Temperature and magnetic field dependence of the superconducting penetration depth in pure and impure aluminum single crystals*

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Measurements at about 0.9 MHz of the temperature dependence of the penetration depth in pure and impure single crystals of aluminum are found to be in substantial agreement with the calculations of Miller, while at the same time showing some anisotropy effects for the pure samples. The effect of a static magnetic field on the surface reactance at this frequency is found to be in substantial agreement for the pure samples with a model proposed by Garfunkel for extreme type-I superconductors. Near the superconducting transition temperature the effect of the static field is compared with the Ginzburg-Landau theory, with only limited success for the impure aluminum sample.

I. INTRODUCTION

The superconducting penetration depth λ defined, for static magnetic fields, by

$$\lambda \equiv \frac{1}{H(0)} \int_0^\infty H(x)\,dx\,, \tag{1}$$

gives the effective depth to which the field penetrates a bulk superconductor. In Eq. (1), $H(x)$ is the magnetic field at a distance x from the surface into the superconductor. Because the interior of the superconductor is shielded from the magnetic field, the direction of the field in the penetration region must be almost parallel to the surface.

The electromagnetic properties of superconductors at fixed angular frequency ω are contained in the complex surface impedance $Z = R + iX$ (R and X are the surface resistance and surface reactance, respectively), defined as

$$Z = 4\pi E(0)/H(0)\,, \tag{2}$$

where $E(x)$ is the electric field at a distance x from the surface. It is sometimes convenient to define a complex skin depth $\delta = \delta_r - i\delta_i$ as in Eq. (1), where the magnetic fields are at angular frequency ω. Thus, $\lambda = \lim_{\omega\to 0}\delta$, and it is easily seen[1] that $4\pi\omega\delta(\omega) = -icZ(\omega)$, where c is the velocity of light.

Measurements of the superconducting penetration depth were initiated soon after the London phenomenological electrodynamics[2] in conjunction with the Gorter-Casimir two-fluid model[3] predicted a temperature-dependent penetration depth given by

$$\lambda(T) = \lambda(0)(1 - t^4)^{-1/2}\,, \tag{3}$$

where t, the reduced temperature, is the ratio of the temperature T to the superconducting transition temperature T_c (i.e., $t \equiv T/T_c$). The penetration depth at absolute zero, $\lambda(0)$, was estimated in the London theory to be of the order of 10^{-5} to 10^{-6} cm for typical superconductors. In the early experiments,[4] Eq. (3) satisfactorily described the results to within the experimental accuracy. However, when the Bardeen-Cooper-Schrieffer (BCS) microscopic theory of superconductivity[5] predicted a slightly different temperature dependence from that given by Eq. (3), Schawlow and Devlin[6] designed an experiment in which they were able to observe the predicted small deviation for superconducting tin. Since then there have been other observations of these small effects in other materials.[7,8]

The study of the effect of a static magnetic field on the superconducting penetration depth was initiated in 1950 by Pippard[9] in measurements of the microwave surface impedance at 10^{10} Hz in tin. The complicated results he obtained in these early experiments could not be explained in terms of the existing theoretical apparatus, but they did lead to a number of other experiments designed to unravel the mystery. Papers by Pippard,[10] Garfunkel,[11] and Budzinski *et al.*[12] give a number of references to the early work and summarize the results.

Budzinski and Garfunkel[13] observed a large magnetic-field dependence of the microwave absorption at photon energies near the energy gap 2Δ in pure aluminum. This led them to propose[13] that the electron energy spectrum was shifted in the presence of a magnetic field by an amount $\vec{p}\cdot\vec{v}$, where $\vec{p}$ is the electron momentum and $\vec{v}$ is the drift velocity associated with the Meissner current which shields the field from the interior of the superconductor. Garfunkel[11] used this shift of energy to calculate the surface impedance for all frequencies. His calculations were quite successful in giving the general features of the high-frequency ($\hbar\omega/\Delta \sim 1$) surface impedance,[12,14] and has had some limited success in explaining the apparent anomalies[10] in the intermediate frequency range ($\hbar\omega/\Delta \sim 10^{-2}$–$10^{-1}$).[10,11,15] In this paper, we

