Evidence for a Mixed State in a Type-I Superconducting Foil in a Transverse Magnetic Field

J. A. CAPE
North American Aviation Science Center, Thousand Oaks, California

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Resistance and magnetization measurements have been carried out as a function of temperature, applied field, and transport current (resistance only) on a series of dilute In–Bi alloy foils ($0.45<\kappa_1<0.75$, thickness $d\geq 0.002$ cm). The data enable the determination of H_{c2}, H_{cB}, $\kappa_1(T)$, and $\kappa_2(T)$ over most of the superconducting temperature range ($t\gtrsim 0.3$). It is found that In–1.25 at.% Bi [$\kappa_1(T_c)\approx 0.64$], and In–1.15 at.% Bi [$\kappa(T_c)\approx 0.62$]—both of which are bulk type-II superconductors at temperatures well below T_c—exhibit mixed-state-like behavior as foils in a transverse applied field at all temperatures below T_c. Specifically, at thicknesses less than approximately 0.005 cm, the transverse-field magnetization exhibits an apparent second-order transition at $H_{c2}<H_{cB}$, the parallel transition field. The $H_{c2}(T)$ and $\kappa_1(T)$ values thus determined are in good agreement with the dirty-limit-theory temperature dependence. Moreover, the $\kappa_2(T)$ values which are determined from the slope of the transverse-field magnetization by the formula

$$-4\pi M=(H_{c2}-H_0)/[(2\kappa_2^2-1)\beta+1]$$

are in good agreement with the κ_1 values near T_c and exhibit similar temperature dependence. It is found that the critical fields of the magnetization curves can be identified with characteristic features of the resistive transition curves.

THE existence of a vortex structure in type-I superconductors was suggested originally by Tinkham[1] for the case of a thin film perpendicular to the magnetic field. Largely because of complications arising from long mean free path and relatively small dimensions, the thin-film picture is characterized by an approximate[2] theoretical description on the one hand and numerous experimental difficulties on the other. As a result the existing experimental evidence in favor of Tinkham's hypothesis is mainly qualitative. For example, the experimental critical fields[3,4] are observed to vary with film thickness qualitatively as predicted by theory.[4]

In the present case, the question of the existence of vortices in type-I superconductors is studied at the opposite extreme, i.e., by measurements on bulk, essentially "dirty limit," alloys where one has considerable confidence in sample preparation and characterization, and in achieving equilibrium behavior, and where a nearly exact and highly successful theory[5] may be expected to apply. Within this framework, we present data on the magnetization and critical fields of type-I In–Bi alloy foils in parallel and perpendicular applied fields. These data are consistent with expectation for a magnetic structure similar to the Abrikosov[6,7] mixed state of type-II superconductors. The requisite condition for this occurrence in a foil appears to be basically (as in the case of thin films) that the critical field H_D appropriate to the *intermediate* state be less than the mixed-state critical field,[8] $H_{c2}=\sqrt{2}\kappa_1 H_{cB}$, where H_{cB} is the "thermodynamic" or bulk type-I critical field. For very thick laminae, $H_D=H_{cB}$, but H_D decreases with decreasing thickness.[9,10] Thus for H_D to be less than H_{c2} in a relatively thick (bulk) foil, it is necessary that H_{c2} be only slightly less than H_{cB}, i.e., κ_1 only slightly less than $1/\sqrt{2}$.

