

## Intrinsic Size Effects in Type-II Superconducting Films

A. S. JOSEPH

*North American Aviation Science Center, Thousand Oaks, California*

AND

W. J. TOMASCH\* AND H. J. FINK\*

*Atoms International, A Division of North American Aviation, Incorporated, Canoga Park, California*

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Null-deflection torque-magnetometer measurements performed on well-annealed low- $\kappa$  type-II superconducting films in nearly parallel magnetic fields suggest that flux entry may be delayed to a field  $H_s$  which can be greater than  $H_c$ . It is found that  $H_s$  depends on the film thickness, the coherence length, and  $\kappa$ . Furthermore, our results indicate that in films having a thickness less than 20 coherence lengths, two critical fields  $H_s$  and  $H_{c3}$  are observed, but not  $H_{c2}$ . The Abrikosov mixed state may be therefore suppressed in favor of a new state which resembles a giant vortex. Such a state is magnetically hysteretic and has been postulated by a recent theory.

**T**ORQUE magnetometer measurements have been performed on well-annealed type-II ( $\kappa \approx 1$ ) superconducting films 1 to 10  $\mu$  thick situated in nearly longitudinal magnetic fields. The results indicate that delayed flux entry and the ability to trap high fields are intrinsic and probably related phenomena which depend on the ratio of film thickness to coherence length. Furthermore, it appears that a state corresponding to a superconducting sheath and a normal core can be retained in zero applied field after suitable field cycling. Possible suppression of the Abrikosov mixed state in these films is discussed and our findings are compared with recent theoretical predictions.

The details of the experimental apparatus<sup>1</sup> and sample preparation<sup>2</sup> have been discussed elsewhere. Films of Sn+~4 at.% In and Pb+~5 at.% Tl were annealed in vacuum for a minimum of 60 h at 180°C. They were then cooled to liquid-helium temperatures in a magnetic field of less than 1 mG which was oriented approximately in the plane of the films. In Figs. 1(a) and 1(c) the torque  $\tau$  is plotted as a function of the magnetic field for a 1  $\mu$  film of Pb+~5 at.% Tl at 4.2°K and a 2  $\mu$  film of Sn+~4 at.% In at 1.2°K. In each instance the angle  $\theta$  between the applied field and the plane of the specimen was kept small (approximately 0.2°) to avoid flux penetration due to the unfavorable demagnetization effects and still maintain acceptable signal-to-noise ratio. The values  $H_{c1}$ ,  $H_{c2}$ , and  $H_{c3}$  indicated in Figs. 1 and 2 come from torque measurements with  $\theta \approx 0.2^\circ$  on 150- $\mu$ -thick well-annealed foils having the same alloy compositions as the films. These values are characteristic of ideal bulk behavior.<sup>3</sup>

Film values for  $H_{c2}$  obtained in transverse fields<sup>4</sup> and the values for  $H_{c3}$  ( $=1.7 H_{c2}$ ) agree well with the bulk values. Unannealed films exhibited  $H_{c3}/H_{c2}$  values as large as 2.4 and the torque above  $H_s$  was noisy and irreproducible. In Figs. 1(b) and 1(d) we plot the quantity  $\tau/H$  corresponding to Figs. 1(a) and 1(c), where  $\tau/H$  is a measure of an effective moment normal to the magnetic field.<sup>5</sup> As the field is initially increased,  $\tau/H$  increases linearly up to a value<sup>6</sup>  $H_s$  greater than  $H_c$ , goes through a maximum, and then decreases rapidly to zero at  $H_{c3}$ . We shall refer to this as the virgin curve. When the field is decreased from values  $H_i$  greater than  $H_s$  a family of straight lines (in  $\tau/H$  versus  $H$ ) parallel to the linear portion of the virgin curve is obtained. Each of these lines is reversible as the field is cycled between zero and the corresponding  $H_i$ . In particular, when  $H_i = H_{c3}$  the lower curves in Fig. 1 are obtained.

The results obtained with a 10  $\mu$  Pb+~5 at.% Tl film for  $\theta \approx 0.2^\circ$  are shown in Fig. 2. In contrast to the behavior of the 1  $\mu$  film (Fig. 1), hysteresis commences at the bulk value of  $H_{c1}$ , and structure in the  $\tau/H$  versus  $H$  curves occurs near the bulk value of  $H_{c2}$ , both for increasing and decreasing field sweeps.