present results[16] at about 0.9 MHz to show that the model is also successful in explaining the static-field dependence of the low-frequency surface reactance ($\hbar\omega/\Delta < 10^{-3}$), or, what is nearly the same thing, the superconducting penetration depth. Aluminum was selected for these studies for several reasons: when pure, it is an extreme type-I superconductor ($\kappa \approx 0.01$), a necessity for comparison with the theory[11]; aluminum has been extensively studied in all frequency regimes[7,12,15]; and it is a convenient material to use in many ways, being available with very high purity, having desirable chemical and metallurgical properties, having a transition temperature and critical field that are readily accessible, and having a relatively small crystalline anisotropy since it has a (face-centered) cubic lattice.

II. EXPERIMENTAL

A. Apparatus

Since the superconducting penetration depth at $T=0$ is of the order of 10^{-5} to 10^{-6} cm, and since the changes of the penetration depth in a magnetic field are only of the order of a few percent, it is necessary to have the sensitivity to detect changes of about 0.2 Å. Because the method used by Schawlow and Devlin[6] is capable of giving this sensitivity, we adapted it for these measurements. This method uses a resonant inductive-capacitive circuit with the superconducting sample as the core of the inductance. The resonant circuit is part of an oscillator, and the measurement consists of determining the resonant frequency of the oscillator. A change in penetration depth causes a change in inductance which in turn causes a change in the resonant frequency. The experiment then consists of controlling and measuring the temperature of the sample, and of applying a uniform static magnetic field, while obtaining accurate measurements of the resonant frequency of the oscillator.

Figure 1 is a schematic diagram of the low-temperature portion of the apparatus, a conventional He^3 refrigerator inside a double Dewar system containing liquid nitrogen and liquid He^4. The aluminum cylinder sample is mechanically attached to the He^3 evaporator and carbon resistance thermometers are attached to the two supports at the two ends of the sample. Thermometers T are calibrated against the vapor pressure[17] of He^3 using the 1962 He^3 temperature scale.[18] Temperature is controlled by a combination of manual control of the He^3 pumping speed and electronic control of the electric current to the heater H. The uniform static magnetic field is applied by the solenoid emersed in the liquid-nitrogen bath. The field from the solenoid is uniform to about 0.2% over the length of the sample and to about 0.02% over the section of the sample covered by the ac coil. Current in the solenoid, and thus the static field, is controlled so that over periods of about 1 h the change is of the order of one part in 10^5. Surrounding the whole Dewar system (but not shown in Fig. 1) is a Helmholtz pair, which is used to cancel the geomagnetic field at the sample to about 0.01 G. Around the sample is the ac coil, which supplies the inductance for the resonant circuit. In order to get maximum sensitivity, it is desirable that the coil be close fitting over the sample, so that the inductance is largely associated with the field that penetrates the sample, rather than the empty space between sample and coil. The coil is wound on a thin-walled (~0.01 cm) epoxy tube (~1.0-cm i.d.) and consists of a single layer of B. & S. gauge No. 36 (diam 0.127-mm) copper wire. (In an earlier version, the coil was wound of niobium wire to reduce its losses, but the inductance of this coil was found to have a larger magnetic-field dependence than that of the copper coil and was therefore discarded.)

