A novel geometry for coherent magnetic breakdown in white tin

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We have measured the transverse magnetoresistance of single-crystal β -tin in the [110] direction in fields up to 60 kOe, and the thermopower in fields up to 150 kOe. For $\mathbf{H} \| [1\overline{10} \pm 20^\circ]$, we observed anomalies in the transverse magnetoresistance and a new set of oscillations in the thermopower. We are persuaded that these results can be explained by magnetic breakdown between the sheets of the Fermi surface in the fourth, fifth and sixth bands near the W point of the Brillouin zone. We have shown that the observed magnetic breakdown has a phase-coherent character.

In the past few years, the electronic properties of lowdimensional and disordered conducting systems have been widely studied by solid-state physicists. In Refs. 1–3 it was shown that a pure metal in a strong magnetic field under coherent magnetic breakdown (CMB) conditions⁴ can behave as a one-dimensional quasirandom system (i.e., incommensurate and disordered), characterized by absolute quantum localization of conduction electrons. Because of the possibility of observing localization-related and nonlinear phenomena under CMB conditions,^{1–4} new attention has been focused on investigating scenarios for magnetic breakdown in metals.

The basic mechanism which gives rise to macroscopic manifestations of magnetic breakdown (MB is an interband effect in which conduction electrons tunnel in a strong magnetic field⁵) is the interference of quasiclassical electron waves scattering off of MB centers in momentum space.⁶ Because of the quantum-interference nature of electron dynamics in metals under MB conditions, small-angle electron scattering has a significant effect on the physical picture of MB.^{6,7} With regard to investigating the properties of metals undergoing MB, the most interesting situation is the intermediate one in which the probability of MB is 0 < w(H) < 1; in this case, quantum wave interference plays an important role.

According to band-structure calculations and investigations of the Fermi surface (FS) geometry of white \tan^{8-15} MB is possible between sheets of the FS in the third and fourth bands near the X and P points of the Brillouin zone (BZ), and between sheets of the FS in the fourth, fifth and sixth bands near the W point (see Figs. 1, 3 below). MB between sheets of the FS in the third and fourth bands has been observed and widely studied experimentally.¹⁶⁻¹⁸ However, the question of whether or not MB occurs at the W point has up until now remained open.

In this article, we report experimental results which indicate that MB is indeed possible near the W point of the BZ. The "anomalous" field and angular dependences of the transverse magnetoresistance (MR) ρ_H are explained by the appearance of a thin layer of open orbits which form as a result of MB beween closed orbits on the fourth- and fifthband sheets of the FS near the W point. We have observed giant quantum oscillations in the transverse magnetic thermopower S_H , which we interpret to be manifestations of MB between orbits of the fourth-, fifth- and sixth-band sheets of the FS in the vicinity of the W point. Analysis of the FS geometry shows that the average direction of an open MB orbit deviates from [001] by an angle of more than 20° and is perpendicular to [110] in momentum space. The width of the MB layer of open orbits $\Delta P/P_F \sim 4 \cdot 10^{-2}$ (ΔP is the width of the MB layer in momentum space, P_F is the Fermi momentum) was estimated by using data on the linear dimensions of the FS of tin.^{14,15} We show that the conductivity of this layer of open MB orbits is extremely sensitive to the density of dislocations and varies with temperature as T^{-3} , which according to Refs. 6, 7 and 19 indicates that MB in this system has a phase-coherent character. Based on the assumption of CMB, we estimate that the breakdown field is given by $H_0 \sim 230$ kOe.

EXPERIMENT

In this paper we investigate the transverse MR $\rho_{yy} \equiv \rho_{H,T}$ and transverse magnetic thermopower S_H as functions of the intensity H and direction (the angle φ) of the magnetic field, the temperature T, and the degreee of plastic deformation ε . The investigations were carried out on single-crystal samples Nos. 1–4 of β -tin (see Table I) with resistivity ratios $\rho_{293 \text{ K}}/\rho_{1 \text{ K}} \approx 2 \cdot 10^5$ and original dislocation densities $N_d \sim 10^6 - 10^7 \text{ cm}^{-2}$. The samples were cut out by an electric-spark machine from a single ingot. After cutting, the deformed samples were etched down to a thickness of 0.1 mm and the surfaces of the samples were annealed in air at T = 110 °C over the course of 50 hours. The axes of the samples (except for No. 2) were oriented along the [110] direction to an accuracy of 0.5°.

