(\varepsilon_p, \varepsilon_{p'})$$

$$+ f(-\varepsilon_{p'} + n\omega_0) \varepsilon_{p'}^{-1} D_{-n_1}(\varepsilon_{p'}, \varepsilon_p) D_{n_2}(\varepsilon_{p'}, \varepsilon_p) \}:$$
(7)

where

$$f(x) = (x + \zeta_p) (x - n_1 \omega_0 + \zeta_{p'}) (x - n \omega_0 + \zeta_p) - \Delta^2 (x + 2\zeta_p + \zeta_{p'} - n_2 \omega_0),$$

$$D_n^{-1} (\varepsilon_p, \varepsilon_{p'}) = (\varepsilon_p + n \omega_0 + i\delta)^2 - \varepsilon_{p'}^2.$$
(8)

The poles in (7) are circled in accordance with the causality principle (see, e.g., $[7_1]$).

Multiplying (7) by -2iep/m and integrating with respect **p**, we obtain the paramagnetic part of the current density

 $\mathbf{j}(t) = \widetilde{\mathbf{j}} + eN_0\mathbf{A}(t)/mc$

where \mathbf{j} is the current density and N_0 is the electron concentration. When integrating with respect to \mathbf{p} and \mathbf{p}' , the terms of (8) that are linear in ζ_p and $\zeta_{p'}$ vanish, since $|\mathbf{p} - p'| \nu_0 \gg \Delta$. As a result of rather prolonged algebraic transformations, the expression for $\mathbf{j}(t)$ can be reduced to the form

$$\mathbf{j}(t) = \frac{eN}{m} \sum_{n} \exp(-in\omega_0 t) \int \frac{d\mathbf{p} \, d\mathbf{q}}{(2\pi)^8} \frac{\mathbf{q} \, |v(\mathbf{q})|^2}{n\omega_0} J_{n_1}(\beta \mathbf{A}_0 \mathbf{q}) J_{n_2}(-\beta \mathbf{A}_0 \mathbf{q}) R_{n_1};$$

$$\mathbf{R}_n = [\varepsilon_p(\varepsilon_p + n\omega_0) - \Delta^2] D_n(\varepsilon_p, \varepsilon_{p-q}) + [\varepsilon_p(\varepsilon_p - n\omega_0) - \Delta^2] D_{-n} \cdot (\varepsilon_p, \varepsilon_{p-q}).$$
(9)

(10)

From the very meaning of the employed approximation $\omega_0 \tau \gg 1$ it is clear that we cannot put n = 0 in (9), i.e., our approximation is not suitable for the determination of the zeroth harmonic. However, multiphoton absorption is described by the first harmonic of the current (n = 1), where there is no divergence.

We integrate first with respect to the vector **p**. From expression (8) for $D_n(\varepsilon_p, \varepsilon_{p-q})$ we see that the main contribution is made by momentum values p satisfying the condition $\varepsilon_{p-q} \approx \varepsilon_p$, i.e.,

 $\cos p q = q/2p_0.$

In a spherical coordinate system with the z axis along the vector **q**, the integral with respect to the polar angle can be easily calculated. As a result we get

$$\mathbf{j}(t) = \sum_{\mathbf{i},n} \mathbf{j}_{n} \exp\left(-in\omega_{0}t\right),$$

$$\mathbf{j}_{i} = \frac{eNm}{8\pi^{2}\omega_{0}} \sum_{\mathbf{i},n} \int \frac{dq}{(2\pi)^{2}} \frac{\mathbf{q} |v(\mathbf{q})|^{2}}{q} \int d\varphi J_{n}(\beta \mathbf{A}_{0}\mathbf{q}) J_{i-n}(-\beta \mathbf{A}_{0}\mathbf{q}) \tilde{R}_{n}, \qquad (11)$$

and the imaginary part of \bar{R}_n is equal to

$$\operatorname{Im} \tilde{R}_{n} = \int \frac{d\xi}{\varepsilon} \left\{ \frac{\varepsilon (\varepsilon - n\omega_{0}) - \Delta^{2}}{\left[(\varepsilon - n\omega_{0})^{2} - \Delta^{2} \right]^{t_{n}}} \operatorname{sign} (\varepsilon - n\omega_{0}) - \frac{\varepsilon (\varepsilon + n\omega_{0}) - \Delta^{2}}{\left[(\varepsilon + n\omega_{0})^{2} - \Delta^{2} \right]^{t_{n}}} \operatorname{sign} (\varepsilon + n\omega_{0}) \right\}.$$
(12)

The limits of integration with respect to ζ are restricted by the requirement that the radicands be positive.

