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ac Susceptibility Transition in Type-II Superconductors and Surface Critical Currents*

B. Bertman and Myron Strongin
Brookhaven National Laboratory, Upton, New York

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The ac susceptibility transition below H_{c3} has been investigated as a function of ac field amplitude, in cylindrical samples of Pb-Bi alloys ($1\leqslant\kappa\leqslant5$). Converting the ac fields to critical surface currents J_c permits comparison with the values calculated by Abrikosov and Park. The present results agree with those of Swartz and Hart, and of Rollins and Silcox. Although above H_{c2} the qualitative behavior is similar to the theoretical calculations, the magnitudes are found to be lower. At H_{c2} a sharp break is found in J_c, as reported previously. $J_c\kappa/H_c$ is found to be independent of κ within experimental error. Surface roughness and slight sample misalignment could have lowered the observed J_c.

INTRODUCTION

In this paper the transition of the ac susceptibility at some steady dc field H_0 is investigated as a function of the ac field amplitude. In previous work[1] investigations of the surface transition at H_{c3} were made by using peak-to-peak fields of 0.04 Oe. In general the measurements in steady dc field indicated complete shielding until the transition at H_{c3}. In the present work extensions of these measurements are made to much higher ac fields and the results are related to the theoretical work of Park,[2] Abrikosov,[3] and Fink and Barnes[4] who predict the surface critical current as a function of dc field. The ac fields were converted to equivalent surface currents and then compared to the above calculations.

EXPERIMENTAL

In these measurements a cylindrical sample $\frac{1}{2}$ in. long and $\frac{1}{16}$ in. in diameter was simultaneously placed in a dc field obtained with a superconducting magnet and in a mutual inductance system. The primary of this mutual inductance supplied ac fields from 0.04 to 90 Oe peak-to-peak. Penetration of the ac field was indicated in these measurements by a change in mutual inductance which reflected changes in χ' and χ'', the real and imaginary parts of the susceptibility. Figure 1 shows a typical transition. Two mutual inductance bridges were used in this null technique. For low ac amplitudes an electronic bridge was used similar to the one described by Pillinger, Jastram, and Daunt.[5] In the higher ac field measurements, a L&N mutual inductometer was used in a Hartshorn bridge circuit.

The samples used were three Pb-Bi alloys of 2, 5, and 13% Bi. The magnetization curves were measured by integrating the output voltages of the secondary coils obtained when the sample was pulled out of the coils in a steady field. The G-L parameter κ of these alloys ranged from 1 to 5.

MEASUREMENTS

In these experiments values of critical ac surface currents as a function of dc field were obtained from measurements of χ' and χ'', the real and imaginary parts of the ac susceptibility. When the ac fields were completely shielded at low fields, $\chi'=-1/4\pi$. Penetration of the ac field was indicated by deviations from $-1/4\pi$. The dc field at the peak in χ'' was arbitrarily taken as an indication of the field at which the transition occurs. In a system where a superconducting sheath goes normal with field, it is found that the peak in χ'' occurs when χ' is halfway between zero penetration (superconducting sample neglecting penetration depth effects) and full penetration (normal metal neglecting eddy current shielding).[6] In these measurements the dc field is varied to obtain the transitions in χ' for different fixed ac fields. A run is made for each fixed ac field. Since the ac surface current is proportional to the applied ac field when the sample is superconducting, the ac field at which the transition occurs is essentially a measure of the ac critical surface current

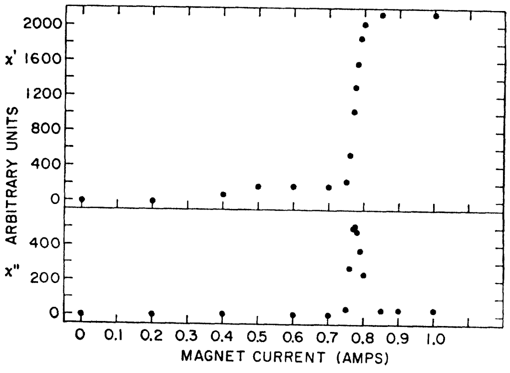

FIG. 1. A typical ac susceptibility transition from superconducting to normal as a function of dc field.

