Magnetoresistance of filamentary cadmium single crystals at 4.2°K

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Results are presented of measurements of the magnetoresistance of thin single crystal samples (whiskers) of cadmium at 4.2°K under conditions of a strong size effect [$l^\infty(4.2°K) \gg d$]. The sample thickness d is between 0.5 and 4.5 μ; the electron mean free path in bulky samples $l^\infty(4.2°K) \approx 200$ μ. For a qualitative comparison of the results with the static skin-effect theory, [1-3] it is assumed that the specular reflection coefficient of the surface depends on the angle of incidence of the electrons on the surface, $p = p(\alpha)$. At weak magnetic field strengths the experimental dependences can then be attributed to a strong influence of specular interaction between the electrons and surface. Satisfactory agreement between the results and the theory in the region of strong magnetic field strength can be obtained if the Fuchs parameter P is assumed to be close to zero.

For the purpose of observing the irregularities predicted by the theory of the static skin effect[1-3], we have investigated filamentary single crystal (whiskers) of zinc[4], having thicknesses d on the order of several microns and $l^\infty(4.2°K) \approx 300$ μ (l^∞ is the electron mean free path in the bulky sample). It was observed that in strong magnetic fields H (2r < d, where r is the average radius of the electron orbit in the magnetic field) the behavior of the transverse magnetoresistance ($\rho^d(H)$ at $\mathbf{H} \perp \mathbf{J}$, where J is the measuring current) can be described qualitatively by the theory of the static skin effect. In weak magnetic fields (2r > d), the experimental results do not agree with the theory of[2], since the resistance $\rho^d(H)$ increases like $H^{2/3}$ in a wide range of magnetic fields. It was suggested that this behavior may be due to specular interaction of the electrons with the surface and to the ensuing quantum surface levels[5].

The purpose of the present paper is to confirm the existence of an unusual behavior of the magnetoresistance of tin samples ($d \ll l^\infty$) in another metal, which we chose to be cadmium, and to present a more realistic physical treatment of the results.

I. SAMPLES AND MEASUREMENT PROCEDURE

Cadmium whiskers in the form of filaments and plates were grown by the method of Sears and Coleman[6]. The initial purity of the material was characterized by a resistivity ratio $\rho(300°K)/\rho(4.2°K) = 7 \cdot 10^3$, corresponding to a mean free path $l^\infty(4.2°K) = 200$ μ. The electric wiring of the samples was the same as in[4]. The distance between the potential contacts of the samples was 400–500 μ. The sample thickness was determined from the resistance at room temperature, when the size effect can be neglected. In the case of filamentary whiskers it was assumed that $d = \sqrt{S}$ (S is the cross section area of the whisker), and for plates $d = S/\Delta$ (Δ is the width of the plate as determined under the microscope). The thicknesses of the investigated samples were 0.5–4.5 μ. Since the width of the potential contact was ~50 μ, the accuracy with which the thickness d was determined should be regarded as being of the order of 20%.

In conditions of strong size effect, $l^\infty \gg d$, the resistance is determined by the product $\rho^\infty l^\infty$, the thickness of the sample, and the specularity of its surface. The specularity of a surface is customarily described by a constant number P ($0 \le P \le 1$) called the specularity coefficient or the Fuchs parameter (for more details see Sec. III). It was shown earlier[7] that P = 0.4–0.6 for cadmium whiskers. The growth conditions and the purity of the initial material were the same in our case as in[7]. A criterion of "faultless" mounting of the samples was taken to be the correspondence (within 20%) of the sample resistivity at 4.2°K to the value P = 0.5 and to the average value $\rho^\infty l^\infty = 2 \times 10^{-11}\,\Omega\text{-cm}^{-2}$. (We note that introduction of physical defects into the sample influences strongly only its residual resistance, and hardly affects the magnetoresistance.)

Thus, the cadmium whiskers satisfy all the conditions necessary for a successful observation of the influence of the sample surface on their magnetoresistance[4], namely: 1) cadmium is a "good" metal with equal numbers of electrons and holes, i.e., the variation of the resistivity of the bulk cadmium samples is given by $\rho^\infty(H) \propto H^2$; 2) the experiments are performed under conditions of strong size effect $l^\infty(4.2°K) \gg d$; 3) the samples have a high specularity coefficient P = 0.5.

The measurement procedure was the same as in[4]. The maximum magnetic-field intensity of the superconducting solenoid reached 70 kOe in the experiments.

