Influence of sample surface on nonlinear cyclotron resonance

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Second-harmonic generation by incidence of an electromagnetic wave of frequency ω on a metal situated in a magnetic field parallel to its surface is considered under conditions of the anomalous skin effect. The case of diffuse reflection of the electrons by the surface is investigated, as well as the case of near-specular reflection. In diffuse reflection, the second-harmonic amplitude has singularities at the frequencies $\omega = 1/2l\Omega$, where Ω is the cyclotron frequency and l is an integer. In the case of almost specular reflection, the cyclotron resonance becomes weaker, and the amplitude of the second harmonic depends substantially on the surface state of the sample (i.e., on the specularity parameter).

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We consider nonlinear reflection of an electromagnetic wave, at the second-harmonic frequency, from a metal situated in a magnetic field parallel to the metal surface, in the case of the anomalous skin effect, when the following inequalities are satisfied

$$\delta/v_F\tau\ll1,\quad \delta\omega/v_F\ll1,\quad \delta/r_H\ll1, \tag{1}$$

where δ is the depth of the skin layer, v_F is the Fermi velocity, r_H is the Larmor radius, τ is the relaxation time, and ω is the frequency of the wave. We assume a non-quantizing magnetic field H, but one that can be classically strong, i.e., the inequality

$$\Omega\tau\gg1 \tag{2}$$

may be satisfied, where Ω is the cyclotron frequency.

As shown earlier,[1] the magnetic field increases in this case the coefficient of nonlinear reflection by many orders. This uncovers a possibility of experimentally investigating second-harmonic generation in the microwave range in typical metals with free-carrier densities $n\sim10^{23}$, in which nonlinearity in the absence of the magnetic field is extremely small. In Ref. 1 was considered diffuse reflection of the electrons by the surface. In the present paper we study both diffuse and near-specular reflection. In the latter case it is assumed, however, that surface scattering of grazing electrons predominates, i.e., that the following inequality holds

$$1-p\gg\left|\frac{\omega+i\tau^{-1}}{\Omega}\right|\left(\frac{\delta}{r_H}\right)^{1/2},$$

where p is the specularity parameter.

In near-specular reflection, owing to the large contribution made to the linear and nonlinear conductivities by the grazing electrons, the amplitude of the reflected second harmonic depends essentially on the state of the surface of the metal, (i.e., on the specularity parameter p), and its oscillations, which are connected with cyclotron resonance are small. It is also shown that the value of the specularity parameter can be determined by measuring the oscillations of the phase of the second harmonic.

In the case of the diffuse reflection, we analyze in greater detail the form of the resonance line and refine some earlier results.[1] The point is that in Ref. 1 we calculated the nonlinear response of an unbounded metal, and allowance for the reflection of the electrons by the surface was reduced only to a certain modification of the linear and nonlinear conductivities. In the linear theory this approach is justified if we are not interested in factors close to unity.[2,3] In our case, however, it yields an incorrect behavior of the amplitude of the second harmonic in an odd resonance $\omega=(l+\frac{1}{2})\Omega$ as $\Omega\tau\to\infty$. At even resonance $\omega=l\Omega$, the asymptotic relation obtained in this manner for the second-harmonic amplitude as a function of the parameter $\Omega\tau$ turns out to be correct.

The first experiment on the observation of nonlinear cyclotron resonance in bismuth was recently performed by Leviev, Ikonnikov, and Gantmakher.[4]

NONLINEAR CONDUCTIVITY OF SECOND ORDER FOR A SEMI-INFINITE CONDUCTOR IN A MAGNETIC FIELD

Let the conductor occupy the half-space $y>0$, and let the magnetic field H be directed along the z axis. We calculate the second-harmonic nonlinear current produced by the electromagnetic field $\mathbf{E}(y,t)$, $\mathbf{H}(y,t)$:

$$\mathbf{E}(y,t)=e^{-i\omega t}\mathbf{E}(y)+\text{c.c.},\quad \mathbf{H}(y,t)=e^{-i\omega t}\mathbf{H}(y)+\text{c.c.} \tag{3}$$

We write down the kinetic equation for the electron distribution function:

$$\frac{\partial f}{\partial t}+v_y\frac{\partial f}{\partial y}+\frac{d\varphi}{dt}\frac{\partial f}{\partial\varphi}+e\mathbf{E}(y,t)\mathbf{v}\frac{\partial f}{\partial\varepsilon}+e\left(E_z(y,t)+\frac{1}{c}[\mathbf{v}\times\mathbf{H}(y,t)]_z\right)\frac{\partial f}{\partial p_z}=-\frac{f-f_0}{\tau}, \tag{4}$$

where φ is the angle variable, ε is the energy, p_z is the projection of the momentum on the z axis, f_0 is the equilibrium distribution function, and e is the electron charge. For simplicity we confine ourselves to the case of an isotropic and quadratic electron dispersion law. Then

$$\frac{d\varphi}{dt}=\Omega+\left(e\mathbf{E}(y,t)+\frac{e}{c}[\mathbf{v}\times\mathbf{H}(y,t)]\right)\frac{\partial\varphi}{\partial\mathbf{p}}, \tag{5}$$

where $\Omega=|eH/mc|$ is the cyclotron frequency

$$\frac{\partial\varphi}{\partial\mathbf{p}}=-\frac{1}{H}\frac{[\mathbf{v}\times\mathbf{H}]}{mv_\perp^2}\quad v_\perp^2=v_x^2+v_y^2. \tag{6}$$

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We write the boundary condition for the distribution function f in the form

$$f(v_y>0, v_x, v_z, y=0)=pf(-v_y, v_x, v_z, y=0)+(1-p)f_0. \quad (7)$$

We shall solve the kinetic equation by an iteration method. In the linear approximation

$$(-i\omega+\tau^{-1})f_1+v_y\frac{\partial f_1}{\partial y}+\Omega\frac{\partial f_1}{\partial \varphi}=-e\mathbf{v}\mathbf{E}(y)\frac{\partial f_0}{\partial \varepsilon}. \quad (8)$$