From preliminary studies of the thickness dependence of $H_\perp$ (the critical field in a perpendicular orientation) for In–1.25 at.% Bi[$\kappa_1(T_{c0})\approx 0.64$] it was found that $H_\perp$ decreased with decreasing thickness but became constant for thicknesses below approximately 0.005 cm. This fact lead to the initial assumption that $H_\perp=H_{c2}$ for $d<0.005$ cm. This assumption was borne out by subsequent data[11] which are reported below for several In–Bi alloy samples rolled into foils of thickness 0.002–0.005 cm. See Fig. 1 for characteristic data. In some cases, the foils were annealed at about 150°C, but this produced no noticable difference in the subsequent

[1] M. Tinkham, Phys. Rev. **129**, 2413 (1963).
[2] The basic difficulty is that the parameters of the theory, i.e., the Ginzburg–Landau parameter κ and the coherence length ξ, are no longer constants for thin films, but — because of boundary scattering, for example — become functions of the film thickness.
[3] E. H. Rhoderick, Proc. Phys. Soc. (London) **79**, 586 (1961). J. Maldy, L. Davadieu, and E. Saint-Maria, Compt. Rend. **B264**, 416 (1967); G. K. Chang and B. Serin, Phys. Rev. **145**, 274 (1966).
[4] G. D. Cody and R. E. Miller, Phys. Rev. Letters **16**, 697 (1966).
[5] K. Maki, Physics **1**, 21 (1964); **1**, 127 (1964); P. de Gennes, Physik Kondensierten Materie **3**, 79 (1964); E. Helfand and N. R. Werthamer, Phys. Rev. **147**, 288 (1966).
[6] A. A. Abrikosov, Zh. Eksperim. i Teor. Fiz. **32**, 1442 (1957) [English transl.: Soviet Phys.—JETP **5**, 1174 (1957)].
[7] G. Lasher, Phys. Rev. **154**, 345 (1967).
[8] A. L. Fetter and P. C. Hohenberg, Phys. Rev. **159**, 330 (1967).
[9] E. A. Davies, Proc. Roy. Soc. (London) **A255**, 407 (1960).
[10] E. Guyon, C. Caroli, and A. Martinet, J. Phys. (Paris) **25**, 683 (1964).
[11] A similar detailed study of In–1.15 at.% Bi led to a similar result. In the latter case, the temperature at which the behavior changes from type I to type II is somewhat lower, being approximately 1.5°K as opposed to about 2.5°K for In–1.25 at.% Bi.

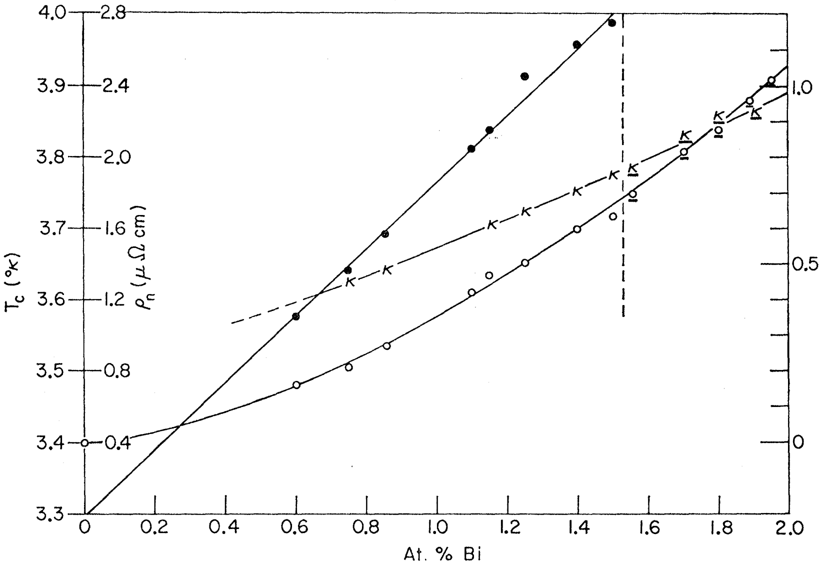

FIG. 1. The resistivity, transition temperature, and κ values of the series of In–Bi alloys used in the present study. The κ values were obtained by extrapolation of the temperature-dependent κ_1 and/or κ_2 values to the limit $T=T_c$. The data to the right of the dashed line with the underlined data points are after T. Kinsel, E. A. Lynton, and B. Serin, Rev. Mod. Phys. **36**, 105 (1964).

measurements. Disk-shaped foil samples (0.7 cm diameter) were placed in a vibrating sample magnetometer for magnetization measurements in both parallel and perpendicular fields. In most instances the foils were measured before and after electroplating with Cr, which served to suppress the superconducting sheath[12] and greatly increased the reversibility of the magnetization curves.

