

(with  $\sigma_{ik}$ ).

The perturbation that induces the single-phonon transitions was taken in the present paper to be only the dynamic strains  $\varepsilon_{ik}$ . They are expressed in terms of products of components of  $u$  and  $q$  [Eq. (5)] that transform like a second-rank symmetric tensor. Yet an active role can be played in principle in the transitions also by dynamic rotations  $\omega = \frac{1}{2}u \times q$  (Refs. 8 and 15), which have the transformation properties of an axial vector. The relative contribution of the strains and of the rotations to the matrix element  $\langle 1|H'|2\rangle$  of a single-phonon transition is determined by the ratio of the deformation potential ( $b$ ) to the transition energy ( $\Delta$ ). In our case  $b \approx 3 \times 10^3 \text{ cm}^{-1}$  and  $\Delta_{\text{max}} \approx 30 \text{ cm}^{-1}$ , therefore the contribution of the rotations to the transition probability is  $\sim (\Delta/b)^2 \approx 10^{-4}$  and can be neglected. In general, the role of the rotations, compared with strain, can be regarded as small for all PPD of the terahertz band, for which  $b \gg \Delta$  always.

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<sup>1</sup>The velocities of the TA and LA phonons were calculated from the elastic constants of  $\text{CaF}_2$  at  $T = 4\text{K}$ .<sup>6</sup> The velocity indicated for the phonon propagation direction  $q \parallel [110]$  is that of the fast FTA phonons.

<sup>2</sup>A perfectly analogous matrix for  $\Gamma_3^+$  with the corresponding parameters  $A$  and  $B$  is given in Ref. 7 [Eq. (6)] for the stresses  $\delta_{ik}$ . The relations between the parameters (3) and (6) of Ref. 7 are  $b = (C_{11} - C_{12})B$  and  $a = (C_{11} + 2C_{12})A$ .

<sup>3</sup>The calculated transition probability in  $\text{CaF}_2:\text{Eu}^{2+}$  crystals at  $\nu = 30 \text{ cm}^{-1}$  and  $T_1^{-1} \approx 6 \times 10^9 \text{ sec}^{-1}$  exceeds by two orders of magnitude, say, the probability of the  $2A \rightarrow \bar{E}$  transition in  $\text{Cr}^{3+}$  ions in ruby ( $\nu = 29 \text{ cm}^{-1}$ ,  $T_1^{-1} = 3 \times 10^9 \text{ sec}^{-1}$ , Ref. 11).

<sup>4</sup>We point out a general analogy between the spatial distribution of acoustic-phonon emission (absorption) and the dis-

tribution of electromagnetic radiation in the case of electric quadrupole transitions. The latter are known<sup>14</sup> to be described by an expression of the type  $|\sum_{ik} e_i q_k Q_{ik}|^2$ , where  $e$  is the electric vector,  $q$  is the wave vector of the electromagnetic wave, and  $Q_{ik} = ex_i x_k$  is the electric quadrupole moment of the transition.

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## Anomalous skin effect on a rough surface in a magnetic field

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The impedance of a metal in a magnetic field parallel to the surface is calculated with allowance for the scattering of the conduction electrons by surface roughnesses. The monotonic part of the impedance is determined by the multiply reflected electrons and depends on the competition of their scattering by bulk or surface defects. The cyclotron oscillations are small, except when the mean free path is very large and in a narrow vicinity of the resonance the contribution of the electrons that do not collide with the surface becomes predominant.

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### 1. INTRODUCTION

The surface impedance of a metal in a magnetic field parallel to the surface depends substantially on the character of the scattering of the conduction electrons

from the sample boundary. The reason is that the influence of the surface comes into play at a distance determined either by the electron mean free path  $l$  or by the Larmor radius  $r$ , i. e., by parameters that can be large compared with the skin-layer depth  $\delta$  (we assume

these conditions to be satisfied).

Yet the theory of cyclotron resonance<sup>1</sup> was initially constructed under the assumption that the electrons are diffusely reflected. In this case the oscillating part of the impedance is comparable with the monotonic one, whereas in experiment it is as a rule smaller by a factor of 100 even for samples with  $l \gg r$ .<sup>2</sup> An attempt to explain this contradiction was made by Chambers,<sup>3</sup> who advanced the hypothesis that the large monotonic part is determined by the contribution of nonresonant points on the electron trajectory. The corresponding quantitative theory was developed by Meierovich,<sup>4</sup> who showed that under conditions of specular reflection from the boundary, the "hopping" electrons (see orbits in Fig. 1) with glancing angles  $\varphi \sim (\delta/r)^{1/2}$ , which spend a long time in the skin layer, determine the large monotonic part of the impedance, which is roughly speaking of the same order as the impedance in the absence of the field. The electrons that do not collide with the boundary but land in the skin layer (Fig. 1, orbit  $v$ ), which we shall name glancing, introduce a relatively small oscillating part.

Thus, the fact that the cyclotron oscillations observed in experiment are small is proof of the almost-specular reflection of the electrons from the boundary. Another proof is the observation, in weak fields,<sup>5</sup> of resonances due to transitions between the quantum surface levels, which arise as a result of the periodicity of the motion of the hopping electrons.

Just as in most kinetic problems, the theory of Ref. 4, makes use of the Fuchs boundary condition

$$f^>(p) = (1-q)f^<(p), \quad (1)$$

which connects, with the aid of the phenomenological diffuseness coefficient  $q$ , the nonequilibrium increment  $f^>$  to the electron distribution of the electrons moving away from the boundary, with  $f^<$  for the reflected electrons;  $f^>$  and  $f^<$  are taken at the same values of the tangential projection of the momentum  $p$  and of the oppositely directed normals. It turns out that, depending on the proximity of the coefficient  $q$  to zero, a distinction must be made between two limiting cases. In one of these cases, an error entered in the calculation of the oscillating increment; this error found its way as well as was rejected in many later papers (the last one of this cycle is Ref. 6). Since the theoretical results depend to a strong degree on the proximity of  $q$  to zero, the question arises of the bearing that these calculations, based on the phenomenological condition (1), have on the real phenomenon.

