Transverse Magnetization of In–Sn Films

P. B. MILLER, B. W. KINGTON,† and D. J. QUINN‡

International Business Machines Corporation, Thomas J. Watson Research Center, Yorktown Heights, New York

INTRODUCTION

Studies of the intermediate state of thick samples (d much larger than the penetration depth) have been successfully interpreted in terms of a first-order transition and may be used to infer the surface energy of a normal–superconducting boundary.[1,2] It has recently been pointed out by Tinkham[3] that experiments on thin films in transverse fields may be interpreted in terms of a second-order transition occurring at an upper critical field (H_2). Several properties of thin films in transverse fields have been observed to change continuously to the normal state value at H_2. These include measurements of flux penetration,[4] thermal conductivity,[5] and the present measurements of magnetic moment. Since the volume fraction of superconducting material may be considerably less than unity near H_2, such a continuous variation gives an ambiguous indication of the order of the transition. A comparison of theory with the data also seems desirable to further establish the order of the transition.

Transverse magnetization curves for relatively thick Sn foils have been measured by Andrew and Lock.[1] The present magnetization curves of thin In–Sn films have a similar shape and may be characterized by two transition fields, a lower field H_1 where the sample enters the intermediate state and the upper field H_2. Typical magnetization curves are shown in Fig. 1. The field H_1 is characterized by the onset of trapping and by a deviation from linear magnetization. The initial slope of the magnetization is determined by an effective demagnetizing factor of the film.[1] The field region between H_1 and the saturation field for trapped flux corresponds to a macroscopically nonuniform intermediate state, as shown by the magneto-optic Faraday rotation and described in Sec. III. In the region near H_2 the field distribution over the film surface is macroscopically uniform and it is in this region that a comparison with theoretical models seems most suitable.

Abrikosov has formulated a model of a second order transition for negative surface energy superconductors based on the Ginzburg–Landau (G–L) theory.[6] His intermediate (or mixed) state near H_2 consists of a periodic arrangement of vortices in the plane perpendicular to the applied field. The model shows good agreement with experimental data on those bulk alloys for which the Ginzburg–Landau parameter $\sqrt{2}\,K > 1$.[7,8]

If a second-order transition also occurs for thin films in transverse fields then an Abrikosov-type model would be appropriate. Tinkham has proposed a similar model based on circular fluxoid vortices.[3] Both of the above models give an identical result for H_2,

$$H_2 = \sqrt{2}\,KH_c\,, \tag{1}$$

where H_c is the bulk critical field. The above relation for H_2 is a general result of the G–L theory for a second-order transition, since it appears as the lowest eigenvalue of the linearized G–L equations with the boundary condition that the order parameter be bounded at infinity. Equation (1) is independent of the detailed nature of the mixed state, i.e., of the fluxoid quantum number.

Resistive transitions of thin films of various thicknesses have been measured by Rhoderick.[9] In addition, Broom and Rhoderick have shown that flux penetration is complete at a lower field than that necessary for restoration of the full normal resistance.[4] They ascribe the difference to the presence of continuous superconducting threads persisting even when the intermediate state has disappeared. Tinkham has analyzed the resistive transitions of Rhoderick and has shown that the temperature dependence of H_2 for the tin films is in accord with Eq. (1). By a comparison of the present magnetic moment measurements with Eq. (1) we may obtain further

† Present address: The Royal Society Mond Laboratory, Cambridge, England.

‡ Present address: United Aircraft Corp., Hartford, Connecticut.

[1] E. R. Andrew and J. M. Lock, Proc. Phys. Soc. (London) **A63**, 13 (1950).

[2] E. A. Davies, Proc. Roy. Soc. (London) **A255**, 407 (1960).

[3] M. Tinkham, Phys. Rev. **129**, 2413 (1963).

[4] R. F. Broom and E. H. Rhoderick, Proc. Phys. Soc. (London) **79**, 586 (1961).

[5] D. E. Morris, Ph.D. thesis, University of California, Berkeley (1962).

[6] A. A. Abrikosov, Zh. Eksperim. i Teor. Fiz. **32**, 1442 (1957) [English transl.: Soviet Phys.—JETP **5**, 1174 (1957)].

[7] B. B. Goodman, IBM J. Res. Develop. **6**, 63 (1962).