The difference between the results of Fig. 1 and those of Fig. 2 apparently stems from the difference in the thickness of the films. If the linear portions of the virgin curves of Figs. 1(b) and 1(d) are associated with the Meissner state, then it is clear that entry of flux into the sample is considerably delayed. As the field is decreased from above  $H_s$ , flux trapping is almost complete. Indeed the lower curves of Fig. 1(b) and 1(d) suggest that the trapped fields are nearly equal to  $H_{c3}$ . In contrast, the 10  $\mu$  sample allows some flux to enter at

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<sup>1</sup> A. S. Joseph and A. C. Thorsen, *Phys. Rev.* **133**, A1546 (1964).

<sup>2</sup> W. J. Tomasch and A. S. Joseph, *Phys. Rev. Letters* **12**, 148 (1964).

<sup>3</sup> The  $H_{c1}$  and  $H_{c2}$  values of the Pb+~5 at.% Tl foils agree with those of G. Bon Mardion, B. B. Goodman, and A. Lacaze, *Phys. Letters* **2**, 321 (1962).

<sup>4</sup> D. Saint-James and P. G. de Gennes, *Phys. Letters* **7**, 306 (1963).

<sup>5</sup> B. K. Sevast'yanov and V. A. Sokolina, *Zh. Eksperim. i Teor. Fiz.* **42**, 1212 (1962) [English transl.: *Soviet Phys.—JETP* **15**, 840 (1962)].

<sup>6</sup> A. S. Joseph and W. J. Tomasch, *Phys. Rev. Letters* **12**, 219 (1964).

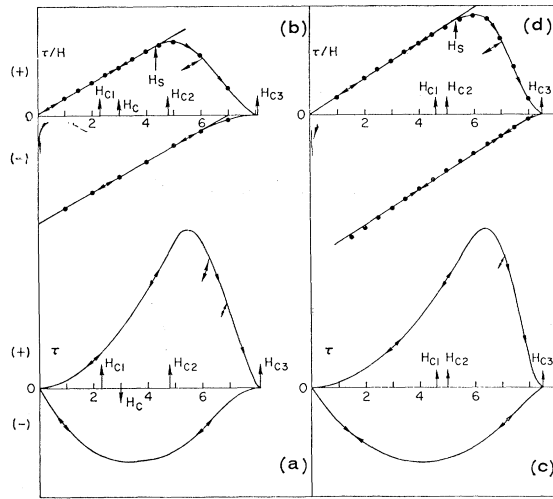


FIG. 1. Plots of  $\tau$  and  $\tau/H$  in arbitrary units as functions of  $H$ . (a) Pb+~5 at.% Tl, 1  $\mu$  thick, 4.2°K; (b) Pb+~5 at.% Tl, 1  $\mu$  thick, 4.2°K; (c) Sn+~4 at.% In, 2  $\mu$  thick, 1.2°K; (d) Sn+~4 at.% In, 2  $\mu$  thick, 1.2°K. Each division on the  $H$  axis is equal to 182 G for (a) and (b) and 61 G for (c) and (d).

$H_{c1}$  as  $H$  is increased and allows flux to escape as the field is decreased. Results on a variety of specimens have indicated that the ability to delay the entry of flux is invariably associated with the ability to trap flux. This suggests that both effects have a common origin.

In bulk superconductors, it is known that for  $H_{c2} < H < H_{c3}$  the magnetization due to induced sheath currents is diamagnetic for increasing fields and paramagnetic for decreasing fields.<sup>7</sup> The magnetization paths which connect the decreasing paramagnetic curve with the increasing diamagnetic curve are reversible parallel straight lines, which have the Meissner effect slope. The applied fields at the connecting points of a