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at some dc field. The first penetration of the ac field is interpreted in terms of the critical ac surface current being obtained and hence the absence of complete shielding. This point will now be discussed further. In previous publications[7] it was indicated that the ac field can be considered to be on a hysteresis loop, as in Fig. 2. In Fig. 2, two ac hysteresis loops above H_{c2} are indicated. The lower field, case *A*, illustrates the hysteresis path for complete diamagnetism. When the ac field amplitude reaches 2 to 4 of the higher field path, the critical current is reached on the backward part of the magnetization curve, and hence further increases in ac field will cause penetration since no further shielding can be obtained after the critical surface current is reached. Hence, the transition starts when the ac amplitude causes the hysteresis path to exceed the ΔH obtained from 2 to 4, on the higher field path. The ac surface currents are obtained from the ac field through the relation $H_{ac}=0.4\pi J_c$, where J_c is in A/cm.

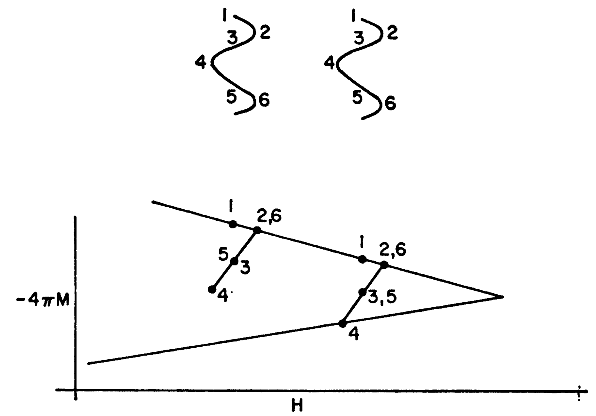

FIG. 2. Illustration of magnetization curve near H_{c3} showing diamagnetic paths followed in an applied ac magnetic field as discussed in the text.

In this analysis it has been assmed that the shielding due to the critical surface currents is the major contribution to M above H_{c2}. If this were not the case, the magnetization above H_{c2} would be proportional to the thickness of the surface layer, i.e., to ξ, the coherence length. This is so small compared to the sample volume, that it could not explain the observed magnetization.