Starting from the results of $[7_1]$, we can show that at nonzero temperatures the integrand in (12) must be multiplied by $\tanh(\varepsilon/2T)$. Multiphoton absorption is described by the imaginary part of \tilde{R}_n , whereas $\operatorname{Re} \tilde{R}_n$ describes the change of the reactions in the electromagnetic field. It is seen from (12) that $\operatorname{Im} \tilde{R}_n$ is an odd function of n at T = 0 and $n \ge 0$

$$\operatorname{Im} \widetilde{R}_{n} = n\omega_{0} \left\{ \frac{x}{1+x} K\left(\frac{1-x}{1+x}\right) - 2(1+x) E\left(\frac{1-x}{1+x}\right) \right\}, \qquad (12')$$

(*E* and *K* are complete elliptic integrals, and $x = 2\Delta/n\omega_{0.0}$) At $n\omega_{0} < 2\Delta$ we always have $\text{Im}\tilde{R}_{n} = 0$. As $n\omega_{0} \rightarrow 2\Delta$, however, the imaginary part $\text{Im}\tilde{R}_{n}$ does not tend to zero, thus indicating that it is necessary to take the collisions more carefully into account at $n\omega_{0} - 2\Delta \approx \tau^{-1}$, and expression (12) is valid only if $n\omega_{0} - 2\Delta > \tau^{-1}$.

We note that a similar situation arises in the theory of Raman scattering of light in superconductors.^[8]

We calculate now the remaining integrals and obtain $\text{Im} j_1$. Integration with respect to $d\varphi$ in (11) obviously yields simply the factor 2π . The integration with respect to the azimuthal angle of the vector **q** in a spherical coordinate system with z axis along **A** is also trivial, inasmuch as it is clear from the symmetry of the problem that **j** is directed along **A**. As a result we obtain

$$\operatorname{Im} j_{1} = \frac{eNm}{4\pi^{3}\omega_{0}\beta A_{0}} \sum_{n>0} n \operatorname{Im} \tilde{R}_{n} \int_{0}^{2p_{0}} dq \, q |v(q)|^{2} \int_{0}^{4} J_{n}^{2}(\beta A_{0}qt) \, dt, \qquad (13)$$

where we have used the recurrence relation for the Bessel functions $xJ_{n+1}(x) + xJ_{n-1}(x) = 2nJ_n(x)$, and also $\operatorname{Im} \tilde{R}_n = -\operatorname{Im} \tilde{R}_{-n}$.

In expression (13) q is in fact that momentum transferred to the electron when scattered through an angle θ by the impurity: $q = 2p_0 \sin(\theta/2)$. We can therefore represent (13) in the form

$$\operatorname{Im} j_{i} = \frac{3eNN_{\circ}}{8\pi^{2}\omega_{\circ}\beta A_{\circ}p_{\circ}} \sum_{n>o} n \operatorname{Im} \tilde{R}_{n} \int |v(\theta)|^{2} do$$
$$\times \int_{\circ}^{4} J_{n}^{2} \left(2\beta p_{\circ}A_{\circ}t \sin \frac{\theta}{2} \right) dt.$$
(13')

At small A_0 the Bessel functions in (13) can be expanded in series, from which it is clear that the criterion for the smallness of the field is the condition

$$eA_{\circ}p_{\circ}/mc\omega_{\circ}\sqrt{n}\approx eA_{\circ}p_{\circ}/mc\sqrt{\omega_{\circ}\Delta}\ll 1,$$
(14)

since $n \approx \Delta/\omega_0$.

Since $4\Sigma n^2 J_n^2(x) = x^2$, for a normal metal where $\text{Im} \tilde{R}_n = -2n\omega_0$, we have

$$\operatorname{Im} j_{1} = -\frac{e^{2}N_{o}A_{o}}{2mc\omega_{o}\tau_{tr}}, \quad \frac{1}{\tau_{tr}} = \frac{mp_{o}}{4\pi}\int (1-\cos\theta) |v(\theta)|^{2} do, \quad (15)$$

where $\tau_{\rm tr}$ is the transport time between collisions.

