points on the curve represent nominal indium concentrations of 0, 1%, 2%, and 3% respectively when read from left to right.

The data of Fig. 3 show that there is a monotonic increase in the exponent of the temperature dependent term for the critical current, but the spread in the exponent for a given residual resistance ratio prevents us from obtaining the exact dependence.

DISCUSSION

The deviation of the indium–tin alloys from the temperature dependence given by Eq. (1) may be due to a different temperature dependence of H_{cb} or λ_L than given by Eq. (2) and Eq. (3).

The temperature dependence of λ of a 3% indium–tin alloy has been discussed by Pippard.[9] Although he could not measure the temperature dependence of λ due to flux penetration he did measure $\delta_i - \lambda$ for two temperatures and found the results in agreement with Eq. 3, where λ and δ_i are the field penetration depth in the superconducting and the normal state, respectively.

Deviations of H_{cb} from the parabolic law are known to exist for pure metals.[10,11] It will be necessary to measure the temperature dependence of H_{cb} of a carefully prepared single crystal of a 3% indium–tin alloy to determine the significance of such deviations.

ACKNOWLEDGMENTS

The author is greatly indebted to R. E. Skoda for the preparation of the tantalum and indium–tin alloy films and for many helpful discussions.

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[10] J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. **108**, 1175 (1957).

[11] E. Maxwell and O. S. Lutes, Phys. Rev. **95**, 333 (1954); M. A. Biondi, A. T. Forrester, M. P. Garfunkel, and C. B. Satterthwaite, Rev. Mod. Phys. **30**, 1109 (1958).

Currents in Thin Superconducting Films: Nonlinear Effects*

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I. INTRODUCTION

The problem of small superconductors—having at least one dimension smaller than ξ_0, the coherence distance of the superconducting wave functions—has been the recent focus of considerable interest, particularly in view of the successes of the Ginzburg–Landau–Gor'kov (GLG) theory in dealing with this situation.

For the case of currents in thin films, of thickness $d \ll \xi_0$, it has been pointed out[1,2] that a rather simple derivation based on the GLG theory leads to an expression for the mass flow density of the form

$$J = \rho_s v[1 - v^2/v_m^2], \qquad (1)$$

where v is the center-of-mass velocity of the electron pairs, ρ_s is an effective equilibrium number of superconducting electrons, and v_m is a combination of parameters of the bulk superconductor having the dimensions of a velocity.[3] The term in brackets represents the extent to which it becomes favorable for the effective number of electrons in the superconducting state to decrease, as a function of the drift velocity v, in order to minimize the free energy.

If a sinusoidal current is induced in a closed superconducting loop, in which the current density can readjust itself freely, it might be expected on the basis of Eq. (1) that harmonics of the fundamental frequency be generated in the current. As has been emphasized by Fulde and Ferrell,[4] who have treated the problem also and in more detail, the analysis is complicated, upon introduction of the magnetic field required to induce the supercurrent, by the large opposing field resulting from the latter.

The recent experiment of Mercereau and Crane[5] immediately suggested itself as an especially useful technique for investigating such a phenomenon; not

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[1] J. Bardeen, Rev. Mod. Phys. **34**, 667 (1962).

[2] C. D. Mitescu, Bull. Am. Phys. Soc. **7**, 609 (1962).

[3] Both v_m and ρ_s are functions of temperature.

[4] P. Fulde and R. Ferrell, University of Maryland Technical Report 299 (unpublished); also P. Fulde, Bull. Am. Phys. Soc. **8**, 307 (1963).

[5] J. E. Mercereau and L. T. Crane, Phys. Rev. Letters **9**, 381 (1962).

only does it give a sharp indication of the critical current limit, but its very close agreement with earlier, stationary, critical persistent current measurements[6] provides assurance that the sample is, indeed, in the superconducting state.

II. EXPERIMENTAL METHOD

The sample, a short cylindrical tin film, is placed in an axial, sinusoidal magnetic field. A 2000-turn pickup coil, inserted within the ring and connected in opposition to a similar, dummy coil at some distance away—the fine balance being provided by an external network—then gives an output signal proportional to the time rate of change of the current in the sample; a tuned, phase-sensitive detector then analyzes the harmonics in the output, after the latter has been amplified by a low-noise differential amplifier. Because of the rather low level of the harmonics observed, it has been found essential to subtract out most of the fundamental frequency component in the output signal by means of a signal proportional to the driving current, fed into the other side of the differential amplifier.