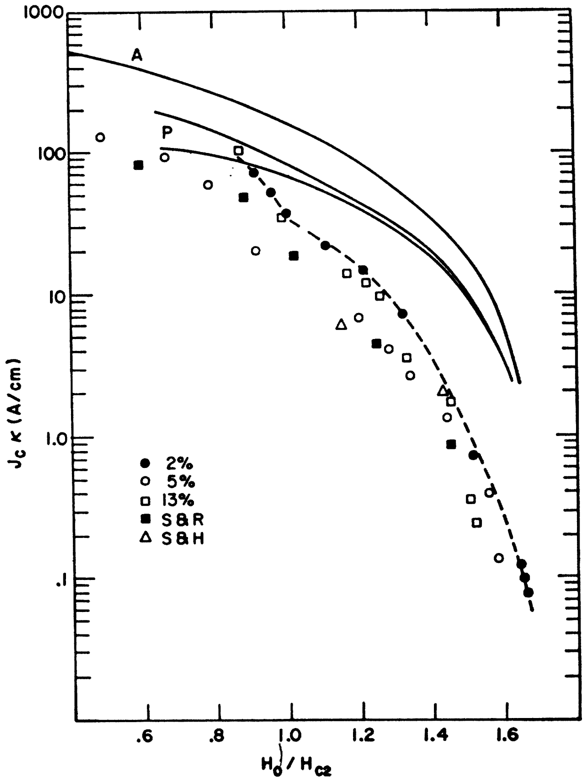

FIG. 3. Critical surface currents multiplied by G-L parameter versus H_0/H_{c2} where H_0 is the applied dc field. The upper solid curves marked *P* are the calculations of Park, the one marked *A* is the calculation of Abrikosov as reported by Park. The data points marked *S* and *R*, and *S* and *H*, refer to the work of Silcox and Rollins and that of Swartz and Hart, respectively. The solid circles are the present data for 2% Bi in Pb which has a κ of 1.03. The 5% has a κ of 1.9 and the κ of the 13% is 4.8. The κ's of *S* and *H* and *S* and *R* samples were 1.75 and 3, respectively. The dotted curve is to illustrate the discontinuity at H_{c2}.

In Fig. 3 the critical surface currents obtained in the present measurements are compared with those calculated by Park from the G-L theory and also Abrikosov's results as reported by Park. Earlier ac measurements by Rollins and Silcox,[8] and some values of the dc surface critical current measurements by Swartz and Hart[9] are included, and are in agreement with the present data.

In some cases ac transitions were found to begin at fields well below H_{c2} (the change in χ' being as much as 10% of the total transition), then to reach a constant value of χ', and finally to undergo the rest of the transition above H_{c2}. The initial part of the transition below H_{c2} was found to be dependent on whether the ends of the samples were in the susceptibility coils and this part of the transition was probably due to the ends of the sample going into the intermediate state. By having the ends of the sample outside the coil this effect could be made negligibly small.

DISCUSSION

The results can be summarized as follows:

1. The measured surface critical current shows the same qualitative field dependence as the calculations of Park and of Abrikosov above H_{c2}, although the mag-

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[8] R. W. Rollins and J. Silcox, Proceedings of the Conference on the Physics of Type II Superconductivity, Western Reserve University, 1964, p. III-32 (unpublished).

[9] P. S. S wartz and H. R. Hart, Jr., Phys. Rev. **137**, A818 (1965).

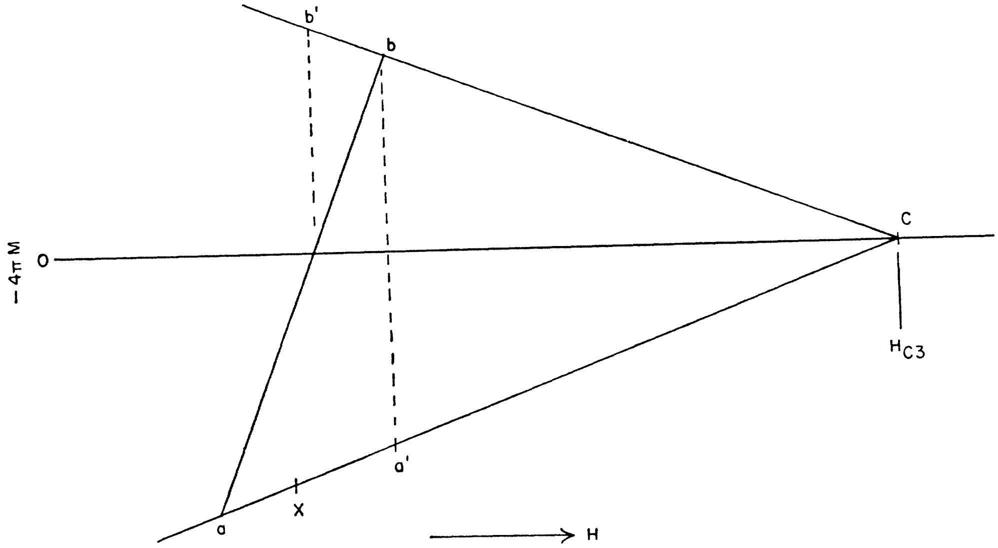

FIG. 4. A schematic magnetization curve near H_{c3} illustrating the discussion of free energy in the text.

nitude and detailed field dependence are different and are in better agreement with Fink and Barnes.