Measurement of the electrical resistivity $\rho_{0,T}$ and the transverse MR were carried out at constant current (0.5–5 A) using the four-probe method with a voltage sensitivity of at least $5 \cdot 10^{-11}$ V (Refs. 20, 21). The measurement error of the resistivity was less than 1%.

In order to obtain $\rho_{H,T}$ and S_H at intermediate temperatures, we used a He vapor pump in our investigations below 4.2 K; above 4.2 K we used a special measurement cell.²² The working temperature in the interval 2–20 K was established and held constant to an accuracy of 0.01 K.

Plastic deformation of the samples was brought about by uniaxial tension at low temperatures. This method of deformation is analogous to that described in Ref. 23. The dislocation density was determined from the electrical resistance at $4.2 \text{ K}.^{24}$

The basic measurements of ρ_{yy} were carried out in fields

TABLE I. Characteristics of samples under study.

Sample	Orientation of Axis	Orientation of faces	ρ _{293 K} /ρ _{4.2 K}	Dimensions, mm ²	Density of Dislocations cm ⁻²
Sn1 Sn2 Sn3 Sn3' Sn3'' Sn3'' Sn4	[110] [110±5°] [110] [110] [110] [110] [110]	$\begin{array}{c} (1\bar{1}0), \ (001) \\ (1\bar{1}0\pm5^{\circ}), \ (001) \\ (1\bar{1}0), \ (001) \end{array}$	$\begin{array}{c} 62\ 000\\ 64\ 000\\ 64\ 000\\ 26\ 000\\ 12\ 500\\ 65\ 000 \end{array}$	$3 \times 3 \times 20$ $3 \times 3 \times 20$ $2 \times 1 \times 26$ $2 \times 1 \times 26$ $2 \times 1 \times 26$ $1 \times 1 \times 13$	$\begin{array}{c} 10^{6}-10^{7}\\ 10^{6}-10^{7}\\ 10^{6}-10^{7}\\ 3\cdot 10^{8} \ (\varepsilon\sim1\%)\\ 1\cdot 10^{9} \ (\varepsilon\sim2\%)\\ 10^{6}-10^{7} \end{array}$

up to 60 kOe, which were created by a superconducting solenoid.²² The magnetic field was uniform to better than 0.1%.

Measurements of the transverse thermopower S_H were carried out for $T \leq 4.2$ K in fields of 100–150 kOe using the scheme described in Ref. 25. The measurements were made at the International Laboratory for Strong Magnetic Fields and Low Temperatures (in the city of Wroclaw, People's Republic of Poland).

RESULTS

In this paper we present the most characteristic results involving the influence of the observed MB near the W point on the kinetic coefficients of β -tin. Measurement of the transverse MR were carried out on samples Sn1, Sn2 and Sn3. In Fig. 1 we present the angular dependences of the MR of samples Sn1, Sn2 at T = 4.2 K and 10 K in a field H = 56kOe ($\omega \tau \sim 700^{11}$ for T = 4.2 K and ~ 20 at 10 K). The angle $\varphi = 0^{\circ}$ corresponds to **H** [[001]], the angle $\varphi = 90^{\circ}$ to H||[110]. The character of our results for the angular dependences of $\rho_{H,T}/\rho_{0,T} = f(\varphi)$ basically agrees with that of data obtained by other investigators, ^{8,16} and is explained by the topology of the FS for tin. For $\varphi \sim 0^{\circ}$ (H near [001]) and $\varphi \sim 18^\circ$, we observed giant MB oscillations in the MR, connected with the δ_1^1 orbit² (MB close to the X-point).^{17,20} The oscillations we observed were used to characterize the quality of the samples.