The solution of Eq. (8) can be easily obtained by the method of characteristics. It can be represented in the form

$$f_1=-\frac{e}{\Omega}\frac{\partial f_0}{\partial \varepsilon}e^{-\beta_1\varphi}\left[1-p\exp\left(-2\beta_1\left(\frac{\pi}{2}-\varphi_0\right)\right)\right]^{-1}$$
$$\times\left\{\int_{\varphi_0}^{\varphi}d\varphi'+p\exp\left(-2\beta_1\left(\frac{\pi}{2}-\varphi_0\right)\right)\int_{\varphi}^{\pi-\varphi_0}d\varphi'\right\}\exp(\beta_1\varphi')$$
$$\times\mathbf{v}(\varphi')\mathbf{E}(y-r_H(\sin\varphi-\sin\varphi')), \quad (9)$$

At $y_0<r_H$ and

$$f_1=-\frac{e}{\Omega}\frac{\partial f_0}{\partial \varepsilon}[e^{2\pi\beta_1}-1]^{-1}\int_{\varphi}^{\varphi+2\pi}d\varphi'\exp(\beta_1(\varphi'-\varphi))$$
$$\times\mathbf{v}(\varphi')\mathbf{E}(y-r_H(\sin\varphi-\sin\varphi')), \quad (10)$$

at $y_0>_H$. Here

$$r_H=v_\perp/\Omega,\quad y_0=y-r_H\sin\varphi,$$
$$\varphi_0=-\arcsin\frac{y_0}{r_H},\quad \beta_1=-i\frac{\omega+i\tau^{-1}}{\Omega}.$$

In the next approximation there appears an increment to the distribution function at the second harmonic frequency, $f_2\exp(-2i\omega t)+$c.c.. For the function f_2 we obtain

$$(-2i\omega+\tau^{-1})f_2+v_y\frac{\partial f_2}{\partial y}+\Omega\frac{\partial f_2}{\partial \varphi}=-e\left(\mathbf{E}(y)+\frac{1}{c}[\mathbf{v}\times\mathbf{H}(y)]\right)\frac{\partial \varphi}{\partial \mathbf{p}}\frac{\partial f_1}{\partial \varphi}$$
$$-e\left(E_z(y)+\frac{1}{c}[\mathbf{v}\times\mathbf{H}(y)]_z\right)\frac{\partial f_1}{\partial p_z}-e\mathbf{E}(y)\mathbf{v}\frac{\partial f_1}{\partial \varepsilon}. \quad (11)$$

Solving Eq. (11), we can calculate the nonlinear current at the second-harmonic frequency $\mathbf{j}^{(2)}(y,t)$ and obtain an expression for the nonlinear conductivity tensor $Q_{\alpha\beta\gamma}(k,k_1,k_2)$, defined in the following manner:

$$j_\alpha^{(2)}(k)=\int_0^\infty dk_1\,dk_2\,Q_{\alpha\beta\gamma}(k,k_1,k_2)E_\beta(k_1)E_\gamma(k_2), \quad (12)$$

where

$$E_\alpha(k)=\frac{1}{\pi}\int_0^\infty dy\,E_\alpha(y)\cos ky,$$

$$j_\alpha^{(2)}(k)=\frac{1}{\pi}\int_0^\infty dy\,j_\alpha^{(2)}(y)\cos ky,$$

$$j_\alpha^{(2)}(y,t)=e^{-2i\omega t}j_\alpha^{(2)}(y)+\text{c.c.}$$

In our case it turns out that the main contribution to the nonlinearity is due to the Lorentz force. Leaving out the very cumbersome calculations, we present only the asymptotic expressions for the nonlinear conductivity tensor $Q_{\alpha\beta\gamma}(k,k_1,k_2)$ in diffuse reflection and in near-specular reflection.

If the reflection is diffuse ($p=0$) and

$$\left|\frac{\omega+i\tau^{-1}}{\Omega}\right|\ll\left(\frac{r_H}{\delta}\right)^{1/2},\quad \omega\tau\ll\left(\frac{r_H}{\delta}\right)^{1/2},$$

then we obtain for the nonlinear conductivity

$$Q_{\alpha\beta\gamma}(k,k_1,k_2)=i\frac{c}{\omega H}\frac{e^2p_F^2}{2\pi^2}b_{\alpha\beta\gamma}$$
$$\times[K(k,k_1,k_2)\operatorname{cth}\beta_2\pi+L(k,k_1,k_2)\operatorname{cth}\beta_1\pi+N(k,k_1,k_2)], \quad (13)$$

where $\beta_2=-i(2\omega+i\tau^{-1})/\Omega$, p_F is the Fermi momentum; the function $K(k,k_1,k_2)$ is defined by the expression

$$K(k,k_1,k_2)=\frac{1}{4k^{1/2}}\left[\frac{(k_1+k_2)^{1/2}}{k_1+k_2-k}+\frac{|k_1-k_2|^{1/2}}{|k_1-k_2|-k}\right], \quad (14)$$

the function $L(k,k_1,k_2)$ is of the form

$$L(k,k_1,k_2)=\frac{k_1^{1/2}}{4}\left[\frac{1}{(k+k_2)^{1/2}(k+k_2-k_1)}+\frac{1}{|k-k_2|^{1/2}(|k-k_2|-k_1)}\right] \quad (15)$$

and finally

$$N(k,k_1,k_2)=-\frac{3+e^{-2\beta_2\pi}}{8}\left(\frac{1}{k-k_1-k_2}+\frac{1}{k-|k_1-k_2|}\right)$$
$$+\frac{3+e^{-2\beta_1\pi}}{8}\left(\frac{1}{k+k_2-k_1}+\frac{1}{|k-k_2|-k_1}\right)$$
$$+\frac{1+e^{-2\beta_1\pi}}{8}\left[\frac{k_1^{1/2}}{(k+k_2)^{1/2}(k_1-k-k_2)}+\frac{k_1^{1/2}}{|k-k_2|^{1/2}(k_1-|k-k_2|)}\right]$$
$$-\frac{1+e^{-2\beta_2\pi}}{8}\left[\frac{(k_1+k_2)^{1/2}}{k^{1/2}(k_1+k_2-k)}+\frac{|k_1-k_2|^{1/2}}{k^{1/2}(|k_1-k_2|-k)}\right]. \quad (16)$$

The tensor $b_{\alpha\beta\gamma}$ in (13) is symmetrical in all the indices α, β, and γ, with $b_{xxx}=b_{xzz}=b_{zxz}=b_{zzx}=1$, and all its remaining components are equal to zero.