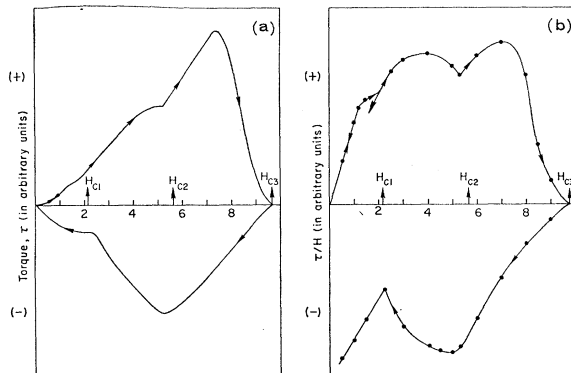


FIG. 2. Plots of  $\tau$  and  $\tau/H$  in arbitrary units as functions of magnetic field. Each division on the  $H$  axis is equal to 182 G, (a) Pb+~5 at.% Tl, 10  $\mu$  thick, 4.2°K; (b) Pb+~5 at.% Tl, 10  $\mu$  thick, 4.2°K.

<sup>7</sup> D. J. Sandiford and D. G. Schweitzer, Phys. Letters **13**, 98 (1964); L. J. Barnes and H. J. Fink, Phys. Rev. **149**, 186 (1966).

given path differ by an amount  $\Delta H$ , and between these fields the currents in the sheath are able to shield completely the interior of the sample and hence conserve the number of fluxoids. Because of the relative magnitudes of contributions from magnetic field energy (volume-dependent) and condensation energy (surface-area-dependent),  $\Delta H$  should be size-dependent. If for the specimens studied in this work  $\Delta H$  approaches  $H_{c3}$ , then one would predict a family of  $\tau/H$  versus  $H$  curves equivalent to those of Fig. 1. Shielding phenomena due to currents in the surface seem to dominate the magnetic behavior of the films as depicted in Fig. 1. When the trapped field for zero applied field is greater than  $H_{c2}$ , it would appear that the specimen is in a state characterized by a superconducting sheath and a normal core. When the trapped field is less than  $H_{c2}$ , it is not obvious from the data whether or not the usual Abrikosov mixed state occurs. It is significant to note, however, that only two critical fields  $H_s$  and  $H_{c3}$  are

TABLE I. Comparison of theoretical and experimental values of  $H_s$ .

Nominal film composition	$\kappa$	$d/2\xi^a$	$(H_s/H_c)_{\text{calc}}^b$	$(H_s/H_c)_{\text{expt}}^b$
Sn+4% In	0.737	9.6	1.10	1.08
Sn+4% In	0.737	1.93	1.60	1.40
Pb+4% Tl	1.02	3.88	1.35	1.37
Pb+5% Tl	1.12	6.17	1.25	1.43
Pb+5% Tl	1.27	2.61	1.87	1.55
Pb+9% Tl	2.0	20.5	1.13	1.35
Pb+8% Tl	1.66	40.5	1.04	1.24
Pb+8% Tl	1.70	9.85	1.25	1.30
Pb+8% Tl	1.70	13.9	1.12	1.21

<sup>a</sup>  $d$  = film thickness;  $\xi$  = coherence length =  $(\phi_0/2\pi H_{c2})^{1/2}$ .

<sup>b</sup>  $H_c$  = thermodynamic critical field;  $H_s$  is defined in Fig. 1.

evident in the data of Fig. 1, with no evidence for an upper field limit for the mixed state (an effective  $H_{c2}$ ) even though the thicknesses of these films are considerably larger than the coherence lengths.

Recent calculations by Fink and Presson<sup>8</sup> have dealt with the question of size effects in type-II superconducting cylinders less than approximately 20 coherence lengths in diameter. While it is not totally correct to compare these results on cylinders with the data of Fig. 1, it is nevertheless interesting to note that a general consistency between the two exists. (1) There is fair agreement between the values for  $H_s$  obtained experimentally (Fig. 1) and theoretically<sup>8</sup> and the results for various films are listed in Table I. In each case  $H_s$  is greater than  $H_c$ . This is especially noteworthy in that for *bulk specimens* it has been previously predicted<sup>9</sup> that  $H_s$  does not exceed  $H_c$ , in contrast to the present results. (2) Fink and Presson conclude that a

<sup>8</sup> H. J. Fink and A. G. Presson, Phys. Rev. **151**, 219 (1966).