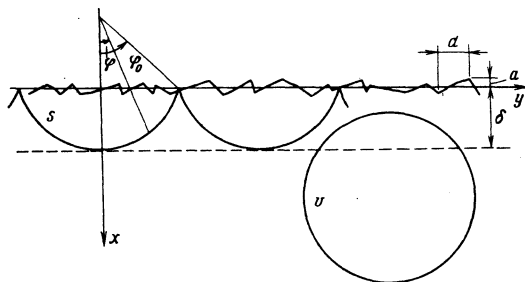


FIG. 1.

In the present paper we use a boundary condition<sup>7</sup> that takes into account scattering by random surface roughnesses

$$f^>(p) = f^<(p) + p_x \int p_x' W(p'-p) [f^<(p') - f^<(p)] \frac{d^2 p'}{\pi^2}. \quad (2)$$

Here  $W(p)$  is the Fourier component of the binary correlator of the roughnesses,  $p_x$  is the modulus of the normal component of the momentum and depends on the tangential component of  $p$  and on the energy  $\epsilon$ . For a quadratic dispersion law (our calculation is made for this case)

$$p_x = (2m\epsilon - p^2)^{1/2}, \quad p_x' = (2m\epsilon - p'^2)^{1/2}.$$

We assume in the estimates that  $W(p)$  is a function that differs from zero in the region  $p < d^{-1}$ , where its value is  $W(0) \sim a^2 d^2$ ;  $a$  and  $d$  have the respective meanings of the average size of the roughness and of the characteristic scale in the tangential direction. Planck's constant is set equal to unity.

Condition (2) reflects correctly the general features of scattering by a surface. It is automatically satisfied by an equilibrium distribution function and ensures the absence of current through the surface. The integral term is an increment to pure specular scattering and vanishes as  $p_x \rightarrow 0$ , i.e., for hopping and glancing electrons. The condition (2) was used earlier<sup>8</sup> to analyze the influence of the roughness on the anomalous skin effect and on the conductivity of thin plates in the absence of a magnetic field.

Although the integral equation to which (2) reduces cannot be solved in general form, it can be easily analyzed in two limiting cases that depend on which of the factors under the integral sign is the sharper function. As already noted, the most important role in the problem is played by small glancing angles  $\varphi \sim (\delta/r)^{1/2}$ . If the electron approaching the surface moves almost parallel to it with  $p \approx p_0$  ( $p_0 = 2m\epsilon_F$ )<sup>1/2</sup> is the Fermi momentum; we are interested in electrons close to the Fermi surface), then after scattering by the roughness we have  $p' \sim |p_0 - 1/d|$ . The corresponding angle

$$\varphi_w \sim \min(1, (p_0 d)^{-1/2})$$

characterizes the width of the function  $W(p'-p)$  at small glancing angles  $p \approx p_0$ . Since the largest contribution to the current is made the effective hopping as well as effective glancing electrons moving within the skin layer, the distribution function changes significantly over angles  $\varphi_f \sim (\delta/r)^{1/2}$ .

When  $f$  is determined for the effective electrons, the integral terms with  $f^<(p')$  can be neglected if  $\varphi_w \gg \varphi_f$ . The result is the Fuchs condition (1) in which, however,  $q$  depends on the angle:

$$q(\varphi) = p_x \int p_x' W(p'-p) \frac{d^2 p'}{\pi^2}. \quad (3)$$

At small  $\varphi$  ( $\varphi < \varphi_w$ )

$$q(\varphi) = 2Q p_x / p_0 \sim 2Q\varphi, \quad (4)$$

where the factor 2 will be found convenient in the equations that follow. An estimate yields

$$Q \sim (a p_0)^2 (p_0 d)^{-1/2} \text{ at } p_0 d \gg 1.$$

The condition  $p_0 d \ll 1$  may be satisfied in semimetals, and then  $Q \sim (adp_0^2)^2$ .

In the other limiting case, i.e., at  $\varphi_w \ll \varphi_f$ , to determine  $f$  in the region  $\varphi > \varphi_w$  it is necessary to expand  $p'_x [f(p') - f(p)]$  under the integral sign in powers of  $(p' - p)_i$ . The first-order term vanishes because  $W(p' - p)$  is odd. The second-order term is proportional to

$$\int (p' - p)_i (p' - p)_j W(p' - p) \frac{d^2 p'_i}{\sigma^2} = \delta_{ij} Q_1, \quad (5)$$

where  $Q_1 \sim (a/d)^2$ . The boundary condition (2) is transformed into the differential equation

$$j^>(p) = j^<(p) + Q_1 p_x [p_x \Delta j^<(p) + 2 \nabla p_x \nabla j^<(p)], \quad (6)$$

where the differentiation is with respect to the tangential components of  $p$  at a given value of the energy  $\varepsilon$ . The boundary condition for (6) is obtained by matching to the region  $\varphi < \varphi_w$ . It can be seen that, in order of magnitude,

$$j^>(p) - j^<(p) \sim Q_1 \varphi_1^{-2} j^<(p). \quad (7)$$

The condition (2) was derived under the assumption that the integral term in it is small compared with unity, i.e.,

$$Q(\delta/r)^{1/2} \ll 1 \quad \text{at } \varphi_w \gg \varphi_f, \quad (8a)$$

$$Q_1 r / \delta \ll 1 \quad \text{at } \varphi_w \ll \varphi_f. \quad (8b)$$

Since we are justified in considering from the Markov point of view only a roughness that is large in the atomic scale, these requirements can be satisfied in a typical metal for effective electrons with small angles  $\varphi_f$ . The condition (2) thus allows us to consider the transition to the specular case. At the same time it permits also an analysis of the diffuse limit if the number  $n$  of the collisions of the electron with the surface is large during the free-path time:

$$n = |l| (\delta r)^{-1/2} \gg 1. \quad (8c)$$

It is easily seen (as will be shown later) that the diffuseness is not small if the product of  $n$  by the diffuseness coefficient in one collision (8a), (8b) is not small.