The principal difference between our data and data

published earlier is the deep "dip" at $\varphi \sim 70^\circ$ mentioned for the first time by the authors of Ref. 20. Our investigations show that this feature is extremely sensitive to the experimental conditions. Thus, as the magnitude of H decreases to 3-5 kOe for T = 4.2 K ($\omega \tau \sim 40$) or the temperature is increased to 10 K at H = 56 kOe ($\omega \tau \sim 20$; Fig. 1, curve 2), the "dip" totally disappears. A plastic deformation of $\varepsilon = \Delta l / l$ $l \sim 2\%$ (see Fig. 2; $\omega \tau \sim 25$, for t = 4.2 K, $\varepsilon \sim 2\%$ and H = 56 kOe) and a deviation of the sample axis from [110] by an angle $> 3^{\circ}$ (Fig. 1, curve 3) also lead to disappearance of the "dip." These observations reveal the reason why this anomaly was not observed in earlier papers^{8,16}: for the samples investigated in Ref. 8 with axes parallel to [110], an estimate of the maximum value of $\omega \tau$ gives the value ~25, while samples analogous to ours which were studied in Ref. 16 had axes inclined from [10] by angles on the order of 3° or more.

In the inset of Fig. 2 we show the fine structure of the "dip" when the vector **H** is rotated exactly in the $[1\overline{10}]$ direction. It is clear that this "dip" consists of two minima separated by an angular interval of about 4°. The depth of the minima, and indeed the "dip" as a whole, strongly depend on the accuracy with which the sample is oriented along [110].

The field dependences of the transverse MR of sample Sn1 at 4.2 K for $\varphi = 90\%$ and $\varphi = 70\%$ are presented in Fig. 3. At $\varphi = 90^{\circ}$ the MR grows with field as H^2 , while for $\varphi = 70^{\circ}$ the MR field dependence deviates from quadratic: $\rho_{H,T} \sim H^4$.



FIG. 1. Angular dependence of the transverse magnetoresistance of β -tin in a magnetic field of 56 kOe: 1—Sn1, T = 4.2 K; 2—Sn1, T = 10 K; 3—Sn2, T = 4.2 K; the inset shows the Brillouin zone.



FIG. 2. Angular dependence of the transverse magnetoresistance of β -tin in a magnetic field of 56 kOe, T = 4.2 K: 1—Sn3, $\varepsilon = 0\%$; 2—Sn3', $\varepsilon = 1\%$; 3—Sn3", $\varepsilon = 2\%$; the inset shows the fine structure of the "dip" at $\varphi = 70^{\circ}$.



FIG. 3. Field dependence of the transverse magnetoresistance of β -tin, sample Sn1, for T = 4.2 K: $1-\varphi = 90^{\circ}$, $2-\varphi = 70^{\circ}$; the inset shows the cross-section of the Fermi surface of tin in the fourth-, fifth- and sixth-band sheets for **H**||[110].

The temperature dependences of the electrical resistivity and transverse MR of sample Sn1 were measured in the interval 2-50 K. The functions $\rho_{H,T} = f(T)$ in a field of 56 kOe for $\varphi = 90\%$ and $\varphi = 70\%$ have a characteristic minimum at $T \sim 20$ K ($\omega \tau \sim 1$)²⁰; for $T \gtrsim 10$ K, these functions coincide (Fig. 4). Analysis of these functions shows that $\rho_{0,T} \sim T^5$ in the interval 4.2-15 K, while $\rho_{H,T}$ ($\varphi = 90^\circ$) $\sim T^{-5}$ below 5 K and is $\sim T^{-3}$ in the interval 7-12 K. The change in the dependences of $\rho_{H,T}$ from T^{-5} to T^{-3} is connected with a magnetically-induced change in the nature of the electron-phonon interaction.²⁶

Analysis of the temperature dependence of the conductivity for $\varphi = 70^{\circ}$ was carried out under the assumption that the total conductivity σ_{tot} in this case is caused both by a contribution from closed orbits and by a contribution σ_{open} from the layer of open orbits. Our results showed that the conductivity of this layer of open orbits in the interval 2–9 K varies as a power law which is close to T^{-3} .



FIG. 4. Temperature dependence of the transverse magnetoresistance of β -tin, sample Sn1: 1—H = 0; 2—H = 56 kOe, $\varphi = 70^{\circ}$; 3—H = 56 kOe, $\varphi = 90^{\circ}$.



