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Muon-spin-rotation measurement of magnetic field penetration and flux pinning in superconducting $EuBa_2Cu_3O_x$

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Muon-spin-rotation measurements have been used to determine the magnetic field penetration depth λ as a function of temperature T in superconducting, sintered $EuBa_2Cu_3O_x$ ($x \approx 7$). The extrapolated value at 0 K, $\lambda(0)$, is 1700 Å. The temperature dependence of $\lambda(T)$ in the interval 3.5 K to T_c is well described by the phenomenological relation $\lambda(T) = \lambda(0)[1-(T/T_c)^4]^{-1/2}$. For $T < T_c$, approximately 85% of the muons stop within the superconducting grains, whereas $\approx 15\%$ reside in normal regions, perhaps at grain boundaries. At $T = 3.5$ K the field-cooled superconductor has expelled only 2.3% of the applied magnetic field, indicating strong flux pinning in the material.

The discovery of superconductivity in oxygen-deficient perovskites[1-4] has generated considerable interest among scientists. Much of this excitement is due to the important technological applications envisioned for superconductors with such a high transition temperature (~90 K). Although much experimental and theoretical research has been devoted to these materials, no clear understanding of the superconducting mechanism has yet emerged. Measurements of physical properties associated with these unusual superconductors are, therefore, necessary. We present muon-spin-rotation results for polycrystalline $EuBa_2Cu_3O_x$ ($x \approx 7$) in the temperature interval 3.5 to 300 K, and extract from the data information on magnetic field penetration and flux trapping.

A sintered ceramic pellet was prepared by the method described in Ref. 5. Briefly, the starting powder was packed loosely and calcined in O_2 for 12 h at 980 °C. It was then pressed and fired at 980 °C for 10 h in flowing O_2, cooled to 400 °C in 8 h, and removed from the oven. The final dimensions of the pill-box-shaped pellet are 2.1 cm in diameter and 0.39 cm in height. With a mass of 7.22 g, the computed density is 5.31 $\mathrm{g\,cm^{-3}}$, yielding a packing fraction of 0.77 (assuming a theoretical density of 6.90 $\mathrm{g\,cm^{-3}}$). Magnetic-susceptibility measurements at 100 Oe on a small portion of this sample indicated 90% shielding and a 31% Meissner effect (at 7 K) with an onset T_c near 95 K. No corrections were made for demagnetization, estimated to be less than a few percent. As determined by magnetization measurements (applied field of 4 kOe), this sample exhibits weakly temperature-dependent paramagnetism in the temperature interval T_c to 300 K. Additional characterization of the sample was done by measuring the conductivity as a function of temperature in an ac-coil apparatus;[6] a sharp superconducting transition has been observed at 93 K by this method. Rutherford backscattering and x-ray diffraction also indicate that the sample is a high-quality, single-phase ceramic specimen.

Standard time-differential muon-spin-rotation (μSR) techniques were used in this study.[7] Polarized positive muons are implanted into the sample one at a time, and the time difference between the implantation and detection of the emitted positron is recorded. Since the muon decays by weak β decay, the positrons are emitted preferentially along the direction of the muon polarization. The number of positrons emitted as a function of angle and time is given by

$$N(\nu,\phi,t) = N_0 \exp(-t/\tau_\mu)[1 + AG_x(t)\cos(\nu t + \phi)] , \quad (1)$$

where τ_μ is the muon lifetime (2.2 μsec), A is the asymmetry, $G_x(t)$ is the envelope of the oscillations, or the transverse depolarization function, and ν is the mean precessing angular frequency of the muon (13.55 MHz $\mathrm{kOe^{-1}}$). For a Gaussian distribution of static, random, local magnetic fields, $G_x(t)$ is given by $\exp[-(\Lambda^2 t^2)/2]$. The quantities of interest are asymmetry A; muon depolarization rate Λ, which is a measure of the width of the local magnetic field distribution at the muon site; and ν, which is proportional to the magnitude of the average local magnetic field.