The copper coil with the sample as a core served as the inductance in the resonance circuit of the oscillator, a transistorized version of the Clapp oscillator[19] having two field effect transistors. In order to isolate the oscillator from the output, to prevent loading, an amplifier stage follows the

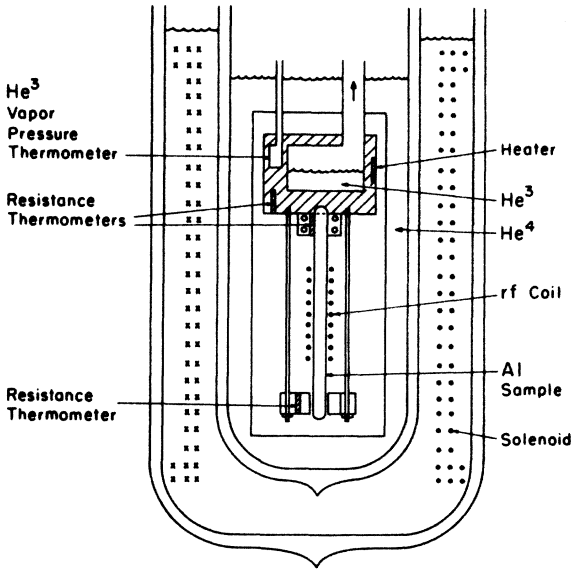

FIG. 1. Schematic diagram of the low-temperature section of the apparatus, showing the sample mounted as the core of the inductance in the resonant circuit. See the text for a description of the details.

oscillator circuit. The frequency of the oscillator, controlled by the selection of capacitor in the resonant circuit, can be measured accurately by counting cycles for 0.100 000 0-, 1.000000-, or 10.000000-sec periods using an electronic counter.[20] The frequency is found to be stable at 800 kHz to within a few hertz over a period of 1 h. Furthermore, the small changes that occur are found to be systematic, drifting at a nearly constant rate. The drift appears to arise from the gradual change in the electrical resistance of the leads between room temperature and the low temperature, as the level of liquid nitrogen falls in the outermost Dewar.

B. Samples

The samples are aluminum,[21] grown into oriented single crystals in vacuum using the temperature-gradient technique. The one impure sample has about 0.2-at.% silver which was added to pure aluminum in the melt. The samples, after growing, were spark machined into cylinders with rounded ends, about 15 cm long and just under 1 cm in diameter. They were then chemically polished,[22] annealed at 550 °C for 24 h to remove strains, and again chemically polished[22] to a mirror-like finish. A measure of the purity of the samples is obtained from the residual resistance ratio (RRR) determined by using an eddy current technique[23] to obtain the sample resistivities at both room temperature and 4.2 K.

C. Measurements

For each of the samples, we first calibrated the thermometers and then measured the resonant frequency of the oscillator as a function of temperature from above the superconducting transition temperature to the lowest temperature we were able to attain. We then carried out the same measurements as a function of static magnetic field up to the critical field.

In order to convert the measurements of frequency to penetration depth or surface reactance it is necessary to know the geometrical relationship between the coil and the sample. Because the penetration depth is so small, it is impossible to obtain this relationship with sufficient accuracy for an absolute determination of the penetration depth. Thus, we are limited to determining changes in the penetration depth from changes in the resonant frequency. We can understand this, for the case of zero static magnetic field, as follows.

If f is the resonant frequency of the oscillator and L is the inductance of the coil, then

$$\delta f/f = -\tfrac{1}{2}\delta L/L \,. \tag{4}$$

In terms of the cross-sectional area of the coil (see Fig. 2) that contains magnetic flux, i.e., $\pi r_c^2 - \pi r^2$, we can determine L and δL. This then yields

$$\frac{\delta L}{L} = -\frac{2\pi r \delta r}{\pi r_c^2 - \pi r^2} \approx \frac{2\pi r \delta\lambda}{A} \,, \tag{5}$$

where λ is the penetration depth and $A \equiv \pi r_c^2 - \pi r^2$. Eliminating $\delta L/L$ and solving for $\delta\lambda$, we get

$$\delta\lambda = (A/\pi r)(\delta f/f) \,. \tag{6}$$

Now $r \approx r_0$, and all that remains is to determine A. Note that

$$A \approx \pi(r_c^2 - r_0^2) \propto L \propto 1/f^2 \,,$$

or

$$A = 1/Bf^2 \,, \tag{7}$$

which defines the proportionality constant $1/B$. To obtain B, we can plot $1/f^2$ as a function of r_0^2 since