As seen from (13), the nonlinear response exhibits singularities of the cyclotron resonance at the frequencies $\omega=l\Omega$ (even resonance) and $\omega=(l+\frac{1}{2})\Omega$ (odd resonance), where l is an integer. We note that satisfaction of the condition $\omega\tau\ll(r_H/\delta)^{1/2}$, which ensures the applicability of expression (13), is possible only near the resonance $\omega=l\Omega$.

If we disregard the reflection of the electrons by the surface of the sample, and assume that the fields are excited in an infinite medium by a current sheath, then the nonlinear conductivity $Q_{\alpha\beta\gamma}(k,k_1,k_2)$ can also be represented in the form (13). We must put here $N\equiv0$,

$$K(k,k_1,k_2)=\frac{k_1+k_2}{(k_1+k_2)^2-k^2}+\frac{|k_1-k_2|}{(k_1-k_2)^2-k^2},$$
$$L(k,k_1,k_2)=\frac{k_1+k_2}{k^2-(k_1+k_2)^2}.$$

On the other hand, if the reflection is close to specular and the following inequalities hold

$$|\beta_{1,2}|(\delta/r_H)^{1/2}\ll1-p\ll|\operatorname{th}\beta_{1,2}\pi|,\ 1, \quad (17)$$

then the asymptotic expression for $Q_{\alpha\beta\gamma}(k,k_1,k_2)$ takes the form

$$Q_{\alpha\beta\gamma}(k,k_1,k_2)=i\frac{c}{\omega H}\frac{e^2p_F^2}{2\pi^2}b_{\alpha\beta\gamma}\frac{1}{1-p}\left[P(k,k_1,k_2)\right.$$
$$\left.+\frac{1-p}{2}R(k,k_1,k_2)\operatorname{cth}\beta_2\pi+\frac{1-p}{2}S(k,k_1,k_2)\operatorname{cth}\beta_1\pi\right], \quad (18)$$

where the function $P(k,k_1,k_2)$ is defined by

$$P(k,k_1,k_2)=-\frac{1}{2}\left[\frac{1}{k-k_1-k_2}+\frac{1}{k-|k_1-k_2|}\right.$$
$$\left.+\frac{1}{k_1-k-k_2}+\frac{1}{k_1-|k-k_2|}\right], \quad (19)$$

$R(k.k_1,k_2)$ is given by

$$R(k,k_1,k_2)=-\frac{1}{2}\left[\frac{1}{k-k_1-k_2}+\frac{1}{k-|k_1-k_2|}\right], \quad (20)$$

and the function $S(k,k_1,k_2)$ is given by

$$S(k,k_1,k_2)=-\frac{1}{2}\left[\frac{1}{k_1-k-k_2}+\frac{1}{k_1-|k-k_2|}\right]. \qquad (21)$$

As seen from the foregoing formulas, the kernel $Q_{\alpha\beta\gamma}(k,k_1,k_2)$ has singularities of the type $k\pm k_1\mp k_2)^{-1}$. In the calculation of the Fourier component of the nonlinear current, these singularities should be integrated in the sense of the principal value.

NONLINEAR REFLECTION FROM THE SURFACE OF A METAL

Assume that an electromagnetic wave is normally incident on a metal occupying the half-space $y>0$. We calculate the amplitude of the reflected electromagnetic wave at the second-harmonic frequency. We consider first the polarization properties of the reflected second harmonic. Since the law governing the penetration of the field into an isotropic metal, both in diffuse reflection and in near-specular reflection, is independent in the linear approximation of the polarization of the field,[5,6] it follows that the polarization of the electric field of the reflected second harmonic coincides with the polarization of the nonlinear current $j_\alpha^{(2)}(y)$. If the incident wave is linearly polarized then, as seen from (13) and (18) and from the explicit expression for the tensor $b_{\alpha\beta\gamma}$, the reflected second harmonic also has nonlinear polarization.

Let the incident wave be linearly polarized, and let ψ_1 be the angle between the electric field of the incident wave and the constant magnetic field. We specify similarly the polarization of the reflected second harmonic by means of the angle ψ_2. It is easy to establish that the angles ψ_1 and ψ_2 are connected by the relation

$$\operatorname{tg}\psi_2={}^1/_2(\operatorname{tg}\psi_1+1/\operatorname{tg}\psi_1). \qquad (22)$$

It follows from (22), in particular, that $|\psi_2|>\pi/4$ at all ψ_1 we assume for the sake of argument that ψ_1 and ψ_2 lie in the interval $(-\pi/2,\pi/2)$]. If the electric field of the incident wave is parallel to the z axis ($\psi_1=0$) or to the x axis ($\psi_1=\pi/2$), then the electric field of the second harmonic is polarized along the x axis ($\psi_2=\pi/2$).

The equation for the second-harmonic field $E^{(2)}(y)$ is of the form

$$-\frac{d^2E^{(2)}}{dy^2}=\frac{8\pi i\omega}{c^2}(j^{(2)}(y)+\hat\sigma(2\omega)E^{(2)}), \qquad (23)$$

where $\hat\sigma(2\omega)$ is the operator of the linear conductivity at the frequency 2ω. We have left out of (23) certain indices, since the operator $\hat\sigma_{\alpha\beta}$ is proportional to the unit tensor $\delta_{\alpha\beta}$ both in diffuse reflection and in near-specular reflection.

