

Mixed-State Hall Effect in an Extreme Type-II Superconductor

R. R. HAKE

Science Center, North American Rockwell Corporation, Thousand Oaks, California

(Received 20 November 1967)

Measurements of mixed-state transverse and longitudinal voltages at $T=1.2^\circ\text{K}$, $0 < H < 39 \text{ kG}$, $0 < J < 800 \text{ A/cm}^2$ on an extremely high- κ type-II alloy superconductor Ti-16 at.% Mo ($\kappa=68$, $T_c=4.2^\circ\text{K}$, zero-temperature upper critical field $H_{c20}=63 \text{ kG}$) show that the high- J , nearly J -independent, mixed-state Hall-angle tangent $\tan \theta_m(H)$ is much larger than the normal-state value $\tan \theta_n(H)$. At the highest applied magnetic field, the reduced Hall-angle tangent $P_m(H/H_{k20}=0.35) = [\tan \theta_m(H=39 \text{ kG})]/[\tan \theta_n(H_{k20})] \approx 0.6$, where $H_{k20}=111 \text{ kG}$ is determined by the measured reduced flux-flow resistivity $\rho_f(H)/\rho_n = H/H_{k20}$. This contrasts with previous results pertinent to pure or low- κ materials where $H_{k20} \approx H_{c20}$: $P_m(0.35) \approx 0.35$ (Bardeen-Stephen theory), (1) (Nozières-Vinen theory), 0.35 (Nb measurements of Fiory and Serin), (2) (Nb-Ta measurements of Niessen *et al.*).

CURRENT interest in the transport properties of quantum fluids has stimulated considerable experimental¹⁻⁸ and theoretical⁹⁻¹³ work on the Hall effect in the mixed state of type-II superconductors. Thus far a wide variety of divergent results has been obtained and the problem remains generally obscure. In the present article we report the first observation of the mixed-state Hall effect in an extremely high Ginzburg-Landau- κ type-II superconductor.

Figure 1 shows mixed-state longitudinal V_L and transverse V_T voltages versus transport current density J for an annealed^{14,15} specimen of Ti-16 at.% Mo. Previous magnetization,¹⁴⁻¹⁶ calorimetric,¹⁷ and resistive^{14,15,18,19} measurements on this material indicate typical¹⁵ extreme type-II mixed-state paramagnetism and afford a good parametric characterization¹⁵: $T_c=$

4.2°K , $\kappa_G=68$, $\rho_n(4.2^\circ\text{K})=1.0 \times 10^{-4} \Omega \text{ cm}$, $R_n(4.2^\circ\text{K}) \approx +1.6 \times 10^{-4} \text{ cm}^3/\text{C}$, $\gamma=7.5 \times 10^3 \text{ erg cm}^{-3} \text{ }^\circ\text{K}^{-2}$, $\xi_0/l \approx 120$, $H_{c20}=63 \text{ kG}$, $H_{c20}^*=99 \text{ kG}$. Here, κ_G is calculated from the Gor'kov-Goodman formula, ρ_n is the normal-state electrical resistivity, R_n is the normal-state Hall coefficient, γ is the electronic specific-heat coefficient, ξ_0 is the BCS²⁰ coherence distance, l is the electron mean free path, H_{c20} is the measured upper critical field extrapolated to $T=0^\circ\text{K}$, and $H_{c20}^*=3.1 \times 10^4 \rho_n \gamma T_c$ is the upper critical field at $T=0^\circ\text{K}$ predicted by dirty-limit theory²¹ which ignores electron spin-applied field interaction. For the geometry indicated in Fig. 1, $V_L \propto E_L$ and $V_T \propto E_T$ arise, respectively, from transverse and longitudinal components of the average flux-velocity vector \mathbf{v} in the plane of the specimen since²² $\mathbf{E} = -c^{-1}(\mathbf{v} \times \mathbf{B})$, where \mathbf{E} is the measured electric field vector (also in the plane of the specimen) and $\mathbf{B} \approx \mathbf{H}$.