Shown in Fig. 1 are the muon depolarization rates Λ as a function of temperature for $EuBa_2Cu_3O_x$ taken in a 1-kOe transverse field. Transverse means that the applied

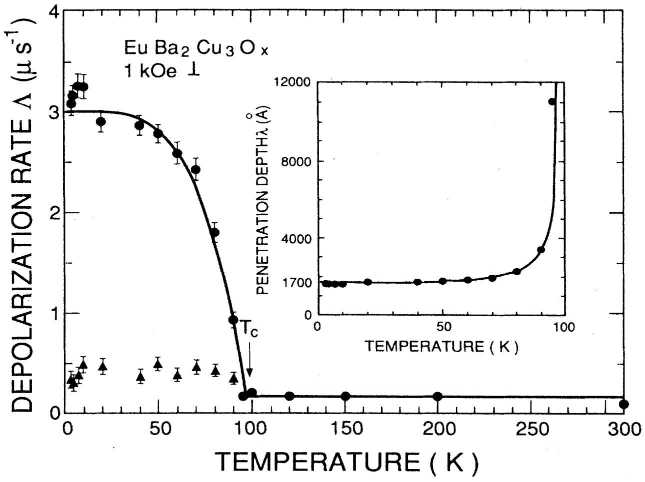

FIG. 1. Temperature dependence of muon Gaussian depolarization rates in $EuBa_2Cu_3O_x$ taken in a 1-kOe transverse field. The triangles and circles correspond to the muon precessional angular frequencies and asymmetries identified with the same symbols in Figs. 2 and 3. See text for a description of the fit to the data. Inset depicts the temperature dependence of the magnetic field penetration depth.

field is perpendicular to the initial muon-spin direction. All data presented here were obtained after field cooling the sample in the measuring field (or in 1 kOe). The μSR data clearly show the existence of two signals for $T < T_c$, but only one signal above T_c. For $T > T_c$, a constant depolarization rate of 0.16 $\mu\mathrm{sec}^{-1}$ is observed. This is approximately the value (~ 0.1 $\mu\mathrm{sec}^{-1}$) observed in $YBa_2Cu_3O_x$ (Ref. 8) and $La_{1.85}Sr_{0.15}CuO_4$ (Ref. 9), and attributed to static dipole moments. The occurrence of two μSR relaxation rates below T_c, taken together with their corresponding frequencies (see Fig. 2), indicates that muons reside at regions with distinctly different local environments. One is associated with normal conducting regions of the sample and is characterized by a nearly constant (~ 0.3–0.5 $\mu\mathrm{sec}^{-1}$) rate for all temperatures down to 3.5 K (see filled triangles of Fig. 1). A majority of the muons stop in those regions corresponding to the superconducting vortex state. In this case, the muons experience a rather inhomogeneous magnetic field distribution resulting in an increased depolarization rate (filled circles of Fig. 1), and a decreased precessional frequency (filled circles of Fig. 2).

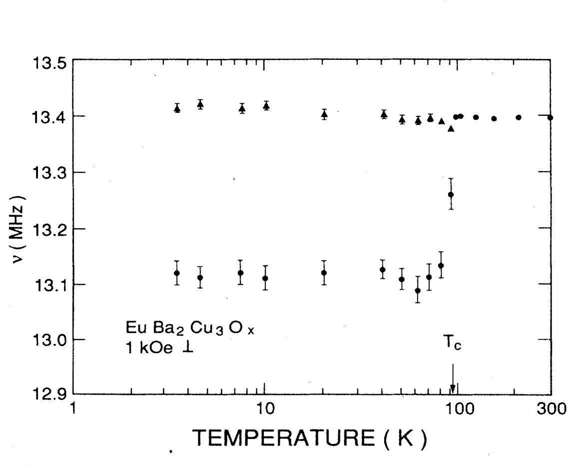

FIG. 2. Temperature dependence of positive muon-spin precessional angular frequencies in $EuBa_2Cu_3O_x$ taken in a 1-kOe transverse field. The corresponding depolarization rates are shown by the same symbols in Fig. 1.