The question of whether these differences are significant will be discussed later in this paper. At present it is well to consider the assumptions of the various calculations. All three calculations assume that the free-energy differences between the normal and superconducting states can be minimized with respect to ψ to obtain the first Ginzberg-Landau equation. However, Fink and Barnes indicate that the free-energy difference between the normal and superconducting state should not be minimized with respect to **A**. This seems reasonable, since because of the persistent surface currents and associated hysteretic properties already discussed,[7] the system consisting of surface sheath and core is no longer in equilibrium (defined as lowest free-energy state) with respect to field. However, Fink and Barnes then assume that $F_s-F_n=0$, where F_s is the free energy of the sample consisting of the superconducting surface sheath plus the core and F_n is the normal free energy of the sample. In the following we show a simple argument that at fields $<H_{c3}$, $F_s-F_n\neq 0$, although near H_{c3}, $F_s-F_n\approx 0$ and the approximation made by Fink and Barnes should be good.

At point c in Fig. 4, $F_s=F_n$, since at H_{c3} the sample is ready to go completely normal. Now consider the free-energy change in going from point c to some other point on the magnetization curve near a' or a. The upper and lower branches of the magnetization curve, say bc or ac, describe irreversible paths and hence one cannot compute the free-energy change because $\Delta Q-T\Delta S\neq 0$. However, previous work[10] has shown that the "diamagnetic" a-b is reversible. Since the process is isothermal $F_{b-a}=-\int_b^a MdH$. However this is also equal to the change from $F_{b-c}+F_{c-a'}+F_{a'-a}$. Now the assumption is made that, because of the symmetry $F_{b-c}=F_{c-a'}$,

$$2F_{c-a'}+F_{a'-a}=-\int_b^a MdH\,,$$

$$F_{c-a'}+\tfrac{1}{2}F_{a'-a}=-\frac{1}{2}\int_b^a MdH\,.$$

$\frac{1}{2}F_{a'-a}$ is the change from a' to some arbitrary point x between a' and a. Hence,

$$F_{c-a'}+F_{a'-x}=-\frac{1}{2}\int_b^a MdH=F_{c-x}\,,$$

$$F_{c-x}=-\frac{1}{8\pi}\int_b^a (H_{b'}-H)dH$$

$$=-\frac{1}{8\pi}\left[H_{b'}H-\frac{H^2}{2}\right]_b^a,$$

$$F_{c-x}=\frac{H_a-H_b}{8\pi}\left[\frac{H_a+H_b}{2}-H_{b'}\right].$$

This is the free-energy change of the system, consisting of sheath plus core in going from c to x. This result is a small positive quantity, since $H_{b'}>(H_a+H_b)/2$. Thus, the free energy of the system, consisting of core plus superconducting sheath, is higher at x than the normal free energy. The small value of this free-energy difference explains why the approximation made by Fink and Barnes should be very good near H_{c3}. However at fields below, say H_{c2}, it would be expected that $F_s-F_n=0$ is a poor assumption.

2. There is no observed κ dependence of $J_c\kappa$ for the

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[10] D. J. Sandiford and D. G. Schweitzer, Phys. Letters 13, 98 (1964).

range of κ investigated ($1<\kappa<5$). In the calculation of Abrikosov and that of Park, $J_c\kappa/H_c$ should be a constant if $\kappa\gg1$. Since H_c can be assumed to be almost constant in these measurements, the result that $J_c\kappa$ is independent of κ suggests that the result of Park[2] that J_c is $H_c/\kappa I(H)$ is true even for $\kappa\approx1$.

3. The measured curves break sharply at $H=H_{c2}$. This has been observed by Swartz and Hart and discussed by them in terms of two separate surface mechanisms. The values of H_{c2} measured in this way were in agreement with those taken from the magnetization curves.