FIG. 5. Angular dependence of the frequency (a) and amplitude (b) of the thermopower oscillations in β -tin (Sn4) for T = 4.2 K; (c) Fourier spectrum of the oscillations for $\varphi = 70^{\circ}$ and T = 1.5 K.

Investigations of the transverse magnetic thermopower S_H were carried out at $T \leq 4.2$ K in fields 100–150 kOe. The dependence of the thermopower on field has an oscillatory character.²⁷ For H close to [001], we observed oscillations in the thermopower, similar to those in the MR, with frequency ~ 2 MHz; these oscillations were due to MB between the third- and fourth-band sheets of the FS of tin. The corresponding branch δ_1^1 is tracked up to $\varphi = 70^\circ$, where its frequency comes to $F_2 \sim 5.7$ MHz (Fig. 5). For $\varphi = 70^\circ$, i.e., in the direction of the anomalous "dip" in the MR, we observed new oscillations in the thermopower with frequency $F_1 \sim 2-$ 3 MHz in fields \gtrsim 100 kOe. No MR oscillations were observed. A plot of the Fourier spectrum of these oscillations in S_H for $\varphi = 70^\circ$ is shown in Fig. 5c, where we can reliably identify 5 groups of frequencies $F_1 - F_5$. The frequency F_1 is absent from the results of measurements of the deHaas-Van Alphen effect and other oscillatory phenomena. The amplitude of these oscillations has a narrow maximum for $\varphi = 70^{\circ}$ (Fig. 5b); in the range 100–150 kOe its magnitude was only 2–4 times smaller than the amplitude of oscillations of δ_1^1 for H||[001]. Our analysis leads us to the following numbers: $F_1 = 2.25$ MHz; $F_2 = 5.7$ MHz; $F_3 \sim 2F_2$; $F_4 \sim 3F_2$; $F_5 = 32.7$ MHz, where F_5 corresponds to the orbit ε_3^1 (Ref. 13). From the thermopower data in the interval 1.5-4.2 K it also follows that $m_1 \approx m_2$ and $m_5 > m_2$, where m is the effective electron mass.

DISCUSSION

White tin is a compensated metal $(n_e = n_h)$, which has two open FS sheets in the (001) plane lying in the fourth and fifth bands and closed electron and hole sheets in the third, fourth and sixth bands.^{14,15} According to theory,²⁸ for all directions of the magnetic field in which open orbits are absent, the transverse MR of white tin $(\omega \tau > 1)$ should vary as $\rho_{H,T} \sim H^2$; this excludes the special case of $\mathbf{H} || [001]$, for which a geometry-induced lack of compensation $(n_e \neq n_h)$ causes the resistivity to saturate. For field directions in which open orbits are observed, the MR is given by the expression

$$\rho_{H, T} = A + BH^2 \cos^2 \alpha, \qquad (1)$$

where α is the angle between the direction of current and the average direction of the open orbits in momentum space (A and B are constants).

Starting from these assumptions, we can explain the appearance of the anomalous "dip" in the MR angular dependence at $\varphi = 70^{\circ}$ and the deviation of the MR field dependence from quadratic (see Figs. 1, 3) only if we assume the presence of open or extended paths whose average direction is perpendicular to the current in momentum space (j || [110]). Given what is known about the topology of the tin FS, such orbits should be absent; however, they can form as a result of MB between the FS sheets in the fourth and fifth bands when the MB occurs between the points 1-1' and 2-2', as shown in Fig. 6. According to band structure calculations,¹² this type of breakdown can occur in white tin near the W point of the Brillouin zone. The average direction of the open MB orbits which contribute to the conductivity (see Fig. 6) lie along the projection of the [001] axis in the plane perpendicular to H, i.e., they deviate from [001] by an angle on the order of 20° in the plane (110). As is clear from the figure, this type of MB may also involve a two-dimensional net of open MB paths when the direction of H lies precisely in the (110) plane. In this net the open paths are oriented at an angle $\sim 40^{\circ}$ relative to one another.