$$1/f^2 = B\pi(r_c^2 - r_0^2) \,, \tag{8}$$

The slope of this graph is $-B\pi$, enabling us to find A as a function of f from Eq. (7). It is sufficient to measure the resonant frequency of the empty coil and the resonant frequency with a sample of known diameter to determine B. However, we found it desirable to check Eq. (8) by using samples of several different diameters. These measurements show that end effects (the field at the end of the coil varies with radius) cause the slope of the $1/f^2$ vs r_0^2 to vary, introducing an 8% error if one uses only the $r_0 = 0$ and one value for $r_0 \leq r_c$. The correction for this error has been made giving us a value of $B = (2.03 \pm 0.04) \times 10^{-11}$ sec^2 cm^{-2}. Another possible source of error in B, namely, the effect of the sample diameter on the distributed capacitance of the coil, seems not to be significant.

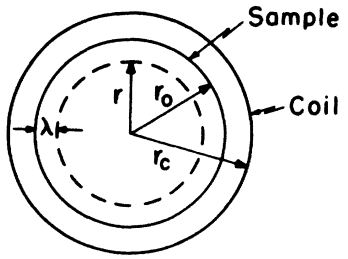

FIG. 2. Schematic diagram showing the cross section of the ac coil and the sample. The dashed curve indicates the distance of penetration of the field. See the text for the calculation of penetration depth changes from frequency changes.

Changes in λ can then be found from

$$\delta\lambda = (\delta f/f)(1/\pi B r_0 f^2) . \quad (9)$$

The argument given above is correct in the absence of a static magnetic field. But in the presence of a static field it is not useful to describe the electromagnetic properties in terms of the penetration depth, since the reactive emf from the coil is given by

$$\mathcal{E} \propto \frac{\partial}{\partial t}(\lambda H) ,$$

where both λ and H vary with the alternating magnetic field. If λ is given in a power series in H^2, i.e.,

$$\lambda = \lambda_0 + \lambda_1 H^2 + \lambda_2 H^4 + \cdots ,$$

then for the simplest case where we only keep terms to H^2 in λ, the apparent measured changes in λ using Eq. (9) would be a factor of 3 too large for the alternating magnetic field parallel to the static field. Thus, for the sake of comparison with the theory[11] we consider changes in the surface reactance δX as a function of static field. The reactance X calculated in the theory[11] implicitly corrects for the field-dependent penetration depth.[24]

We have investigated several possible extraneous sources of field dependence of the resonant frequency. We found that with a coil wound of superconducting niobium there was a large unpredictable effect of the field on the resonant frequency. This was probably related to the mechanical forces on the superconducting wire resulting from trapped flux in the wire. At any rate, we switched to a copper coil to avoid this problem. To be sure that there were no similar problems with the copper wire we ran a simple test. An empty coil of two layers of B. & S. gauge No. 36 copper wire wound in opposite directions was used to check the changes in frequency as a function of temperature and magnetic field. We found that the changes were so small that they would have negligible effect on the experimental results. We also checked the sensitivity of the coil form by making a solid coil core of the same epoxy. We found no temperature or field dependence. Another possible effect is that of mechanical stress on the sample from the field. At the largest field we used (about 75 G) the change in radius of the samples should be less than 0.03 Å, smaller, by a factor of about 10 than our measuring sensitivity.

In order to measure the transition temperature T_c, we observed the rapid change in penetration depth with temperature approaching T_c from below. The change in slope is so abrupt that the transition temperature is determined to within about 1 mK. The ac power dissipated in the copper coil and in the sample just above T_c is negligible from the point of view of introducing temperature errors.