## 2. GENERAL SOLUTION

In the solution of the kinetic equation

$$v_x \frac{\partial f}{\partial x} + \Omega \frac{\partial f}{\partial \varphi} + \left( \frac{1}{\tau} - i\omega \right) f = -e v E(x) \frac{\partial f_0}{\partial \varepsilon} \quad (9)$$

[ $E(x)$  is the electric field in the metal] it is customary to use in momentum space the following variables: the energy  $\varepsilon$ , the momentum projection  $p_x$  on the magnetic field, the rotation angle  $\varphi = \Omega t$  around the magnetic field, an angle connected with the time  $t$  by the equations of motion in the magnetic field:  $dv_x/dt = -\Omega v_y$  and  $dv_y/dt = \Omega v_x$ , where  $\Omega = eH/mc$  is the cyclotron frequency. It is convenient to use  $v_x = -v_1 \sin \varphi$ ,  $v_y = v_1 \cos \varphi$ , and  $v_1 = (v_0^2 - v_x^2)^{1/2}$ . In terms of these variables, the phase-space element is  $d^3 p = m d\varepsilon dp_x d\varphi$ . In the calculation of the current, the integral with respect to  $\varepsilon$  is evaluated with the aid of  $\partial f_0 / \partial \varepsilon = -\delta(\varepsilon - \varepsilon_F)$ , so that we can put  $\varepsilon = \varepsilon_F$  in all the quantities that depend on  $\varepsilon$ . We use the complex mean free path  $v_0/l = \tau^{-1} - i\omega$  and the Larmor radius  $r = v_0/\Omega$ .

If the characteristic of Eq. (9)

$$x = \Omega^{-1} \int v_x d\varphi = \Omega^{-1} v_y(\varphi) + \text{const} \quad (10)$$

does not cross the sample boundary  $x=0$ , then the solution of (9) is obtained from the requirement of periodicity with respect to  $\varphi$  with period  $2\pi$ :

$$f(\varphi) = \int_{-\pi}^{\varphi} d\varphi' v(\varphi') E \{ x + [v_y(\varphi') - v_y(\varphi)] \Omega^{-1} \} \exp[r(\varphi' - \varphi)/l], \quad (11)$$

the factor  $-e \Omega^{-1} \partial f_0 / \partial \varepsilon$  will be written in the expression for the current.

If the characteristic crosses the boundary, then the constant in (10) can be taken in the form  $-\Omega^{-1} v_y(\varphi_0)$ , where  $\varphi_0$  is the angle of approach to the surface,  $\varphi \leq \varphi_0 \leq \pi$ . The region corresponding to colliding electrons is shown shaded in Fig. 2. The solution takes here the form

$$f(\varphi) = e^{-r\varphi/l} \left[ \int_{-\pi}^{\varphi} d\varphi' u(\varphi', \varphi_0) e^{r\varphi'/l} + F(-\Omega^{-1} v_y(\varphi_0)) \right], \quad (12)$$

$$u(\varphi', \varphi) = v(\varphi') E \{ \Omega^{-1} [v_y(\varphi') - v_y(\varphi)] \}.$$

The arbitrary function  $F$  is obtained from the boundary condition (2), the left-hand side of which contains  $f(-\varphi_0)$  and the right side  $f(\varphi_0)$ . Using (12), we express  $f(-\varphi_0)$  in terms of  $f(\varphi_0)$  and substitute in (2). We obtain

$$[e^{2r\varphi_0/l} - 1] f(\varphi_0) - \int_{-\pi}^{\varphi_0} d\varphi' u(\varphi', \varphi_0) e^{r(\varphi_0 + \varphi')/l} = p_x \int_{-\pi}^{\varphi_0} p_x W(p' - p) [f(\varphi') - f(\varphi_0)] \frac{d^2 p'_i}{\pi^2}; \quad \varphi = \varphi_0 > 0. \quad (13)$$

Solving (13), we obtain  $f(\varphi)$  and calculate the current

$$j_x(x) = \frac{e^2 m}{4\pi^2 \Omega} \int_{-\pi}^{\varphi} dp_x \int_{-\pi}^{\varphi} d\varphi v_x(\varphi) f(\varphi), \quad (14)$$

where the integration with respect to  $\varphi$  within the limit of the shaded region should be carried out with the function (12), and with the function (11) outside this region.

Continuing in even fashion the electric field and the current into the region  $x < 0$  and changing over the Fourier components, we write down the connection between the current and the field in the form

$$j_x(k) = \sigma_{ij}(k) \mathcal{E}_j(k) + \int_{-\pi}^{\pi} \sigma_{ij}(kk') \mathcal{E}_j(k') \frac{dk'}{\pi}; \quad (15)$$

we see that in terms of the employed coordinates the conductivity tensor  $\sigma_{ij}(k)$ ,  $\sigma_{ij}(kk')$  is diagonal.