4. The critical surface currents are found to agree within experimental error with the dc results of Swartz and Hart[9] (in fact, the present values may be slightly greater) on flat plates. This would appear to be an important point since our data were on cylinders in which the volume of trapped field would be much greater than in flat plates. This observation might indicate that the kinetic energy of the superconducting electrons in the surface layer and the field energy associated with this layer are the pertinent factors in determining the critical current of the surface. That is, the energy associated with the trapped flux in the core of the sample does not significantly affect the value of the critical surface currents. Hence, the agreement between the data on plates and on cylinders. This is not surprising since it can be shown in the case of solid or hollow cylinders of type-I superconductors that the current flowing within a penetration depth of the surface at H_c can be calculated by setting the kinetic energy per unit volume of the surface electrons equal to the gap energy per unit volume.[11]

This calculation shows that the kinetic energy per unit volume of the superelectrons at the surface is

$$\text{K.E.}=\tfrac{1}{2}(4\pi\lambda^2/c^2)J_s^2=\tfrac{1}{2}mv^2N,$$

where $J_s=H_cc/4\pi\lambda$ is the current flowing on the surface. We can say that $J_s=i/l\lambda$ and therefore $i=lH_cc/4\pi$. One can show that the total field energy due to this current is $(H_c^2/8\pi)V$. For instance the field energy of the current is $1/2(Li^2/c^2)$ but $L=4\pi A/l$ so

$$\frac{Li^2}{2c^2}=\frac{l^2c^2H_c^2}{2(4\pi)^2c^2}\,\frac{4\pi A}{l}=\frac{H_c^2}{8\pi}V.$$

For a solid sample this would give the expected results, i.e., the field energy at H_c is equal to the condensation energy of the whole sample.

In the hollow cylinder case, the same arguments will yield a value of J equal to that calculated above. However, the field energy set up by this current is no longer equal to the condensation energy of the actual superconducting volume, and of course, current can flow on the inner wall as well as the outer one in this case. Hence, the same J_c is obtained even though the actual volume of superconductor and the trapped field are different.

Some particular features of the present measurement should be emphasized in connection with a detailed comparison with theory. Since cylindrical samples are being used, it is very difficult to achieve exact alignment between the sample axis and the magnetic field. Swartz and Hart's data show that a deviation of 5° can decrease the critical surface current by a factor of 10. It is estimated that the present maximum angle was $<3°$. The theoretical case was also calculated for a perfect surface, whereas the surfaces in the present study were not subjected to any special treatment and might be expected to display regions where the field would not be parallel to the surface of the metal. The value of H_{c3} is, of course, less sensitive to angle than the current, since $H_{c3}/H_{c2}=1.7$ in the parallel case and 1 in the perpendicular case. On the other hand, J_c can change many orders of magnitude with angle.

A question arises as to the definition of surface critical currents in these particular measurements. Since the penetration of the ac field into the sample is measured, the model illustrated in Fig. 1 would imply that the critical current is reached when ac penetration starts (i.e., when χ' first deviates from $-1/4\pi$). This is indeed true; however, if the sample axis is not exactly parallel to the field or if any surface roughness exists, part of the cylindrical surface will experience a perpendicular field component and hence will go normal at a field lower than the part of the sheath that is parallel to the field. In this case the high-field part of the transition would be more meaningful. In most cases the transitions are fairly sharp in field and the above considerations do not lead to large uncertainties in the J_c at a given field. The ranges from beginning to end of the transitions have been considered, and use of any point in the range leads to no qualitative difference.

The shape of the surface critical current curve at low currents indicates how small the ac field must be, so that the transition dc field will not be reduced appreciably. In the previous work at ac fields of 0.04 Oe peak-to-peak, corresponding to a J_c of ≈0.003 A/cm, we estimated that the transitions were within a percent of H_{c3}.

