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Field variation of the penetration depth in ceramic $Y_1Ba_2Cu_3O_y$

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The radio-frequency penetration depth is found to increase quadratically with applied dc magnetic field B, i.e., $\Delta \lambda_{eff} = k(T)B^2$, between 4.2 and 85 K in samples of ceramic Y₁Ba₂Cu₃O_y. The parameter k(T) rises steeply with increasing temperature T, and even at 4.2 K is orders of magnitude larger than a Ginzburg-Landau prediction. Instead, the B^2 dependence and the large values of k can be quantitatively explained in terms of the dc flux dependence of the highfrequency response of a Josephson junction network, with typical junction area about 6 μm^2 . The analysis provides an explanation of the observed correspondence between $\lambda_{eff}(T)$, k(T), and the threshold field for irreversible flux entry.

Knowledge of the penetration depth and surface impedance of the new oxide superconductors¹ is important both for the elucidation of the fundamental mechanism for superconductivity in these materials and as a guide for practical applications in high-frequency devices. It is increasingly evident that in their response to dc and highfrequency electromagnetic fields, the oxide superconductors in ceramic form display properties substantially different than those of conventional bulk superconductors. Measurements^{2,3} of the high-frequency penetration depth and surface resistance display anomalous temperature dependences and unusually high values, and it is suspected that these may not be representative of the intrinsic properties of the superconductor, but rather may be dominated by intergranular coupling complicated by the intrinsic anisotropy.⁴ In this paper, we describe the results of experiments measuring the effect of a static magnetic field on the radio-frequency penetration depth which afford a sensitive means of distinguishing between the intrinsic response of superconducting grains and the response of the intergranular Josephson coupling.

The experiments studied the effects of a static magnetic field B on the radio-frequency penetration depth λ_{eff} of ceramic $Y_1Ba_2Cu_3O_{\nu}$. In the temperature range 4.2-85 K, we find that the penetration depth increases quadratically with B, and the data can be represented as $\Delta \lambda_{\rm eff}$ $=k(t)B^2$, where k increases strongly with the reduced temperature t. The values of k are about 5 orders of magnitude larger than would be expected on the basis of a simple Ginzburg-Landau theory, which might be expected to apply to the intrinsic behavior of individual superconducting grains. Instead a model based upon the response of a Josephson network, which also yields a B^2 dependence, is quantitatively consistent with the data. The analysis also explains the observed proportionality near T_c between the zero-field $\lambda_{eff}(t)$, and the parameter k(t)which represents the field dependence, and provides clear evidence that the high-frequency dynamic response of the ceramic superconductors is dominated by intergranular coupling.

The experimental procedure has been described previously.² Disks 1-cm-diam by 1-mm-thick were placed in the tank coil of a tunnel diode oscillator whose frequency f was measured as a function of external field at various fixed temperatures. The samples were single phase, as confirmed by powder x-ray diffraction analysis, densities varied between 93 and 95% of the theoretical value, and the dc resistivities showed transition widths of less than 1 K. In all the work reported here, f was typically about 1 MHz. At fixed temperature, changes in the resonant frequency can be related to changes in the penetration depth with sensitivity of about 100 Å. While there have been several experiments which have studied the field dependence of the microwave absorption, ⁵ to our knowledge these are the first experiments on the field dependence of the penetration depth.

In earlier experiments² on the zero-field temperature dependence of λ_{eff} , we found that $\lambda_{\text{eff}}(t)$ rises extremely rapidly for reduced temperatures near T_c , and reaches values orders of magnitude beyond values for typical BCS superconductors. Yet at lower temperatures, λ_{eff} shrinks to levels compatible with the submicron values found in muon-spin-resonance⁶ measurements and elsewhere.^{3,7} The entire temperature dependence is in clear disagreement with the BCS theory, and one suspects that this is due to the granular nature of the ceramic superconductor.

In the presence of a dc magnetic field, we observe large changes of λ_{eff} even for modest fields. Typical results for $\Delta\lambda_{\text{eff}}$ are shown in Fig. 1, where the shifts in penetration depth at T=77 K are plotted against B^2 . As Fig. 1 demonstrates, λ_{eff} increases as B^2 . Samples were cooled to the measuring temperature in zero field, and the data were completely reversible provided that a small threshold field $B_1(T)$ was never exceeded. However, hysteresis was observed if B was increased further, as shown in Fig. 2. Here B_1 is about 4 G, which is consistent with many other experiments showing flux penetration at very low fields for the ceramic materials.⁸ These values are much lower than the estimated H_{c1} for single crystals of the oxide superconductors,⁹ which indicates that the hysteretic behavior is not associated with conventional (intragranular) vortices. In any event, the behavior observed for fields less than B_1 strongly suggests that we can treat the sample as if it were in a classical Meissner state, except for the details of the field distributions in the penetration layer.

