Flux Trapping of rf Fields in Superconductors*

C. R. HADEN† AND W. H. HARTWIG
Department of Electrical Engineering, The University of Texas, Austin, Texas
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Measurements on superconductive resonant circuits in the frequency range of 10–1000 Mc/sec indicate that flux trapping of the rf magnetic fields is responsible for a residual loss, not predicted by surface-resistance theories. A simple model, based on flux trapping at local sites, predicts such a loss for both type I and type II materials. The surface resistance due to flux trapping is shown to be proportional to frequency, the density of trapping centers, the penetration depth, and a "threshold" function of the rf magnetic field. Experimental data are given for several materials, verifying the salient features of the model. It is also concluded that the flux-trapping centers are of a common nature.

I. INTRODUCTION

PAST studies of the surface resistance of superconductors indicate the existence of a residual loss well below T_c.[1–3] This loss is not predicted by either the quantum or phenomenological theories for surface resistance.[4–8] A study of this loss has been made through use of high-Q resonant circuits at frequencies near 100 Mc/sec. Comparison of the authors' experimental data and a simple model for flux trapping at surface imperfections[9] indicates that flux trapping is responsible for the observed losses.

Several investigators have observed flux-trapping losses at very low frequencies in type II superconductors.[10–12] These losses are, to some extent, explained by the Bean-London theory,[13,14] but it does not account for losses observed for maximum fields less than the bulk critical value.[9,10] Moreover, the Bean-London theory assumes the features characteristic of type II superconductors, and hence is not applicable to type I superconductors. Hysteresis effects have long been observed in such materials, however, including lead and tin.[15–18] Trapping can apparently occur near dislocations or other imperfections in the surface, even in well-annealed samples. Buchhold has given a qualitative picture which applies to this type of trapping.[19] Although the flux-trapping loss is small in type I materials, particularly if they are well-annealed, it becomes quite noticeable in high-Q resonant circiuts, where the Q is limited by the loss. Such losses in lead cavities at 3 kMc/sec have been observed by Fairbank *et al.*,[1] and were attributed to flux trapping. It is the purpose of this research to clarify the features of flux trapping at low fields in type-I as well as type-II superconductors and to demonstrate that flux trapping can account for the rf losses observed.

II. MATHEMATICAL REPRESENTATION

Flux trapping in type-I materials and in type-II materials must be described on a local basis. Such a description is difficult because of the many different types of local conditions that are possible. Buchold[19] has shown qualitatively how surface roughness, voids, etc. can cause local field distortion or provide circulating currents which exceed the local critical value. One of these forms, the void, offers a simple picture of local flux trapping and can be described in simple mathematical terms.

Consider a void which exists in the surface of a superconductor. Any time change in the flux linking the void creates a circulating current around it. Type II conditions are assumed in the immediate vicinity of the void due to local stresses.[20] This allows application of the critical-current-field independence of the Bean-London theory[13,14] on a local basis. Using a current cycle that increases linearly through the local critical magnetic field value H_{cL} and reaches a maximum of H_m in each direction, a hysteresis loop is swept out. The

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† Presently at the University of Oklahoma, Norman, Oklahoma.

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power loss at the local trapping center is the energy loss per cycle times the frequency f.

$$P_L=4fKH_{cL}(H_m-H_{cL}), \quad (1)$$

where K is a constant.

The total power loss, per unit surface area, is obtained through multiplying the local power loss by the density of trapping centers N and integrating into the material for a distance over which the local critical field is exceeded.

$$P_T=4fKH_{cL}\int_0^{x1} N(x)(H_{m0}e^{-x/\lambda}-H_{cL})dx, \quad (2)$$

where H_{m0} is the maximum surface field, λ is the field penetration depth, x is the distance into the material, and

$$H_{m0}e^{-x_1/d}=H_{cL}. \quad (3)$$

This assumes, as a first approximation, that H_{cL} is constant throughout the material. If it is assumed that $N(x)$ is independent of x, then the total power loss is given by Eq. (4):

$$P_T=4KfN\lambda H_{cL}[H_{m0}-H_{cL}(1+\ln H_{m0}/H_{cL})]. \quad (4)$$

The equivalent surface resistance is

$$R_S=8KfN\lambda(H_{cL}/H_{m0}^2)\times[H_{m0}-H_{cL}(1+\ln H_{m0}/H_{cL})]. \quad (5)$$