Equation (23) should be solved with the boundary condition

$$\left.\frac{dE^{(2)}}{dy}\right|_{y=+0}=-\frac{2i\omega}{c}\left.E^{(2)}\right|_{y=0}. \qquad (24)$$

We continue the solution of Eq. (23) into the half-space $y<0$ is even fashion. We then get in place of (23)

$$-\frac{d^2E^{(2)}}{dy^2}=\frac{8\pi i\omega}{c^2}(j^{(2)}(|y|)+\hat\sigma(2\omega)E^{(2)})+\frac{4i\omega}{c}E^{(2)}(0)\delta(y). \qquad (25)$$

We introduce now the current sheet $I(y,t)=I_0\delta(y)e^{-2i\omega t}$, which produces in the linear approximation a field $\mathscr{E}(y)e^{-2i\omega t}$, with $\mathscr{E}(0)=1$. Applying the reciprocity theorem for the fields $\mathscr{E}^{(2)}(y)$, $\mathscr{E}(y)$ and for the corresponding currents, we obtain the following expression for the amplitude of the reflected second harmonic:

$$E^{(2)}(0)=-\frac{4\pi}{c}\frac{\zeta(2\omega)}{1+\zeta(2\omega)}\int_0^\infty dy\, j^{(2)}(y)\mathscr{E}(y), \qquad (26)$$

where $\zeta(\omega)$ is the surface impedance. The function $\mathscr{E}(y)$ describes in the linear approximation, in accord with the foregoing, the penetration of a field of frequency 2ω into a metal.

Inasmuch as in our case $|\zeta|\ll 1$ we have

$$E^{(2)}(0)=-\frac{4\pi}{c}\zeta(2\omega)\int_0^\infty dy\, j^{(2)}(y)\mathscr{E}(y),\quad \mathscr{E}(0)=1 \qquad (27)$$

or

$$E^{(2)}(0)=-\frac{8\pi^2}{c}\zeta(2\omega)\int_0^\infty dk\,dk_1\,dk_2\,Q(k,k_1,k_2)\mathscr{E}(k)E(k_1)E(k_2), \qquad (28)$$

where $\mathscr{E}(k)$ is the cosine-transform of the function $\mathscr{E}(y)$. In the derivation of (28) we have also assumed that the incident wave is polarized parallel or pendicular to the magnetic field. The quantity $Q(k,k_1,k_2)$ in (28) should be taken to mean one of the components Q_{xxx}, $Q_{xzz}(Q_{xxx}\equiv Q_{xzz})$.

There is a known[6,7] asymptotically exact-expression for the functions $E(k)$ and $\mathscr{E}(k)$ in the two situations of interest to us. The kernel $Q(k,k_1,k_2)$ was calculated by us in the preceding section, so that the problem of second-harmonic generation has in principle been solved. However, the Fourier component of the field $E(k)$ is represented in the form of rather complicated contour integrals, so that the exact calculation of the integrals with respect to k, k_1, and k_2 in (28) is quite difficult.

We investigate first the case of diffuse reflection. We represent the function $E(k)$ in the form

$$E(k)=i\frac{\omega}{2c}\frac{E(0)}{\zeta(\omega)}\delta_1^2e_1(k\delta_1), \qquad (29)$$

where $E(0)$ is the electric field of the first harmonic on the surface of the metal, and

$$\delta_1=\delta(\omega)=\delta_0(\omega)\,|1-e^{-2\pi\beta_1}|^{1/3}$$

is the depth of the skin layer at the frequency ω, $\delta_0(\omega)=(c^2e^{-2}p_F^{-2}\omega^{-1})^{1/2}$.

For the function $\mathscr{E}(k)$ we have similarly

$$\mathscr{E}(k)=i\frac{\omega}{c}\frac{1}{\zeta(2\omega)}\delta_2^2e_2(k\delta_2), \qquad (30)$$

where $\delta_2=\delta(2\omega)$ is the depth of the skin layer at the frequency 2ω. Substituting (29) and (30) in the integral equation[5] for $E(k)$, we obtain equations for the functions $e_{1,2}(k)$:

$$k^2e_{1,2}(k)-i\frac{|1-e^{-2\pi\beta_{1,2}}|}{1-e^{-2\pi\beta_{1,2}}}\left(\frac{e_{1,2}(k)}{k}-\int_0^\infty dk'\,\Lambda_{1,2}(k,k')e_{1,2}(k')\right)=-\frac{2}{\pi}, \qquad (31)$$

where

$$\Lambda_{1,2}(k,k')=\frac{(1+e^{-2\pi\beta_{1,2}})^2}{4\pi}\frac{1}{(kk')^{1/2}(k+k')}+\frac{(1-e^{-2\pi\beta_{1,2}})(3+e^{-2\pi\beta_{1,2}})}{2\pi^2}\frac{\ln(k/k')}{k^2-k'^2}. \qquad (32)$$

The dimensionless functions $e_{1,2}(k)$ satisfy the normalization conditions

$$\int_0^\infty dk\, e_1(k) = -i\frac{\zeta(\omega)c}{\delta_1\omega} \sim 1,$$
$$\int_0^\infty dk\, e_2(k) = -i\frac{\zeta(2\omega)c}{2\delta_2\omega} \sim 1. \quad (33)$$

It follows from the solution obtained by the Hartmann and Luttinger[7] that the functions $e_{1,2}(k)$ vanish in power-law fashion as $k\to 0$, and tend to zero in inverse proportion to k^2 as $k\to\infty$. In the region $k\sim 1$ the characteristic values of the functions $e_{1,2}(k)$ are also of the order of unity.

