Quantum oscillations of the amplitude of the second harmonic of sound in a conductor

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Quantum oscillations of the second harmonic amplitude of sound in a conductor located in a magnetic field are considered. These are due to oscillations of the nonlinear electron susceptibility. It is shown that the only condition for the existence of strong nonlinear susceptibility oscillations is the presence of a quantizing magnetic field. Quantum oscillations of the second harmonic amplitude can be observed in the propagation of sound along the magnetic field, as well as perpendicular to it. In the latter case, oscillations of geometric resonance may be superimposed on the "ordinary" quantum oscillations. The calculation is performed for an arbitrary electron spectrum and a closed Fermi surface.

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It is known that the sound absorption coefficient in a conductor located in a quantizing magnetic field can undergo strong oscillations with the magnetic field—the so-called giant quantum oscillations.[1] At the present time, this effect is widely used for the study of Fermi surfaces of metals. In the works of Demikhovskiĭ and the author,[2,3] the existence of another effect has been pointed out—strong oscillations of the nonlinear susceptibility in the propagation of sound along the magnetic field, which can lead to oscillations of the amplitude of the second harmonic of the sound. The second-harmonic oscillations were then discovered experimentally.[4] However, the conditions for the existence of these oscillations have not previously been considered. In contrast with[2,3], where only the case of an isotropic and quadratic spectrum was considered, we give a calculation here for a closed Fermi surface and an arbitrary spectrum. We have shown that strong quantum oscillations of the second harmonic can be observed even when $\mathbf{q} \perp \mathbf{H}$ ($\mathbf{q}$ is the sound wave vector, $\mathbf{H}$ the constant magnetic field strength). In this case, oscillations of geometric resonance can be superimposed on the "ordinary" quantum oscillations. In contrast to the quantum oscillations of the absorption, the presence of a quantizing field is actually the only condition for the existence of strong second-harmonic oscillations.

1. For a description of the interaction of electrons with sound, we use the simplest model of a deformed potential. The Hamiltonian of the interaction of an electron with the lattice is of the form

$$\hat{V}(t) = \Lambda \operatorname{div} \mathbf{u},$$

where Λ is the deformation potential constant, which we shall for simplicity assume to be independent of the momentum; $\mathbf{u}$ is the lattice displacement vector.

We now write down the equation of motion of the lattice:

$$\rho_0 \partial^2 \mathbf{u}/\partial t^2 = \lambda \Delta \mathbf{u} + \Lambda \nabla n, \tag{1}$$

where ρ_0 is the density of the crystal, λ the elastic modumodulus, and n the electron concentration. The concentration of electrons n depend nonlinearly on the lattice displacement vector $\mathbf{u}$, and this fact leads to the generation of higher harmonics of the sound. In the absence of a magnetic field, electron nonlinearity in metals gives a contribution to the generation of harmonics that is of the same order as the lattice and "geometric" nonlinearities. However, we shall be interested only in the electron nonlinearity; therefore, we use the linear equation of elasticity theory (1).

Let a sound wave propagate in the sample with frequency ω, wave number $\mathbf{q}$ and amplitude $\mathbf{u}$:

$$\mathbf{u}(\mathbf{r}, t) = \mathbf{u} e^{i\mathbf{q}\mathbf{r} - i\omega t} + \text{c.c.}$$

Assuming the amplitude of the fundamental u to be given, we find the amplitude of the second harmonic $\mathbf{u}^{(2)}$ from (1), recognizing that $\mathbf{u}^{(2)} = 0$ on the boundary of the sample:

$$u^{(2)} = \beta L, \tag{2}$$

where L is the distance from the sample boundary,

$$\beta = (2\lambda)^{-1} \Lambda^3 \chi^{(2)}(2\omega, 2\mathbf{q}) (\mathbf{q}\mathbf{u})^2,$$

$\chi^{(2)}(2\omega, 2\mathbf{q})$ is the nonlinear susceptibility, defined by the relation

$$\delta n^{(2)} = \chi^{(2)}(2\omega, 2\mathbf{q}) V^2 e^{2i(\mathbf{q}\mathbf{r} - \omega t)} + \text{c.c.}$$

$\delta n^{(2)}$ is the correction to the electron concentration at the frequency of the second harmonic, $V = i\Lambda \mathbf{q}\mathbf{u}$. The expression (2) is valid if the conditions of synchronism are satisfied at the distance L, and $\Gamma L \ll 1$ (Γ is the damping coefficient).