The dependences of f and N in Eq. (5) are not related to the shape of the hysteresis curve. They would be obtained with any local model. The λ dependence would also result from any power relation between local energy loss and H_{m0}. These three dependences are not necessarily intrinsic functions of the local model chosen. The field dependence, on the other hand, is sensitive to the shape of the hysteresis curve. This shape, in turn, is a function of the local conditions assumed. In the simple model suggested, analysis of Eq. (4) reveals an approximately H_m^2 loss for the region $2.5H_{cL}<H_m<8H_{cL}$. This leads to a linear equivalent resistance. Outside this range the slope is lower and for high fields the loss mechanism might not have been observable in the experiments conducted. The assumed uniform distribution of trapping centers is not the only one possible, and almost any field dependence could be obtained with assumptions which manipulate the shape of the hysteresis loop. Only the f, N, and λ dependence are invariant with the model for the nature of the flux-trapping center. A more general form for the equivalent surface resistance is Eq. (6).

$$R_s=K'fN\lambda(t)F(H_{m0},H_{cL})=K'fN\lambda_0[1-t^4]^{-1/2}F(H_{m0},H_{cL}) \quad (6)$$

$$R_s=R_0(f,N,\lambda_0,H_{m0},H_{cL})[1-t^4]^{-1/2},$$

where F is some function of the magnetic field, K' is a constant, t is the reduced temperature, λ_0 is the penetration depth at 0°K, and the empirical form[21] for $\lambda(t)$ is assumed.

This model is limited in its concept and probably would not account for the large scale trapping at high fields in type-II materials. However, since the mechanism is placed on a local basis, it can account for the rf losses observed at low fields in both type-I and type-II materials. The small temperature dependence predicted here can only be observed if flux trapping is the dominant loss rather than the superconducting surface resistance. This is not in conflict with the existing theories for surface resistance. The existence of this temperature function is supported indirectly by the dc measurements of Budnick *et al.*[22] which indicate this same dependence for the percentage of flux trapped in their specimens.

III. MEASUREMENT TECHNIQUE

The experimental method consists of a pulse measurement of the quality factor Q of a resonant circuit constructed from the superconducting material under study. The circuit is constructed from metallic foils wound on Teflon tubes such that a minimum of Teflon is in the electric field. The dielectric loss tangent for Teflon at 4.2°K has been determined by Hartwig and Grissom as $1.3(10^{-6})$,[23] so that dielectric losses are negligible. The resonant circuit is enclosed in a lead-plated radiation shield can. Residual losses in the superconducting lead have been shown to be at least an order of magnitude below those in any of the materials measured. It is thus possible to make the following approximation with a high degree of accuracy.

$$Q_n/Q_s\approx R_s/R_n=r. \quad (7)$$

The formation of this ratio eliminates the necessity of an absolute measurement and provides for the cancellation of geometric factors inherent in the circuit Q.

The Q is determined through an rf-pulse measurement. Pulses of rf energy, with envelope rise and fall times of about 0.1 μsec, are fed into the resonant circuit. The circuit energy decays with a time constant, $Q/\pi f$, which is of the order of 10 msec. The output waveform is amplified, detected, and photographed, so that the Q may be determined graphically. An Empire Devices Model NF105 radio receiver is used for amplification and detection, and a Hewlett-Packard 175A oscilloscope is used for recording.

IV. PREPARATION OF CIRCUITS

The bulk of the experimental data were taken on alloys from the In-Sn system. The indium content was

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limited to 5 at.%, in order to maintain a single-phase composition.[24] The tin was supplied by American Smelting and Refining Company and was 99.99% pure. The indium used was 99.999% pure and was obtained from the Texas Instruments Company. The materials were weighed on a chemical balance and melted in Pyrex. After slow "in-tube" cooling, the ingots were rolled into foils at room temperature and then cut into circuit form. They were cold-worked in this manner to assure heavy lattice distortion.[25] The foils were etched lightly in a 50% sulfuric acid solution shortly before each test run and between annealings.