Defining k(t) by $\Delta\lambda_{\text{eff}} = k(t)B^2$, we exhibit its excep-



FIG. 1. Change $\Delta \lambda_{\text{eff}}$ of penetration depth with applied dc magnetic field *B* at 74 K. The quadratic dependence was typical of data between 4.2 and 85 K, at low fields. The dashed line is a guide to the eye.

tional size and dramatic increase with temperature in the semilogarithmic plot of Fig. 3. As the figure shows, k extrapolates to about 150 Å/G² at T=0, which is roughly 200 000 times larger than is found for the conventional superconductor tin.^{10,11} The two principal features of our results, viz. the increase of λ_{eff} and its quadratic dependence on *B*, are both consistent with elementary considerations; application of *B* "pushes" the superconductor towards the normal state, depressing the order parameter and increasing λ . Also, symmetry requires that the leading term must be quadratic. From the Ginzburg-Landau equations,¹² ignoring spatial variation of the order parameter ψ , one gets $\Delta |\psi| \sim A^2 \sim B^2$. Therefore, since $\lambda \sim |\psi|^{-1}$, we have $\Delta\lambda \sim B^2$ directly. In experiments in which there are both dc and rf fields, the total vector po-



FIG. 2. "High"-field effects on the penetration depth at 85 K, displayed as $\Delta \lambda_{\text{eff}}$ vs *B*. Note the hysteresis due to flux inclusion at modest field values. The dashed lines are guides to the eye.

tential is $\mathbf{A} = \mathbf{A}_{dc} + \mathbf{A}_{rf}$, with $A_{rf} \ll A_{dc}$. For quadratic variations of the order parameter $|\psi| = |\psi_0| (1 - \alpha A^2)$, the rf part of the supercurrent is given by

$$\mathbf{J}_{\rm rf} = -\frac{c}{4\pi\lambda^2} [\mathbf{A}_{\rm rf} - \alpha A_{\rm dc}^2 \mathbf{A}_{\rm rf} - 2\alpha (\mathbf{A}_{\rm dc} \cdot \mathbf{A}_{\rm rf}) \mathbf{A}_{\rm dc}].$$

In the present case, the fields are parallel, and taking α from standard Ginzburg-Landau theory one finds

$$k_{\rm GL}(t) = \frac{\lambda(t)}{6H_c(t)^2} = \frac{\lambda(0)}{6(1-t^2)(1-t^4)^{1/2}},$$
 (1)

where the second form invokes the conventional Gorter-Casimir extension to low temperatures. This expression gives $k_{GL}(0) = 7 \times 10^{-4} \text{ Å/G}^2$ for the elemental BCS superconductor tin, and both the magnitude and character of the field dependence are in reasonable agreement with the observed behavior.^{10,11}

In the case of the oxide superconductors, $\lambda(0)$ is about 0.5 μ m (Refs. 6 and 7) and the thermodynamic critical field $H_c(0)$ is estimated to be roughly 10³ G.¹³ Therefore k_{GL} is about 10⁻³ Å/G², i.e., many orders-of-magnitude below the values shown in Fig. 3. It is unlikely that inclusion of further details of the microscopic mechanisms, ¹¹ such as the density of states, or of nonequilibrium effects, will reconcile the orders-of-magnitude discrepancy between the present data and the simple Ginzburg-Landau treatment.

The failure of this approach, in which the quadratic behavior arises from the intrinsic properties of the superconductor, suggests that the observed result is not due to the individual grains. Rather, as many workers have proposed, the ceramic must be viewed as a composite of many grains, whose response is substantially different from the intrinsic behavior. This suggests a simple picture which incorporates the various experimental results: An array of grains coupled by small Josephson junctions, in which the local rf current is driven by the intergranular rf electric field, modulated by the dc flux in the junction. It is as if



FIG. 3. Temperature dependence of the parameter k(t) vs $t = T/T_c$, where $\Delta \lambda_{\text{eff}} = k(t)B^2$. The inset displays both k(t) and $\lambda_{\text{eff}}(t)$ vs t, and shows the close proportionality between k (squares) and λ_{eff} (stars). The lines are guides to the eye.

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the sample behaves as a "large Josephson junction," with the usual Josephson penetration depth playing the role of λ_{eff} .