II. MEASUREMENT RESULTS

1. Determination of the Orientation of the Whisker Axes

A direct determination of the orientations of the whisker axes is a difficult methodological problem. However, since the main results do not depend on the orientation, we confine ourselves, as in the case of zinc[4], to an indirect determination method. To this end we used the anisotropy of the sample resistance in a magnetic field H = 60 kOe, which can be regarded as strong (2r ≪ d) for almost all the measured samples. It was shown in[8] that in spite of the strong size effect, the main features of the resistivity anisotropy in a strong magnetic field should be preserved for metals with open Fermi surfaces.

Cadmium has an open Fermi surface[9]. The anisotropy of the magnetoresistance of bulky samples of cadmium was investigated in a number of studies[9-11]. A comparison of the anisotropy of the resistance of the whiskers with the anisotropy of bulky samples makes it possible to draw some conclusions concerning the

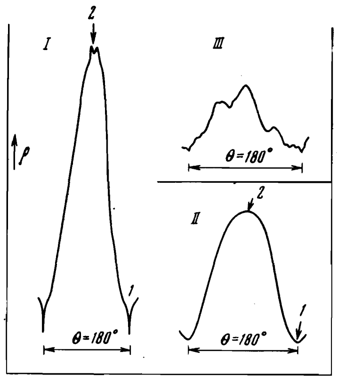

FIG. 1. Anisotropy of transverse magnetoresistance of Cd whiskers in a field H = 60 kOe at T = 4.2°K. The arabic numerals mark the directions for which the magnetoresistance was investigated in greatest detail.

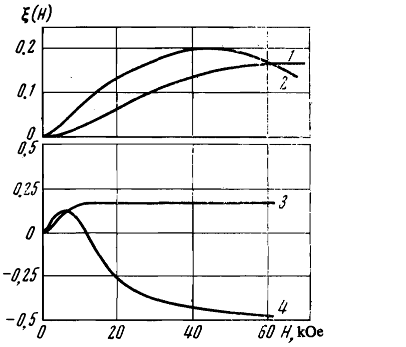

FIG. 2. Longitudinal magnetoresistance Cd whiskers, T = 4.2°K; 1–d = 0.5 μ, 2–d = 1 μ, 3–d = 4.5 μ, 4–d = 2 μ, Δ = 9 μ (Δ is the width of the plate).

orientation of the whisker axes. The problem is facilitated by the fact that the whiskers grow only along certain selected directions (three or four directions) with a low sum of crystallographic indices. It is known from the published data, e.g., that the axis of zinc whiskers lie in the $(0\bar{1}10)$ plane and have directions $[\bar{2}110]$, $[\bar{2}111]$, $[\bar{2}112]$ and $[\bar{2}113]$[12,13]. With respect to cadmium whiskers it is known that at the very least the directions $[\bar{2}110]$ and $[\bar{2}113]$ are realized for them[13,14].

All the whiskers of cadmium investigated by us belong to three anisotropy groups. Typical rotation diagrams $\rho^d_H(\theta)$ for each of them are shown in Fig. 1 (θ is the angle of rotation of the magnetic field in a plane perpendicular to the whisker axis).

The first type of anisotropy is possessed by all plate-like and some filamentary whiskers. Comparison with bulky samples shows that the axis of these whiskers are parallel to the (0001) plane, and seem to have the orientation $[\bar{2}110]$. The second type, with weaker resistivity anisotropy, can be assigned an orientation $[\bar{2}111]$. Finally, the third type, with still weaker anisotropy but having a larger number of details, should be connected with a whisker axis orientation along $[\bar{2}113]$. Regardless of the whisker-axis orientation, the magnetic field is parallel to the (0001) plane for the peak resistivity minima marked 1 in Fig. 1.

To study the magnetoresistance we used only whiskers having a resistance anisotropy of the first type (plates) and of the second type (filamentary whiskers).

2. Longitudinal Magnetoresistance

The measurements have shown that the magnitude and character of the longitudinal resistivity of the Cd whiskers, as in the case of zinc, depend strongly on the angle between the field and the current. We therefore used the same precautions to attain the condition H ∥ J as in[4]. Nonetheless, the results still had a noticeable scatter. Therefore the curves shown in Fig. 2 should be regarded only as a qualitative illustration of the behavior of the longitudinal resistance, the main feature of which reduce to the following: The initial growth of the resistivity is always given by

$$\xi(H) = \frac{\Delta\rho^d(H)}{\rho^d(0)} = \frac{\rho_d(H)-\rho^d(0)}{\rho^d(0)} \propto H^n,$$