V. VERIFICATION OF FREQUENCY DEPENDENCE

The foil-Teflon configuration has multiple resonances for a given foil length, so that a relatively wide range of frequencies was measured. Other sets of frequencies were obtained by shortening the foil. The normal surface resistance of In-Sn alloys is known to be classical in the frequency range used, hence the resistance ratio R_s/R_n should be proportional to $f^{1/2}$. The frequency dependence has been verified experimentally as is demonstrated by Fig. 1. This indicates that the residual loss is truly of a hysteresis nature.

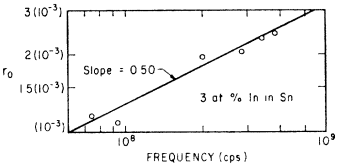

FIG. 1. Residual-resistance ratio plotted versus frequency. The theoretical slope is arbitrarily drawn through the experimental points, showing substantial agreement.

VI. VERIFICATION OF THE DEPENDENCE UPON TRAPPING-CENTER DENSITY

Measurement of the effect of trapping-center density presented some difficulty, since the exact nature of the trapping-center is undetermined. Two general classes of center were deemed possible, one being connected with impurities and the other with deformity of the material. The first of these was investigated by running tests on a set of samples, each having a different In content. As demonstrated in Fig. 2, little dependence upon In content was observed.

The effects of deformity were determined through a series of annealing experiments. The unannealed sample was subjected to a temperature run so that r could be obtained. The circiut was then annealed for successive time periods at 200°C in an argon atmosphere. After each annealing, r was again determined. The results of such an experiment for a 4 at.% In-Sn sample are shown

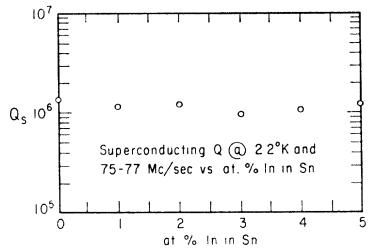

FIG. 2. Superconducting Q for several circuits plotted against atomic percentage of indium content. Little variation is noted.

in Fig. 3. The resistance ratio decreases exponentially with annealing time, indicating verification for the linear dependence upon trapping-center density. A decrease in deformity during annealing is a rate process and would exhibit the classical exponential behavior, $N=N_0\exp(-t/\eta)$, where η is a characteristic time.[26] The characteristic time for the annealing process appears to be about 96 h at this annealing temperature. A single "bad" point is attributable to failure to clean the surface properly before this particular test.

VII. VERIFICATION OF THE TEMPERATURE DEPENDENCE

The temperature dependence predicted on the hysteresis model had been verified by the authors for several materials.[27] The resonant frequencies of the various circuits were kept in the range of 75–77 Mc/sec to avoid frequency-dependent effects. In order to increase resolution in analysis of the temperature data, the extrapolated value at 0°K is subtracted from the experimental data, and the difference is normalized, as indicated in Eq. (8).

$$[r-r(0)]/r(0)=(1-t^4)^{-1/2}-1=\psi(t). \qquad (8)$$

A value of $r(0)$ is set by fitting the data to Eq. (8) at the lowest temperature points where the sensitivity is greatest. At intermediate and higher temperatures

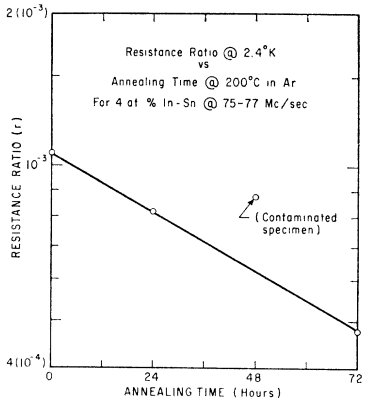

FIG. 3. Plot of resistance ratio at a fixed temperature versus annealing time exhibits an exponential dependence having a characteristic time of 96 h at 200°C.