[8] T. Kinsel, E. A. Lynton, and B. Serin, Phys. Letters **3**, 30 (1962).

[9] E. H. Rhoderick, Proc. Roy. Soc. (London) **A267**, 231 (1962).

evidence for the order of the transition. In addition the slopes of the magnetization curves near H_2 may be used to infer some additional properties of the mixed state.

EXPERIMENTAL DETAILS

The samples were prepared from 99.999% Sn and In supplied by the Consolidated Mining and Smelting Company of Canada. The substrates used were rectangular sections of polished polycrystalline quartz. Sample materials were placed in Ta crucibles, which had been previously degassed, and after melting were also degassed to a pressure of approximately 5×10^{-8} Torr. The crucible temperature for all the evaporations was 1370°C and the background pressure on the average was 5×10^{-6} Torr. The alloy concentrations were controlled by evaporating to completion known amounts of the constituents. Films were deposited onto liquid nitrogen cooled substrates.

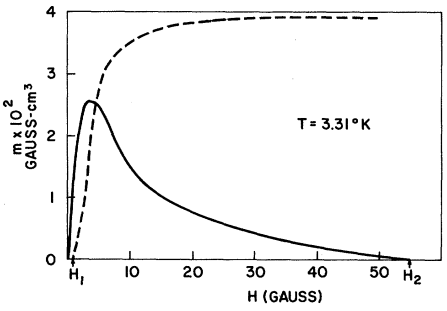

FIG. 1. Typical magnetization curve (sample No. 4). The magnetic moment is plotted versus the applied field. The dotted curve is the negative of the moment in zero field (trapped flux) after turning off an applied field H.

Magnetization measurements were obtained by the general method described by Shoenberg.[10] The present apparatus employs two pairs of flat coils as the sensing device, with the sample movable from between one pair to between the other, which are wound in series opposition, thereby inducing a signal. Each coil has 1750 turns of No. 43 Formvar-coated copper wire and at helium temperature has a resistance of 4 Ω. The four coils are connected in series with a galvonometer, giving an over-all sensitivity of 1.19×10^{-3} G cm³/mm of deflection. Calibration of the instrument was achieved by measuring the galvonometer deflection due to a known current in a one-turn coil having the same outside dimensions as the samples. A more detailed description of the apparatus will be available.[11] Within the limits of the experiment the last 10% of the magnetization decreases linearly with field. For this reason a least squares fit of the data in that region was used to determine the zero intercept viz. H_2.

Sample thicknesses were calculated from measurements of the resistance at 300° and 4.2°K and the film area. An additional interferometric technique was used to check the thickness of several of the films and gave agreement with the resistance method. The concentrations were determined by x-ray fluorescence. Helium-temperature to room-temperature resistance ratios were measured for several of the samples and were found to be quite linear with the analyzed concentrations. Likewise the superconducting transition temperature T_c was found for the pure and the most highly concentrated alloy by noting the temperature at which all the voltage across the sample due to a 1-mA measuring current disappeared. The T_c values of the remaining specimens were then calculated by assuming a linear dependence of T_c on concentration.

The field H_1 at which entrance into the intermediate state occurs was found by locating the initial appearance of trapped flux. This was accomplished by measuring the moment of the sample in zero field after each incremental increase in applied field.

Faraday rotation pictures were taken by means of conventional polarizer and analyzer techniques with the polarized beam deflected by prisms into a cryostat where the plane of polarization is rotated by a polished cerium metaphosphate disk placed on the thin film sample. This apparatus is described in detail by Haering *et al.*[12]

RESULTS

A typical magnetization curve is shown in Fig. 1. The field marked H_1 may be defined by either the onset of flux trapping or the initial deviation from linear magnetization. At the field H_2 the sample becomes magnetically normal. The trapped flux starts to increase at H_1 and attains a maximum value M_s.

The Faraday rotation photographs were taken with the analyzer set at extinction so regions of zero field appear black. The intermediate state domain size is beyond the resolution of the instrument (viz.) 10^{-1} cm.[12] A macroscopically nonuniform pattern was seen in the photographs in the region of the magnetization curve between H_1 and the field where the trapped flux saturates (H_s). Above H_s the photo-

[10] D. Shoenberg, Phys. Soc. Cambridge Conference Rept. 2, 85, 93 (1947).