Solving next Maxwell's equation

$$k^2 \mathcal{E}_i(k) + 2E'_i(x=0) = 4\pi i \omega c^{-2} j_i(k), \quad (16)$$

we obtain the surface impedance

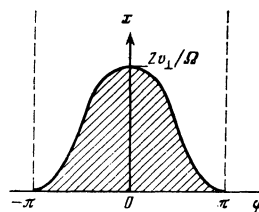


FIG. 2.

$$Z_i = \frac{4i\omega}{c^2 E_i'(0)} \int_0^\infty dk \mathcal{E}_i(k). \quad (17)$$

This program can be used in the two limiting cases referred to in the Introduction.

### 3. LOW EFFECTIVENESS ANGLES $\varphi_r \ll \varphi_W$

In this case we can introduce the angle-dependent diffuseness coefficient (3), (4). Equations (13)–(16) can then be solved exactly. Leaving out of (13) the small term with  $f(\varphi')$ , we obtain

$$f(\varphi_0) = [e^{2r\varphi_0/l} - 1 + q(\varphi_0)]^{-1} \int_{-\varphi_0}^{\varphi_0} u(\varphi', \varphi_0) e^{r(\varphi_0 + \varphi')/l} d\varphi'.$$

We then obtain with the aid of (12)

$$f(\varphi) = \int_0^\pi F d\varphi' + [1 - (1 - q(\varphi_0)) e^{-2r\varphi_0/l}]^{-1} \int_{-\varphi_0}^{\varphi_0} F d\omega', \quad (18)$$

$$F = v(\varphi') E \{ (v_y(\varphi') - v_y(\varphi_0)) / \Omega \} e^{r(\varphi' - \varphi_0)/l}; \\ v_y(\varphi_0) = v_y(\varphi) - x\Omega.$$

It is important that the solution (18) goes over continuously into (11) when we go over from electrons that collide with the surface to non-colliding ones. To verify this, it suffices to represent the integral in (11) in the form

$$\int_{-\infty}^{\varphi_0} \dots = \int_{-\infty}^{\varphi_0} \dots + \sum_{n=0}^{\infty} \int_{\pi(1-2n)}^{\pi(1-2n)} \dots$$

and use the periodicity of the integrand. We obtain

$$\int_{-\infty}^{\varphi_0} \dots = \int_{-\infty}^{\varphi_0} \dots + [1 - e^{-2r\pi/l}]^{-1} \int_{-\pi}^{\pi} \dots \quad (19)$$

The transition considered corresponds to  $\varphi_0 \rightarrow \pi$ . It is seen now that (18) goes over into (11), since  $q(\pi) \rightarrow 0$  (as  $\varphi_0 \rightarrow \pi$  the projection  $p_x \rightarrow 0$ ). We emphasize that this continuity of the distribution function is a consequence of the specular reflection of the glancing electrons that have orbits of the  $v$  type and approach the surface tangentially.

In the calculation of the Fourier component of the current, the integral with respect to  $x$  over the unshaded region (see Fig. 2) is taken of the function (11), and of (18) over the shaded region. We shall instead integrate (11) with infinite limits, omitting expression (11) in the shaded region. The first integral yields the conductivity of an infinite sample. Its asymptotic form is known:

$$\sigma_{ij}^s(k) = \delta_{ij} \frac{3\pi\sigma_0}{4kl} \operatorname{cth} \frac{\pi r}{l}, \quad (20)$$

where  $\sigma_0 = N_e e^2 l p_0^{-1}$ ; here and elsewhere  $k > 0$ .

Expression (20) does not depend on the polarization of the field and is valid in the case  $kr \gg 1$ ,  $(r/k)^{1/2} / |l| \ll 1$ . The last condition allows us to disregard the change of the field during the time of motion of the electrons within the limits of the skin layer. This condition is equivalent to (8) with  $k = \delta^{-1}$ . We note that the "bulk" conductivity (20) receives equal contributions from the hopping electrons on the orbit  $s$  at the point  $\varphi = 0$  and by the electrons glancing on the  $v$  orbit at the point  $\varphi = \pi$ .

We change over in the integral over the shaded re-

gion from  $z$  to the variable  $\varphi_0$ , using the last Eq. of (18). Using (19), we get

$$\sigma_{ij}(kk') = \frac{3\sigma_0 r^2}{2\pi l v_0^4} \int_{-\varphi_0}^{\varphi_0} dv_z \int_0^\pi d\varphi_0 |v_x(\varphi_0)| \int_{-\varphi_0}^{\varphi_0} d\varphi v_i(\varphi) \left\{ [1 - (1 - q(\varphi_0)) e^{-2r\varphi_0/l}]^{-1} \right. \\ \left. \times \int_{-\varphi_0}^{\varphi_0} G d\varphi' - (1 - e^{-2r\pi/l})^{-1} \int_{-\pi}^{\pi} G d\varphi' + \int_{-\varphi_0}^{\varphi_0} G d\varphi' \right\}, \quad (21)$$

$$G = v_j(\varphi') z(k\varphi\varphi_0) z(k'\varphi'\varphi_0) e^{r(\varphi' - \varphi)/l}, \\ z(k\varphi\varphi_0) = \cos[k(v_x(\varphi) - v_x(\varphi_0)) / \Omega].$$

The integration with respect to  $\varphi$ ,  $\varphi'$ , and  $\varphi_0$  can be easily carried out by the saddle point method. The saddle points are  $\varphi_0 = 0$  and  $\pi$ .