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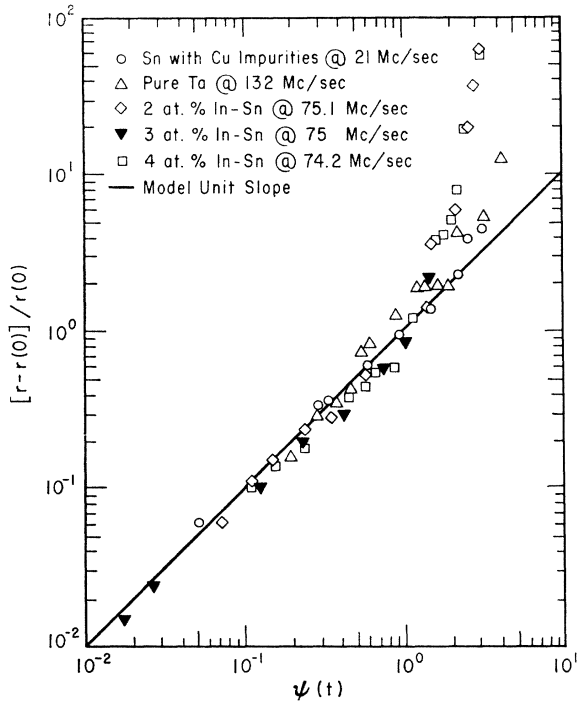

FIG. 4. Agreement of experimental data, for a number of materials, with the model temperature dependence.

any discrepancy in $r(0)$ is not observable. Attention is called to the choice of scales on all curves which reflects the resolution present in the data.

Good agreement between experiment and the predicted dependence is demonstrated for various materials at reduced temperatures down to 0.5 in Fig. 4. Lower temperatures were not required because of the flux-trapping dominance over much of this range. The data diverge from the predicted straight line for large ψ because of the dominance of the ordinary surface resistance near $t=1$. They appear to fit the surface resistance equation derived from the London[6,28] theory as shown in Fig. 6. The resistance ratio is given by Eq. (9).

$$r=[\alpha\phi/(1+\alpha^2\phi^2)\{(1+\alpha^2\phi^2)^{1/2}-1\}]^{1/2}, \quad (9)$$

where $\alpha=2\pi f\tau$, $\phi=t^4/(1-t^4)$, and τ is the relaxation time of normal electrons. The "experimental" points are obtained by subtracting out the flux-trapping model, using a projected value of the hysteresis loss at 0°K. An attempt has also been made to fit the data to the BCS theory,[29,30] which is reflected in simplified form by the Pippard[31] empirical formula, Eq. (10).

$$r=r(f)t^4(1-t^2)/(1-t^4)^2+r(0), \quad (10)$$

where $r(f)$ is a function of frequency only and $r(0)$ is a residual loss independent of temperature.

The region dominated by the residual loss and not the loss due to ordinary surface resistance is important to this discussion. In the frequency range of these experiments the residual loss dominates the surface resistance

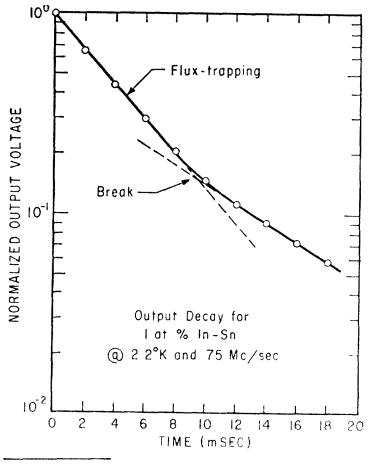

FIG. 5. Plot of decay of normalized output voltage with time, substantiating the existence of a local critical field.