The samples used have been short, tin films, evaporated at liquid nitrogen temperatures, on 1-cm-diam, fused quartz substrates.

As the experiments have been performed within a few hundred millidegrees of the critical temperature of the ring, the earth's field was nulled out in the experimental volume.

The reaching of the critical limit in the ring is indicated by the very abrupt appearance of a spike in the output waveform corresponding to a sudden limiting of the circulating current. As it will be seen below, it is equally observable in the behavior of the harmonics.

III. OBSERVATIONS

1. General Characteristics

While the main supercurrent is predominantly linear, the presence of odd harmonics, specifically the third and fifth, has been clearly observed below the critical current level. As the external driving field is increased through the critical limit, both harmonics show an extremely sharp decrease followed by a very steep rise, this coinciding with the visual appearance of the spike in the output waveform. On analyzing the signal phase-sensitively, two orthogonal components of each harmonic current are observed. In the case of the third harmonic, for example, a component I_3', in phase with the fundamental current I_1,[7] merely shows the much steeper rise. At the critical field, the other component I_3'' in quadrature with I_3' actually drops through zero and reverses its sign before rising steeply, indicating a change in the character of the nonlinearity at the saturation.

The critical currents do, in fact, obey fairly closely the $(T_c - T_{cI})^{\frac{3}{2}}$ law—where T_c is the ultimate critical temperature of the film and T_{cI}, the transition temperature for current I_1—predicted by both the BCS and GLG theories.

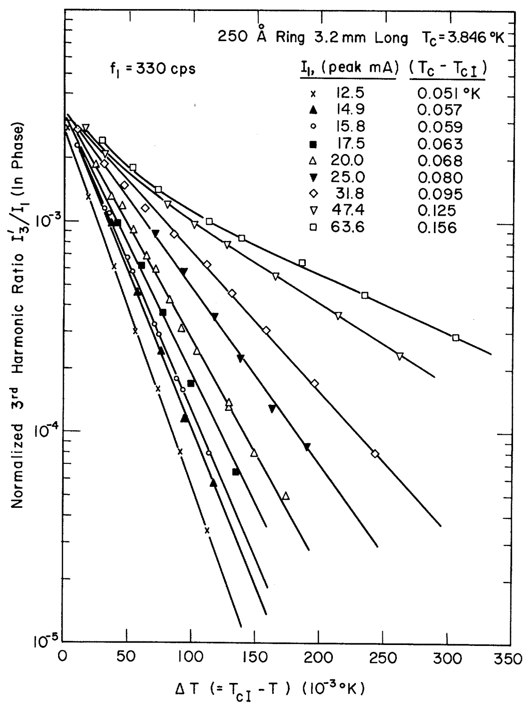

FIG. 1. Temperature dependence of the normalized, in-phase, third harmonic component I_3'/I_1 for various currents. Note the monotonic decrease in logarithmic slope α_I' with current, as well as the convergence to the same relative level of harmonic at the critical limit.

The transition through the critical limit into the "saturation" region appears to be of a thermodynamically reversible nature since the identical curve is reproduced both for increasing and decreasing drive currents. As a matter of fact, one can hold any point on the steeply rising transition curve and observe clearly an occasional oscillating mode of the bath-temperature control, less than a millidegree in amplitude. More remarkably perhaps, the curve of harmonic against drive current at constant tempera-

[6] J. E. Mercereau and T. K. Hunt, Phys. Rev. Letters 8, 243 (1962).

[7] That is, of the nature of sin $3\omega t$ for a fundamental current sin ωt.

ture shows no hysteresis whether it is obtained by working down from above the critical region, or by cycling above T_c, and then obtaining the curve for gradually increasing amplitudes (to guard against the effect of any trapped field).

2. Quantitative Functional Dependence below the Critical Region

Rather surprisingly, contrary to what might be inferred from Eq. (1), the curves of amplitude against drive current at constant temperature *did not show a cubic behavior* even at low amplitude, down to the limits of sensitivity of the experiment.