The measurements consisted of two parts: the change in penetration depth with temperature; and the change in surface reactance with magnetic field. The change with temperature was carried out in zero magnetic field (actually, only known to be less than 0.01 G). Measurements were taken both with increasing and decreasing temperature. There was no observable hysteresis except for about 1 mK at T_c, which we attribute to supercooling in the decreasing temperature data. In an attempt to discover whether trapped flux might cause some difficulty, we drove a pure aluminum sample normal with a large field at low temperature and then reduced the field to zero. There was no apparent change in the temperature dependence indicating that there was no flux trapped in the sample. The field dependence of the surface reactance was determined by increasing the field at fixed temperature. At the lower temperatures these measurements were easily made, but near T_c small variations in temperature cause large changes in penetration depth, overwhelming the small field dependence. Thus, our measurements were taken only up to $t \equiv T/T_c = 0.96$.

III. RESULTS

A. Temperature dependence of the penetration depth

In Fig. 3 we show a plot of the frequency of the oscillator as a function of the absolute temperature with a pure-aluminum sample (Al-11) as the core of the coil. The data for the other samples look very much the same, except that in the case of the impure sample (Al-12) the transition temperature

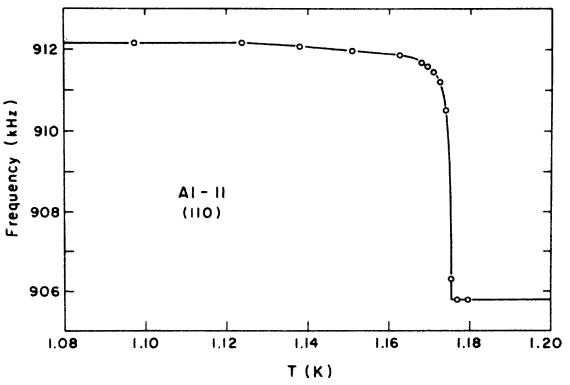

FIG. 3. Oscillator frequency vs sample temperature for pure aluminum sample Al-11. Note the very sharp change in frequency at $T = T_c$.

TABLE I. Properties of the aluminum samples.

Sample	at. % Ag	Crystal direction of axis	Residual resistance ratio	Superconducting transition temperature T_c (K)	Sample radius at 4.2 K in cm	κ	Theoretical $\lambda(0)$ in Å [a]
Al-9	0	[100]	4100±500	1.177±0.002	0.492	0.01	544
Al-10	0	[111]	4200±500	1.176±0.002	0.493	0.01	544
Al-11	0	[110]	4000±500	1.176±0.002	0.493	0.01	544
Al-12	0.2	[100]	12.8±1.0	1.128±0.002	0.491	0.075	723

[a] Theoretical values for the penetration depth were obtained from the calculations by Miller in Ref. 25, by using the values $\lambda_L(0)=160$ Å and $\xi_0=16\,000$ Å appropriate for aluminum.

is lower, and the total change in frequency is about four times as large indicating that the penetration depth in the normal state is larger. We collect the characteristic data for all four samples in Table I.

Because the London[2] theory in conjunction with the Gorter-Casimir[3] two-fluid model has only a slightly different temperature dependence than that of the microscopic theory[5] it is convenient to use Eq. (3) for comparison. Defining $z=(1-t^4)^{-1/2}$, we can calculate the change in λ from the data and Eq. (6). If we had one value of λ, say $\lambda(0)$, we could then construct a graph of $\lambda=\lambda(z)$. Although the experiment does not give us the absolute value for $\lambda(0)$, we choose values to agree with the theoretical predictions of Miller,[25] using for the London penetration depth[5] $\lambda_L(0)=160$ Å and the BCS coherence length $\xi_0=16\,000$ Å. With these selections, which are in essential agreement with previous measurements, at least in the pure cases, we can now plot λ as a function of z. But it is necessary to repeat that only the differences between points are determined in this experiment, and the values at $T=0$ are arbitrary from the point of view of the experiment. With this qualification, we now plot the curves of λ vs z: for the three pure samples in Figs. 4(a)–(c); and for the impure sample in Fig. 4(d). In each figure is shown a theoretical[25] curve for a sample having the same electron free path. Note that the graphs do not have a constant slope close to $z=1$ ($t=0$), curving in a manner similar to the theoretical curve. However, one limitation must be made; namely, the appearance of curvature or the absence of it in the experiment depends somewhat on the value taken for T_c. A shift of only a few millikelvin can change the appearance of the curvature. Thus, we do not believe that the detailed shapes of these surves are completely reliable, although they do seem to generally agree with the form of the deviation from Eq. (3) predicted by the microscopic theory.[5]