A Gaussian depolarization function described by $P(t) = \exp[-(\Lambda^2 t^2)/2]$ yields the best fit to our data. Assuming a square Abrikosov lattice for the vortex state ($H_{c1} < H_{\rm app} < H_{c2}$), the magnetic field inhomogeneity is given by[10]

$$\langle |\Delta H|^2 \rangle = [(H_{\rm app}\phi)/(4\pi\lambda^2)][1 + (4\pi^2\lambda^2 H_{\rm app})/\phi]^{-1} = \Lambda^2/\gamma_\mu^2 , \qquad (2)$$

where Λ is the muon depolarization rate, γ_μ is the muon gyromagnetic ratio ($2\pi \times 13.55$ MHz kOe^{-1}), λ is the magnetic field penetration depth, and ϕ is the flux quantum (2×10^{-7} Oe cm^2). Thus, the magnetic field penetration depth can be determined from an experimental measurement of the muon-spin depolarization rate. This assumes of course that $EuBa_2Cu_3O_x$ is a usual type-II superconductor. By extrapolating Λ to 0 K and subtracting the constant relaxation rate 0.16 $\mu\mathrm{sec}^{-1}$, we find $\Lambda(0)$ to be 2.84 $\mu\mathrm{sec}^{-1}$. Substituting this value into Eq. (2) yields the magnetic field penetration depth at 0 K, $\lambda(0) = 1700$ Å. The temperature dependence of the penetration depth is usually represented by the empirical relation[11] $\lambda(T) = \lambda(0)[1-(T/T_c)^4]^{-1/2}$. Substituting this expression with $\lambda(0) = 1700$ Å into Eq. (2) and using our experimental values of $\Lambda(T)$, the penetration depths, as shown by the filled circles in the inset of Fig. 1, are obtained. The solid line in the inset is the temperature-dependent penetration depth calculated from $\lambda(T) = \lambda(0)[1-(T/T_c)^4]^{-1/2}$, with $\lambda(0) = 1700$ Å and a best-fit value of $T_c = 97$ K.

By substituting the empirical expression into Eq. (2), one finds that, to a first approximation, the vortex-state relaxation can be described by $\Lambda(T) \propto [1-(T/T_c)^4]$. Thus, an attempt has been made to fit the muon depolarization data to an expression of the form $\Lambda(T) = a[1-(T/T_c)^4] + b$. The solid line in Fig. 1 is the result of this fit. For $T > T_c$, $\Lambda(T) = 0.16$; and for $T < T_c$, $\Lambda(T) = 2.85[1-(T/T_c)^4] + 0.16$, with T in K, Λ in $\mu\mathrm{sec}^{-1}$, and $T_c = 97$ K.

Shown in Figs. 2 and 3 are the muon precessional frequencies and asymmetries for $EuBa_2Cu_3O_x$ taken in a 1-kOe transverse field. The filled triangles and circles correspond to the muon depolarization rates identified by the same symbols in Fig. 1. Surprisingly, the muon precessional frequency associated with the vortex state (filled circles of Fig. 2) does not decrease with decreasing temperature as expected; it does, however, show a sharp drop at T_c. In fact, for $3.5\ \mathrm{K} < T < T_c$, this frequency is nearly constant with $\nu = 13.1$ MHz, implying a local magnetic field of 966 Oe. Thus, the sample expels very little flux when field cooled in 1 kOe. Measurements of the Meissner fraction for single-crystal $YBa_2Cu_3O_x$ in a 1-kOe field show also that practically no flux is expelled.[12] Of course, in the absence of such strong flux pinning, a significant de-

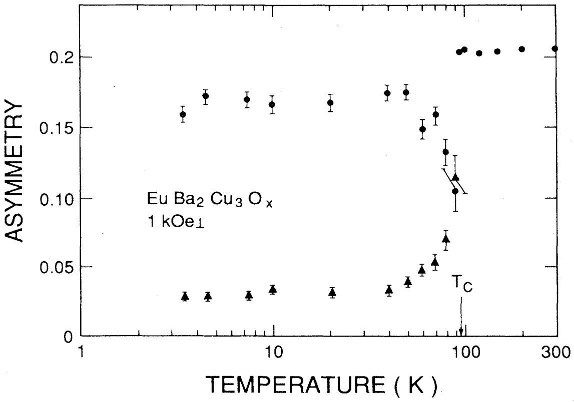

FIG. 3. Temperature dependence of positive muon asymmetries in $EuBa_2Cu_3O_x$ taken in a 1-kOe transverse field. The corresponding precessional angular frequencies and relaxation rates are shown by the same symbols in Figs. 1 and 2.