where $n \approx 2$. The exponent n then begins to decrease to $n = 0$, which is reached at the maximum of the resistivity in a field $H = H_{max}$. The resistivity increment at the maximum is approximately 10–20% of the resistivity in $\rho^d(0)$ in a zero field. Owing to a large scatter of the results, we were unable to find a regular dependence of H_{max} on the thickness. It can only be stated that the product H^d_{max} for ranges from 4 to 6 kOe-cm for whiskers with axis along $[\bar{2}111]$ and from 1.5 to 3.0 kOe-cm for whiskers along $[\bar{2}110]$. The resistivity starts to decrease after the maximum is reached. There were samples, however, which were faultless from the point of view of wiring and orientation in the field, but whose resistivity remains constant in fields $H > H_{max}$ (Fig. 2, curve 3).

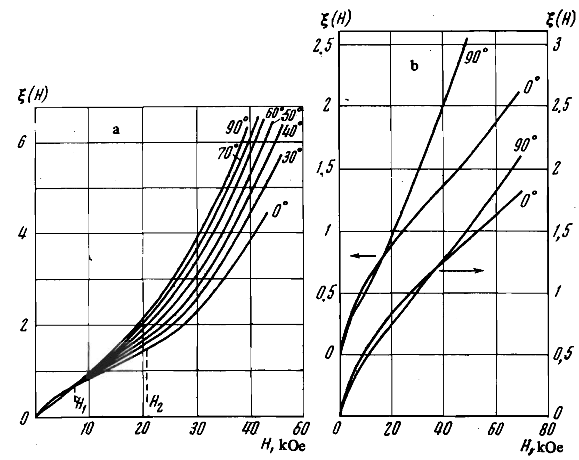

FIG. 3. Transverse magnetoresistance of filamentary cadmium whisker, T = 4.2°K; the curves are marked with the angle between the direction 1 (see Fig. 1, type II) and the magnetic field. a–d = 1.9 μ, b–d = 0.9 μ for the upper two curves (left-hand ordinate scale), and d = 0.5 μ for the lower two curves (right-hand ordinate scale).

3. Transverse Magnetoresistance

The irregularities observed in the behavior of the magnetoresistance of zinc whiskers[4] were observed also in cadmium and were most strongly pronounced in the thinnest of the measured samples. Just as in zinc, the main features of the $\Delta\rho^d(H)/\rho^d(0) \equiv \xi(H)$ curves at $H \perp J$ do not depend on the shape of the sample or on the orientation of the field relative to the crystallographic directions. Figures 3–5 show the results of the measurements for the thinnest cadmium whiskers.

The general form of the $\xi(H)$ dependence in different magnetic-field intervals can be described in the following manner:

1) In the initial magnetic-field region $0 < H \leq H_0$, the resistivity increases like $\xi(H) \propto H^n$ with $n = 2$.

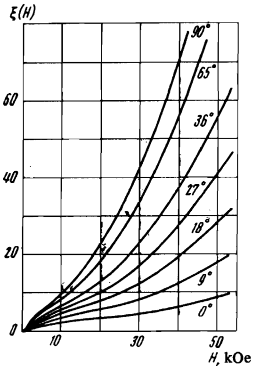

FIG. 4

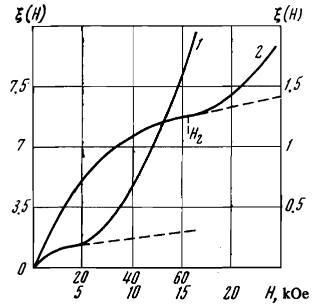

FIG. 5

FIG. 4. Transverse magnetoresistance of Cd plate whisker ($d = 2\,\mu$, $\Delta = 143\,\mu$), T = 4.2°K. The curves are marked by the angles between the direction 1 (see Fig. 1, type I) and the magnetic field.

FIG. 5. Curve from Fig. 4 for the 0° direction in enlarged scale: 1—ordinate axis on the left, values of magnetic field on the upper scale. 2—ordinates on the right, magnetic field values on the lower scale.

For samples of thickness in the micron range this region ends at approximately $H_0 = 1$ kOe, and is not discernible in the figures.

2) $H_0 \leq H \leq H_1$. For all the measured samples we can state that $\xi(H) \propto H^n$ with $n < 1$. For the thinnest samples with $d \leq 2\,\mu$, the $\rho(H)$ curves agree best with $n = 2/3$.