As will be shown below, the nonlinear susceptibility $\chi^{(2)}(2\omega, 2\mathbf{q})$ in the quantizing magnetic field can undergo strong oscillations, which leads to oscillations of the sound second harmonic.

2. We proceed to the calculation of the nonlinear susceptibility. We shall start out from the equation for the single-particle density matrix $\hat{\rho}$:

$$\partial \hat{\rho}/\partial t + i[\hat{H}, \hat{\rho}] = \hat{I}(\hat{\rho}), \tag{3}$$

where $\hat{H} = \hat{H}_0 + \hat{V}(t)$, $\hat{H}_0$ is the Hamiltonian of the electron in the quantizing magnetic field and $\hat{I}(\hat{\rho})$ is the collision integral. The τ approximation is often used for approximation of the collision integral, and is written in the form

$$\hat{I}(\hat{\rho}) = -(\hat{\rho} - \hat{\rho}_0(\hat{H}_0))/\tau, \tag{4}$$

where $\hat{\rho}_0(\hat{H}_0)$ is the equilibrium density matrix and τ is the relaxation time. However, such a collision integral, as is well known, does not conserve the number of particles and its use in the calculation of the reaction of the system to a longitudinal wave can lead to incorrect results.

The collision integral, which describes the relaxation in terms of an "instantaneous-equilibrium" density matrix is physically more justified:

$$\hat{I}(\hat{\rho}) = -(\hat{\rho} - \hat{\rho}_0(\hat{H}, \zeta))/\tau. \quad (5)$$

The operator $\hat{\rho}_0(\hat{H}, \zeta)$ satisfies the equation

$$[\hat{H} - \zeta, \hat{\rho}_0(\hat{H}, \zeta)] = 0 \quad (6)$$

with boundary condition $\hat{\rho}_0(\hat{H}, \zeta) = \rho_0(\hat{H}_0)$ if $\hat{V}(t) = 0$. The parameter ζ is the nonequilibrium contribution to the chemical potential, determined from the condition.

$$\mathrm{Sp}\,\{\hat{n}(\mathbf{r})(\hat{\rho} - \hat{\rho}_0(\hat{H}, \zeta))\} = 0, \quad (7)$$

where $n(\mathbf{r})$ is the particle density operator.

If the perturbation $\hat{V}$ does not depend on time, then $\zeta = 0$ and the solution of (3) will be

$$\hat{\rho} = \hat{\rho}_0(\hat{H}) = \hat{\rho}_0(\hat{H}_0 + \hat{V}).$$

The collision integral (5), by virtue of the condition (7), preserves the number of particles. Such a collision integral is also used frequently in the literature. It was applied, for example, to the calculation of the linear conductivity tensor in a quantizing magnetic field. [5] We shall also use the collision integral in the form (5)); however, in the specific cases considered by us, the parameter ζ is small and can be neglected. Therefore, we shall immediately set $\zeta = 0$, so that we do not have to write out more cumbersome general formulas.

Let $\hat{V}(t)$ depend on the time according to the law

$$\hat{V}(t) = \hat{V}e^{-i\omega t} + \mathrm{H.c.}$$

The operator $\hat{\rho}_0(\hat{H})$ is now expanded in powers of $\hat{V}$:

$$\hat{\rho}_0(\hat{H}) = \hat{\rho}_0(H_0) + e^{-i\omega t}\hat{\rho}_0^{(1)} + e^{-2i\omega t}\hat{\rho}_0^{(2)} + \ldots \quad (8)$$