EXPERIMENTAL RESULTS

Detailed studies were carried out on the resistive transitions in numerous samples with κ values in the range 0.45–0.75. As a result of these studies, we have been led to a number of conclusions which are described in the following.

Case I: κ_1, only slightly less than $1/\sqrt{2}$, is illustrated in Fig. 2 for In–1.25 at.% Bi ($\kappa \approx 0.64$).

(a) For a bare foil aligned parallel to the applied field H_0, as the current is decreased, a point is reached where the resistance *onset* field becomes constant even though the high-field tail of the transition curve continues to vary (see curves A′–C′ of Fig. 2). We believe this low-current-limit onset field should be associated with H_{c3} rather than a value associated with the restoration of total normal-state resistance. By adhering to this identification, as will be noted in the following, good agreement with the theoretical ratio 1.69 for H_{c3}/H_{c2} was observed consistently.[13] The onset field was strongly dependent on angle of the foil with the field as expected so that considerable care was taken in achieving accurate alignment. The tail above H_{c3} in the present data is not understood. Possibly it arises from surface inhomogeneities and/or irregularity.

(b) For the plated foils, apparently the low-current-limit resistance onset again marks the parallel transition field which in this case is H_{cB}, the bulk first-order transition field. (See curves A–E, Fig. 2, and note that the notation is that $\hat{H}$ denotes a plated sample.) Again there are current-dependent tails at the high-field end, possibly because of incomplete suppression of the surface superconductivity.

(c) When the foils were perpendicular to the applied field, the bare foils, particularly, showed marked sensitivity to transport current density at values below 10^2 A cm^{-2}, probably because of sheath superconductivity on the edges. The plated foils, on the other hand (Fig. 2, curves A″–D″), showed a well-defined knee in the resistance curves at $R=R_n$ and $H=H_\perp$. Below the knee one always observed a current-dependent tail, probably due to flux flow.[14] The position of the knee, $H_\perp$, for the case shown, $d<0.005$, and $\kappa_1(T)\simeq 0.7$, is believed to determine H_{c2}. This is the principal hypothesis of this paper and evidence for it is summarized below, particularly in the data of Table I.

Case II: $\kappa_1 > 1/\sqrt{2}$. The parallel unplated transitions reveal H_{c3} as in case *I*. For the plated samples, both the parallel and perpendicular transition curves are current-dependent and appear to approach the normal-state resistance at H_{c2}. The current dependence is similar to curves A″–D″ of Fig. 2 and probably is associated with pinning effects on the flux-flow resistance.

Case III: $0.418<\kappa_1<1/\sqrt{2}$ and the samples are sufficiently thick so that $H_D \approx H_{cB}$. In this case the unplated

[12] D. Saint-James and P. G. de Gennes, Phys. Letters **7**, 306 (1963). The plating was effected by electrolysis from solution. Interferometric studies indicated that the Cr films were substantially less than 500 Å in all cases.

[13] This leads us to conjecture that in previous works, where higher ratios were deduced from resistive data, the value of H_{c3} was determined not from resistance onset but from the high end of the tail near $R=R_n$.

[14] J. A. Cape and I. F. Silvera (unpublished).

TABLE I. Data for In–1.25 at. % Bi ($T_c \approx 3.65$°K).