It follows from (15) that in the first approximation in ν/ω_0 , in normal metals, only single-photon absorption is possible. Therefore in this approximation multiphoton absorption is connected entirely with the specifics of the superconducting state. We note also that only in the linear approximation is the absorption expressed in terms of $\tau_{\rm tr}$; *n*-photon absorption is obtained from a more complicated integral of the scattering potential. At large *n*, the absorption is determined mainly by the amplitude of the back scattering through an angle $\theta = \pi$.

3. So far we have assumed scattering from the surface to close to specular, which enabled us to reduce the problem of absorption in films to the problem of an infinite semiconductor. In diffuse reflection this is no longer possible. To understand how to proceed in this case, we consider the spatial Fourier component of the Green's function (2)

$$G(\mathbf{p},t,t') = G_0(\mathbf{p},t-t') \exp\left\{-\frac{ie}{mc}\int_{t'} \mathbf{A}(\tau) \mathbf{p} \, d\tau\right\}.$$
 (16)

It is evident that the influence of the homogeneous field reduces simply to an additional dependence of the phase of the wave function on the time. In diffuse reflection from the surface, however, the phase synchronism is lost after a time d/v_x where v_x is the phase velocity of the wave in a direction perpendicular to the film. This means that in (16) the difference t - t' cannot exceed d/v_x , and the times that are significant for absorption are $t - t' \approx \omega_0^{-1}$. In the case of diffuse reflection from the surface the expression (16) can therefore be replaced by

$$G(\mathbf{p},t,t') = G_{\mathbf{o}}(\mathbf{p},t-t') \exp\left\{-\frac{ie}{mc}\int_{t'}^{t} \mathbf{A}(\tau) \mathbf{p}S(\mathbf{p}) d\tau\right\},$$
 (16')

where the function $S(\mathbf{p})$ is equal to unity at $p_{\mathbf{z}}/p < d\omega_0/\nu_0$ and to zero in the opposite case.

Expression (16) corresponds to the fact that we disregard the weak influence of the field on those electrons that collide with the boundary during a time on the order of ω_0^{-1} . Expression (16) can be obtained more rigorously in the following manner. It is known that the Green's functions of a superconductor in the absence of a field can be expressed in terms of the single-electron eigenfunctions $\psi_n(\mathbf{r})$ (which we assume to be real), e.g.,

$$G_{0}(x,x') = \sum_{n,m} \int \frac{d\omega}{2\pi} \exp\left[-i\omega(t-t')\right] \psi_{n}(\mathbf{r}) \psi_{m}(\mathbf{r}') \frac{\omega + \xi_{n}}{\omega^{2} - (\varepsilon_{n} - i\delta)^{2}}, \quad (17)$$

where ζ_n is reckoned from the Fermi surface and $\varepsilon_n^2 = \zeta_n^2 + \Delta^2$. In first-order approximation in the field the correction to $G_0(x, x')$ takes the form

$$\delta G(x, x') = -\frac{e}{mc} \sum_{n, m, \omega_0} \int \frac{d\omega}{2\pi} \psi_n(\mathbf{r}) \psi_m(\mathbf{r}') \mathbf{A}(\omega_0) \mathbf{p}_{nm}$$

$$\times \frac{\omega(\omega - \omega_0) + \xi_n \xi_m + \Delta^2 + \omega \xi_m + \xi_n(\omega - \omega_0)}{[\omega^2 - (\varepsilon_n - i\delta)^2][(\omega - \omega_0)^2 - (\varepsilon_m - i\delta)^2]} \exp\{-i\omega(t - t') + i\omega_0 t'\}, \quad (17')$$

where $\mathbf{A}(\omega_0)$ is the Fourier component of the field and \mathbf{p}_{nm} is the matrix element of the momentum operator.