Solving Eq. (6) by the iteration method, we get

$$\hat{\rho}_{0\lambda\lambda'}^{(1)} = \hat{V}_{\lambda\lambda'}\frac{f_\lambda - f_{\lambda'}}{\varepsilon_\lambda - \varepsilon_{\lambda'}}, \quad (9)$$

$$\hat{\rho}_{0\lambda\lambda'}^{(2)} = \sum_{\lambda''}\frac{\hat{V}_{\lambda\lambda''}\hat{V}_{\lambda''\lambda'}}{\varepsilon_\lambda - \varepsilon_{\lambda'}}\left\{\frac{f_\lambda - f_{\lambda''}}{\varepsilon_\lambda - \varepsilon_{\lambda''}} - \frac{f_{\lambda''} - f_{\lambda'}}{\varepsilon_{\lambda''} - \varepsilon_{\lambda'}}\right\}, \quad (10)$$

where $\hat{\rho}_{0\lambda\lambda'}^{(1)}$ and $\hat{V}_{\lambda\lambda'}$ are the matrix elements in the wave functions of an electron in the magnetic field, λ is the complete set of quantum numbers of the electron, ϵ_λ the eigenvalues of the energy, f_λ the equilibrium distribution function of the electrons.

We write down the density matrix $\hat{\rho}$ in the form

$$\hat{\rho} = \hat{\rho}_0(\hat{H}) + e^{-i\omega t}\hat{\rho}^{(1)} + e^{-2i\omega t}\hat{\rho}^{(2)} + \ldots \quad (11)$$

Solving Eq. (3) in the linear approximation, we get

$$\hat{\rho}_{\lambda\lambda'}^{(1)} = \frac{\omega\hat{V}_{\lambda\lambda'}}{\varepsilon_\lambda - \varepsilon_{\lambda'} - \omega - i\tau^{-1}}\cdot\frac{f_\lambda - f_{\lambda'}}{\varepsilon_\lambda - \varepsilon_{\lambda'}}. \quad (12)$$

In the next approximation, we have

$$\hat{\rho}_{\lambda\lambda'}^{(2)} = \sum_{\lambda''}\left\{\frac{2\omega}{\varepsilon_\lambda - \varepsilon_{\lambda'} - 2\omega - i\tau^{-1}}\frac{\hat{V}_{\lambda\lambda''}\hat{V}_{\lambda''\lambda'}}{\varepsilon_\lambda - \varepsilon_{\lambda'}}\left[\frac{f_\lambda - f_{\lambda''}}{\varepsilon_\lambda - \varepsilon_{\lambda''}} - \frac{f_{\lambda''} - f_{\lambda'}}{\varepsilon_{\lambda''} - \varepsilon_{\lambda'}}\right]\right.$$
$$+\frac{\omega\hat{V}_{\lambda\lambda''}\hat{V}_{\lambda''\lambda'}}{\varepsilon_\lambda - \varepsilon_{\lambda'} - 2\omega - i\tau^{-1}}\left[\frac{1}{\varepsilon_\lambda - \varepsilon_{\lambda''} - \omega - i\tau^{-1}}\frac{f_\lambda - f_{\lambda''}}{\varepsilon_\lambda - \varepsilon_{\lambda''}}\right. \quad (13)$$
$$\left.\left. - \frac{1}{\varepsilon_{\lambda''} - \varepsilon_{\lambda'} - \omega - i\tau^{-1}}\frac{f_{\lambda''} - f_{\lambda'}}{\varepsilon_{\lambda''} - \varepsilon_{\lambda'}}\right]\right\}.$$

