

Direct Observation of a Flux Line Lattice Field Distribution across an $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ surface by Low Energy Muons

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The technique of low energy muon spin rotation has been used to measure the microscopic field distribution in the vortex state of a 700 nm thick $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ film. By varying the implantation energy of the muons we were able to monitor the spatial evolution of the magnetic field distribution as the flux lines emerge through the surface of the superconducting film. The results are in excellent agreement with calculations based on the London model and clearly demonstrate the potential of the new technique for thin film and surface studies.

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Because of the great sensitivity of the positive muon as a microscopic magnetic probe the technique of muon spin rotation or relaxation (μSR) is a powerful tool for studying the internal distribution of magnetic fields within solids. In the context of high temperature superconducting cuprates, μSR experiments have provided important contributions to a better understanding of the physics of the vortex state [1,2], superfluid density [3,4], and the complex interplay between magnetism and superconductivity [5]. So far, a major drawback of the technique was its limitation to bulk studies of matter due to the high kinetic energy of the positive muon (4.2 MeV). This large energy scale is a consequence of the kinetics of the pion decay, from which the muons originate, and results in implantation depths on the order of several hundred μm .

The recent development of a beam of polarized low energy μ^+ [6] allows the extension of the μSR technique to thin film studies (low energy μSR or LE- μSR). The beam is based on the moderation of an intense beam of 4.2 MeV muons in a condensed van der Waals gas layer, such as Ne, Ar, or N_2 [7,8]. Specific properties of these solids tend to suppress the energy loss mechanisms responsible for the thermalization of the muon, resulting in a high probability for the emission of epithermal muons with a kinetic energy of about 15 eV [9]. The conservation of the spin polarization during the moderation process [6] opens the possibility to use these epithermal muons as a source of a beam of tunable energy between 15 eV and 30 keV. The corresponding implantation depth of these muons in solids ranges from a fraction of a nm up to some hundred nm.

In this paper we report the first LE- μSR measurements on high temperature superconductor films. From studies of the microscopic field distribution within the flux line lattice in an $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ film, we were able to derive the magnetic penetration depth. By varying the energy of the

slow muon beam we were able to adjust the implantation depth into the $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ film and thus to measure the evolution of the field distribution on a nanometer scale as the vortex lines emerge from the surface of the film. To access the region outside the superconductor a second $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ film was grown under exactly the same conditions and covered *ex situ* with a 70 nm thick Ag layer. By stopping the muons within this Ag layer we are also able to monitor the spatial evolution of the field distribution outside the superconductor.

The measurements were performed on 700 nm thick, *c*-axis oriented, epitaxial $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ films grown by thermal coevaporation on LaAlO_3 (2 in. diameter) substrates [10]. The quality of the films can be considered homogeneous over the whole film area [11]. The films exhibited sharp transitions into the superconducting state at $T_c = 87.5$ K and critical current densities $j_c(77\text{ K}) > 10^6$ A/cm².

The LE- μSR experiments were performed at the $\pi\text{E}3$ beam line at the Paul Scherrer Institut in Villigen, Switzerland by using the LE- μ^+ apparatus described in detail in Ref. [8]. The epithermal muons are extracted from the moderator region by applying a high voltage of up to 20 kV to the electrically insulated substrate and are transported by electrostatic lenses and a mirror to the sample. The final kinetic energy of the muons can be varied by applying an accelerating or decelerating potential of up to 12.5 kV to the film, which is mounted via a sapphire plate on a continuous-flow helium cryostat. In the experiment a magnetic field is applied perpendicular to the initial muon spin polarization (transverse field experiment) and parallel to the *c* axis of the $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ film.

In a magnetic field, $\text{YBa}_2\text{Cu}_3\text{O}_7$ behaves as an extreme type-II superconductor with vortex cores of diameter $\xi_{\text{ab}} \approx 2$ nm, and with the magnetic field varying spatially on the scale of the penetration depth $\lambda_{\text{ab}} \approx 150$ nm.