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Note added in manuscript. In this paper we used the field at which the maximum in χ'' occurred as an indication of the dc field at which the susceptibility transition occurred. For this dc field we then said the critical current was approximated by $H_{\text{ac}}=0.4\pi J_c$. We also indicated towards the end of the paper that the

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[11] See, e.g., M. Tinkham, *Low Temperature Physics* (Gordon and Breach Science Publishers, Inc., New York), p. 217.

first penetration of the ac field into the sample defines the dc field at which the critical current, in the model illustrated in Fig. 2, is reached. However, because of experimental factors such as precise identification of the transition point and other points discussed in the paper, we felt the χ'' peak was the best "all-around" point to use for the transition. After submission of this paper Fink,[12] and Rollins and Silcox[13] have discussed the χ' and χ'' transitions in great detail. From Fink's work one gets the relation that $h_c=0.385\ H_{ac}$. Hence the critical current values on this argument should be given by $0.385\ H_{ac}=0.4\pi J_c$. This would then reduce all our current values by over a factor of 2.5. h_c is defined by Fink as the vertical height from $4\pi M=0$ to point (2,6) in the high-field hysteresis loop (curve B) in Fig. 2. This is of course just the point at which any increase in dc field will cause χ' to deviate from $-1/4\pi$, as discussed in the paper.

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Thermal Conductivity of Thick Pure Lead Films for the Study of Surface Superconductivity*†

T. SEIDEL‡ AND HANS MEISSNER

Department of Physics, Stevens Institute of Technology, Hoboken, New Jersey

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The differential thermal conductivity of lead films in a magnetic field parallel to the plane of the film has been measured in the temperature range 1.2 to 4.2°K. The thicknesses of the films (from 2500 to 7000 Å) have been selected so that the expected volume of the superconducting surface regions is an appreciable volume fraction of the sample. The fraction of material remaining in the surface superconducting state just above the film's critical field is related to the measured thermal conductivity via a simple phenomenological model. The analysis allows a heuristic determination of the product of the thickness of the superconducting surfaces and the square of the order parameter at the surface. The results are compared with the recent calculations of Fink and Kessinger. Well defined values of H_c and H_{c3} are obtained. Some hysteresis is observed near H_c in a decreasing magnetic field, and the role of phonons in the heat transport is manifest, but its contributions above H_c is negligible when compared with the electronic contribution to the thermal conductivity.

I. INTRODUCTION

SURFACE superconductivity[1] was predicted by Saint James and de Gennes for type-II superconductors[2] for magnetic fields up to $1.69H_{c2}$. At H_{c2} the magnetic field completely fills the interior of a type-II superconductor, keeping the interior in the normal state. The Saint James–de Gennes superconducting surface layer is expected to have a thickness of the order of the superconducting coherence[3] distance ξ. The existence of surface superconductivity in type-II superconductors is well established.[4]

Remnant superconducting properties have also been observed in type-I superconductors above the bulk thermodynamic critical field H_c. Among the indications of this phenomenon were the microwave surface resistance[5] on pure bulk lead, the magnetization[6] in dilute alloys of the Bi-Pb system, and electron tunneling[7] in thick pure films of lead. These experiments have been interpreted in terms of surface superconductivity mainly on the basis of the observation that superconductivity exists for external fields up to $H\approx 1.7\ H_{c2}$, where even for a type-I superconductor H_{c2} is taken as $\sqrt{2}\kappa_{G.L.}H_c$. ($\kappa_{G.L.}$ is the dimensionless Ginsburg-Landau parameter.) The purpose of this work is to investigate the superconductivity of pure lead below and above H_c by measurements of the thermal and electrical conductivity. The significance of the thermal conductivity approach is that it may be used to demonstrate directly that the thickness of the superconducting region is of the order of the coherence distance.[8]

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‡ Present address: RCA Laboratories, Princeton, New Jersey.

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