Using Eqs. (10) and (13), we get for the nonlinear susceptibility $\chi^{(2)}(2\omega, 2\mathbf{q})$

$$\chi^{(2)}(2\omega, 2\mathbf{q}) = \sum_{\lambda,\lambda',\lambda''}\langle\lambda'|e^{-2i\mathbf{q}\mathbf{r}}|\lambda\rangle\langle\lambda|e^{i\mathbf{q}\mathbf{r}}|\lambda''\rangle\langle\lambda''|e^{i\mathbf{q}\mathbf{r}}|\lambda'\rangle$$
$$\times\left\{\frac{1}{\varepsilon_\lambda - \varepsilon_{\lambda'}}\left[\frac{f_\lambda - f_{\lambda''}}{\varepsilon_\lambda - \varepsilon_{\lambda''}} - \frac{f_{\lambda''} - f_{\lambda'}}{\varepsilon_{\lambda''} - \varepsilon_{\lambda'}}\right]\right.$$
$$+\frac{2\omega}{\varepsilon_\lambda - \varepsilon_{\lambda'} - 2\omega - i\tau^{-1}}\frac{1}{\varepsilon_\lambda - \varepsilon_{\lambda'}}\left[\frac{f_\lambda - f_{\lambda''}}{\varepsilon_\lambda - \varepsilon_{\lambda''}} - \frac{f_{\lambda''} - f_{\lambda'}}{\varepsilon_{\lambda''} - \varepsilon_{\lambda'}}\right] \quad (14)$$
$$\left.+\frac{\omega}{\varepsilon_\lambda - \varepsilon_{\lambda'} - 2\omega - i\tau^{-1}}\left[\frac{1}{\varepsilon_\lambda - \varepsilon_{\lambda''} - \omega - i\tau^{-1}}\frac{f_\lambda - f_{\lambda''}}{\varepsilon_\lambda - \varepsilon_{\lambda''}} - \frac{1}{\varepsilon_{\lambda''} - \varepsilon_{\lambda'} - \omega - i\tau^{-1}}\frac{f_{\lambda''} - f_{\lambda'}}{\varepsilon_{\lambda''} - \varepsilon_{\lambda'}}\right]\right\}$$

3. We first consider in more detail the case in which the sound is propagated along the magnetic field. We shall assume the magnetic field to be parallel to the z axis. If the Fermi surface is closed, then the energy ϵ_λ depends only on the two quantum numbers $\epsilon_\lambda = \epsilon_n(p_z)$, p_z is the projection of the momentum on the direction of the magnetic field, and n = 0, 1, 2, ... (for simplicity, we shall not take spin splitting into account). Let the condition

$$qR \ll 1, \quad (15)$$

be satisfied, where R is the Larmor radius. In this case we have for the matrix elements

$$\langle n, p_z, p_x|e^{i\mathbf{q}\mathbf{r}}|n', p_z', p_x'\rangle = \delta_{nn'}\delta_{p_x p_x'}\delta_{p_z, p_z'+q_z}. \quad (16)$$

In several special cases, for example, for an isotropic and quadratic spectrum, the expression (16) is accurate and its validity is not connected with satisfaction of the inequality (15).

In addition, we shall assume that the following inequalities are satisfied:

$$q^2/mT \ll 1, \quad ms^2 \ll T, \quad (17)$$

where m is the effective mass, T the temperature, s the speed of sound; then (14) can be transformed into the expression

$$\chi^{(2)}(2\omega, 2\mathbf{q}) = \frac{eH}{2\pi^2 c}\sum_n\int dp_z\left\{\frac{1}{2}\frac{\partial^2 f}{\partial\varepsilon^2} + \frac{\omega}{2v_n(p_z)q - 2\omega - i\tau^{-1}}\frac{\partial^2 f}{\partial\varepsilon^2}\right.$$
$$\left.+\frac{\omega}{2v_n(p_z)q - 2\omega - i\tau^{-1}}\frac{\partial}{\partial p_z}\frac{q}{v_n(p_z)q - \omega - i\tau^{-1}}\frac{\partial f}{\partial\varepsilon}\right\}, \quad (18)$$

where $v_n(p_z) = \partial\epsilon_n/\partial p_z$ is the velocity, e the charge of the electron (absolute value) c the speed of sound. We obtain this formula by solving the one-dimensional "classical" kinetic equation.