		Resistive				Magnetic				
T(°K)	$H_{\|\|}/1.69$ (G)	H_{c2} dirty-limit (G)	$\hat{H}_{\|\|}$ [a] (G)	$\hat{H}_{\perp}$ (G)	κ_{1R} [b]	$H_{\perp M}$ (G)	$H_{\|\|M}$ (G)	κ_{1M} [c]	$\kappa_{2M}^{\|\|}$	$\kappa_{2M}^{\perp}$ [d]
3.56	14	14	16	···	0.62	···	17	···	e	0.67
3.47	29	29	32	28.6	0.64	28	31	0.64	e	0.67
3.37	43	44	47.5	43	0.65	42	46	0.65	e	0.69
3.16	74	74.6	79	73.5	0.66	72	77	0.66	e	0.72
2.83	125	124	130	125	0.68	120	124	0.68	e	0.74
2.45	178	177	178	176	0.71[e]	173	174	0.70	0.71	0.74
2.00	236	237	235	235	0.71[e]	232	232	0.71[e]	0.72	0.75
1.43	299	303	296	298	0.71[e]	···	295	···	0.73	···
1.21	···	···	···	···	···	···	···	···	0.735	···
1.20	···	···	···	···	···	···	···	···	···	0.78

[a] $\hat{H}_{\|}$ denotes Cr plated sample parallel to the applied field.
[b] $\kappa_{1R} \equiv H_{\| R}/1.69\sqrt{2}\hat{H}_{\| R}$.
[c] $\kappa_{1M} \equiv H_{\perp M}/\sqrt{2}H_{\| M}$.
[d] Evaluated using $\beta_v = 1.16$ (Ref. 20).
[e] In–1.25 at.% Bi is apparently changed from type I to type II at about 2.5°K.

parallel transitions again give H_{c3} in the low-current limit. However, the current carrying capacity of the sheath falls off strongly as κ becomes smaller. For example, the resistance onset field was current-independent only for $j < 10^{-5}$ A/cm of surface for In–0.86 at.% Bi ($\kappa \approx 0.5$). The plated sample transitions in this case appear to be determined by H_{cB} for both parallel and transverse orientations. In a parallel field, the resistance onset is at H_{cB} and appears similar to case Ib. The perpendicular transition exhibits a well-defined knee at H_{cB} with current-dependent curves not unlike those of Fig. 2, case Ic. These could be due to intermediate-state flux flow.

An evident conclusion from the foregoing is that using thick foils ($H_D \approx H_{cB}$) parallel field resistance measurements of plated and nonplated samples permit precise determination of H_{cB} and H_{c3}, respectively, and thus provide an independent measure of κ_1. Such results for In–1.25 at.% Bi are shown in Table I, where the data are also compared with theoretical values derived from the dirty-limit, weak-coupling approximation of the microscopic theory.[5] The theoretical critical-field curves are prescribed completely by specifying the transition temperature T_{c0} and the initial slope $(dH_{\text{crit}}/dT)_{T_{c0}}$. Therefore, some additional effort in the form of repeated measurements near T_{c0} was made to get good values for those numbers though this curve fitting procedure is obviously not the optimum one. The leading values in the experimental critical-field columns, then, together with the transition temperature, determine all the values shown in the theoretical columns. One notes that the agreement for H_{c2} between