The $V_L(J)$ curve of Fig. 1 displays the usual^{23,24} annealed specimen, nearly free-flux-flow form with a small critical $J_c(H)$ for flux depinning and a nearly constant $(\partial V_L/\partial J)_H \propto \rho_f(H)$ over a relatively wide²⁴ J range, where ρ_f is the flux-flow resistivity. Figure 2 shows that for low $H/H_{k20} \leq 0.35$ and low $T/T_c=0.26$,

$$\rho_f(H)/\rho_n = H/H_{k20}. \quad (1)$$

In the present case the "Kim field"²³ $H_{k20}=111 \pm 5 \text{ kG}$ is close to the H_{c20}^* of paramagnetic-limitation-ignoring theory,²¹ as previously shown for this¹⁹ and other^{23,25} high- κ_G materials. Equation (1) evidently implies²³ a mixed-state effective vortex-core to specimen-volume ratio of $\approx H/H_{k20}$ at low reduced H and T . For low- κ_G materials there is no electron-spin-induced paramagnetic

¹ W. A. Reed, E. Fawcett, and Y. B. Kim, Phys. Rev. Letters **14**, 790 (1965).

² A. K. Niessen and F. A. Staas, Phys. Letters **15**, 26 (1956); see also, F. A. Staas, A. K. Niessen, W. F. Druyvesteyn, and J. v. Suchtelen, *ibid.* **13**, 293 (1964).

³ F. A. Staas, A. K. Niessen, and W. F. Druyvesteyn, Phys. Letters **17**, 231 (1965).

⁴ A. K. Niessen, F. A. Staas, and C. H. Weijsenfeld, Phys. Letters **25A**, 33 (1967).

⁵ W. F. Druyvesteyn, G. J. van Gorp, and C. A. A. J. Greebe, Phys. Letters **22**, 248 (1966).

⁶ B. W. Maxfield, Solid State Commun. **5**, 585 (1967).

⁷ J. Baixeras and S. J. Williamson, Solid State Commun. **5**, 599 (1967); S. J. Williamson and J. Baixeras, in Proceedings of the Conference on the Physics of Solids in Intense Magnetic Fields, Crete, 1967 (to be published).

⁸ A. T. Fiory and B. Serin, Phys. Letters **25A**, 557 (1967).

⁹ J. Bardeen and M. J. Stephen, Phys. Rev. **140**, A1197 (1965).

¹⁰ A. G. van Vijfeijken and A. K. Niessen, Phys. Letters **16**, 23 (1965); Philips Res. Rept. **20**, 505 (1965).

¹¹ P. Nozières and W. F. Vinen, Phil. Mag. **14**, 667 (1966).

¹² W. F. Vinen and A. C. Warren, Proc. Phys. Soc. (London) **91**, 409 (1967); **91**, 399 (1967).

¹³ C. Caroli and K. Maki, Phys. Rev. **164**, 591 (1967).

¹⁴ R. R. Hake, Phys. Rev. Letters **15**, 865 (1965).

¹⁵ R. R. Hake, Phys. Rev. **158**, 356 (1967).

¹⁶ J. A. Cape, Phys. Rev. **148**, 257 (1966).

¹⁷ L. J. Barnes and R. R. Hake, Phys. Rev. **153**, 435 (1967); Ann. Acad. Sci. Fennicae **VIA**, 78 (1966).

¹⁸ R. R. Hake, D. H. Leslie, and T. G. Berlincourt, J. Phys. Chem. Solids **20**, 177 (1961).

¹⁹ R. R. Hake, Bull. Am. Phys. Soc. **11**, 480 (1966); in Proceedings of the Tenth International Conference on Low Temperature Physics, 1966 (to be published).

²⁰ J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. **108**, 1175 (1955).

²¹ K. Maki, Physics **1**, 21 (1964); P. G. de Gennes, Physik Kondensierten Materie **3**, 79 (1964); E. Helfand and N. R. Werthamer, Phys. Rev. **147**, 288 (1966).

²² B. D. Josephson, Phys. Letters **16**, 242 (1965); H. G. B. Casimir, *ibid.* **17**, 177 (1965).