3) $H_1 \leq H \leq H_2$. The $\xi(H)$ dependence can be adequately approximated by the straight line

$$\xi(H) = \xi(H_1) + a(H - H_1). \quad (1)$$

4) $H > H_2$. In this region the resistance can be represented as a sum of three terms (independent of the field, linear, and quadratic in the field):

$$\xi(H) = \xi(H_2) + a_1(H - H_2) + b(H - H_2)^2. \quad (2)$$

We note a few other features of the $\xi(H)$ curves.

a) H_1 and H_2 do not depend on the field orientation and differ from each other in general by a factor 2—3. For platelike whiskers with $[\bar{2}110]$ direction and with H ∥ (0001) (field parallel to the plate surface) the values of H_1 and H_2 are practically equal. Therefore the $\xi(H)$ curves have an inflection point in this case (Fig. 5).

b) The product $H_1 d$ for samples of different thickness and orientation lies in the range 3—5 Oe-cm at H ∥ (0001).

c) The slope of the linear section of the resistance growth in the interval from H_1 to H_2 always coincides with the slope of the linear component of the resistance in fields $H > H_2$, i.e., $a = a_1$ in (1) and (2).

d) For $[\bar{2}111]$ whiskers, the resistance anisotropies in weak and strong fields have opposite signs. This is manifest, in particular, in a crossing of the $\xi(H)$ curves in Fig. 3.

III. DISCUSSION OF RESULTS

We note first that the results obtained for cadmium agree with those obtained for zinc[4], and we can therefore conclude that the scattering of the electrons by the surface is indeed predominant in cadmium samples of micron thicknesses.

In a longitudinal magnetic field, just as for zinc, the resistivity increases, goes through a minimum, and then tends to a constant value with increasing field. The question of the limiting resistivity remains open since, on the one hand, the condition $r \ll d$ is not reached in our experiment, and on the other hand there is never assurance that the condition J ∥ H is satisfied with sufficient accuracy in a given measurement.

The product $H_{max}d$ for samples of different thickness and shape is equal to several units of Oe-cm. We have noted earlier[4] that these results do not agree with the conclusion of Azbel's qualitative theory[2], which predicts a growth of filament resistivity in weak fields a maximum plate resistivity at $r \approx \sqrt{ld}$. Inasmuch as $l/d \approx 10^2$ in our experiments, this maximum should have been observed at $r \gg d$. The dimensions of the Fermi surface of cadmium allow us to assume that the average radius of the electron orbits in a magnetic field correspond to a product Hr on the order of several units of Oe-cm. This means that the maximum is observed under the condition $r \approx d$. This disagreement with the theory can be attributed to the appreciable influence of the character of reflection from the boundaries on the resistivity in weak fields. The behavior of longitudinal magnetoresistance of thin plates in the presence of partial specular reflection of the electrons from the surface was computer-calculated recently[15] using a very simple isotropic model of the Fermi surface. It was found that the position of the resistance maximum shifts towards stronger fields when the fraction of the specular reflection is increased.

Although the shape of the Fermi surface of cadmium is far from spherical, it is of interest to present, for comparison, the results of a numerical calculation under conditions close to the experimental ones. Thus, at $l/d = 10^2$ and $P = 0.8$ the maximum should be located at $d/r \approx 0.3$, with a ratio $\rho^d(H_{max})/\rho^d(0) = 1.15$. These values are close to the experimental ones (Fig. 2, curve 4). Unfortunately, there are no analogous calculations for filaments at present.

From the quantitative and qualitative points of view, the results for the transverse magnetoresistance of zinc and cadmium whiskers coincide. Therefore, just as for zinc, we assume that the field H_2 corresponds to the condition $2r = d$.

For a physical interpretation of the behavior of $\rho^d(H)$ near strong magnetic fields, which was determined in[4] for zinc whiskers, it was assumed that the surface quality of a sample can be described by the constant number P ($0 \leq P \leq 1$) (the Fuchs parameter), which can be interpreted as the probability of specular reflection and is independent of the angle of incidence of the electrons on the surface[16]. This has shown that for $H > H_2$ the linear component of the magnetoresistance in (2) is connected with the "specular" reflection and is proportional to the coefficient P, while the quadratic component is connected with the "diffuse" reflection and is proportional to $1 - P$. From the experimental values of $\rho^d(H)$ we obtained for zinc $P \approx 0.5$ for filaments and plates. This value agrees with the coefficient P determined by another method[17]. The same value of P can be obtained also for filamentary cadmium whiskers.