By virtue of the second inequality (17), the largest contribution will be made by the first term in the curly brackets of Eq. (18), and we get

$$\chi^{(2)} = \frac{eH}{2\pi^2 c}\sum_n\int dp_z\frac{1}{2}\frac{\partial^2 f}{\partial\varepsilon^2}. \quad (19)$$

The physical meaning of Eq. (19) is extremely simple. Actually, the condition $ms^2 \ll T$ means that the electrons on the Landau level with maximum $n = n_F$, which interact resonantly with the sound, are not degenerate. At the same time, the small contribution $s(T/m)^{-1/2}$—the ratio of the sound speed to the thermal speed of the electrons—is the parameter of the adiabaticity for electrons on the Landau level with $n = n_F$ (on other levels, such parameters are the ratios s/v_{nF}, where v_{nF} is the Fermi velocity on the n-th level). Because of this, the nonlinear reaction of the electrons is identical with the static one and can be obtained from the expansion of $\hat{\rho}_0(\hat{H}_0 + V)$ in powers of V, i.e., from $\hat{\rho}_0^{(2)}$. It is by calculating the nonlinear susceptibility with the help of the operator $\hat{\rho}_0^{(2)}$ for small q that we obtain (19). It is clear that, inasmuch as the nonlinear susceptibility is identical with the static value, it does not depend on the scattering (of course, if the condition $\Omega\tau \gg 1$ is satisfied, Ω being the cyclotron resonance) and Eq. (19) remains valid even for $ql < 1$ (l is the free path length). On the other hand, the expression (19) is valid even in the case in which $\tau = \infty$.

We now estimate the magnitude of the nonlinearity. For $\Omega \gg T$ and upon satisfaction of the resonance condition $\mu/\Omega = n_F$, where μ is the chemical potential, the basic contribution to the nonlinearity is made by the component with $n = n_F$. Calculating it, we obtain

$$\chi^{(2)} \sim \frac{eH}{2\pi^2 c} \frac{(2mT)^{1/2}}{T^2}, \tag{20}$$

which can exceed appreciably the value of the nonlinearity without a magnetic field:

$$\chi^{(2)}(H=0) \infty m^2/2\pi^2 p_F, \tag{21}$$

where p_F is the Fermi momentum. Summation over n in (19) can be carried out by using the Poisson formula. After standard calculations, we obtain the following expression for the oscillating part of $\chi^{(2)}$:

$$\chi_{\text{osc}}^{(2)} = -\frac{eH}{\pi c} \sum_{k=1}^{\infty} \sum_m k^{1/2} \Omega^{-2}(p_z^m) \left(\frac{c}{2\pi eH} \left| \frac{\partial^2 S_m}{\partial p_z^2} \right| \right)^{-1/2} \times \psi(\lambda_m k) \sin\left(k \frac{cS_m}{eH} \pm \frac{\pi}{4} \right), \tag{22}$$

where

$$\psi(z) = z/\text{sh}\, z, \quad \lambda_m = 2\pi^2 T/\Omega(p_z^m),$$

S_m is the area of the extremal cross section of the Fermi surface, and p_z^m is the momentum corresponding to this cross section. The nonoscillating part is the same as in the absence of a magnetic field.

If $\Omega \gg 2\pi^2 T$ and if the resonance condition $cS_m/eH = 2\pi n_F$ is satisfied, all the harmonics in (22) are added in phase and the contribution to the sum over k is made by a large number of harmonics with $k \lesssim \Omega/2\pi^2 T$. Replacing the summation over k by integration, we again obtain the contribution of a single component with $n = n_F$ in the initial sum over n (20). Thus we see that the nonlinear susceptibility $\chi^{(2)}$ in a quantizing field can execute strong, essentially nonsinusoidal oscillations with the magnetic field, which one can call giant quantum oscillations.

We also note that, as follows from (19), the nonlinear susceptibility $\chi^{(2)}$ can be expressed in terms of the linear $\chi^{(1)}$ and the total number of particles n_0:

$$\chi^{(2)} = -\frac{1}{2} \frac{\partial \chi^{(1)}}{\partial \mu} = \frac{1}{2} \frac{\partial^2 n_0}{\partial \mu^2}. \tag{23}$$

The oscillations of the linear susceptibility in the situation that we have considered, in which the inequality (17) is satisfied, are of course small.