[11] D. J. Quinn (to be published).

[12] R. R. Haering, A. M. Toxen, P. B. Miller, W. P. Dumke, and B. W. Kington, Solid State Electronics 6, 365 (1963).

graphs indicate the field is uniformly penetrating the film. The circular symmetry was maintained in the Faraday patterns indicating their dependence on the samples macroscopic geometry rather than on local physical defects. Similar patterns were obtained independent of the alloy concentration in the thickness range studied.

The initial slope of the magnetization is given by (1), (11) to be

$$K/K_0 = (1/3)(w/d) , \qquad (2)$$

where K is the film susceptibility and $K_0 = (4\pi)^{-1}$. Our data are in good agreement with this relation. We find the field H_1 to be two orders of magnitude larger than that predicted by the simple demagnetizing ratio of width to thickness. This difference can be understood when one takes into account the penetration effect which determines the maximum current density at the edge of the film. This current density has been calculated by Marcus[13] with the inclusion of the penetration effects and is given by

$$J_m = 1.23\, c/4\pi\lambda(w/d)^{\frac{1}{2}}H . \qquad (3)$$

The importance of penetration in determining the current density may be qualitatively understood as an averaging over a distance λ of the very sharply peaked field distribution at the edge, which would be present in the absence of penetration. Assuming that the film enters the intermediate state when $J_m = J_c = cH_c/4\pi\lambda$ we get an expression for H_1 from Eq. (3). This relation gives the order of magnitude and temperature dependence observed in our data. Torque measurements of the magnetic moment of thin Sn and In films[14] give an H_1 which is two orders of magnitude lower than that found in the present work. The value for the parallel critical field obtained by these torque measurements are only half of those reported by Toxen.[15] These differences are not well understood. The present results for H_1 also imply that the alignment problem in a parallel field is not nearly as critical as the demagnetizing factor criterion. To prevent entry into the intermediate state in a parallel field one needs to keep the perpendicular component less than H_1 which is of the order of several gauss.

Several properties of thin films in a transverse field change continuously to the normal state at H_2.[4,5] Similar measurements on bulk specimens also show a continuous transition near H_2.[1] Hence the order of the transition cannot be experimentally inferred from continuity arguments. However, the existence of a second-order transition may be directly inferred from a comparison of the experimental data with Eq. (1), since the relation may be derived from the G–L equations under the general assumption of a second-order transition together with the boundary condition that the order parameter be bounded at infinity. The Ginzburg–Landau parameter K was computed as a function of alloy concentration and temperature from the bulk critical field measurements of Quinn[11] and the penetration depth measurements of Toxen.[15] These values of K for 3000-Å films are listed in Table I. The temperature dependence of H_2/H_c for a pure In film is shown in Fig. 2. Figure 3 shows the concentration dependence of H_2. Small corrections have been made to the data points for Fig. 2 to adjust it to a thickness of 3000-Å and Fig. 3 to a reduced temperature of 0.9 by use of Eq. (1). The agreement of the data with Eq. (1) in absolute value, temperature dependence and impurity dependence is strong evidence of a second-order transi-

TABLE I. Summary of film properties.

Sample No.	Thickness d Å	at. % Sn	T_c K°	t	H_2 gauss	$\frac{4\pi M(H)}{(H_2 - H)V}$ ($\times 10^3$)	K
1	2300	0	3.40	0.708	69.5	1.3	0.26
				0.891	21.0	1.1	
				0.956	6.7	1.4	
				0.894	20.6	1.1	
2	2550	2.3	3.67	0.901	34.2	0.24	0.38
3	2800	3.0	3.75	0.884	48.5	0.07	0.43
4	3000	3.3	3.78	0.875	51.2	0.11	0.45
5	1970	6.6	4.16	0.891	57.5	0.08	0.71
6	3000	7.1	4.22	0.911	71.2	0.05	0.76

[13] P. M. Marcus (to be published).

[14] B. K. Sevastyanov, Zh. Eksperim. i Teor. Fiz. **40**, 52 (1961) [English transl.: Soviet Phys.—JETP **13**, 35 (1961)].