Our estimate of the width of the layer of open MB orbits $\Delta P/P_F$ was made based on detailed data concerning the linear dimensions of the FS given in Refs. 14, 15. In our case the parameters which limit the width of this layer are the size of the necks of the fourth-band FS sheets, the thickness of the hole sheet along the line ΓH in this band, the length and diameter of the "tubes" which connect the "bulbs" of the electron sheet in the fifth band, and the angular separation



FIG. 6. Cross-section of the Fermi surface of β -tin for $\mathbf{H} || [1\overline{10}] \pm 20^{\circ}$] in the (110) plane; the open MB path is directed along the projection of the [001] axis on the plane perpendicular to \mathbf{H} , and is perpendicular to [110].

between the two minima of the "dip." This estimate gives a layer width $\Delta P/P_F$ on the order of $4 \cdot 10^{-2}$.

We note that a layer of open orbits of width $\Delta P / P_F \sim 10^{-2}$ fully determines the conductivity if $\Delta P / P_F > (r/l)^2$ (Ref. 28). Here r is the Larmor radius $(r = cP_F/eH)$, while l is the mean free path of an electron (in the white-tin samples we studied, l is on the order of 0.8 mm at 4.2 K). Simple estimates show that in our case a layer of open orbits of width $\Delta P / P_F \sim 4 \cdot 10^{-2}$ should fully determine the conductivity even in fields ~ 2 kOe, if the openness has a geometric origin, i.e., $\rho_{H,T}$ could conceivably saturate even at fields $H \gtrsim 2$ kOe, whereas for MB involving an open-orbit layer of similar width saturation of $\rho_{H,T}$ should begin at fields on the order of H_0 . Hence, this latter situation would be clear evidence of the magnetic-breakdown character of the observed open orbits.

In order to estimate the breakdown fields H_0 ($w = \exp(-H_0/H)$) from the experimental data, we used expressions which describe the conductivity of the one-dimensional MB layer of open trajectories in the two limiting cases of stochastic (SMB) and coherent (CMB) magnetic breakdown^{29,30}:

$$\sigma_{\rm SMB} = \frac{\Delta P}{P_F} \frac{w}{1 - w} \frac{\sigma_0}{\omega \tau}, \qquad (2)$$

$$\sigma_{\rm CMB} = \frac{\Delta P}{P_F} w \sigma_0 \frac{\tau_a}{\tau_{\rm imp}},\tag{3}$$

where σ_0 is the metal conductivity for H = 0; τ_d is the coherence lifetime of an electron, which is connected with scattering by dislocations and which determines the Dingle temperature; $\tau_{\rm imp}$ is the electron lifetime connected with scattering by impurities. Here, $\sigma_{\rm MB}$ is determined from the expression

$$\sigma_{\rm MB} = \sigma_{\rm tot} - \sigma_{\rm closed} \tag{4}$$

where $\sigma_{\text{tot}} = 1/\rho_H^{70}$ is the total conductivity due to contributions from the layer of open MB orbits and the closed orbits for $\varphi = 70^\circ$; $\sigma_{\text{closed}} = 1/\rho_{H(70)}^{\text{closed}}$ is the conductivity due to closed orbits for $\varphi = 70^\circ$. It follows from the experimental data that in the absence of MB we have $\rho_{H(70)}^{\text{closed}} \approx \rho_{H(90)}^{\text{closed}}$ (Fig. 1, curve 2).

An estimate of the breakdown field (when breakdown occurs at H = 56 kOe and T = 4.2 K) in the case of CMB gives a value of $H_0 \sim 40$ kOe. However, this value of H_0 is clearly contradicted by the experimental data we have obtained; the absence of saturation of ρ_{yy} for $\varphi = 70^{\circ}$ and H = 56 kOe and the observed dependence $\sigma_{\rm MB} \sim T^{-3}$ (Ref. 19), the more so because in our samples at fields 40-60 kOe we have the relation $d_{\text{disloc}} = N_d^{-1/2} \gtrsim r (d_{\text{disloc}} \text{ is the aver-}$ age distance between dislocations). From our data on the thermopower it also follows that the breakdown field is at least $\gtrsim 100$ kOe. An estimate of the breakdown field in the CMB case for these experimental conditions and a value τ_d / $\tau_{\rm imp} \sim 0.1$ gives a field $H_0 \sim 230$ kOe. The value of τ_d was estimated according to Ref. 7, giving $\tau_d \sim N_d^{-1/2} v_F^{-1} \approx 10^{-11}$ sec (where $N_d \sim 10^6$ cm⁻², and v_F is the electron Fermi velocity). The impurity lifetime in our tin samples was $\sim 10^{-10}$ sec. The large value of H_0 serves to explain the absence of MB oscillations in the MR in our experiments, while the value of the conductivity of the MB layer attests to the coherent character of the MB.