Substituting (13), (29) and (30) in (28), we obtain

$$E^{(2)}(0) = -\frac{1}{H}\frac{e^2p_F^2\omega}{c^2}\frac{\omega}{c}\frac{\delta_1^3\delta_2}{\zeta^2(\omega)}[c_2 \operatorname{cth}\beta_2\pi + c_1\operatorname{cth}\beta_1\pi + c_0](E(0))^2, \quad (34)$$

where

$$c_2 = \int_0^\infty dk\, dk_1\, dk_2\, e_1(k_1)e_1(k_2)e_2(k)K(ak, k_1, k_2), \quad (35)$$
$$c_1 = \int_0^\infty dk\, dk_1\, dk_2\, e_1(k_1)e_1(k_2)e_2(k)L(ak, k_1, k_2),$$
$$c_0 = \int_0^\infty dk\, dk_1\, dk_2\, e_1(k_1)e_1(k_2)e_2(k)N(ak, k_1, k_2),$$
$$a = \delta_1/\delta_2.$$

We assume next that $\Omega\tau \gg 1$. Far from resonance, and also in the case of even resonance $\omega = l\Omega$ we have $a\sim 1$. In formulas (35), in the essential region of integration $k\lesssim 1, k_{1,2}\lesssim 1$, the characteristic values of the integrands are of the order of unity, and consequently $|c_2|\sim|c_1|\sim|c_0|\sim 0.1-1$. Taking this into account we obtain an estimate for the amplitude of the reflected second harmonic[1]:

$$E^{(2)}(0) \sim \frac{1}{H}\frac{c}{\delta_1\omega}(E(0))^2. \quad (36)$$

Inasmuch as δ_1 near the resonance decrease by a factor $(\Omega\tau)^{1/2}$, the amplitude of the reflected second harmonic at resonance increases in proportion to $(\Omega\tau)^{1/3}$ at a fixed value of the first-harmonic field on the surface of the metal, and decreases in proportion to $(\Omega\tau)^{-1/2}$ at a fixed value of the amplitude of the incident wave.

We proceed now to study the odd resonance $\omega = (l + \frac{1}{2})\Omega$. In this case, as seen from (34), to determine the character of the singularity of the amplitude of the reflected second harmonic it is necessary to know the behavior of the coefficient c_2 near resonance. At the resonance $\omega = (l+\frac{1}{2})\Omega$, the parameter $a\gg 1$, so that we need to obtain the asymptotic expression for c_2 at large values of a. From the formulas obtained above it follows that

$$c_2 = \frac{1}{4}\int_0^\infty dk\, dk_1\, dk_2 \frac{e_1(k_1)e_1(k_2)e_2(k)}{(ak)^{1/2}} \times\left[\frac{(k_1+k_2)^{1/2}}{k_1+k_2-ak} + \frac{|k_1-k_2|^{1/2}}{|k_1-k_2|-ak}\right]. \quad (37)$$

We rewrite the integral of (37) as follows:

$$c_2 = -\frac{1}{4a^{3/2}}\int_0^\infty dk_1\, dk_2 \int_0^\infty dk \frac{e_1(k_1)e_1(k_2)e_2(k)}{k^{1/2}} \times\left[\frac{(k_1+k_2)^{1/2}}{k-a^{-1}(k_1+k_2)} + \frac{|k_1-k_2|^{1/2}}{k-a^{-1}|k_1-k_2|}\right] = \frac{F(a)}{a^{3/2}}. \quad (38)$$

The behavior of $F(a)$ as $a\to\infty$ depends on the behavior of the function $e_2(k)$ at small k [we recall that at large k the function $e_{1,2}(k)$ always decreases like k^{-2}]. As follows from the solution of Hartmann and Luttinger,[7] at the exact resonance $\omega = (l+\frac{1}{2})\Omega$ the following asymptotic expansion is valid for the function $e_2(k)$ as $k\to 0$:

$$e_2(k) = dk^\nu + o(k^{1/2}), \quad (39)$$

where $\nu = (2/\pi\Omega\tau)^{1/2}$ and the constant $d\sim 1$. The parameter a in this case is equal to $(\Omega\tau/\pi)^{1/2}$. Taking the foregoing into account, we can verify that the integral $F(a)$ remains finite as $\Omega\tau\to\infty$. Consequently

$$|c_2| \propto a^{-3/2}. \quad (40)$$

Thus, from (34) and (40) we find that the amplitude of the reflected second harmonic at odd resonance increases in proportion to $(\Omega\tau)^{1/6}$.

We note that if we calculate the nonlinear current $j^{(2)}(y)$ by using the nonlinear response of an infinite medium and taking into account the reflection of the electrons by the surface only by modifying somewhat the resonant factors in the linear and nonlinear conductivity (this method is valid in the linear theory[2,3]), then we can find that there is no resonance at the frequency $\omega = (l+\frac{1}{2})\Omega$. In fact, we have for c_2 in this case

$$c_2 = \int_0^\infty dk\, dk_1\, dk_2\, e_1(k_1)e_1(k_2)e_2(k) \times\left[\frac{k_1+k_2}{(k_1+k_2)^2-a^2k^2} + \frac{|k_1-k_2|}{(k_1-k_2)^2-a^2k^2}\right], \quad (41)$$
$$e_{1,2}(k) \propto k/(k^3-i).$$

At large a this yields $c_2 \propto a^{-2}$. We see thus that $E^{(2)}(0)$ remains finite at $\omega = (l+\frac{1}{2})\Omega$. We have shown above, however, that this is not the case, and that $E^{(2)}$ increases at $\omega = (l+\frac{1}{2})\Omega$ in proportion to $(\Omega\tau)^{1/6}$. Near an odd resonance it is therefore necessary to take correct account of the boundary condition for the distribution function even in the case of diffuse reflection.