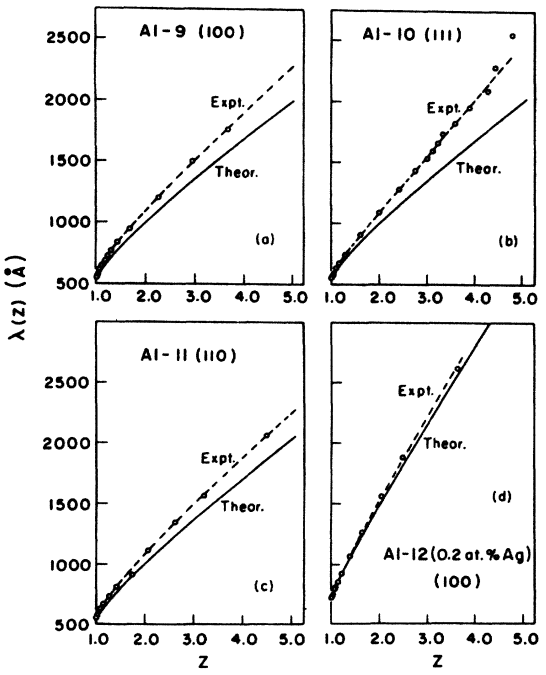

FIG. 4. Penetration depth λ vs $Z=(1-t^4)^{-1/2}$ for the four aluminum samples. The solid curves are from the calculations of Miller in Ref. 25. Since the absolute value of $\lambda(0)$ could not be determined in the experiment, the experimental data are plotted to agree with the theory at $Z=1$ ($T=0$).

B. Static-magnetic-field dependence of the surface reactance

For the reasons discussed in Sec. II, in the presence of a static magnetic field, it is better to refer to the surface reactance X rather than the penetration depth. We have made measurements of changes in surface reactance as a function of static magnetic field at reduced temperatures of $t=0.30$, 0.60, 0.84, and 0.96. These results are normalized by finding the fractional increase of the reactance over the zero-field value at each temperature. The normalized data are then plot-

ted in Figs. 5(a)–(d). Note that the change in surface reactance (and thus the penetration depth) increases with increasing field for all samples although the results are rather complicated, having a somewhat different form at different temperatures.

IV. DISCUSSION

A. Temperature dependence of the penetration depth

The experimental curves of Fig. 4 show the expected deviation from Eq. (3) predicted by BCS. Although, we are somewhat unsure of the extent of the deviation as mentioned above, we do believe that these results along with the results of Tedrow *et al.*[7] establish the existence of a BCS-like deviation from Eq. (3) for aluminum in contrast to the results reported by McLean[26] in his microwave experiments on pure aluminum. The curves for the three pure samples all show some small discrepancy with the theory and with each other. We attribute this to crystalline anisotropy, although it is also possible that the quality of the sample surfaces has some effect. The impure sample, Fig. 4(d), agrees remarkably well with the theory.