4. Now let the sound be propagated perpendicular to the magnetic field: $\mathbf{q} \perp \mathbf{H}$. Here, just as above, we shall assume that the inequality $qR \ll 1$ is satisfied. Then, taking into account only the matrix elements that are diagonal in the quantum number n, since the remaining matrix elements are small, and taking in (14) the limit as $q_z \to 0$, we get

$$\chi^{(2)}(2\omega, 2\mathbf{q}) = \frac{eH}{2\pi^2 c} \sum_n \int dp_z \left\{ \frac{1}{2} \frac{\partial^2 f}{\partial \varepsilon^2} - \frac{1}{2} \frac{\omega}{\omega + i(2\tau)^{-1}} \frac{\partial^2 f}{\partial \varepsilon^2} \right\}. \tag{24}$$

If $\omega\tau \ll 1$ then we again get

$$\chi^{(2)} = \frac{eH}{2\pi^2 c} \sum_n \int dp_z \frac{1}{2} \frac{\partial^2 f}{\partial \varepsilon^2},$$

and in the case in which $\omega\tau \to \infty$, the expression (24) vanishes. Thus, we see that, to obtain the correct static limit for the susceptibility $\chi^{(2)}(2\omega, 2q)$ in this case, we must take the scattering into account, in contrast to the situation when the sound is propagated along the magnetic field, while use of the collision integral (4) would have led to an incorrect result. The nonlinear susceptibility $\chi^{(2)}(2\omega, 2q)$ thus calculated goes to zero as $qR \to 0$, independent of the values of the parameter $\omega\tau$.

It follows from Eq. (24) that the nonlinear susceptibility in the low-frequency region $\omega\tau \ll 1$ can execute giant quantum oscillations, just as for $\mathbf{q} \parallel \mathbf{H}$. At large values of the parameter $\omega\tau$ the oscillations are weakened, as follows from (24). However, we must note that in this case, when $\mathbf{q} \perp \mathbf{H}$ and $\omega\tau \gg 1$, the model of interaction of electrons with the sound that we have used is unsatisfactory, since it is necessary to take the electromagnetic field into account. It appears, however, that a weakening of the oscillations for large values of $\omega\tau$ is the correct result. We shall not consider this question further.

We now proceed to the general case, when the parameter qR can take on arbitrary values. In the quasiclassical approximation $n_F \gg 1$ and $q \ll p_F$, the following expression is valid for the matrix element $\langle \lambda | e^{i\mathbf{q}\cdot\mathbf{r}} | \lambda' \rangle$:

$$\langle n, p_z, p_x + q_x | e^{i\mathbf{q}\mathbf{r}} | n', p_z, p_x \rangle = J_l(\mathbf{q}, \varepsilon_n, p_z) = \frac{1}{T_{\text{cl}}} \int_0^{T_{\text{cl}}} dt \exp\left\{ i\mathbf{q} \int_0^t \mathbf{v}(t_1)\, dt_1 + il\Omega t \right\}, \tag{25}$$

where $l = n' - n$, $\mathbf{v}(t) = \mathbf{v}(\epsilon_n, p_z, t)$ is the velocity and $T_{\text{cl}} = 2\pi/\Omega$ is the period. The integral (25) is calculated from the classical trajectory. The matrix element $J_l(q)$ is exponentially small if $l \gg qR$; therefore the important contribution is made by terms with $l \lesssim qR \ll n_F$, inasmuch as $q \ll p_F$. Consequently, we can assume that

$$\varepsilon_{n+l}(p_z) - \varepsilon_n(p_z) = l\Omega(p_z).$$

We will again consider only the case $\omega\tau \ll 1$. Then the basic contribution to the nonlinear susceptibility is made by terms that are independent of frequency, obtained from the expansion of the instantaneous-equilibrium density matrix $\hat{\rho}_0(\hat{H})$ in powers of $\hat{V}$. We calculate the susceptibility by using the Poisson summation formula. As usual, we transform to integration over ϵ and p. Since the important contribution to the integral over ϵ is made by the small region near the Fermi surface, we can extend the integration to $-\infty$. The integral over p_z is calculated by the saddle-point method. After some transformations, the oscillating part of the susceptibility can be represented in the form