However, the results for cadmium plate whiskers cannot be described with the aid of the Fuchs parameter. Indeed, from the ratio of the linear and quadratic parts of the $\xi(H)$ curve in strong fields (Fig. 5) it can be concluded that the parameter P is smaller for plates than for filaments. This seems little likely[17]. Moreover, from the form of $\xi(H)$ in weak fields $(2r > d)$ it follows that the "specularity" is larger for plates than for filaments (the field H_1 and H_2 practically coincide (Fig. 5)). This forces us to modify the physical interpretation of the results in the region of strong fields.

This modification is based on a more realistic approach to the allowance for the interaction of the electrons with the metal surface. We take this interaction into account with the aid of the relation $p = p(\alpha)$, where $p(\alpha)$ is the probability that an electron incident on the surface at an angle α conserves the tangential component of the quasimomentum after reflection. The value of p lies in the range $1 \leq p \leq 0$ when α is varied from 0 to $\pi/2$.

The simplest form of $p(\alpha)$ was proposed in[18,19]:

$$p(\alpha)=1, \quad 0\leqslant\alpha\leqslant\alpha_0; \qquad p(\alpha)=0, \quad \alpha_0<\alpha\leqslant\pi/2. \tag{3}$$

According to this step-function relation, the electrons arriving at the surface at angles smaller than α_0 are reflected from it only specularly, and all the remaining electrons are reflected only diffusely.

The relation $p = p(\alpha)$ explains why the metal whiskers have unexpectedly large values of Fuchs parameter $(P = 0.5-0.7)$, namely, in thin samples $(l^\infty/d >> 1)$ the principal role in the conductivity is played by electrons traveling at a small angle to the surface, for which the specularity coefficient is close to unity.

The dependence of the resistivity on the ratio l^∞/d was calculated in[18] under the assumption of a step function $p(\alpha)$ in the form (3). A comparison of the results of these calculations with analogous calculations, assuming P to be independent of α, shows that at $l^\infty/d = 10^2$ values $P = 0.5-0.8$ of the Fuchs parameter correspond to angles $\alpha_0 \sim 5-10^\circ$. The latter is not surprising for such perfect samples as whiskers.

The existence of the $p(\alpha)$ relation leads inevitably to a dependence of the Fuchs parameter on the ratio l^∞/d. At the present time there are no correct measurements capable of ascertaining whether this is indeed the case. Nonetheless, the function $p(\alpha)$ describes the behavior of the magnetoresistance in strong fields better than the Fuchs parameter. Moreover, the regions of weak and strong fields can be treated from a unified point of view. (A function $p = p(\alpha)$ in the form of the step function (3) was proposed in[4] for the region $2r > d$.)

In very weak magnetic fields $(r \gg d)$ the electrons colliding with the surface can be divided into two groups. In the first the electrons collide with any one of the surfaces and are reflected from it specularly (owing to the small incidence angle), and in the second they collide with two surfaces. In this group there are both "diffuse" and "specular" electrons. The electrons of the first group and the "specular" electrons of the second group move on "quasi-open" periodic trajectories made up of segments of closed orbits. The effective mean free path of this motion is $l_{\text{eff}} \approx l^\infty$. For the "diffuse" electrons of the second group we have $l_{\text{eff}} \approx d$. With increasing magnetic field, the maximum angle of incidence in the first group increases and reaches α_0. At this instant, the "specular" electrons of the second group vanish, and "diffuse" electrons with $l_{\text{eff}} \approx \sqrt{rd}$ [2] (from the second group) appear in the first group. For "good" polyvalent metals it is easy to estimate that at angles $\alpha_0 \sim 5-10^\circ$ and at sample thicknesses $\sim 1\ \mu$ this instant sets in in fields on the order of $H_0 \approx 100-200$ Oe. (We shall henceforth disregard the field region $H < H_0$.)

It is seen from the foregoing that in fields $H > H_0$ the principal role in the conductivity is played by the electrons of the first group. In this group, the relative number of "specular" electrons decreases with increasing $H > H_0$, and the resistance becomes more and more "diffuse". In the language of the Fuchs parameter, this means a decrease of P. The field H_1 can then be treated as the field above which one can neglect the contribution of the "specular" electrons to the conductivity of the sample.