B. Surface reactance in a static magnetic field

In order to compare the experimental results of Figs. 5(a)–(c) with the calculations of Garfunkel,[11] it is necessary to find the relationship between the critical magnetic field H_c and the pair-breaking field[27] H_0 of the theory.[11] Budzinski *et al.*[12] find from microwave-absorption experiments that $\alpha_{\text{expt}} \equiv H_0/H_c \approx 0.76$ to fit their data on aluminum. We adopt this same value to reduce the theoretical curves for comparison with experiment, and show the theoretical curves in Fig. 6(a) in comparison with the experimental data in Figs. 6(b)–(d). We plot the fractional change in surface reactance as a function of temperature for reduced magnetic field values $h \equiv H/H_c = 0.3$, 0.5, 0.6, and 0.7. The agreement both in magnitude and general shape is excellent for Al-9 and is not too bad for the other two pure samples Al-10 and Al-11. Clearly there are crystalline anisotropy effects which have not been accounted for in the theory. Nevertheless, the general agreement adds to our confidence in the theory[11] in this frequency region.

We now consider the results for the impure sample shown in Fig. 5(d). The fractional change in the surface reactance as a function of reduced magnetic field is much larger than for the pure samples although it shows some of the same characteristics. As can be seen from Table I, κ for this sample is 0.075, more than seven times larger than for the pure specimens. For this

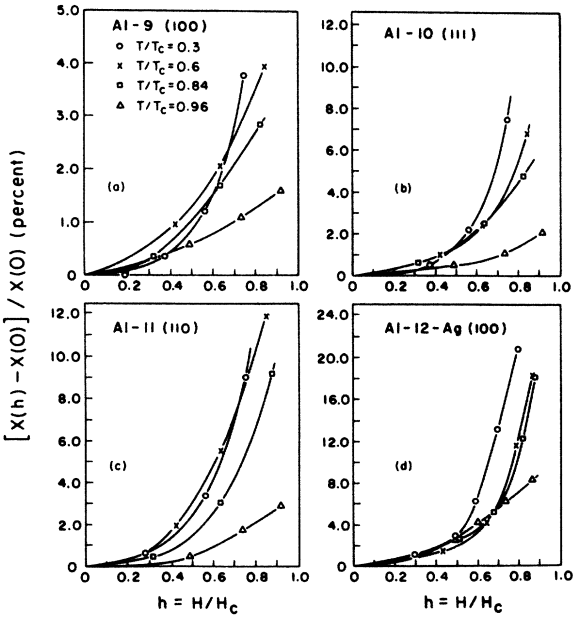

FIG. 5. Fractional change in surface reactance as a function of reduced static magnetic field for the aluminum samples at various temperatures.

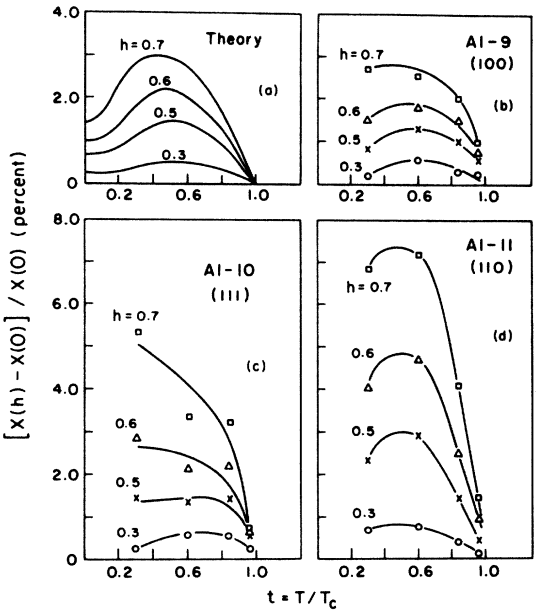

FIG. 6. Fractional change in surface reactance as a function of reduced temperature at various values of fixed reduced static magnetic field. The theoretical curves from Ref. 11 are shown in (a) and are to be compared with the experimental curves for the pure samples in (b), (c), and (d).