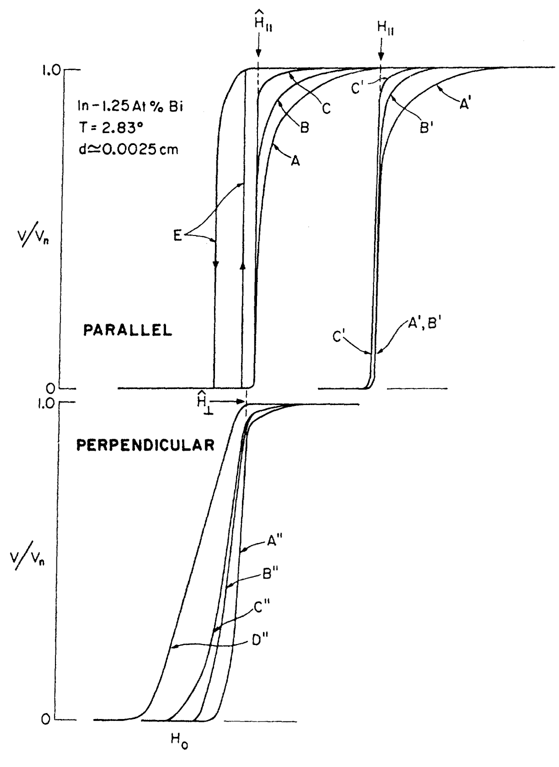

FIG. 2. Normalized resistive voltages in In–1=25 at.% Bi ($\kappa_1 \approx 0.64$). For curves A–E, and A′–C′, the foil is parallel to the applied field. For curves A–E, the surface is Cr plated and the current densities are 0.4, 20, 200, and 2000 A cm^{-2}, respectively. For A′–C′ the sample is as rolled and the current densities are 0.1, 0.3, and 5.0 A cm^{-2}. For curves A″–D″, the foil is plated and perpendicular to H_0. The J values are 0.4, 2.0, 20.0, and 200 A cm^{-2}. Note that $\hat{H}_{\perp}$ is slightly less than $\hat{H}_{\|}$.

the dirty-limit-theory values and the values derived from $H_{c3} \equiv H_{||}$ are very good. One may conclude that the critical fields and κ_1 (for $\kappa_1 > 0.418$) values may be measured fairly accurately and easily by resistive techniques with the use of Cr plating to quench the superconducting sheath.

A typical example of the magnetometer data is shown in Fig. 3. One notes that the magnetic moment behavior in a parallel field is characteristic of a type-I superconductor whereas in the perpendicular case, M vanishes at $H_{\perp M}$ significantly less than H_{cB}. Values of H_{cB} and H_{c2} thus obtained were in very good agreement (within 2%) with those obtained from the resistance data (Fig. 2). A second noteworthy feature is that the perpendicular magnetization curve is nearly reversible in the vicinity of $H_{\perp M}$. The reversibility in the perpendicular case was observed to be markedly dependent on the presence of Cr on the foil surface, particularly on the edges of the foil. The behavior is strongly suggestive of induced surface currents in the sheath as has been noted frequently by others.[15] The addition of Cr to the foil edges served to decrease symmetrically the angle subtended by the increasing-field and decreasing-field magnetization curves as shown in Fig. 3. Accordingly, the line bisecting this angle at $H_{\perp M}$ in Fig. 3 was assumed to represent the equilibrium magnetization.

Values of $\kappa_1 \equiv H_{c2}/\sqrt{2}H_{cB}$ were computed from $\hat{H}_{\perp M}/\sqrt{2}\hat{H}_{||M}$, the critical fields of the magnetization curves, and from $H_{||R}/1.69\sqrt{2}\hat{H}_{||R}$, the critical fields of the resistance data. (Here the carat over H denotes a plated sample.) These values are recorded in Table I. From the slopes of the magnetization curves the parameter κ_2 was determined through the generalized Abrikosov[6] formula

$$-4\pi M = (H_{c2} - H)/(2\kappa_2^2 - 1)\beta_v. \quad (1)$$

In applying Eq. (1), it is necessary to note that H is not generally the externally applied field H_0, but depends on the shape of the sample. As a result, for bulk foils, for which the thickness-to-diameter ratio is negligible compared to unity, it can be shown that Eq. (1) takes the forms[16]

$$-4\pi M = (H_{c2} - H_0)/(2\kappa_2^2 - 1)\beta_v, \quad (\text{foil} \,||\, \mathbf{H}_0) \quad (2)$$

$$-4\pi M = (H_{c2} - H_0)/[(2\kappa_2^2 - 1)\beta_v + 1], \quad (\text{foil} \perp \mathbf{H}_0). \quad (3)$$

[15] L. J. Barnes and H. J. Fink, Phys. Letters **20**, 583 (1966). See also R. W. Rollins and J. Silcox, Phys. Rev. **155**, 404 (1967), and references therein.