$$\chi_{\text{osc}}^{(2)} = \frac{eH}{\pi^2 c} \sum_m \sum_{ls} \sum_{k=1}^{\infty} \int d\varepsilon\, f(\varepsilon) \frac{1}{l\Omega} \{ [F_{ls}^k(\varepsilon - (l+s)\Omega) \operatorname{Re} e^{2\pi i k n(\varepsilon - (l+s)\Omega) \pm i\pi/4} - F_{ls}^k(\varepsilon - l\Omega) \operatorname{Re} e^{2\pi i k n(\varepsilon - l\Omega) \pm i\pi/4}] \frac{1}{s\Omega} + \frac{1}{(l+s)\Omega} [F_{ls}^k(\varepsilon) \operatorname{Re} e^{2\pi i k n(\varepsilon) \pm i\pi/4} - F_{ls}^k(\varepsilon - (l+s)\Omega) \operatorname{Re} e^{2\pi i k n(\varepsilon - (l+s)\Omega) \pm i\pi/4}] \}_{p_z = p_z^m}, \tag{26}$$

where

$$f(\varepsilon) = (e^{(\varepsilon-\mu)/T} + 1)^{-1},$$

$$F_{ls}^k(\varepsilon) = \left(k \left| \frac{\partial^2 n}{\partial p_z^2} \right| \right)^{-1/2} \frac{dn}{d\varepsilon} w_{ls}(\varepsilon, \mathbf{q}),$$

$$w_{ls}(\varepsilon, \mathbf{q}) = J_l(\varepsilon, -2\mathbf{q}) J_s(\varepsilon, \mathbf{q}) J_{-l-s}(\varepsilon, \mathbf{q}).$$

In Eq. (26) the summation is over all l and s, including $l = 0$ and $l + s = 0$. The corresponding terms are taken in the sense of the limit as $l \to 0$, $s \to 0$ and $l + s \to 0$.

The functions $F_{ls}^k(\epsilon)$ are smooth functions of the energy; therefore they can be expanded in series:

$$F_{ls}^{k}(\varepsilon-l\Omega)=F_{ls}^{k}(\varepsilon)-\frac{dF_{ls}^{k}(\varepsilon)}{d\varepsilon}l\Omega+\frac{1}{2}\frac{d^2F_{ls}^{k}(\varepsilon)}{d\varepsilon^2}(l\Omega)^2. \quad (27)$$

Upon substitution of (27) in (26), we see that differences of the rapidly oscillating exponents appear, of the form

$$\frac{1}{l\Omega}(e^{2\pi ikn(\varepsilon-l\Omega)}-e^{2\pi ikn(\varepsilon)}).$$

Recognizing that $n(\epsilon - l\Omega) = n(\epsilon) - l$, we obtain

$$\frac{1}{l\Omega}(e^{2\pi ikn(\varepsilon-l\Omega)}-e^{2\pi ikn(\varepsilon)})=-\delta_{l0}\frac{d}{d\varepsilon}e^{2\pi ikn(\varepsilon)}.$$

It is therefore clear that the basic contribution to Eq. (26) is made by terms with $l = s = 0$; therefore, only the rapidly oscillating exponential $e^{2\pi ikn(\epsilon)}$ is differentiated twice. Taking this into consideration, and carrying out integration over ϵ, we get

$$\chi_{osc}^{(2)}=-\frac{eH}{\pi c}\sum_{k=1}^{\infty}\sum_{m}k^{1/2}\Omega^{-2}(p_z^m)w_{00}(q,p_z^m)\times\left(\frac{c}{2\pi eH}\left|\frac{\partial^2 s_m}{\partial p_z^2}\right|\right)^{-1/2}\psi(\lambda_m k)\sin\left(k\frac{cs_m}{eH}\pm\frac{\pi}{4}\right). \quad (28)$$

Equation (28) differs from (22) only in the factor w_{00}.