Summation in (17') over all states with given energy ζ_n and ζ_m can be carried out with the aid of a quasiclassical method.^[9,10] The product $\psi_n(\mathbf{r})\psi_m(\mathbf{r}')$ will be represented in the form of a matrix element of the operator $f(\mathbf{\hat{r}}) = \delta(\mathbf{\hat{r}} - \mathbf{r}') \exp[i\mathbf{\hat{p}}(\mathbf{r} - \mathbf{r}')]$:

$$\psi_n(\mathbf{r})\psi_m(\mathbf{r}')=f_{mn}.$$

We then have in the quasiclassical approximation^[10]

$$\mathbf{J}(\mathbf{r},\mathbf{r}') = \sum_{m}' \psi_{n}(\mathbf{r}) \psi_{m}(\mathbf{r}') \mathbf{p}_{nm}$$

$$\frac{1}{2\pi v'} \int_{-\infty}^{\infty} dt \exp\left[-i(\zeta_{n} - \zeta_{m})t\right] \langle n|\hat{\mathbf{p}}(t)f(\hat{\mathbf{r}}(0))|n\rangle, \qquad (18)$$

where the summation over *m* is carried out over all states with given energy, $\nu' = mp_0/2\pi^2$, and the integral with respect to time is taken along a classical electron trajectory that goes through the point with coordinate r' at t = 0.

The summation over all n with a given energy means summation over all such trajectories. In the case of diffuse reflection, the momentum p(t) after collision with the wall will have equally probable directions, and only the section of the trajectory bounded by the two collisions closest to the instant t=0 will contribute to the integral in (18). (We assume that $A_z=0$ and therefore $p_z=0$ in (18).) Calculating the integral with respect to time, we obtain

$$\mathbf{J}(\mathbf{r},\mathbf{r}') = \sum \exp\left[i\mathbf{p}\left(\mathbf{r}-\mathbf{r}'\right) + \frac{iz'}{v_z}(\zeta_n - \zeta_m)\right] \delta_{v_z}\left(\zeta_n - \zeta_m\right)$$
$$\times \delta_{v_z}\left(x\right) = \frac{1}{\pi v' x} \sin\left(\frac{dx}{2|v_z|}\right), \tag{19}$$

where it is assumed that the film is bounded by the planes $z = \pm d/2$ and $v_g = p_g/m$.

In the case of specular reflection, the integral in (18) extends over the entire time axis, and $\delta_{ve}(\zeta_n - \zeta_m)$ in (19) is replaced by $\delta(\zeta_n - \zeta_m)$. Then expression (17') coincides with $\delta G(x, x')$ for an infinite superconductor and the summation of all orders in the field yields, obviously, (16). Since the significant values are $\zeta_n - \zeta_m \approx \omega_0$, then the function $\delta_{vs}(\zeta_n - \zeta_m)$ for momenta **p** such that $\omega_0 d/v_z > 1$ can be replaced by a δ -function, and the Green's function in the field acquires the form (16). At $\omega_0 d/v_s < 1$ the influence of the field on the Green's function is negligible, since $\delta_{vs}(\omega_0)$ is small. It is this which is expressed in (16'). The character of the reflection depends generally speaking on the incidence angle. In rough approximation it can be assumed that there exists a certain incidence angle θ_0 such that at $\theta > \theta_0$ the reflection is specular and at $\theta < \theta_0$ it is diffuse. With this reflection law we can assume in (16') that $S(\mathbf{p})$ is equal to unity when

 $p_z/p < \alpha = \max \{\cos \theta_0, d\omega_0/v_0\}$

and to zero in the opposite case.

The expression for the linear-in-concentration correction to the Green's function coincides fully with (4)if we substitute in the argument of the Bessel function in (6)

$$\beta(\mathbf{A}_{0}\mathbf{p})S(\mathbf{p}) - \beta(\mathbf{A}_{0}, \mathbf{p}-\mathbf{q})S(\mathbf{p}-\mathbf{q}) = \beta \widetilde{\mathbf{A}}_{0}\mathbf{q}.$$
(20)

It follows therefore that for the current density we obtain a formula that coincides with (11) if we replace in the latter A_0 by \tilde{A}_0 and calculate the integral with respect to φ under the condition

$$\cos\theta = \cos \mathbf{pq} = q/2p_0.$$

However, the remaining integrals cannot be calculated in the general case and we consider the case of small α .