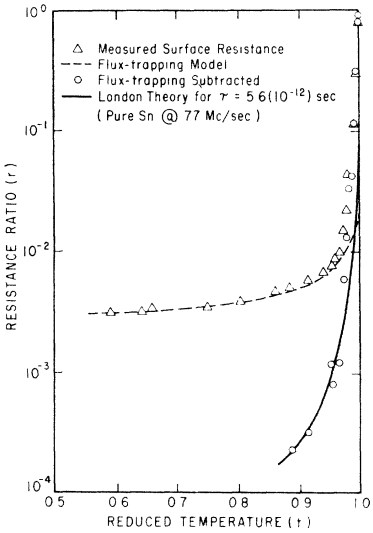

FIG. 6. Plot demonstrating agreement of experimental data with London theory after flux-trapping effect is subtracted.

predicted by either London or Pippard for $t<0.9$. Below $t=0.9$ the flux-trapping model is nearly independent of temperature, but the increased sensitivity of these experiments supports the relation between the residual loss and the penetration depth. Since the data fits the other features of the flux-trapping model as well as the temperature dependence, the use of the temperature function appears well justified.

VIII. FIELD DEPENDENCE OF THE LOSS

The field dependence predicted from the simple hysteresis model is very strongly a function of the model assumptions. In turn, the correct form for the assumptions is somewhat arbitrary. These facts are reflected

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in the partial disagreement of experimental results and the model predictions. Inspection of Fig. 5 reveals that the circuit Q, and therefore the equivalent resistance, are independent of the magnitude of the decaying output over the upper portion of the decay range. Exact determination of the field intensity was not possible. It is obvious, however, that because the Q exceeded 10^6, the bulk critical field was not exceeded. As the output level continues to decrease a sudden break, or change in the time constant, is observed. This is taken as corresponding to the "effective local critical field" predicated in the model, and provides it with further substantiation. The abruptness of the change to lower loss indicates that most of the trapping centers are very much alike, a welcome feature but not expected on the basis of the model. The nature of the loss mechanism below the local critical field is as yet undetermined.

IX. CONCLUSION

The existence of rf flux trapping is confirmed through verification of the predicted model dependences. More study is required before the nature of the flux-trapping center is understood and the magnitude of residual resistance can be predicted. The direction of the investigation seems clear from these efforts. A promising mechanism for flux trapping is associated with background magnetic fields trapped in the superconductor during the cooling process. This will shortly be reported on by J. M. Victor and the authors. The existence of an effective local critical field, as demonstrated by experiment, the abruptness of the Q change as the circuit energy decays, coupled with the annealing and impurity experiments, indicate that the trapping is dominated by sites with a common nature.

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Energy Levels of Trivalent Gadolinium and Ionic Contributions to the Ground-State Splitting*

B. G. WYBOURNE†
Argonne National Laboratory, Argonne, Illinois
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This paper represents an attempt to estimate the ionic contributions to the ground-state splitting of the trivalent gadolinium ion in a lanthanum ethyl sulfate lattice. The energy levels of the $4f^7$ configuration are calculated and compared with those observed experimentally. The calculated ground-state g factor is found to deviate significantly from its measured value. An attempt has been made to estimate the contributions to the ground-state splitting for eight different mechanisms: (1) a fourth-order mechanism that is *linear* in the crystal-field strength and *cubic* in the spin-orbit interaction; (2) fourth-order mechanisms that are *quadratic* in both the crystal-field strength and the spin-orbit interaction; (3) a third-order spin-spin mechanism acting within the $4f^7$ configuration; (4) Pryce's second-order spin-spin configuration mixing mechanism; (5) a second-order relativistic mechanism; (6) mechanisms involving configuration mixing by the crystal field; (7) mechanisms involving nonlinear electrostatically correlated crystal-field interactions; (8) fifth-order configuration interaction mechanisms. The total contribution due to these mechanisms is found to be *twice* the magnitude of the observed splitting and of *opposite* sign. It is suggested that no purely ionic model can account for the observed splitting and that the correct explanation must involve the detailed interaction of the gadolinium ion with its ligands. A semiqualitative explanation of some of the intensity features of the crystal and solution spectra of trivalent gadolinium is attempted.

I. INTRODUCTION

THE lack of an adequate interpretation of the ground-state splittings of ions having a half-filled shell of electrons has constituted a serious problem in crystal field theory. While considerable progress[1–10] has been made in the interpretation of the splittings for the transition ions, comparatively little progress

* Based on work performed under the auspices of the U. S. Atomic Energy Commission.
† Present address: Physics Department, University of Canterbury, Christchurch, New Zealand.

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