For $qR \ll 1$, we have $w_{00} = 1$; if $qR > 1$, then w_{00} executes geometric-resonance oscillations. We calculate the matrix element $J_0(\mathbf{q})$ in the limit $qR \gg 1$, using the saddle-point method. There are at least two points on the trajectory of the extremal cross section where $\mathbf{q}\cdot\mathbf{v} = 0$, the vicinities of which also give the principal contribution to the integral (25). For simplicity, we shall assume that there are two such points, and denote them by $t_{(1)}$ and $t_{(2)}$. We choose the origin so that $t_{(1)} = 0$. Then, calculating the integral (25), we get

$$J_0(\mathbf{q})=\frac{1}{T_{cl}}\left[\exp\left\{i\mathbf{q}\int_0^{t_{(2)}}\mathbf{v}(t)\,dt+i\frac{\pi}{4}\operatorname{sign}\mathbf{qv}'_{(2)}\right\}\left|\frac{2\pi}{\mathbf{qv}'_{(2)}}\right|^{1/2}+\exp\left\{i\frac{\pi}{4}\operatorname{sign}\mathbf{qv}'_{(1)}\right\}\left|\frac{2\pi}{\mathbf{qv}'_{(1)}}\right|^{1/2}\right], \quad (29)$$

where $\mathbf{v}' = d\mathbf{v}/dt$.

We assume that $\mathbf{q}\cdot\mathbf{v}' \neq 0$. Let the vector $\mathbf{q}$ be directed along the x axis. Then

$$\mathbf{q}\int_0^{t_{(2)}}\mathbf{v}(t)\,dt=\frac{qc}{eH}[p_y(t_{(1)})-p_y(t_{(2)})].$$

It is thus seen that w_{00} at $qR \gg 1$ is an oscillating function of $1/H$ with a period

$$\Delta(1/H)=2\pi e/qc\,(p_y(t_{(1)})-p_y(t_{(2)}))_{p_z=p_z^m}. \quad (30)$$

Thus, for $qR > 1$, the oscillations of geometric resonance are superimposed on the ordinary quantum oscillations. We also note that, generally speaking, the period of the geometric-resonance oscillations considered by us is not the same as the period of the geometric-resonance oscillations in sound absorption. Actually, the absorption oscillation period is the extremal difference $(p_y(t_{(1)}) - p_y(t_{(2)}))_{extr}$, whereas (30) contains this difference taken on the extremal section, i.e., on the section with extremal area. These quantities can coincide in special cases.

According to (29), the amplitude of oscillations of the nonlinear susceptibility falls off with increase in qR as $(qR)^{-3/2}$. If at the saddle points $t_{(1)}$ and $t_{(2)}$ $\mathbf{q}\cdot\mathbf{v}' = 0$, then this decrease is much slower—like $(qR)^{-1}$, as can be verified by calculating the asymptotic matrix elements.

For the nonoscillating part of $\chi^{(2)}$, we get

$$\chi^{(2)}=\frac{2}{(2\pi)^3}\int d^3\mathbf{p}\,\frac{1}{2}\frac{\partial^2 f}{\partial\varepsilon^2}, \quad (31)$$

which is identical with the nonlinear susceptibility without a field. In obtaining Eq. (31), we have assumed that

$$\sum_{ls}w_{ls}=1.$$

Thus, we have shown that the nonlinear susceptibility can undergo strong quantum oscillations, and the only condition for their existence is the presence of a quantizing field. For observation of giant quantum oscillations, as is well known, satisfaction of the condition

$$q_z l(\Omega/\mu)^{1/2}\gg 1, \quad (32)$$

is necessary. This is a rather strong limitation on the purity of the metal and the sound frequency. For observation of nonlinear oscillations, it is more convenient to use comparatively low frequencies, since the sound damping coefficient Γ decreases with decrease in frequency, and the condition $\Gamma d \ll 1$ (d is the thickness of the sample) can be obtained for rather large samples.

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