When $2r = d$ is reached all the surface-scattering processes become stationary. The Fuchs parameter assumes its nonzero minimal value $P_{\min} \ll P_{H=0}$ and does not change any more. Therefore the experimentally-observed unusual dependence of the resistance on the magnetic field in the region $2r < d$ must be attributed to a decrease in the contribution of the "specular" electrons to the total conductivity with increasing field. In strong magnetic fields, $2r < d$, the resistance should have in the main a "diffuse" character, and $\Delta\rho^d(H) \propto H^2 d$ [8]. The experimental results agree with this dependence of the resistance on the thickness[4].

Allowance for the quantum surface levels that are produced in specular interaction between electrons and a surface in a magnetic field[5] does not change essentially the foregoing treatment of the experimental results. In the initial region of the magnetic field $H < H_0$, the number of surface levels N in the sample increases with increasing field. The number N is determined by the condition that the sample thickness d be equal to the height Δz_n of the arc of the stationary orbit[20]:

$$\Delta z_n \propto H^{-1/3} n^{2/3}, \quad n=1, 2, \ldots$$

The discrete angles of incidence of the electrons on the surface increase simultaneously[21]

$$\alpha_n \propto (Hn)^{1/3}, \quad n=1, 2, \ldots$$

In the field $H = H_0$, the number of quantum surface levels in the sample reaches a maximum value ($N = N_{\max}$, $\alpha_{N_{\max}} = \alpha_0$), since the levels with angles $\alpha_n > \alpha_0$ are completely smeared out. In fields $H > H_0$ the number of levels N begins to decrease, since the number of orbits with angles $\alpha_n < \alpha_0$ decreases. In comparison with the classical approach, this means a faster decrease of the contribution of the "specular" electrons to the sample conductivity. In strong magnetic fields, an instant is reached when $\alpha_1 > \alpha_0$. The "specular" electrons disappear completely. The resistance is determined by the diffuse interaction of the electrons with the surface, and the Fuchs parameter P vanishes.

Nothing changes qualitatively if account is taken of electrons with incidence angles $\alpha > (180^\circ - \alpha_0)$.

On the basis of the qualitative considerations, we can make a few quantitative estimates. First, we call attention to the following: in strong fields the experimental results (2) can be represented by the expression

$$\xi(H)=\xi(H_2)+a(H-H_2)+b(H-H_2)^2=Ah^2+Bh+C, \tag{4}$$

where $h = d/r$ and A, a, B, b, and C are constants.

Next, the tangent to the trinomial (4) at the point $H = 2$ ($2r = d$, $H = H_2$) is always a continuation of linear function $\xi(H)$ in fields $H > H_1$. Finally, the constant B is negative. Its value is several times smaller for filamentary cadmium whiskers than for plate whiskers, so that we can assume $B = 0$ for completely diffuse reflection.

Let $B = 0$; then $\xi(H) = Ah^2 + C$. This form of the dependence should result from the decrease in the relative number of near-surface electrons, by a factor $4r/d$ (with allowance for the two surfaces of the sample). It follows therefore that the linear relation $\xi(H) = A_1 h$ should hold in weak fields ($h < 2$) and for completely diffuse reflection. To check this conclusion, it is necessary to perform measurements on samples with $P_{H=0} = 0$ or of samples in which $P_{H=0}$ is varied in succession. No such measurements have been performed so far. Nor is there an exact theoretical analysis of the behavior of the resistance at different characters of electron-surface interaction in the region of weak fields ($r \gg d$) and intermediate fields ($r \gtrsim d$).

The resistance at the point $h = 2$ and the constant C can be obtained from the condition that the curves $Ah^2 + C$ and $A_1 h$ be joined at the point $h = 2$ (see above). We find $\xi(H) = 8A$ and $C = 4A$. The theory of the static skin effect yields for strong fields[8]

$$\Delta\rho^d(H) \approx \rho^\infty l^\infty \frac{d}{(2r)^2} = \frac{\rho^\infty l^\infty h^2}{4d} \approx \rho^d(0)\frac{h^2}{4}.$$

Comparing the coefficients of h^2, we get $A \approx 1/4$. Therefore $C = 1$ and $\xi(H) \approx 2$. These values, obtained under rather rough assumptions, are close to the experimental values at $H = H_2$—see Fig. 3. (See Fig. 9 of[4] for zinc.)

Specularity of the sample surface should lead to an increase in the total conductivity, in comparison with the case of complete diffuse reflection. In the region of weak magnetic fields, its influence should be strong and should lead, in particular, to an increase of the field H_1 and to a decrease of the slope of the straight line in fields $H > H_1$. In the region of not too strong fields $2r < d$, the influence of the specular reflection should remain the same. This seems to explain the appearance of an additional term, linear in h, in expression (4).

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