²³ Y. B. Kim, C. F. Hempstead, and A. R. Strnad, Phys. Rev. **139**, A1163 (1965).

²⁴ W. C. H. Joiner, Phys. Rev. Letters **19**, 895 (1967).

²⁵ Y. Shapira and L. J. Neuringer, Phys. Rev. **140**, A1638 (1965).

limitation on the upper critical field, and H_{k20} as defined in Eq. (1) is approximately equal to the measured H_{c20} .^{12,23}

The $V_T(J)$ curves of Fig. 1 allow determination of the Hall voltage

$$V_H = 0.25[V_T(J_+, H_+) - V_T(J_+, H_-) - V_T(J_-, H_+) + V_T(J_-, H_-)] \quad (2)$$

which reverses with H and with J . It is apparent from Fig. 1 that $V_T(J)$ also contains a non-Hall component which reverses with J but not with H .²⁶ That this component is primarily due to defect-guided flux motion in the longitudinal direction^{2,7} is suggested in the present work by prominent structure in $V_T(\varphi)$ (where φ is the angle shown in Fig. 1 between H and the specimen's wide surface) which reverses only with J , is uncorrelated with similar microvolt structure in $V_L(\varphi)$, and depends critically on cold-work and annealing history.

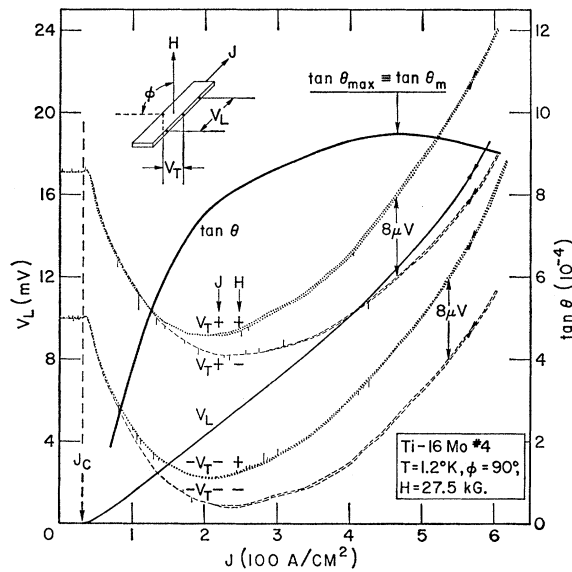


FIG. 1. Typical X-Y recorder tracings of longitudinal V_L and transverse V_T voltages versus longitudinal transport current density J for an annealed, polycrystalline, flat-strip specimen of Ti-16 at.% Mo. For clarity, zero shifts have been made on the V_T traces—below J_c all V_T and V_L voltages are zero. The Hall-angle tangent $\tan\theta(J)$ is related to $V_T(J)$ and $V_L(J)$ by Eqs. (2) and (5). [Specimen dimensions are length 2.0 cm, width $W=0.31$ cm, thickness 0.023 cm. Indium-coated copper current lugs were pressed against the wide surfaces over 0.3 cm at each end. The four fine-copper-wire potential leads were held against the specimen edges by beryllium-copper-strip springs. Small spikes on the $V_T(J)$ traces are due to slight contact instabilities. V_L contact spacing was $L=0.97$ cm. Potential measurements in the 4.2°K normal state indicated that longitudinal misalignment spacing of the V_T contacts was less than 2.3×10^{-4} cm.]

²⁶ The introduction of a small fraction f of the V_L voltage into the measured V_T due to transverse contact misalignment results in a non-Hall voltage of this sort, but measurements of the linear $V_T(J)$ and $V_L(J)$ in the normal state at 4.2°K indicate that this misalignment voltage is negligible ($f=2.4 \times 10^{-4}$).