In our investigations of the transverse magnetic thermopower S_H , we observed a new group of giant quantum oscillations in S_H at a frequency $F_1 = 2-3$ MHz in the region of anomalous transverse MR for $\varphi = 70^\circ$. From the data on the frequency F_1 and the effective mass m_1 it follows that a small cross-section of the FS enters into this effect. A similar nonextremal cross-section appears for the geometry of this experiment in the sixth-band FS sheet. Consequently, our results can be connected with MB between closed orbits of the fourth-, fifth- and sixth-band sheets of the FS and with coherence effects which give rise to the minimum for small orbits.

In order to elucidate in more detail the character of the observed MB, we have thoroughly analyzed the temperature dependence of the conductivity of the one-dimensional layer of open orbits which forms as a result of the MB. Some simple transformations show that the temperature-dependent part of the conductivity of this layer can be expressed in the following way:

$$\sigma_{\mathrm{MB}}^{ep} = \sigma_{\mathrm{MB}} \sigma_{\mathrm{MB}}^{\mathrm{imp}} / (\sigma_{\mathrm{MB}}^{\mathrm{imp}} - \sigma_{\mathrm{MB}}) ,$$

where $\sigma_{\rm MB}$ is defined according to (4) while $\sigma_{\rm MB}^{\rm imp}$ is the conductivity of the MB orbits in the absence of electron-phonon scattering, i.e., for $T \rightarrow 0$. The analysis was carried out for H = 56 K (Fig. 4), while $\sigma_{\rm MB}^{\rm imp}$ was determined at T = 1.5 K. We found that the conductivity $\sigma_{\rm MB}^{ep}$ varied as $T^{-3 \pm 0.3}$. According to Ref. 19, this indicates that the MB we observed has a phase-coherent character.

In summary, the totality of the data we have obtained for $\rho_{H,T}$ and S_H , along with the analysis of this data, allow us to say that we have observed MB in β -tin in the vicinity of the W-point of the Brillouin zone between fourth-, fifth- and sixth-band sheets of the FS. This MB leads to the formation of a narrow layer of open orbits in which the average direction of an open orbit deviates from [001] by an angle of 20° in the (110) plane. The following facts suggest that this magnetic breakdown has a coherent character: the high sensitivity of the MR to changes in the dislocation density, the character of the MR temperature dependence at $\varphi = 70^\circ$, the observation of giant quantum oscillations in the thermopower, and estimates of the magnitude of the conductivity of the layer of open MB orbits.

We also note that ours is the first observation of a twodimensional MB network of open orbits for a direction of magnetic field which does not lie along a high-order symmetry axis.

The character of the observed MB in compensated high-purity β -tin (the coherence and conductivity for $\varphi = 70^{\circ}$ are basically determined by MB) allow us to carry out a search for the phenomena of localized-electron conductivity and departures from Ohm's law predicted in the papers by Slutskin.^{2,4}

We wish to express our gratitude to A. A. Slutskin for useful discussions of the results of this paper, and to M. Glinski for help in measuring the thermopower. ¹⁾The ratio $\omega \tau = (H/\rho_{0:T})(n_{\text{eff}}cc)^{-1}$ was used to estimate the value of $\omega \tau$, where ω is the cyclotron frequency, τ is the transport lifetime of an electron, n_{eff} is the number of charge carriers in 1 cm⁻³, e is the electron charge, and c is the velocity of light. The quantity n_{eff} was determined from data on the temperature dependence (at the minimum of the curve $\rho_{H,T} = f(T), \omega \tau \sim 1$). This estimate of $\omega \tau$ corresponds to large orbits in the fourth- and fifth-band sheets of the FS, since it is just these groups of charge carriers which give the primary contribution to the conductivity. ²⁾Using the notation employed in Ref. 13.

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