On the other hand, far from the resonance, $\omega = (l+\frac{1}{2})\Omega$, and also near the even resonance $\omega = l\Omega$, the calculation of the nonlinear current $j^{(2)}(y)$ without allowance for the reflection of the electrons by the surface yields an incorrect estimate of the amplitude of the reflected second harmonic. This is not at all surprising. The point is that when no account is taken of the reflection of the electrons, the obtained estimate for the density of the nonlinear current $j^{(2)}(y)$ is valid only at $y\sim\delta_1$. On the other hand, in the case of the odd resonance $\omega = \delta_2 \ll \delta_1$, the inequality $\delta_2\ll\delta_1$ is satisfied and, as seen from (27), an important factor in the calculation of the amplitude of the reflected second harmonic is the behavior of the nonlinear current $j^{(2)}(y)$ at distances $y\sim\delta_2\ll\delta_1$. To obtain the correct expression for the density of the nonlinear current $j^{(2)}(y)$ at $y\ll\delta_1$ it is necessary, as it turns out, to take correct account of the reflection of the electrons from the sample boundary.

In the foregoing discussion of the cyclotron resonance we have assumed the parameter $\Omega\tau$ to be large. However, satisfaction of the condition $\Omega\tau\gg 1$ is not essential in our case. In fact, the condition for the applicability of the expression obtained by us for the nonlinear conductivity $Q_{\alpha\beta\gamma}(k, k_1, k_2)$ [Eqs. (13)-(16)], and also for the applicability of the expression for the amplitude of the reflected second harmonic (34), which bounds the magnetic field from below, is the following inequality:

$$\left|\frac{\omega+i\tau^{-1}}{\Omega}\right| \ll \left(\frac{r_H}{\delta}\right)^{1/2}. \tag{42}$$

The inequality (42) means that the time required for the electron to pass through the skin layer is much shorter than the free path time or the period of the electromagnetic field. In metals with skin-layer depth $\sim 10^{-5}$ cm, at frequencies $\omega/2\pi \sim 10$ GHz, this inequality is satisfied up to magnetic fields of several dozen oersteds. The parameter $\Omega\tau$ in such fields is quite small.

On the other hand, if the parameter $\Omega\tau$ is not very large, the dependence of the amplitude of the second harmonic on the magnetic field near the resonances $\omega = l\Omega/2$ can differ substantially from that obtained by us on the basis of the asymptotic expansion in the parameter $(\Omega\tau)^{-1}$.

For a more detailed study of the dependence of the amplitude of the second harmonic on the magnetic field it is necessary to calculate the values of c_2, c_1, and c_0, which enter in expression (34). Inasmuch as an analytic calculation of these quantities is hardly possible, we have employed numerical methods. We have not used for the functions $e_{1,2}(k)$ the expressions obtained by Hartmann and Luttinger,[7] but solved the integral equations (31) directly by quadratures. To verify the solutions obtained in this manner we used the normalization conditions (33). After finding the Fourier components $e_{1,2}(k)$, we calculated c_2, c_1, and c_0 as functions of the magnetic field. At $\omega\tau = 5$ and $\omega = \Omega$ we obtained

$$c_2 = 0.075 - i0.132, \quad c_1 = -0.072 + i0.123,$$
$$c_0 = 0.035 - i0.054$$

(the error is approximately 0.003).

Figure 1 shows the power of the reflected second harmonic (in arbitrary units) against the constant magnetic field at a constant amplitude of the first harmonic incident on the sample, i.e., at a constant first-harmonic magnetic-field amplitude on the surface of the metal. The parameter $\omega\tau = 5$. It is seen from the figure that there is a rather large maximum in the odd resonance $\omega = 3\Omega/2$ and a much smaller maximum on the fundamental resonance $\omega = \Omega$. According to the results obtained above, however, which are based on an asymptotic expansion in the parameter $(\Omega\tau)^{-1}$, a minimum of the intensity of the second harmonic should be observed near the fundamental resonance, for in this case the depth of the skin layer and the amplitude of the electric field of the first harmonic on the surface of the metal decrease.

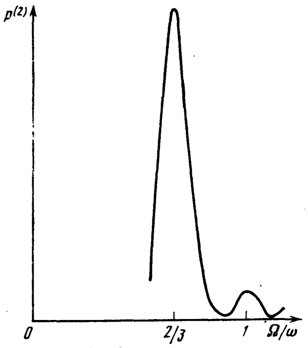

FIG. 1. Dependence of the second-harmonic power on a magnetic field in the case of diffuse reflection at a constant amplitude of the first harmonic incident on the sample. The parameter $\omega\tau = 5$.

The shape of the resonance curve observed in the experiment on bismuth[4] differs substantially from that shown in Fig. 1. Namely, strong spikes of the intensity of the second harmonic appeared in Ref. 4 for the even resonances, and weaker one for the odd resonances. This difference, as noted in Ref. 4, can be attributed, on the one hand, to the fact that the linear theory used by us (in the case of diffuse reflection) yields a stronger change of the surface impedance and of the skin-layer depth than are observed in experiment, and on the other hand to the different values of the parameter $\omega\tau$. We recall that, as shown in Ref. 1, when the inequality $\omega\tau \gg (r_H/\delta)^{1/2}$ is satisfied the nonlinearity has a much stronger singularity in the even resonance than in the odd one. This situation will be considered by us separately.

We proceed now to the case of near-specular reflection, i.e., we assume that the inequalities (17) are satisfied. In this situation the main contribution to the linear and nonlinear conductivity is made by electrons that graze along the surface.