It follows from geometrical considerations that

$$p_{z}/p = \cos \theta \cos \theta_{1} - \sin \theta \sin \theta_{1} \sin \varphi,$$

where θ_1 is the angle between the normal to the film (the z axis) and the vector **q**. As $\alpha - 0$, the function $S(\mathbf{p})$ differs from zero for angles $\varphi_0 - \delta \varphi < \varphi < \varphi_0 + \delta \varphi$ such that

$$\sin \varphi_{\bullet} = \frac{q}{(4p_{\bullet}^{2}-q)^{\frac{1}{2}}} \operatorname{ctg} \theta_{i}, \quad \delta \varphi < \min \left\{ 1, \frac{\alpha}{(\sin^{2} \theta_{i} - q^{2}/4p_{\bullet}^{2})^{\frac{1}{2}}} \right\}, \quad (21)$$

$$\mathbf{A}_{0}\mathbf{p} = \frac{p_{0}}{q\sin^{2}\theta_{1}} \left\{ \frac{\mathbf{q}}{2p_{0}} (\mathbf{A}_{0}\mathbf{q}) \pm [\mathbf{A}_{0} \times \mathbf{q}]_{z} \left(\sin^{2}\theta_{1} - \frac{q^{2}}{4p_{0}^{2}} \right)^{\frac{1}{2}} \right\} = \mathbf{A}_{0}^{\pm}\mathbf{q}; \quad (22)$$

at $\varphi = \varphi_0$ the plus and minus signs correspond to the two values of the angles φ_0 in (21). Analogously, $S(\mathbf{p} - \mathbf{q})$ differs from zero at

 $\sin \varphi_0 = -q \operatorname{ctg} \theta_1 / (4p_0^2 - q^2)^{\frac{1}{2}}$

and $\mathbf{A}_0 \cdot (\mathbf{p} - \mathbf{q})$ is simply equal to (22) with the sign reversed. Therefore after integrating with respect to $d\varphi$ we obtain

$$j_{i} = \frac{\alpha e N m}{2\pi^{2} \omega_{0}} \sum_{n} \tilde{R}_{n} \int \frac{d\mathbf{q}}{(2\pi)^{3}} \frac{\mathbf{q} | v(q) |^{2}}{q} \{ J_{n} (\beta \mathbf{A}_{0}^{+} \mathbf{q}) \\ \times J_{i-n} (-\beta \mathbf{A}_{0}^{+} \mathbf{q}) + J_{n} (\beta \mathbf{A}_{0}^{-} \mathbf{q}) J_{i-n} (-\beta \mathbf{A}_{0}^{-} \mathbf{q}) \} (\sin^{2} \theta_{i} - q^{2}/4 p_{0}^{2})^{-1/i}.$$
(23)

Let the vector **A** have only one component $A_x = A_0$. It is easily seen that

$$\mathbf{A_0}^+ \mathbf{q} = A_0 p_0 \cos(\varphi_1 - \varphi_0'), \quad \mathbf{tg} \, \varphi_0' = \left(\frac{4 p_0^2}{q^2} \sin^2 \theta_1 - 1\right)^{\frac{1}{2}},$$

 φ_1 is the azimuthal angle in a spherical coordinate system with z axis normal to the film. A similar expression is valid also for $\mathbf{A}_0 \cdot \mathbf{q}$. Therefore the integral with respect to $d\varphi_1$ in (23) of the functions that depend on φ_1 yields the factor

$$2\cos\varphi_0'\int_0^{2\pi}d\varphi_1\cos\varphi_1J_n(\beta A_0p_0\cos\varphi_1)J_{1-n}(-\beta A_0p_0\cos\varphi_1).$$
(24)

The integration with respect to θ_1 is now trivial, and after a number of transformations, which have already been carried out in connection with (13), we obtain ultimately

$$\operatorname{Im} j_{i} = \frac{3}{\pi} \sum_{n \geq 0} \frac{e N_{0} \alpha n \operatorname{Im} \overline{R}_{n}}{\omega_{0} \tau_{ir} \beta A_{0} p} \int_{0}^{2\pi} J_{n}^{2} (\beta A_{0} p_{0} \cos \varphi) d\varphi.$$
(25)

We see that the answer contains only τ_{tr} , and for a normal metal in the case of diffuse reflection only single-photon absorption is possible, just as in specular reflection.¹⁾ The parameter in the case of completely diffuse reflection is proportional to the film thickness. On the other hand, if there exists an incidence-angle region at which the reflection is close to specular, then for small film thicknesses the dependence of j_1 on ddrops out. We note also that in the linear approximation in the field the absorption will take place also in the absence of impurities, as follows from (17').