[16] J. A. Cape and J. M. Zimmerman, Phys. Rev. **153**, 416 (1967); I. O. Kulik, Zh. Eksperim. i Teor. Fiz. Pis'ma v Redaktsiyu **3**, 395 (1966) [English transl.:—JETP Letters **3**, 259 (1966)].

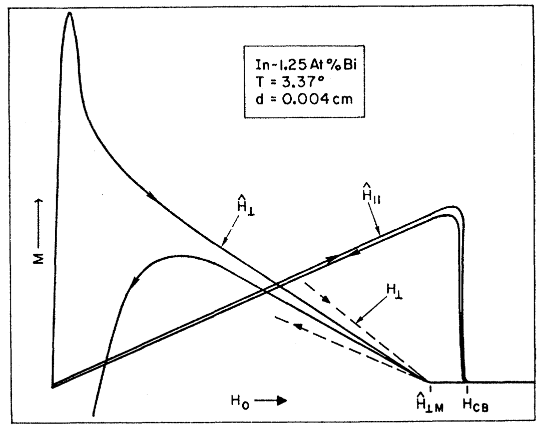

FIG. 3. Magnetic moment of an In–1.25 at.% Bi foil in parallel ($H_{||}$) and perpendicular ($H_\perp$) applied magnetic field. The notation $\hat{H}$ denotes a sample plated with Cr. The dashed lines near $\hat{H}_{\perp M}$ show the behavior of an unplated sample. It is argued in the text that the perpendicular magnetization near $H_\perp$ is that of a vortex-lattice mixed state.

One is then able to obtain two independent values of κ_2 from the magnetization measurements. This feature proves to be valuable in the present case inasmuch as In–1.25 at.% Bi becomes type II below about 2.5°K, and we are thus able to compare the respective κ_2 values.

Table I summarizes the resistive and magnetic data on In–1.25 at.% Bi. The conclusions that may be drawn from these data are largely evident. (1) Both the resistance and magnetic data suggest that In–1.25 at.% Bi is type II below roughly 2.0 to 2.5°K. Note, for example, that κ_{1R} (as defined in Table I) increases to $0.71 \approx 1/\sqrt{2}$ and then levels off. This would be expected to happen when $\hat{H}_{||R}$ becomes equal to H_{c2}. (2) Both κ_1 and κ_2 increase with decreasing temperature as predicted by dirty-limit theory.[5,17] In fact, as should be expected, the temperature dependence of "H_{c2}" (i.e., both $H_{||}/1.69$ and $\hat{H}_{\perp M}$) is fitted quite precisely with a dirty-limit theoretical curve.[18] Dirty-limit theory further predicts that $\kappa_1(T_c) = \kappa_2(T_c)$ and that the two parameters differ by no more than about 3% [19] at any temperature below T_c. In this connection one notes that (3) the κ_2 values derived from Eqs. (2) and (3) and the slopes of the parallel and perpendicular magnetization curves (using $\beta_v = 1.16$)[20] are nearly equal and are also consistent with the observed κ_1 values. Over the entire temperature range, the $\kappa_2^\perp$ values appear to be consistently larger than the other κ's by what is—to within experi-

[17] C. Caroli, M. Cyrot, and P. G. de Gennes, Solid State Commun. **4**, 17 (1966).

[18] For In–1.25 at.% Bi, using a free-electron model, we estimate $l/\xi_0 \sim 0.05$.

[19] For the dependence on mean free path, see G. Eilenberger, Phys. Rev. **153**, 584 (1967).

[20] W. H. Kleiner, L. M. Roth, and S. H. Autler, Phys. Rev. **133**, A1227 (1964).

mental scatter—a constant factor. This discrepancy, however, is within the corresponding experimental uncertainty[21] in the slopes of the perpendicular magnetization curves.

Conclusion (3) is most significant insofar as it indicates that the present data are entirely consistent with the "mixed-state" hypothesis; i.e., that a uniformly flux-penetrated state, terminating at H_{c2}, occurs in the present case for a bulk foil perpendicular to H_0 even though κ is less than $1/\sqrt{2}$. Data entirely similar to but less detailed than those of Table I have been observed for In–1.15 at.% Bi, where κ is still smaller and the transition to type II is at a still lower temperature.