We represent the Fourier component $E(k)$ of the first-harmonic field in the form

$$E(k) = i\frac{\omega}{2c}\frac{E(0)}{\zeta(\omega)}\delta_0^2(\omega)e_1(k\delta_0(\omega)). \tag{43}$$

For the surface impedance $\zeta(\omega)$ we have according to Kaner *et al.*[6]

$$\zeta(\omega) = \frac{\sqrt{3}\,\omega\delta_0(\omega)}{2c}e^{-\pi i/3}\left(1+\frac{1-p}{2}\coth\beta_1\pi\right)^{-1/2}. \tag{44}$$

The quantity $\delta(\omega)$ is the depth of the skin layer without allowance for the small contribution of the resonant electrons:

$$\delta_0(\omega) = \left[\frac{c^2(1-p)}{e^2 p_F^2 \omega}\right]^{1/3}. \tag{45}$$

Substituting (43) in the integral equation for $E(k)$,[6] we obtain an integral equation for $e_1(k)$. It is of the form

$$k^2 e_1(k) - i\left(1+\frac{1-p}{2}\coth\beta_1\pi\right)\left(\frac{e_1(k)}{k} - \frac{2}{\pi^2}\int_0^\infty dk'\frac{\ln(k/k')}{k^2-k'^2}e_1(k')\right) = -\frac{2}{\pi}. \tag{46}$$

We expand the function $e_1(k)$ in powers of the small parameter $\frac{1}{2}(1-p)\coth\beta_1\pi$. Confining ourselves to the first two terms, we have

$$e_1(k) = e_0(k) + \frac{(1-p)\coth\beta_1\pi}{2}\tilde{e}(k). \tag{47}$$

In the zeroth order in the parameter $\frac{1}{2}(1-p)\coth\beta_1\pi$ we get from (46) an equation for $e_0(k)$:

$$k^2 e_0(k) - i\frac{e_0(k)}{k} + \frac{2i}{\pi^2}\int_0^\infty dk' \frac{\ln(k/k')}{k^2-k'^2} e_0(k') = -\frac{2}{\pi}. \quad (48)$$

Taking into account the terms of first order of smallness in the resonance parameter, we obtain an equation for $\bar{e}(k)$:

$$k^2\bar{e}(k) - i\frac{\bar{e}(k)}{k} + \frac{2i}{\pi^2}\int_0 dk' \frac{\ln(k/k')}{k^2-k'^2}\bar{e}(k') = \frac{2}{\pi} + k^2 e_0(k). \quad (49)$$

From (48) and (49) we see that the functions $e_0(k)$ and $\bar{e}(k)$ do not depend on the magnetic field or on any other parameters, and are consequently universal functions.

For $\mathscr{E}(k)$ we have similarly

$$\mathscr{E}(k) = i\frac{\omega}{c}\frac{1}{\zeta(2\omega)}\delta_0^2(2\omega)e_2(k\delta_0(2\omega)),$$
$$e_2(k) = e_0(k) + \frac{(1-p)\coth\beta_2\pi}{2}\bar{e}(k). \quad (50)$$

As follows from the solution obtained by Hartmann and Luttinger,[7] the functions $e_{1,2}(k)$ and $e_0(k)$ tend to 0 like $k^{1/2}$ as $k \to 0$, and decrease like k^{-2} as $k \to \infty$. Obviously, the function $\bar{e}(k)$ also decreases like k^{-2} as $k \to \infty$, and tends to 0 no more slowly than $k^{1/2}$ as $k \to 0$. In the region $k \sim 1$ the typical values of the functions $e_0(k)$ and $\bar{e}_0(k)$ are also of the order of unity.

Integrating (43) with respect to k from 0 to ∞ and expanding in powers of the small resonant increment, we easily obtain the normalization conditions for the functions $e_0(k)$ and $e(k)$:

$$\int_0^\infty dk e_0(k) = -3\int_0^\infty dk\bar{e}(k) = -i\frac{\sqrt{3}}{2}e^{-i\pi/3}. \quad (51)$$

Substituting (18), (43), and (50) in (28) and expanding in powers of the small parameters $\frac{1}{2}(1-p)\coth\beta_{1,2}\pi$ we obtain for the amplitude of the reflected second harmonic

$$E^{(2)}(0) = -\lambda\zeta_0(\omega)\frac{(H(0))^2}{H}\left[1+\lambda_1\frac{1-p}{2}\coth\beta_1\pi + \lambda_2\frac{1-p}{2}\coth\beta_2\pi\right], \quad (52)$$

where $H(0)$ is the amplitude of the magnetic field of the first harmonic on the surface of the metal,

$$\zeta_0(\omega) = \frac{\sqrt{3}\,\omega\delta_0(\omega)}{2c}e^{-i\pi/3},$$

and the quantities λ, λ_1, and λ_2 are defined by the following expressions:

$$\lambda = \frac{2^{2/3}}{\sqrt{3}}e^{+i\pi/3}\int_0^\infty dk dk_1 dk_2 P(2^{1/3}k, k_1, k_2)e_0(k)e_0(k_1)e_0(k_2), \quad (53)$$

$$\lambda_1 = \frac{2^{2/3}}{\lambda\sqrt{3}}e^{i\pi/3}\int_0^\infty dk dk_1 dk_2\{S(2^{1/3}k, k_1, k_2)e_0(k)e_0(k_1)e_0(k_2)$$
$$+P(2^{1/3}k, k_1, k_2)e_0(k)\bar{e}(k_1)e_0(k_2) + P(2^{1/3}k, k_1, k_2)e_0(k)e_0(k_1)\bar{e}(k_2)\}, \quad (54)$$

$$\lambda_2 = \frac{2^{2/3}}{\lambda\sqrt{3}}e^{i\pi/3}\int_0^\infty dk dk_1 dk_2\{R(2^{1/3}k, k_1, k_2)e_0(k)e_0(k_1)e_0(k_2)$$
$$+P(2^{1/3}k, k_1, k_2)\bar{e}(k)e_0(k_1)e_0(k_2)\}. \quad (55)$$

By virtue of the foregoing, the quantities λ, λ_1, and λ_2 are constants independent of the magnetic field and of any other parameter of the problem. It is quite difficult to calculate these constants analytically. It is possible to show, however, that λ_1 and λ_2 satisfy the relation

$$\lambda_1 + \lambda_2 = -1/3. \quad (56)$$

In fact, let $\omega = l\Omega$. We rewrite the Fourier components $E(k)$ and $\mathscr{E}(k)$ in a form somewhat different from (43)

$$E(k) = i\frac{\omega}{2c}\frac{E(0)}{\zeta(\omega)}\delta^2(\omega)\tilde{e}_1(k\delta(\omega)),$$
$$\mathscr{E}(k) = i\frac{\omega}{c}\frac{1}{\zeta(2\omega)}\delta^2(2\omega)\tilde{e}_2(k\delta(2\omega)); \quad (57)$$