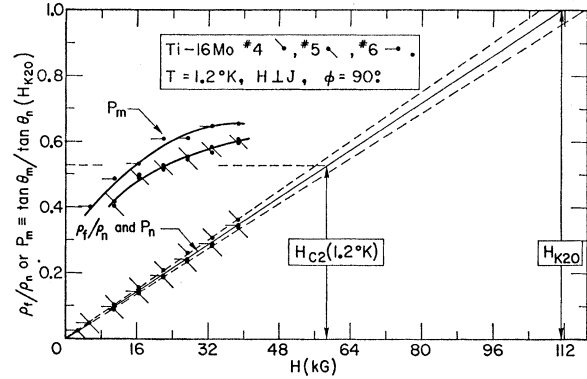


FIG. 2. The reduced mixed- and normal-state Hall-angle tangents P_m and P_n [see Eqs. (6) and (7)], and the reduced mixed-state flux-flow resistivity ρ_f/ρ_n , all versus the applied magnetic field H for three annealed Ti-16 at.% Mo specimens. $H_{c2}(1.2^\circ\text{K})$ is the magnetization-measured upper critical field at 1.2°K of Ref. 15 and H_{k20} is defined by Eq. (1). P_n is actually determined at 4.2°K but it is assumed that P_n is temperature-independent below 4.2°K.

Despite marked variation of guided-motion transverse voltages with φ [up to $\pm 1100\%$ of $V_H(\varphi=90^\circ)$ with average peak half-width $\approx 2^\circ$], we observe

$$V_H(\varphi, H=39 \text{ kG}, J=280 \text{ A/cm}^2) \propto \sin\varphi \quad (3)$$

for $80^\circ < \varphi < 300^\circ$, just as in the normal state, strongly suggesting that V_H is not greatly perturbed^{4,7,8} from the free-flux-flow value in the present annealed specimens by guided flux motion at high (J, H) . We also observe

$$V_L(\varphi, H=39 \text{ kG}, J=280 \text{ A/cm}^2) = V_L(\varphi=90^\circ) \quad (4)$$

to within 8%.

Figure 1 also shows the Hall-angle tangent

$$\tan\theta \equiv E_H/E_L = V_H L/V_L W, \quad (5)$$

where L and W are, respectively, the distances between the longitudinal and transverse contacts. At high J , $\tan\theta(J)$ displays a tendency to saturate near the maximum value $\tan\theta_{\max}$, which we identify as the measured mixed-state Hall-angle tangent $\tan\theta_m$ and interpret as a lower limit^{1,4,7} to the free-flux-flow $\tan\theta_m$ of theoretical interest. The decrease of $\tan\theta(J)$ at high J and H as shown in Fig. 1 always occurs in the nonlinear $V_L(J)$ region where interpretation is somewhat clouded by the possibility of Joule heating at the current contacts. At lower $H < 17$ kG neither prominent high- J nonlinearities in $V_L(J)$ nor actual maxima in $\tan\theta(J)$ were observed, although $\tan\theta(J)$ always became nearly J -independent at the highest $J \leq 790$ A/cm².

Figure 2 shows the reduced mixed-state Hall-angle tangent

$$P_m \equiv [\tan\theta_m(H)]/[\tan\theta_n(H_{k20})], \quad (6)$$

and the reduced normal-state Hall-angle tangent²⁷

$$P_n \equiv [\tan\theta_n(H)] / [\tan\theta_n(H_{k20})]. \quad (7)$$

The data are normalized at H_{k20} [defined by Eq. (1)] rather than H_{c20} in order to allow a more legitimate comparison of our data with the results of previous experiment¹⁻⁸ and theory⁹⁻¹³ which apply to the low- H domain where electron-spin interaction with H is presumably unimportant and $H_{k20} = H_{c20}$. Strictly speaking, the P_m data of Fig. 2 represent lower limits^{1,4,7} and show that at low H/H_{k20} the Hall-angle tangent is much larger in the mixed state than in the normal state, i.e., $P_m \gg P_n$, in agreement with the results of the Philips workers²⁻⁴ on low- κ_G Nb-Ta alloys. Actually, in view of the discussion centering on Eqs. (3) and (4), we believe that our result

$$P_m(H/H_{k20}=0.35) \approx 0.6, \quad (8)$$

²⁷ The normal-state Hall data agree with Ref. 18. The normal-state $\tan\theta_n(H)$ of Fig. 2 is derived from J -independent $\tan\theta_n(J)$ and $\tan\theta_n(H)$ data taken at $16 < H < 39$ kG and $T = 4.2^\circ\text{K}$, where both the mixed state and the sheath (Ref. 15) are suppressed, assuming that the observed direct proportionality of $\tan\theta_n$ to H holds for $0 < H < H_{c20}^*$ and that $\tan\theta_n$ is T -independent below 4.2°K .