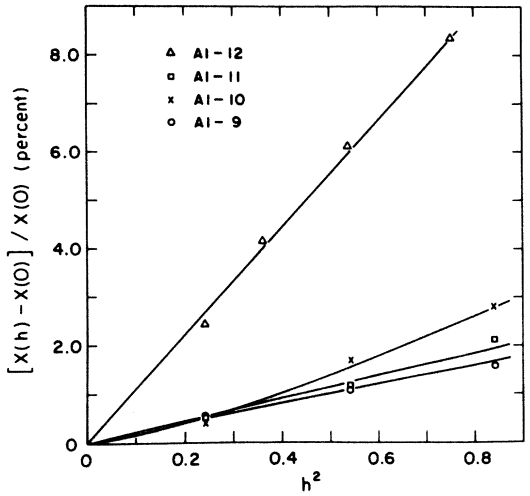

FIG. 7. Fractional change in surface reactance vs the square of the reduced field near T_c. The Ginzburg-Landau theory (Ref. 28) gives a straight line for these curves with a smaller slope.

reason, it is not quite appropriate to compare the results with the Garfunkel model[11] which was developed for the small-κ limit. The theory of Ginzburg and Landau[28] (GL), which applies near T_c,[29] predicts a change in surface reactance which increases with increasing κ. Although our measurements have not been taken quite close enough to T_c for the GL theory to be applicable[29] for the pure samples, we compare the results with the predictions for all the samples. Since the largest value of κ among our samples is 0.075, the GL[28] theory gives within experimental accuracy

$$\Delta X/X=[X(h)-X(0)]/X(0)=ah^2\,, \tag{10}$$

where for the ac and dc fields parallel,

$$a=3\kappa(\kappa+2\sqrt{2})/8(\kappa+\sqrt{2})^2\,. \tag{11}$$

In Fig. 7, we plot $\Delta X/X$ vs h^2 at $t=0.96$ for the four samples. We see that, to the accuracy of the data, Eq. (10) is satisfied for the impure sample Al-12, but is not quite so well satisfied for the pure samples. Nevertheless, we can find a value for a from the initial slopes of each of the curves. For the impure sample, the slope gives $a\approx 0.1$. This compares with the theoretical value from Eq. (11) of 0.04; not good agreement, but it is of about the right order of magnitude. In the case of the pure samples, the experimental value of about 0.02 is four times larger than the theoretical value of 0.005, in even worse agreement with the Ginzburg-Landau[28] prediction. The primary reasons for this disagreement are that none of the data are close enough to T_c for a proper comparison with the GL theory,[28] and the effects of the Garfunkel model[11] have been ignored.

Notes added in proof. (a) In a recent paper[30] Josephson reports on some old measurements of the magnetic field dependence of the surface reactance in tin at 174 MHz. His results do not seem to fit any present model, although some of the features of his results (e.g., the existance of a decrease in surface reactance with increasing static field) are also features of the Garfunkel model.[11] Of course, the case of tin is complicated in that it is intermediate in the sense that it has a κ value of about 0.2, requiring both Ginzburg-Landau and $\vec{p}\cdot\vec{v}$ corrections. It is also possible that the inclusion of energy-gap anisotropy in the $\vec{p}\cdot\vec{v}$ corrections can cause the Garfunkel model to show the anomalous features of the Josephson results. (b) In addition to the measurements presented in this paper we obtained results on a second [100] pure crystal in which measurements were made with the static field both parallel and perpendicular to the axis of the sample. The parallel measurements reproduced the results of sample Al-9 indicating that the results are not very sensitive to surface preparation. The perpendicular case, as explained in Sec. II can be related to the parallel case in a simple way if λ is quadratic in H. For this case (i.e., λ quadratic in H) the cylindrical geometry we used would give a change in surface reactance in the perpendicular case $\frac{2}{3}$ of that in the parallel case. Since experimentally we found the change to be greater than $\frac{2}{3}$ and to depend on both temperature and field, we conclude that terms of higher order than H^2 are necessary to describe the field dependence of λ. This conclusion was also clear from the curves of the three pure samples in Fig. 7.

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