Here $\delta(\omega)$ is the depth of the skin layer with allowance for the resonant electrons:

$$\delta(\omega) = \delta_0(\omega)(1+{}^1/_2(1-p)\coth\beta_1\pi)^{-1/3}.$$

Since $\omega = l\Omega$, it follows that $\delta(\omega)$ is a real quantity and furthermore $\delta(\omega)/\delta(2\omega) = \delta_0(\omega)/\delta_0(2\omega) = 2^{1/2}$. It is also easy to see that the functions $\tilde{e}_{1,2}(k)$ satisfy the same integral equation as $e_0(k)$, so that $\tilde{e}_1(k) = \tilde{e}_2(k) \equiv e_0(k)$.

Substituting (18) and (57) in (28) and taking also into account the relation

$$P(k, k_1, k_2) = R(k, k_1, k_2) + S(k, k_1, k_2),$$

we obtain for the amplitude of the reflected second harmonic at $\omega = l\Omega$

$$E^{(2)}(0) = -\lambda\zeta(\omega)(H(0))^2/H. \quad (58)$$

Expanding in this formula the impedance $\zeta(\omega)$ in terms of the small resonant increment and comparing the result with (52), we arrive in fact at (56).

To calculate the constants λ, λ_1, and λ_2, we have used numerical methods. Just as in the case of diffuse reflection, we solved first the integral equations (48) and (49) by the method of quadrature formulas, and verified the normalization conditions (51), after which we calculated the integrals (53)–(55). Within the limits of error, all three constants, λ, λ_1, and λ_2 turned out to be real (or more readily, the imaginary parts of λ, λ_1, and λ_2 are exactly equal to zero). As a result we obtained

$$\lambda = 0.054,\ \lambda_1 = 0.25,\ \lambda_2 = -0.58$$

(the relative error is approximately 5%). It is seen that relation (56) is satisfied.

We have thus shown that in the case of near-specular reflection, i.e., when the inequalities (17) are satisfied, the amplitude of the reflected second harmonic is described by the expression (52), and the values of constants λ, λ_1, and λ_2 in (52) are given above. It is seen from (52) that as a result of the large contribution to the linear and nonlinear conductivity of the electrons that glance along the surface, the power of the radiated second harmonic depends substantially on the specularity coefficient and therefore its measurement can yield information on the character of the reflection of the electrons by the surface of the sample. In addition, it follows from (52) that in the principal approximation the second-harmonic power decreases with increasing magnetic field like H^{-2}, and its oscillations, which are connected with cyclotron resonance, are small.

It is also possible to determine the specularity coefficient by measuring the oscillations of the phase of the second harmonic $\Delta\varphi^{(2)}$. In fact, from (52), recognizing that $|\Delta\varphi^{(2)}| \ll 1$, we obtain

$$\begin{aligned} \Delta\varphi^{(2)} &= {}^1/_2(1-p)\chi(\omega\tau, H), \\ \chi(\omega\tau, H) &= \lambda_1 \operatorname{Im}\coth\beta_1\pi + \lambda_2 \operatorname{Im}\operatorname{cth}\beta_2\pi. \end{aligned} \tag{59}$$

From (59) we see that, measuring the oscillations of the phase, we can in principle determine both the specularity coefficient p and the free path time τ. In fact, the relaxation time can be determined from the damping of the oscillations of the phase in the magnetic-field region where the parameter $\Omega\tau$ is not large. After determining τ, we obtain directly also the specularity coefficient. Figure 2 shows the dependence of the phase of the second harmonic on the magnetic field at $\omega\tau=3$.

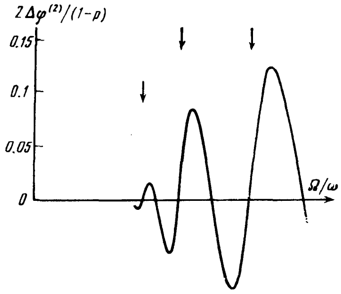

FIG. 2. Oscillations of the phase of the second harmonic in the case of near-specular reflection at a constant amplitude of the first harmonic incident on the sample. The arrows mark the positions of the resonances $\Omega/\omega=1$, 2/3, and 1/2. The parameter $\omega\tau=3$.

Thus, the second-harmonic generation method can supplement the presently employed methods of investigating the electron spectrum and the surface properties of metals. In addition, measurement of the second harmonic can yield also new information on the electron spectrum. In fact, in the case of a complicated dispersion law the nonlinear conductivity $Q_{\alpha\beta\gamma}(k, k_1, k_2)$ can be obtained from (13) and (18) by making the substitution

$$\frac{e^2 p_F^2}{2\pi^2} b_{\alpha\beta\gamma} \to \frac{e^2}{2\pi^3}\int_0^{2\pi} \frac{d\varphi}{K(\varphi)} \frac{n_\alpha n_\beta n_\gamma}{n_x}$$

where n_α is a unit vector normal to the Fermi surface, $K(\varphi) \equiv K(\theta=\pi/2, \varphi)$ is the Gaussian curvature of the Fermi surface. θ and φ are the polar and azimuthal angles of the normal vector n_α, and the polar axis coincides with the y axis.

In this case, for example for diffuse reflection, the nonlinear conductivity acquires tensors of the form

$$\int_0^{2\pi} \frac{d\varphi}{K(\varphi)} \frac{n_\alpha n_\beta n_\gamma}{n_x} \coth\left\{-\frac{i\pi}{\Omega}(s\omega+i\tau^{-1})\right\}, \quad s=1,2.$$

Generally speaking, such expressions do not enter in the linear conductivity, so that observation of the second harmonic can yield additional information for the reconstruction of the electron dispersion law. We are unable however, to dwell here in greater detail on the effects connected with the anisotropy of the electron spectrum.

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