We consider first hopping electrons, i.e., the point  $\varphi_0 = 0$ . The first term in the curly brackets, in which the important role is played by small  $\varphi_0 \sim (\delta/r)^{1/2}$ , makes the principal monotonic part of the conductivity. At small  $\varphi_0$

$$1 - [1 - q(\varphi_0)] e^{-2r\varphi_0/l} = \frac{2r\varphi_0}{l} \left( 1 + \frac{l}{r} Q \right) \sim n^{-1} + q(\varphi_0) \ll 1. \quad (22)$$

The monotonic part of the conductivity

$$\sigma_{ij}^{(0)}(kk') = (1 + lQ_i/r)^{-1} \sigma_{ij}^m(kk') \quad (23)$$

differs by a factor  $(1 + lQ_i/r)^{-1}$ , which depends on the on the magnetic field and on the mean free path, on the specular-reflection conductivity  $\sigma_{ij}^m(kk')$ . The quantity  $\sigma^m$  is defined by the integral

$$\sigma_{ij}^m(kk') = \delta_{ij} \frac{3\sigma_0 r}{16\pi v_0^4} \int_{-\varphi_0}^{\varphi_0} v_\perp v_z^2(\varphi=0) dv_z \sum_{\pm k, k'} \int_0^\pi d\varphi_0 \int_{-\varphi_0}^{\varphi_0} d\varphi \int_{-\varphi_0}^{\varphi_0} d\varphi' \\ \times \exp[i\lambda(\varphi^2 - \varphi_0^2) - i\lambda'(\varphi'^2 - \varphi_0^2)]; \\ \lambda = kv_\perp / 2\Omega, \quad \lambda' = k'v_\perp / 2\Omega.$$

After making the substitutions  $\varphi = \varphi_0 x$  and  $\varphi' = \varphi_0 y$  we calculate first the integral with respect to  $\varphi_0$ . The subsequent integrations with respect to  $x$ ,  $y$ , and  $v_z$  are trivial:

$$\sigma_{ij}^m(kk') = \delta_{ij} \frac{3\sigma_0 r^{-1/2}}{2^{1/2} \pi^{1/2}} \int_0^\infty \frac{v_z^2(0) dv_z}{v_0^{3/2} v_\perp^{1/2}} \sum_{\pm k, k'} \int_0^1 \frac{dx dy}{[-ik(x^2 - 1) + ik'(y^2 - 1)]^{1/2}} \\ = -\delta_{ij} A c_i \sigma_0 r^{-1/2} (kk')^{-1/2} (|k+k'|^{-1/2} - |k-k'|^{-1/2}), \quad (24)$$

where

$$c_i = 1, \quad c_y = 3/2, \quad A = 3 \cdot 2^{1/2} \pi^2 / 5 \Gamma^2(1/4).$$

Since  $Q \sim v_\perp$  in (22) [see (4)], (23) is an interpolation formula. After integrating with respect to  $v_z$  we see that the value of  $Q_i$  in (23) is smaller by approximately a factor  $\pi^{1/2}$  for the case  $lQ_i \gg r$  than for the opposite case. In addition,  $Q_i$  depends on the field polarization:  $Q_y \approx 2Q_x$ . We shall neglect these quantities hereafter.

The last two terms in (21) lead to the oscillating part

$$\sigma_{ij, \text{osc}}^s(kk') = \delta_{ij} \frac{3\sigma_0}{2\pi l} \operatorname{cth} \frac{\pi r}{l} \int_0^\pi \frac{v_z^2(0) dv_z}{v_0^2 v_\perp} \\ \times \sum_{\pm k, k'} \int_0^1 dx \int_{-\infty}^{+\infty} dy [k'(y^2 - 1) - k(x^2 - 1) + i0]^{-2} \\ = -\frac{3\pi^2 \sigma_0}{8l|k|} \operatorname{cth} \frac{\pi r}{l} [\delta(k-k') + \delta(k+k')] \delta_{ij}. \quad (25)$$

To obtain the last expression it is convenient first to integrate with respect to  $y$  and then with respect to  $x$  and symmetrize the result relative to  $k$  and  $k'$ .

We see that following the integration in (15) expression (25) cancels out completely the contribution of the point  $\varphi = 0$  to the bulk conductivity (20). This fundamental cancellation should always take place under conditions of cyclotron resonance. It has a simple physical meaning: the point  $\varphi = 0$  cannot contribute to the cyclotron resonance, inasmuch as on the  $s$  orbits such electrons are nonresonant, and on the  $v$  orbits they move outside the skin layer, and any point, except  $\varphi = \pi$ , has an equally probable position on such a trajectory.

The oscillating term (20) is large compared with the monotonic (23) if the following condition is satisfied:

$$|(\delta r)^{1/2} l^{-1} (1 + Ql/r) \operatorname{cth}(\pi r/l)| \gg 1. \quad (26)$$

The coefficient of the hyperbolic cotangent is under our conditions (8) and (8a) a small parameter. The ratio of the scattering by the surface and in the interior is given by the quantity  $Ql/r$ . At a large number of impact, the influence of the surface prevails is

$$Q|l|/r = nq(Qr) \gg 1.$$

Thus, the condition (26) can be satisfied only if  $\Omega\tau \gg 1$  in a small vicinity of the resonance points  $\omega = N\Omega$ , and it can be expressed in the form

$$\frac{\omega\tau}{\pi} \left(\frac{\delta}{r}\right)^{1/2} \left[ \frac{1 + (Q\Omega/\omega)^2}{1 + \tau^2(\omega - N\Omega)^2} \right]^{1/2} \gg 1. \quad (26')$$

We consider now the contribution of the glancing electrons with  $\varphi_0 \approx \pi$  to the "surface" conductivity (21). The factor preceding the first integral in the curly brackets takes in the vicinity of the point  $\varphi_0 = \pi$  ( $\varphi_0 = \pi - \psi_0, \psi_0 \sim (\delta/r)^{1/2}$ ) the form

$$[e^{2\pi r/l} - 1 + 2Q\psi - 2r\psi/l]^{-1} e^{2\pi r/l}. \quad (22')$$

The subsequent calculations are different in the weak and strong resonance cases distinguished by the condition (26). If the resonance is weak, then we must omit from (22') the terms that depend on the angle  $\psi$ . The terms in the curly brackets of (21) are then effectively cancelled out, and we get an integral with respect to  $\varphi'$  from  $\varphi_0$  to  $\pi$  (and an analogous integral from  $-\pi$  to  $-\varphi_0$ ) with a resonant denominator. The change of variables  $\varphi = \pm(\pi - \psi)$  yields an integral that differs from (25) in the integration limits, which are from 1 to  $\infty$  with respect to  $x$  and from 0 to 1 with respect to  $y$ . Calculating this integral and symmetrizing relative to  $k$  and  $k'$ , we verify that it vanishes. This means that the correction of the resonant value of the conductivity  $\frac{1}{2}\sigma^v$  is small in terms of the parameter (26) under the conditions of weak resonance.