The current density j_1 obtained from (17') turns out to be proportional to the film thickness d. In the higherorder approximations in the field there appear, obviously, powers of d. Thus, for two-photon absorption j_1 will be proportional to d^3 , since three functions $\delta_{vz}(\zeta_n - \zeta_m)$ appear.

Expression (25) yields the first power of d for n-photon absorption. Therefore at large n and small thicknesses the effect considered by us is the principal one, all the more because in specular reflection, for large incidence angles, j_1 may in general be independent of the film thickness. The function

$$f(y) = \frac{1}{y^2} \sum_{n>0} n \operatorname{Im} \tilde{R}_n \int_0^{\pi/2} J_n^2(y \cos \varphi) \, d\varphi,$$
 (26)

where $y = eA_0p_0/mc\omega_0$, which is proportional to the dissipative part of the conductivity of the film, as seen from (25), is shown in Fig. 1 at $2\Delta/\omega_0 = 20.5$. It is seen that f(y) undergoes a sharp rise at $y \approx 2\Delta/\omega_0$ and rapidly reaches its limiting value $\sim \pi/8$, which coincides with the value of f(y) at $\Delta = 0$. This indicates that at large field amplitudes the influence of the gap on the absorption is negligible.

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The conductivity of thin superconducting films in a strong high-frequency field was investigated by Rose and Sherril, ^[11] who found that the dependences of dissipative conductivity on the amplitude is a curve with a maximum. The films investigated by them, however, do not satisfy apparently the condition $\nu \ll \omega_0$, so that the theoretical experimental result may not be in agreement in ^[11].

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¹⁾We disregard here the possible multiphoton absorption connected with multiple collisions with the walls, which is proportional to higher powers of d (see below).

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Effect of hybridization on the phase transition to the state of an excitonic ferromagnet

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A phase transition to the state of an excitonic ferromagnet {B. A. Volkov and Yu. V. Kopaev, Pis'ma Zh. Eksp. Teor. Fiz. **19**, 168 (1974) [JETP Lett. **19**, 104 (1974)]; Volkov, Kopaev, and A. I. Rusinov, Zh. Eksp. Teor. Fiz. **68**, 1899 (1975) [Sov. Phys. JETP **41**, 952 (1975)] is considered within the framework of the electron spectrum model previously proposed for semiconductors of the IV-VI group {Volkov and Kopaev, Zh. Eksp. Teor. Fiz. **64**, 2184 (1973) [Sov. Phys. JETP **37**, 1103 (1973)]}. It is shown that the transition may be one of either first or second order, depending on the degree of doping and the relation between interelectron interaction constants. Allowance for hybridization does not violate the symmetry between the singlet and triplet pairing, and the phase transition occurs only at finite coupling constants. The model can by used to explain the magnetic properties of some doped narrow-band semiconductors.

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1. INTRODUCTION

It is known^[1] that a system with a single-electron spectrum consisting of two inverted overlapping bands whose extrema lie at one point of the Brillouin zone is unstable to electron-hole pairing that leads to the appearance of a gap in the electron spectrum and to a transition to the state of an excitonic dielectric (ED). Depending on the ratio of the values of the electron-phonon and nucleon interactions, the electron-hole pairing can be realized either in a singlet or in a triplet state. If the constant of the effective interaction that is responsible for the pairing in the singlet state exceeds the corresponding value for the triplet pairing, then the instability leads to the appearance of ED in the singlet state, this being accompanied by the appearance of a charge-density wave and consequently by a structural transformation.^[2] In the opposite case, the system goes

over into an ED in the triplet state; this produces a spin-density wave and the new phase is antiferromagnetic.

As shown by Volkov and Kopaev, ^[4] for a doped ED, at definite ratios of the interelectron interaction constants, the triplet and singlet pairings can coexist. This lifts the spin degeneracy and leads to the appearance of a spontaneous magnetic moment, inasmuch as at nonzero doping the number of electrons is not equal to the number of holes. This phenomenon is called excitonic ferromagnetism (EF) and has been investigated in detail by Volkov, Kopaev, and Rusinov.^[5]

The EF model was proposed to explain the magnetic properties of certain narrow-band semiconductors, as well as metals of the iron group. The phase transition into the ED in IV-VI semiconductors (compounds of the