[15] A. M. Toxen, Phys. Rev. **127**, 382 (1962); A. M. Toxen and M. J. Burns, Phys. Rev. **130**, 1808 (1963).

tion. The same temperature dependence has also been observed for pure Sn films.[3,9]

At low reduced temperatures (<.7) consecutive measurements of the magnetization at the same field near H_2 varied widely. This scatter, however, did not set in at any particular field, thus indicating its source to be the sample and not the instrumentation. It might be speculated that this scatter is due to the metastability of the states near H_2.

Although the transition field H_2 is independent of the detailed structure of the state, the slope at H_2 may be used to infer some of its properties. The Abrikosov model for the state assumes a fluxoid quantum number of one and no variation of order

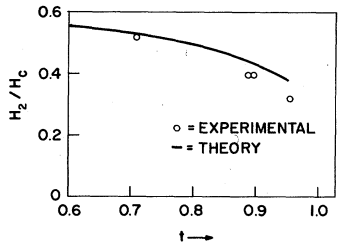

FIG. 2. Temperature dependence of H_2/H_c for pure In film. The solid curve is the G–L theory.

parameter in the field direction and predicts a slope given by[6]

$$-4\pi M/H_2 - H = 1/1.18(2K^2 - 1)\,, \qquad (4)$$

where M is the magnetic moment per unit volume. The above relation is in good agreement with the bulk data.[8] The circular fluxoid model discussed by Tinkham is also based on a fluxoid quantum number of one and gives a slope of the same magnitude. The present data (Table I) indicates a slope several orders of magnitude larger than these models. This large

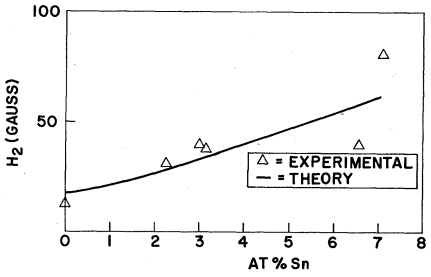

FIG. 3. Concentration dependence of H_2. The solid line is the G–L theory.

slope cannot be accounted for in terms of any model with current loops of small area (i.e., fluxoid quantum number one), and suggests the investigation of models with current loops of large area creating large magnetic moments (i.e., large fluxoid quantum numbers).

ACKNOWLEDGMENTS

We would like to thank R. R. Haering, P. M. Marcus, and A. M. Toxen for their helpful discussions and suggestions on these topics. The technical assistance of R. J. Parker and A. C. Burgess were of great value to this work.

Discussion 7

GOODMAN: I am just wondering if I have understood properly that your magnetization goes to zero at the upper transition field with a much steeper gradient than you expect. Is that right?

P. B. MILLER, *IBM Research Center:* The gradient is steeper than is given by the Abrikosov theory which says that the slope of magnetization is one over some constant divided by the field.

GOODMAN: So this means that the area under your magnetization curve is much larger than the difference in free energies between the superconducting and normal states.

MILLER: That is correct. It is a highly irreversible transition, unlike bulk material. I would like to emphasize that even though this is a highly irreversible transition, the Ginzburg–Landau equation still holds—the equation $H_2 = \sqrt{2}\,\kappa H_c$. It appears as the lowest eigenvalue and it makes no difference whether the state is metastatic or stable. You can't have any superconductivity above that field.

M. TINKHAM, *University of California:* Two short comments: First, although the matter of the magnetization slopes being so large can be looked at in terms of loops larger than those corresponding to unit fluxoid, I think it is simpler to think of it as a certain amount of current of a macroscopic sort circulating around the film, as in the Bean model. The point is that, in determining the magnetic moment, the radius of the loop comes in strongly. Hence a relatively small current circulating over the whole surface of the film would give a moment which would swamp out that of the small vortices even if there were much more kinetic energy associated with the vortices. So this measurement is, I think we all agree, not much good at showing any details about the vortices.

MILLER: Well, you can call the large circulating current that you postulate a vortex of large fluxoid quantum number.

TINKHAM: I'll concede that, but I think it's just macroscopic current and the quantum numbers are so large that the correspondence principle can be applied. The other point I want to make very briefly is that, in most of your films, kappa is less than $1/\sqrt{2}$, so this is a rather unusual case similar to some of the things that I have talked about. You get type II behavior even when the critical field is less than the bulk critical field; it is imposed by the film geometry.