obtained at the highest $H = 39$ kG, is reasonably close to the free-flux-flow P_m of theoretical interest. Assuming this to be the case and that other Hall measurements reflect the free-flux-flow condition, there appears to be no quantitative agreement of our results with previous experimental or theoretical work: $P_m(0.35) \approx 2$ (low- κ_G Nb-Ta),^{4,28} 0.35 (pure Nb),^{1,5,8} 1 (pure Nb),⁶ 0.35 (Bardeen-Stephen theory),⁹ 1 (Nozières-Vinen theory).¹¹ It is not clear whether the disagreement of our work with previous results can be attributed to electron spin-applied field interaction, to the high κ_G of our alloy, to its extreme "dirtiness" (high ξ_0/l), or to some combination of these factors.

We thank D. M. Sellman for able assistance with the measurements; H. Nadler, R. G. Herron, and P. Q. Sauers for careful specimen preparation; C. G. Rhodes and R. A. Spurling for metallography; S. J. Williamson for valuable discussions; and A. S. Joseph for generously allowing our use of his 22-in. Varian magnet facilities.

²⁸ Using the present techniques we have corroborated this result for annealed Nb-50 at.% Ta. For this alloy the J dependence of $\tan\theta$ is less than that shown in Fig. 1.

Superconductivity in Granular Aluminum Films

ROGER W. COHEN AND B. ABELES

RCA Laboratories, Princeton, New Jersey

(Received 13 November 1967)

A detailed description is given of the preparation of granular aluminum films (grain sizes less than 40 Å) with stable superconducting transition temperatures T_c up to 3.7°K. We present new experimental data of energy gaps, parallel critical fields, and critical current densities. The results are in good agreement with the predictions of the theories of granular superconductors. However, it is found that none of the mechanisms offered so far for the enhancement of T_c can satisfactorily account for all the properties of the enhanced T_c granular films.

I. INTRODUCTION

IT has been known for many years that some superconductors, when composed of small grains (20 to 200 Å), have superconducting transition temperatures T_c appreciably higher than the ordinary values. Such films were prepared¹⁻³ by evaporation on to substrates held at 4.2°K. However, upon warming to room temperature, grain growth occurred, and the T_c 's returned to the usual values. More recently, high T_c films of the same metals were prepared by evaporation in an

oxygen atmosphere.^{4,5} The observed increases in the T_c 's were of the same magnitude as those obtained by the low-temperature evaporation technique, but they did not change upon storing at room temperature. Furthermore, these films exhibited very high critical magnetic fields and very high normal resistivities.⁶ It is believed that these properties resulted from the precipitation of oxygen at the grain boundaries in the form of oxide. The oxide (a) prevented recrystallization and grain growth, thereby stabilizing T_c , and

¹ A. Shalnikov, *Nature* **142**, 74 (1938); *Zh. Eksperim. i Teor. Fiz.* **10**, 630 (1940).

² W. Buckel and R. Hilsch, *Z. Physik* **131**, 420 (1952); **138**, 109 (1954).

³ N. V. Zavaritzkii, *Dokl. Akad. Nauk SSSR* **82**, 229 (1952).

⁴ B. Abeles, R. W. Cohen, and G. W. Cullen, *Phys. Rev. Letters* **17**, 632 (1966).

⁵ R. W. Cohen, B. Abeles, and G. S. Weisbarth, *Phys. Rev. Letters* **18**, 336 (1967).

⁶ B. Abeles, Roger W. Cohen, and W. Stowell, *Phys. Rev. Letters* **18**, 902 (1967).