Thus, the conductivity under conditions of weak resonance is determined by the relations

$$\sigma_{ij}(k) = \frac{1}{2}\sigma_{ij}^v(k); \quad \sigma_{ij}(kk') = (1 + Ql/r)^{-1} \sigma_{ij}^m(kk'), \quad (27)$$

where  $\sigma^v$  and  $\sigma^m$  are given by (20) and (24), and the estimate of  $Q$  follows from (4).

If the resonance is strong, then in a small vicinity of the resonance, where the condition (26) is satisfied, we must set the exponentials in (22') equal to unity. The contribution of the point  $\varphi_0 = \pi$  to the first term in the curly brackets is given by the integral

$$\begin{aligned} \sigma_{ij}^m(kk') &= \delta_{ij} \frac{3\sigma_0 r^{-1/2}}{2^{1/2} \pi^{1/2} (-1 + lQ/r)} \sum_{x, k, k'} \int_0^\infty \frac{dv_z}{v_0^{1/2} v_\perp^{1/2}} \int_0^\infty \frac{dx dy}{[-ik(x^2 - 1) + ik'(y^2 - 1)]^{1/2}} \\ &= -\frac{Ac_i \sigma_0 r^{-1/2}}{\pi (-1 + lQ/r) k^h w^h} \left[ \frac{1}{(1+u)^h} \ln \frac{[1 + (1+u^{-1})^h][1 + (1+u)^h]}{[1 - (1+u^{-1})^h][1 - (1+u)^h]} \right. \\ &\quad \left. - \frac{1}{(1-u)^h} \left( \ln \frac{1 + (1-u)^h}{1 - (1-u)^h} + 2 \operatorname{arctg}(u^{-1} - 1)^h \right) \right], \quad (28) \end{aligned}$$

where  $u = k'/k$ . The second term in the curly brackets makes no contribution, as can be verified by a calculation similar to (25). We see that the conductivity in the vicinity of a strong resonance is given by

$$\sigma_{ij}(k) = \frac{1}{2}\sigma_{ij}^v(k), \quad \sigma_{ij}(kk') = \sigma_{ij}^v(kk') + \sigma_{ij}^m(kk'), \quad (29)$$

where  $\sigma^v$ ,  $\sigma^o$ , and  $\sigma^m$  are given by (20), (23), (24), and (28).

The most typical experimental situation is described by (27), when the condition (26) does not hold and the resonance is weak. In the principal approximation, the impedance depends monotonically on the magnetic field. Equations (15) and (16) are made dimensionless with the aid of the parameter

$$\begin{aligned} k_i &= \left[ \frac{4\pi\omega\sigma_0}{ic^2} \frac{Ac_i}{r^h (1 + lQ/r)} \right]^{1/2} = \left[ \frac{\omega_p^2 \Omega^h Ac_i}{c^2 v_0^h} \right]^{1/2} [1 + (1/\omega\tau + Q\Omega/\omega)^2]^{-1/2} e^{-2i\theta/5}, \\ \theta &= \operatorname{arctg}(1/\omega\tau + Q\Omega/\omega); \quad \omega_{p1} = (4\pi N_e e^2/m)^{1/2}, \quad (30) \end{aligned}$$

where  $\omega_{p1}$  is the plasma frequency. With the aid of (30) we obtain the skin-layer thickness  $\delta_i = |k_i|^{-1}$ .

The monotonic part of the impedance is

$$Z_i^{(0)} = g_1 4\pi\omega/ic^2 k_i. \quad (31)$$

The small oscillating increment is calculated by the perturbation method:

$$\begin{aligned} Z_i^{(1)} &= Z_i^{(0)} g_2 \frac{3\pi\omega}{8Ac_i} \left(\frac{\delta_i}{v_0\Omega}\right)^{1/2} \left[ 1 + \left(\frac{1}{\omega\tau} + \frac{Q\Omega}{\omega}\right)^2 \right]^{1/2} \\ &\quad \times \operatorname{cth}\left(\frac{\pi r}{l}\right) \exp\left(i\frac{\pi}{2} + i\frac{6}{5}\theta\right). \quad (32) \end{aligned}$$

The constant  $g_1$ , and with it also  $g_2$ , can be found by the Harman-Luttinger method<sup>9</sup>:

$$\begin{aligned} g_1 &= \left(\frac{5\pi}{4}\right)^{1/2} \pi / \Gamma^2\left(\frac{2}{5}\right) \sin\frac{2\pi}{5} = 0.88, \\ g_2 &= \left(\frac{2}{5\pi}\right)^{1/2} \frac{\sin(2\pi/5)}{2^{1/2}} \int_0^\infty x \operatorname{cth}\frac{\pi x}{2} \\ &\quad \times \left| \Gamma\left(\frac{1+2ix}{5}\right) \Gamma\left(\frac{3+2ix}{5}\right) \right|^2 = 0.65. \quad (33) \end{aligned}$$