One may question whether an intermediate-state surface-energy model can also be used to explain the present data. For example, for the slope of the magnetization curve near the critical field, Andrew and Lock[22] have derived the expression

$$4\pi(dM/dH)=1+6.1(\Delta/d)^{2/3}. \qquad (4)$$

Values of Δ (the characteristic length associated with the interphase surface energy) chosen to fit the observed slopes were found to be consistently nearly an order of magnitude larger than predicted by the Ginzburg-Landau theory.[23] Moreover, the transition field associated with Eq. (4) is supposed to be thickness-dependent. The present data, however, appear to be characteristic of a thickness-*independent* behavior and in fact indicate that the transition field is H_{c2}, not H_D. In conclusion, therefore, it appears that the mixed state, not the intermediate state, is the appropriate model to explain the present observations.

[21] This comes about because the perpendicular magnetization is at present calibrated against the *parallel* Meissner slope as is the parallel magnetization. Experiments with saturated ferromagnetic foils indicate that the calibration is correct to within a few percent, but this is the same magnitude as the discrepancy needed to be resolved here.

[22] E. R. Andrew and J. M. Lock, Proc. Roy. Soc. (London) **A63**, 13 (1949).

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I wish to thank D. H. Leslie for technical assistance, J. W. Savage for alloy preparation, W. M. Robertson for interferometric studies of the Cr films, and T. G. Berlincourt and R. R. Hake for their careful criticism of the manuscript.

[23] V. L. Ginzburg, Physica **24**, S42 (1958).

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Flux-Transport Noise in Type-II Superconductors

G. J. VAN GURP

Philips Research Laboratories, N. V. Philips' Gloeilampenfabrieken, Eindhoven, Netherlands

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The transport of magnetic flux in type-II superconductors under the influence of a transport current is shown to give rise to a noise voltage, superposed on the dc voltage. This flux-flow noise is analogous to shot noise limited by transit-time effects, and is caused by the motion of discrete units of flux between the edges of the superconductor. If the flux is in a circular motion, so that no transit of flux between the edges takes place, no flux-flow noise can be detected. It is shown from measurements of the frequency spectrum of the noise voltage in vanadium foils that the dc voltage is built up of voltage pulses generated during flux motion, whose duration is equal to the transit time of the flux units. Because of interaction with pinning centers, a fraction of the flux does not take part in the motion. This fraction is found to increase with decreasing temperature and current. It increases with magnetic field in cold-rolled vanadium that exhibits a resistance minimum, but decreases with field in annealed vanadium, where such a minimum is not present. The moving flux units are bundles of flux lines. The average number of flux quanta in a bundle depends on pinning conditions, as is shown for vanadium foils and an indium-thallium crystal. This number decreases with increasing temperature, current, and magnetic field. It is also found to fluctuate owing to interaction with the pinned fraction. At high fields and currents, flux transport in annealed material is in the form of single flux quanta. When the power dissipation in the specimen is increased, a noise voltage with approximately $1/f^b$ frequency dependence is found, with $2<b<3$. This flicker noise, as we call it, is shown to be generated by temperature fluctuations in the foil. These fluctuations are caused by nucleate boiling of liquid helium, and they virtually disappear below the helium λ point. Some flicker noise is left there, due to pressure variations above the liquid.

1. INTRODUCTION

If a type-II superconductor is subjected to a magnetic field greater than a critical value H_{c1} and if a transport current is passed that is greater than a critical value J_c, a dc voltage can be measured. This voltage is generally accepted as being due to the viscous motion of supercurrent vortices containing a flux quantum, under the influence of a Lorentz force.[1] The vortices are extended in the magnetic field direction and are therefore called vortex lines or flux lines.

The direct experimental evidence for this motion,

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