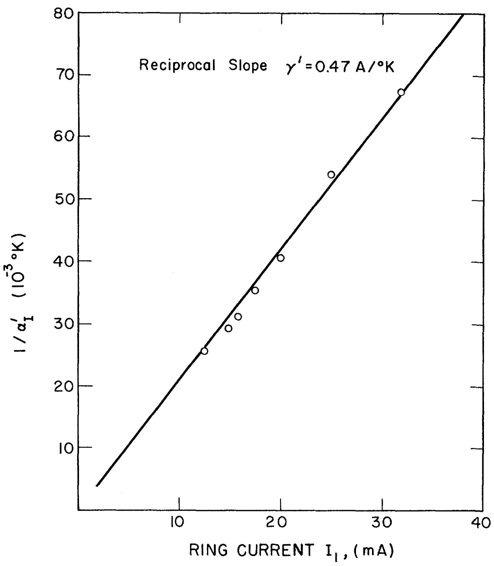

FIG. 2. Variation of the reciprocal of the logarithmic slope α_I' with current. A relationship $\alpha_I' = \gamma'/I_1$ is observed.

A simple dependence was found, however, for the third harmonic amplitude as a function of temperature at constant drive. Figure 1 shows a semilogarithmic plot of the normalized, in-phase component I_3'/I_1 against $\Delta T(=T_{cI} - T)$. This choice of abscissa serves to emphasize the rather interesting characteristic that all curves extrapolate to the *same relative level of harmonic at their respective transition.* They form a family of curves of the form $A'e^{-\alpha_I'\Delta T}$, the out-of-phase component I_3'' also giving a similar set $A''e^{-\alpha_I''\Delta T}$—where α_I' and α_I'' are the logarithmic slopes.[8] A'' is quite comparable to A' in magnitude. A deviation from straight-line behavior is observed in Fig. 1, for higher currents, roughly in the same range where the critical currents for this film might be thought to depart from the $(T_c - T_{cI})^{\frac{3}{2}}$ law.

[8] It should be pointed out that for the experimental range of temperatures, the dependence could equally well be exponential in $-\Delta(1/T)$ as in ΔT.

Figure 2 shows the dependence of the reciprocal of α_I' on the fundamental current (or the external driving field). It is observed that α_I' is inversely proportional to the linear current, giving a final dependence of the type $e^{-\gamma\Delta T/I_1}$. Again, the out-of-phase component behaves quite similarly, but as its coefficient for the particular film $\gamma'' = 0.68$ Å/°K is appreciably larger than $\gamma'(=0.48$Å/°K), it drops off much more rapidly.

IV. CONCLUSIONS

A number of interesting inferences can be drawn. First of all, the reversibility of the curve on passing through the critical region seems to indicate a second-order phase transition probably of the nature of a Type II superconductor.[9]

The absence of even harmonics merely verifies the expected symmetry of the conduction mechanism on current reversal.

On the other hand, the presence of an out-of-phase component in the third harmonic is of particular interest, since it suggests an asymmetry in the acceleration–deceleration cycle of the supercurrent. Such asymmetry might result from a local depairing, and dissipative scattering of superelectrons as they are accelerated in a nonequilibrium manner to a high velocity. This would be more likely to occur near the peak of a velocity cycle and might thus possibly explain the $e^{-\gamma\Delta T/I_1}$ dependence of the harmonics as a "velocity-induced" disorder. It would also be in keeping with the interpretation that when the depairing has reached a certain limit—indicated by a certain relative level of harmonics—a "catastrophic," current-limiting depairing sets in.

Such dissipation would not be incompatible with the existence of stationary, persistent currents, as it would occur only while electrons are being accelerated. If the harmonics are indeed due to such a cycling dissipation, they should show an increase with frequency but no change in slope.[10] While our presently available data is insufficient to be conclusive, there are indications that this might be the case.

[9] H. London (see Proceedings of this Conference) suggests the same conclusion for the case of measurements on niobium films, near their transition temperature.