In the opposite limiting case when the condition (26) can be satisfied, the resonance is strong. In the vicinity of the resonance it is necessary to use formulas (29) for the conductivity, and the principal term is  $\frac{1}{2}\sigma^v$ . We obtain the corresponding values of the reciprocal of the penetration depth

$$\begin{aligned} k_0 &= \left[ \frac{3\pi\omega_{p1}^2 \omega}{4c^2 v_0} \right]^{1/2} [1 - e^{-2\pi r/l}]^{-1/2} \\ &= \left(\frac{3\omega_{p1}^2 \omega}{8c^2 v_0}\right)^{1/2} \left(\frac{1}{\Omega\tau} - i\frac{\omega - N\Omega}{\Omega}\right)^{-1/2} \quad (34) \end{aligned}$$

and the impedance

$$Z^{(0)} = \frac{8\pi\omega}{3^h c^2 k_0} (1 - i3^h). \quad (35)$$

The correction due to the second expression of (29)

is

$$Z_i^{(1)} = \frac{2^2 \omega^2}{c^4} \int_0^{\pi} \sigma_i(kk') \left[ k^2 - \frac{4\pi i \omega}{c^2} \sigma_i(k) \right]^{-1} \left[ k'^2 - \frac{4\pi i \omega}{c^2} \sigma_i(k') \right]^{-1} dk dk'$$

An estimate of this integral

$$Z_i^{(1)} \sim Z_i^{(0)} \frac{lk_0^{1/2}}{r^{1/2} \text{cth}(\pi r/l)} \left( \frac{g_3}{-1+lQ/r} + \frac{g_4}{1+lQ/r} \right)$$

shows that it is small to the extent that condition (26) holds;  $g_3$  and  $g_4$  are coefficients of the order of unity.

#### 4. LARGE EFFECTIVENESS ANGLES $\varphi_f \gg \varphi_w$

In this case the integrand in (13) must be expanded in powers of  $p' - p$  in analogy with (6) and (7):

$$(e^{2\pi\varphi/l} - 1) f - Q_i p_x (p_x \Delta p_x f + 2\nabla f \nabla p_x) = \int_{-\varphi}^{\varphi} d\varphi' u(\varphi', \varphi) e^{i(\varphi' + \varphi)/l}. \quad (36)$$

We were unable to obtain the solution of this equation. It can be estimated, however, without difficulty. The rate of change of  $f$  with changing angle  $\varphi$  is determined by the right-hand side, the characteristic angle interval of which is  $\varphi_f \sim (\delta/r)^{1/2}$ . Therefore an estimate of the differential term yields

$$Q_i \partial^2 f / \partial \varphi^2 \sim Q_i (r/\delta) f.$$

For hopping electrons  $\varphi \sim \varphi_f \ll 1$ , and we obtain in the left-hand side of (36)

$$2 \frac{(r\delta)^{1/2}}{l} (1 + Q_i l r^{1/2} \delta^{-1/2}).$$

The influence of the roughness is determined by multiplying the number  $n$  of the impact by the diffuseness  $Q_i r/\delta$  in one collision with the surface. The contribution of the hopping electrons to the conductivity is thus

$$\sigma^0 = \delta^{-1} \sigma(k, k' \sim \delta^{-1}) = \sigma_0 (\delta/r)^{1/2} (1 + Q_i l r^{1/2} \delta^{-1/2})^{-1}. \quad (37)$$

The factor  $(\delta/r)^{1/2}$  is determined by the smallness of the angle interval of the effective electrons.

The distribution function of the glancing electrons that collide with the surface ( $\varphi \sim \pi - (\delta/r)^{1/2}$ ) contains the factor

$$\{e^{2\pi\varphi/l} - 1 + [Q_i r/\delta - 2(r\delta)^{1/2} l^{-1}]\}^{-1}.$$

The quantity in the square brackets is small in our approximation [see (8b) and (8a)]. We therefore encounter here the same two cases of weak and strong resonance, distinguishable by the condition (27), in which, however, we must make the substitution

$$Q \rightarrow Q_i (r/\delta)^{1/2}. \quad (38)$$

If the resonance is weak, then the monotonic part of the conductivity is given by (37), and the oscillating part is equal to  $\frac{1}{2}\sigma^0$  [see (20)]. In the opposite case the term  $\frac{1}{2}\sigma^0$  is the principal one, while the reciprocal depth of penetration and the impedance are given by (34) and (35).

#### 5. DISCUSSION OF RESULTS

We now compare our results with those obtained in the traditional approach in which the phenomenological diffuseness coefficient  $q$  is used,<sup>6</sup> and which leads to the following qualitative formula

$$\delta^{-3} = \frac{4\pi\omega\sigma_0}{ic^2 l} \left\{ \text{cth} \frac{\pi r}{l} + \left[ \frac{(\delta r)^{1/2}}{l} + \frac{q}{2-q} \right]^{-1} \right\}, \quad (39)$$

from which we determine  $\delta$  and then the impedance  $Z = 4\pi\omega\delta/ic^2$ . The expression in the curly brackets is the ratio of the total conductivity to  $\sigma_0\delta/l$ . Therefore adding for  $k$  and  $k' \sim \delta^{-1}$

$$\sigma(k) + \delta^{-1} \sigma(kk')$$

and taking (27) and (37) into consideration, we see that the expression in the square brackets of (39) must be replaced by

$$(\delta r)^{1/2} / l + (\delta/r)^{1/2} Q, \quad \text{if } p_0 d \ll r/\delta, \quad (40a)$$

$$(\delta r)^{1/2} / l + (r/\delta) Q_i, \quad \text{if } p_0 d \gg r/\delta. \quad (40b)$$

We note that both terms in (40) are assumed small within the framework of our treatment, but their ratio can be arbitrary.