A measure of justification for this idea is provided by examining the expression¹² for the current through a single small junction of thickness d and area w^2

$$I_{\rm rf} = 2J_c w^2 \frac{\sin(\pi \Phi/\Phi_0) \sin\phi}{\pi \Phi/\Phi_0} \,. \tag{2}$$

Here ϕ is the phase difference in the absence of the dc flux, and it represents the currents driven by the intergrain rf electric field E_{rf} . For $E_{rf}(z,t) = E(z) \exp(-i\omega t)$, $\phi = \phi_0 - 2edE_{rf}/i\hbar\omega$, and since E_{rf} is small we expand $\sin\phi$ to get

$$I_{\rm rf} = -\frac{2w^2 J_c e d}{i\hbar\omega} \cos\phi_0 \left(\frac{\sin\pi\Phi/\Phi_0}{\pi\Phi/\Phi_0}\right) E_{\rm rf} \,. \tag{3}$$

If λ_{eff} is much larger than the junction size, then locally averaged quantities can be defined which obey a pseudo-London relation $\mathbf{J}_{\text{rf}}(z) = -(c/i\omega)\sigma(z)\mathbf{A}_{\text{rf}}(z)$ where σ (and consequently λ_{eff}) depends on the dc magnetic field B(z) via the Josephson factor $\sin(\pi\Phi/\Phi_0)/(\pi\Phi/\Phi_0)$. The resulting electrodynamics is very complicated, and beyond the scope of this communication. However, the weak-field case can be solved exactly under the assumption that spatial dependence is the same for the dc and (1 MHz) rf fields. Combining the above equation with Maxwell's equations leads directly to an equation of the form $d^2A/dz^2 = a_1A - a_3A^3$, from which one gets the slope at the sample surface by simple integration. Substituting values for a_1 and a_3 , one finds¹⁴

$$\lambda_{\text{eff}} = \lambda_{\text{eff}}(B=0) \left[1 + \frac{1}{6} \left(\frac{\pi SB_{\text{dc}}}{2\Phi_0} \right)^2 + \cdots \right], \qquad (4)$$

where S is the effective junction area and $\lambda_{\text{eff}}(B=0) \sim J_c^{-1/2}$. The model is highly simplified, of course, particularly in its neglect of interference of currents flowing in neighboring junctions and the substitution of typical values for average ones. Nevertheless it should serve as a reasonable projection for the outcome of a proper theory. According to this picture, the effective penetration depth will be sensitively dependent on B as a consequence of the Josephson factor, with $k(t) = \lambda_{\text{eff}}(t)(\pi S/2\sqrt{6}\Phi_0)^2$.

Furthermore, both k and λ_{eff} should be very large, due to the high tunneling resistances (small J_c 's) and small filling factor of the junctions; and the temperature depen-

dence of k will be a mixture of the dependences of both J_c and λ_{intr} (the intrinsic penetration depth of the grains) on t. This last feature reflects the fact that the effective junction thickness $d = d_0 + 2\lambda_{intr}(t)$, where d_0 is the geometric thickness. If we assume that λ_{intr} takes the BCS form, then its effect will be observable only extremely close to t=1. Therefore, we should expect to find that k and λ_{eff} are essentially proportional except at very high reduced temperatures, where k will rise more steeply. While the data are limited, these are just the features exhibited in the inset to Fig. 3.

Taking k(0) from Fig. 3 and assuming $\lambda_{eff}(0)$ $\approx \lambda_{intr}(0) \approx 0.5 \ \mu m$, we find $S = 6 \ \mu m^2$ or that the length $L \approx S/2\lambda_{\text{intr}} = 6 \ \mu \text{m}$ for a typical junction. This result is quite compatible with the 10-20 μ m observed grain size, and in agreement with the findings of Blazey et al. for finer-grained samples.⁵ This picture is also consistent with the modest fields at which flux entry occurs. In a network of superconducting current loops, once a link is driven normal by the dc field, flux conservation tends to prevent restoration of the original loop topology when the field is subsequently reduced. The onset of hysteresis occurs when $\Phi = \Phi_0$ at the weakest junctions near the surface of the sample, notwithstanding the smearing of the fine structure of the Josephson pattern. Taking S from above we find $B_1 = \phi_0/S = 3.3$ G, which is consistent with the experimental results. Once flux entry occurs, the rf response is dominated by the motion of these trapped current loops or "vortices," which are presumably pinned to the intergranular regions.⁴ The higher-field results (Fig. 2) probe the dynamics of these "vortices," and further studies of this region are currently underway.

In conclusion, we note that the analysis presented here is able to explain the large magnitudes of the effects observed in the experiments, and in addition it provides an explanation of the relation between k(t) and $\lambda_{\text{eff}}(t)$. Further, the analysis shows how the same junction dimensions enter into the observed quadratic dependence and the threshold field B_1 for irreversible flux entry. Thus the Josephson array picture is able to correlate the zero-field response represented by λ_{eff} , the field dependence embodied in the parameter k, and the critical field B_1 .

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¹⁴The complete solution is

$$A(z) = \gamma^{-1/2} \left(1 - \frac{\kappa_0 - \exp(-2z/a_1^{1/2})}{\kappa_0 + \exp(-2z/a_1^{1/2})} \right)^{1/2}$$

where $\gamma = a_3/2a_1$ and

$$\kappa_0 = [1 + (1 - \gamma A_0^2)^{1/2}] / [1 - (1 - \gamma A_0^2)^{1/2}]$$