<sup>9</sup> P. G. de Gennes, Solid State Commun. **3**, 127 (1965); H. J. Fink, Phys. Letters **20**, 356 (1966).

pure Abrikosov mixed state (having no surface sheath) is energetically less favorable in these cylinders than the lowest energy of a new state (called the giant-vortex state) which is very similar to a superconducting sheath. In this case, they predict only two critical fields  $H_s$  and  $H_{c3}$ , which is consistent with the data of Fig. 1. The magnetic properties of this predicted state are hysteretic, and large field trapping should be an intrinsic characteristic. However, trapped fields presently observed (at zero applied field) are about 50% larger than predicted for the cylinders.

In conclusion, we have presented experimental evidence that intrinsic size effects occur in type-II ( $\kappa \approx 1$ )

superconducting films as thick as sixteen coherence lengths, the magnetic properties of such films differing appreciably from bulk behavior. Some aspects of recent theoretical predictions on thin superconducting cylinders appear to be consistent with the present experimental results.

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## Quantum States and Transitions in Weakly Connected Superconducting Rings

A. H. SILVER AND J. E. ZIMMERMAN

*Scientific Laboratory, Ford Motor Company, Dearborn, Michigan*

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This paper reports the results of an experimental and phenomenological investigation of the static and dynamic behavior of weakly connected superconducting rings. The configuration is essentially a macroscopic superconducting ring of inductance  $L$  incorporating a point contact as a weak link which determines the critical supercurrent in the ring,  $i_c$ . A phenomenological model for the stationary quantum states of the system as a function of an applied field is developed. The dynamic behavior is obtained directly from the time dependence of the applied fields. Experiments demonstrating both the stationary and the time-dependent magnetic behavior are described. The stationary behavior was obtained with a magnetometer incorporating a weakly connected ring as a sensor to measure the flux through the ring under test. The experimental results confirm the phenomenological model if the critical current  $i_c$  is greater than  $\Phi_0/2L(1+\gamma)$ , where  $\Phi_0 = h/2e$  is the flux quantum and  $\gamma$  is a material and geometric parameter which is usually small compared to unity. In the regime  $Li_c > \Phi_0/2(1+\gamma)$ , the quantum states are discrete, and the transitions between states are well defined and irreversible. If the critical current is not too large, the transitions generally occur only between adjacent states; that is,  $\Delta k = \pm 1$ . At large critical current, multiple quantum jumps are observed. On the other hand, if  $Li_c < \Phi_0/2(1+\gamma)$ , the quantum states merge into one another continuously and reversibly. In this case the magnetic behavior in the neighborhood of the half-quantum points is related to the depairing or gapless regime in superconductivity. Measurements of the ac properties of the weakly connected ring at 30 MHz are interpreted directly in terms of the static properties under the influence of a time-varying applied field. In fact, no qualitative corrections to the theory are expected up to frequencies of the order of the superconducting energy gap.

### I. INTRODUCTION

**T**HIS paper reports the results of an experimental and phenomenological study of weakly connected superconducting rings. The system under study is a macroscopic superconducting ring incorporating one weak link which closes the circuit. We present a phenomenological model which describes the electromagnetic properties of this ring as a function of an applied external field. The model is first presented as a linear theory and later we show the nonlinearities which must be introduced to be in agreement with the data. We present data showing the electromagnetic behavior of the ring as a function of applied field including a measurement of the magnetic flux or magnetic moment

in the ring and direct observations of the transitions which the system will undergo.

In his phenomenological theory of superconductivity, London<sup>1</sup> first proposed that the fluxoid was a constant of the motion for a superconductor. He also proposed that the constants which the fluxoid would assume might be quantized in units of  $h/e$ . This ultimately led to the discovery in 1961 of the flux quantum by Doll and Näbauer<sup>2</sup> and by Deaver and Fairbank,<sup>3</sup> with the one modification that the flux quantum was  $h/2e$

<sup>1</sup> F. London, *Superfluids* (John Wiley & Sons, Inc., New York, 1950), Vol. 1.

<sup>2</sup> R. Doll and M. Näbauer, *Phys. Rev. Letters* **7**, 51 (1961).

<sup>3</sup> B. S. Deaver and W. M. Fairbank, *Phys. Rev. Letters* **7**, 43 (1961).