crease in ν as T decreases should be seen. From this result we conclude that strong flux pinning is indeed the case in our sample. Note also that ν associated with the normal state is approximately constant for all temperatures; the value is 13.4 MHz and corresponds to a field of 989 Oe. Due to demagnetizing effects, this local field is less than the applied field of 1 kOe. Nevertheless, these results indicate that only a small fraction of the magnetic field is excluded from the muon site corresponding to the superconducting region. From our susceptibility measurements we find that in a 100-Oe field 31% of the flux is expelled (Meissner effect), a value much larger than observed in our 1-kOe μSR data (2.3%). However, in a 1-kOe field the Meissner experiment shows that only about 7% of the external field is expelled, which is qualitatively consistent with our μSR measurements.

Our recent μSR results[13] on single-phase $GdBa_2Cu_3O_x$ also show the existence of two distinct muon stopping regions, one normal and the other superconducting. In contrast to our $EuBa_2Cu_3O_x$ data where the muon precessional angular frequency is approximately constant, the $GdBa_2Cu_3O_x$ data show a smooth decrease in frequency with decreasing T. One expects this behavior because the magnitude of flux expulsion should be temperature dependent, with more flux being expelled at lower temperature. Because this behavior is not observed in $EuBa_2Cu_3O_x$ we conclude that the flux-pinning forces are greater in $EuBa_2Cu_3O_x$ than in $GdBa_2Cu_3O_x$. This assumes that the muon stopping sites, which have not yet been accurately determined, are the same in these very similar materials.

As shown in Fig. 3, the muon asymmetry for $T > T_c$ is nearly constant with $A = 0.205$. For $T < T_c$ two asymmetries are seen which correspond to muons stopping in normal and superconducting regions of the material. At temperatures much below T_c the approximately constant asymmetry values are 0.03 and 0.17, respectively. Note, however, that no asymmetry is lost, i.e., the sum of the asymmetries associated with the normal and superconducting regions ($T < T_c$) is equal to the initial asymmetry ($T > T_c$). This implies that $\approx 15\%$ of the muons stop in normal conducting regions and $\approx 85\%$ come to rest in superconducting volumes of the sample.

A tentative explanation for the occurrence of these two regions is the following. First we note that the Fourier power associated with the normal and superconducting fractions is distributed such that $\approx 15\%$ of the signal is attributable to muons stopping in normal regions of the sample and $\approx 85\%$ due to muons located in superconducting volumes, in agreement with the asymmetry results. Thus, we suggest that ≈ 15 % of the stopped muons reside at grain boundaries where the local magnetic field is approximately equal to the applied magnetic field, 1 kOe in the present case, and that $\approx 85\%$ of the muons reside within the superconducting grains where they experience a reduced local magnetic field due to flux expulsion, albeit weak. Because the muons do not diffuse at these temperatures, many more are expected to stop within the grains ($\approx 85\%$) than at the boundaries ($\approx 15\%$). As the superconducting state is entered some small fraction of the magnetic field is expelled from the grains, thus, enhancing the field at the grain boundaries. Since only 2.3% of the flux is expelled and distributed around the perimeter of a typical grain boundary (~ 10 μm), no appreciable shift in the muon precessional frequency associated with the normal regions is expected. As shown in Fig. 2 (triangles), there is only a hint of an increase in the frequency for $T < T_c$.

Summarizing, we conclude that two distinct muon stopping regions exist in $EuBa_2Cu_3O_x$. One corresponds to normal conducting volumes of the sample, which we tentatively associate with grain boundaries, and the other corresponds to superconducting regions, which are just the $EuBa_2Cu_3O_x$ grains. Only 2.3% of the external magnetic field is expelled from the superconducting volume at 3.5 K when field cooled in a 1-kOe field. The magnetic field penetration depth extrapolated to 0 K is 1700 Å, with a temperature dependence well described by the phenomenological relation $\lambda(T) = \lambda(0)[1-(T/T_c)^4]^{-1/2}$.

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