[10] Such increase should be linear in frequency unless the depairing has an associated relaxation time comparable to the period of the applied field; in that case, at corresponding and lower frequencies, a change from linearity would be expected. The observation of such a time of relaxation to a thermodynamically stable state would be of appreciable interest as it could account for the negative result of one of the experiments on flux quantization [J. E. Mercereau and L. L. Vant-Hull, Bull. Am. Phys. Soc. 6, 121 (1961)].

Discussion 48

MEISSNER: (Discussing first two papers). We have looked at the complete transition in pulsed currents for a small strip over a ground plane and it turned out that, even at currents about 4 or 5 times the current which restores half the normal resistance, we find that the thin film still is partially superconducting. Now a remark to the last paper (Mitescu's). I think one has to be very careful. You will always get some field lines breaking through the film. I do not believe that you really can have your field lines truly all parallel to the film but that some will break through and you will have the effects which Tinkham observed and described as flux inclusions. It's not quite clear that these are necessarily always resistanceless if you get this breakthrough in an alternating field.

C. D. MITESCU, *California Institute of Technology:* The fact that the persistent currents which have been observed in stationary d.c. conditions are comparable to our critical currents makes the presence of ordinary, normal resistance rather unlikely. I don't think that flux leakage could account for the observed very sharp exponential dependence on the current.

MEIKLEJOHN: I might make a remark on one of Meissner's remarks. It depends on just how sharp your transition is as to whether or not this resistance at which you make your measurement is important. We did not find any difference in our temperature dependence if we went down to a tenth of this voltage sensitivity which would be a different resistance level.

BEAN: I'd like to ask Dr. Mitescu if he has checked for the absence of static hysteresis in the magnetic properties of his ring when he has less than complete penetration. The reason I ask this is, if I understand the theory, there is no hysteresis at low frequencies because the Ginzburg–Landau theory does not have strong inertial terms. If one does have hysteresis due to the irreversible or partially irreversible motion of flux lines as Professor Meissner mentioned, then one would generate harmonics in the sense that I showed in my talk on the first day.

MITESCU: There is no hysteresis in going above the critical transition or coming down through it. One point which might argue against the flux line motion is that, if I remember rightly, in Dr. Bean's paper the dependence in his model was quadratic in the amplitude. I think one would expect some kind of power law in the case of flux line motion, unless it is a very strange kind of motion.

DOUGLASS: I have a question to Dr. Meiklejohn. In regard to the exponents on Δt, I noticed some of your points were very close to the transition temperature, a millidegree or thereabouts. These points strongly influence the slope of these curves and any small error in the transition temperature could appreciably change the value of the exponent. I'd like to know what the width of the transitions were.

MEIKLEJOHN: It is true that the actual magnitude of the critical temperature can influence these slopes. However, we were as high as 50 mdeg in our measurements and the points fartherest away from the critical temperature were not greatly influenced by changing slightly the critical temperature. The widths of the transition varied from about 4 mdeg to about 16 mdeg in the various films that were measured.

Temperature Dependence of the Critical Fields of Thin Superconducting Films

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I. INTRODUCTION

To interpret and understand critical magnetic field measurements on superconducting films, a theory is needed which includes both strong-field effects and nonlocal effects–strong field, to describe phenomena occurring at the critical field; nonlocal, to adequately describe thickness and mean free path effects.

In a previous paper[1] (hereafter referred to as I), a simple strong-field, nonlocal theoretical model was presented which relates the critical magnetic fields of superconducting films to the kernel of the current–vector–potential relationship for any theory of superconductivity. In I, the theoretical model was worked out in detail for the Pippard nonlocal kernel with specular boundary conditions in the superconductive state. Comparison of the theoretical model to critical field data for pure indium films indicated that the theoretical model predicted quite well the thickness dependence of critical field at temperatures near the critical temperature. In a second paper[2] (hereafter referred to as II), the theoretical model was compared to critical field measurements made on dilute alloy films of indium containing up to 5 at.% tin. In II, it was shown that the theory using the Pippard kernel predicted quite well the variation of critical field with mean free path at temperatures near T_c.

In this paper, we review the theoretical model and

[1] A. M. Toxen, Phys. Rev. **127**, 382 (1962).

[2] A. M. Toxen and M. J. Burns, Phys. Rev. **130**, 1808 (1963).