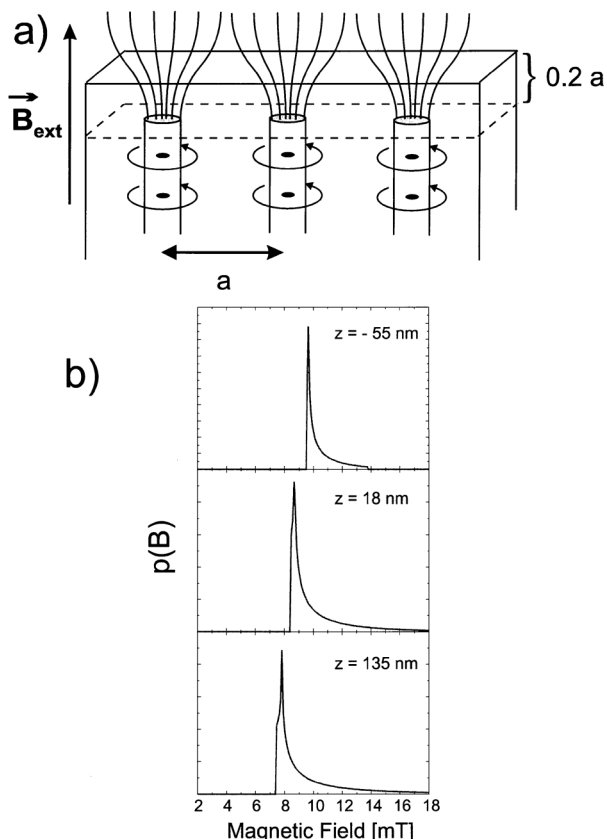


FIG. 1. (a) Sketch of the principle of the experiment, showing how the flux lines emerge through the surface of the superconducting film. (b) Simulated field distributions for $\lambda_{ab} = 150$ nm and $B_{ext} = 10.4$ mT as a function of the implantation depth z into the $YBa_2Cu_3O_{7-\delta}$ film. The negative z value corresponds to a field distribution outside the superconducting film.

Figure 1(a) shows schematically how the vortex lines emerge from the bulk across the surface of a $YBa_2Cu_3O_{7-\delta}$ film. Inside the film the vortex cores are separated by a distance $a = \sqrt{(2/\sqrt{3})(\Phi_0/B)} = 1546 \text{ nm}/\sqrt{B_{ext} [\text{mT}]}$, where Φ_0 is the flux quantum. The field distribution near the surface of the superconductor is obtained by a development of the simplified treatment given by [12]. We take the surface of the superconductor as the plane $z = 0$; inside the superconductor ($z > 0$), we assume that the London equation is a sufficiently accurate description of the spatial variation of $B_z(x, y, z)$, with source terms representing the flux line cores, whereas on the outside B_z obeys Laplace's equation,

$$-\nabla^2 B_z - \frac{\partial^2 B_z}{\partial z^2} + \frac{B_z}{\lambda^2} \Theta(z) = \frac{\Theta_0}{\lambda^2} \Theta(z) \sum_{\mathbf{R}} \delta(\mathbf{r} - \mathbf{R}).$$

Here, \mathbf{r} is a two-dimensional vector in the xy plane, \mathbf{R} are the vortex positions, ∇^2 is the two-dimensional Laplacian, and Θ is a step function, unity for $z > 0$ and zero for $z < 0$. Currents flow only in the xy plane, so the penetration depth λ is λ_{ab} for our geometry. Fourier

transforming in the xy plane, we obtain an equation for the components of the field $B_z(\mathbf{k}, z)$, which are only nonzero for \mathbf{k} equal to a reciprocal lattice vector of the flux lattice. Solutions are easily obtained for $z > 0$ or $z < 0$, and after matching at the boundary and to the known situation at infinity, we obtain

$$B_z(\mathbf{k}, z) = \frac{B_0}{\lambda^2} \frac{\Theta(-z)}{\Lambda(\Lambda + k)} e^{kz} + \frac{B_0}{\lambda^2} \frac{\Theta(z)}{\Lambda^2} \left[1 - \frac{k}{\Lambda + k} e^{-\Lambda z} \right],$$

where $\Lambda^2 = k^2 + 1/\lambda^2$, and B_0 is the average field.