Thus, the quantity named the diffuseness coefficient by Kaner *et al.*<sup>6</sup> actually depends on the magnetic field and on the depth of the skin layer. This is the consequence of the dependence of the surface scattering on the glancing angle. The very strong dependence on the proximity of the diffuseness coefficient  $q$  to zero thus vanishes. The ratio of the two terms of (40), which is determined by the competition between the scattering in the bulk and on the surface, is for example in the case (40a), of the order of  $lQ/r$ . For a surface with  $ap_0 \sim 10$  and  $p_0 d \sim 10^2$  we have  $Q \sim 10$ , and this competition is perfectly realistic.

The ratio of  $\text{coth}(\pi r/l)$  to the term in the square brackets in (39) is determined by our parameter (26), and can be neglected everywhere except in a narrow vicinity of the resonance at very large  $\Omega\tau$ . Leaving out this ratio, we obtain  $\delta$ . Its plot is shown in Fig. 3 for the most general case. Region I corresponds to a weak magnetic field and is beyond the scope of our treatment, since condition (8) is not satisfied here. In region II the effectiveness angles are small,  $\varphi_f \ll \varphi_w$ , and the order of magnitude of the impedance is

$$Z^{(0)} = \frac{4\pi\omega}{ic^2} \left( \frac{ic^2 v_0 (Q+r/l)}{\omega \omega_p r^{1/2}} \right)^{1/2}; \quad l^{-1} = \frac{-i\omega + \tau^{-1}}{v_0},$$

while its exact value is given by (30) and (31). Region III corresponds to relative large effectiveness angles  $\varphi_f \gg \varphi_w$ , and the impedance here is

$$Z^{(0)} = \frac{4\pi\omega}{ic^2} \left( \frac{ic^2 v_0}{\omega \omega_p l^2} r Q_i \right)^{1/2}.$$

Region IV corresponds to strong magnetic fields, which are not considered here, with the orbit located inside the skin layer.

At weak resonance, when (39) must be expanded in powers of the hyperbolic cotangent, the oscillating in-

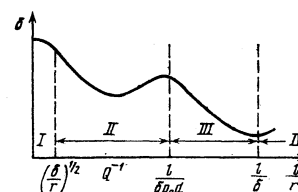


FIG. 3.

crement is given by (32), with the substitution (38) necessary in the region  $\varphi_f \gg \varphi_w$ . A strong resonance is described in a somewhat more complicated manner. Outside the narrow resonance region, the resonance is weak as before. In the immediate vicinity of the resonance, however, where the condition (26) is satisfied, it is necessary to use Eqs. (34) and (35).

We note in conclusion that the study of the anomalous skin effect in a magnetic field has been shown here to be a most effective method of investigating the surface scattering of conduction electrons. The calculations presented apply to an isotropic metal. This restriction, however, is not essential, since the current is determined by a narrow "strip" on the Fermi surface, where  $v_x = 0$ . The specularity of reflection of such electrons and the vanishing of the diffuseness coefficient in accord with Eq. (4) are simple consequences of the indistinguishability of the electronic states near this strip. In the other limiting case ( $\varphi_f \gg \varphi_w$ ) we used in general form only the sharp directivity of the scattering indicatrix.

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## Orientalional phase transitions in ammonium bromide at high pressures

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The phase diagram and isobaric heat capacity of  $\text{NH}_4\text{Br}$  are investigated up to 2.65 kbar using an improved adiabatic calorimetric technique. The pressure is maintained constant to within  $10^{-2}$  bar. The coordinates of the triple point are  $T_{tr} = 203.35 \pm 0.15$  °K and  $P_{tr} = 1730 \pm 20$  bar. The coordinates of the tricritical point on the  $\delta$ - $\beta$  transition line are  $T_{ic} = 207.9 \pm 0.3$  °K and  $P_{ic} = 2250 \pm 35$  bar. The heat of the  $\delta$ - $\gamma$  transition is found to vanish in the vicinity of the triple point, which can thus be regarded as a "distorted" bicritical point.

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### I. INTRODUCTION

A large number of experimental<sup>1-13</sup> and theoretical<sup>14-18</sup> papers are devoted to the study of the phase transitions in ammonium halides. Ammonium halides are of interest because of the nature of the phase transitions in these compounds changes with pressure. Ammonium halides have all important advantage over ferroelectrics and magnets: they have no depolarizing and demagnetizing fields complicating the interpretation of the experimental data. Ammonium bromide is especially attractive; in this compound, there are three types of orientational phase transitions in a relatively accessible pressure range.

Two orientations of the  $\text{NH}_4^+$  tetrahedron are possible within the crystal lattice. On the  $\text{NH}_4$  phase diagram (Fig. 1) there are the orientationally disordered  $\beta$  phase, the  $\delta$  phase with parallel ordering of the ammonium

ions, and the  $\gamma$  phase with antiparallel ordering. As has been shown<sup>14-18</sup> both types of ordering are due to the ammonium octupole interaction: direct interaction leads to parallel ordering, and indirect interaction (through the hydrogen-halogen dipole) leads to antiparallel ordering. The interaction potentials depend primarily on the polarizability of the halide ion, which is a strong function of pressure.

Previously available data on the  $\text{NH}_4\text{Br}$  phase diagram have been very crude. Thus, in Ref. 6 the pressure at the tricritical point<sup>1)</sup> on the  $\delta$ - $\beta$  transition line is indicated to be in the interval 2.5-3.65 kbar, and the uncertainty in the triple point coordinates amounted to several hundreds of bars. We have set ourselves the task of significantly refining the nature of the phase diagram and investigating the isobaric heat capacity of  $\text{NH}_4\text{Br}$  up to pressures of 2.6 kbar with an accuracy that is no