The x and y components of B may be obtained from the z components for each \mathbf{k} , using Maxwell's equations, and bearing in mind that currents flow only in the xy plane. Performing the Fourier sum at any z , we may calculate $\mathbf{B}(x, y; z)$. It is of interest to note that the z variation of all Fourier components is controlled by Λ or k , which have values always equal to or larger than $2\pi/a_0$, where a_0 is the flux lattice plane spacing. Hence the deviations of the field distribution from uniform (outside) or bulk flux lattice (inside) die away to zero over the short distance $\sim a_0/2\pi$ from the boundary at $z = 0$.

The field variation $B(x, y; z)$ within the triangular vortex lattice produces a distinctive asymmetric field distribution $p(B)$. It shows a pronounced tail towards high fields arising from regions within the vortex lattice close to the vortex cores, a cusp which corresponds to the most probable field B_{sad} at the saddle point between adjacent vortices, and a cutoff on the low field side corresponding to the field minimum at the point which is most remote from the vortex cores. This can be clearly seen from Fig. 1(b) in which we display the calculated field distributions for $z = -55$ nm, 18 nm, and 135 nm, respectively. As the flux lines emerge towards and through the surface of the $YBa_2Cu_3O_{7-\delta}$ film, B_{sad} moves towards the applied field, and the overall width of the field distribution diminishes. On the outside of the superconducting film the field distribution is still asymmetric but the tail towards high fields is drastically reduced.

For the measurement of the field distribution, the $YBa_2Cu_3O_{7-\delta}$ film is cooled to $T = 20$ K in an external field at 10.4 mT. A muon, implanted at a certain (x, y) and a depth z in the sample, precesses about the local magnetic field at a rate determined by the magnitude of $B(x, y)$ at this z . Hence the probability distribution of muon precession frequencies at z , which is extracted from the time evolution of the muon spin polarization $P(t)$ via maximum entropy techniques [13,14], reflects the probability distribution of $B(x, y)$.

It should be noted that the *amplitude* of the precessing component depends on the angle between the spin of the incoming muon and the local field. In a bulk superconductor, under our conditions, the local field, although varying with x and y , is always in the z direction

and hence perpendicular to the spin of the incoming muon. However, as we see in Fig. 1(a), near the surface, the lines of field splay out. We have corrected for this effect, but, in fact, it is always small under the range of conditions of our experiments.

For the proper interpretation of our measurements the knowledge of the range and range distribution of the low energy muons in matter is necessary. No experimental data are available, and we calculated the implantation profile by using a Monte Carlo code which simulates the transport of particles in matter (TRIM.SP, transport of ions in matter [15]). We assessed the reliability of these simulations in a separate experiment in which we investigated various M/SiO_2 samples (M = thin metal layer of Al, Cu, or Au). By implanting μ^+ with different energies the implantation depth can be continuously varied through the metal/ SiO_2 interface. In a transverse magnetic field μSR measurement the fraction of μ^+ stopping in the metal layer can easily be distinguished from the fraction stopping in SiO_2 , where nearly all μ^+ form muonium which precesses about 100 times faster. We find good agreement between the measured fractions of μ^+ stopped in the metal layers and the theoretical predictions [16]. These results indicate that on the basis of the TRIM.SP code, reliable values for the implantation profile of μ^+ in matter can be obtained.

In Fig. 2 we display three representative field distributions, which were obtained by implanting muons with energies of 29 and 3 keV into the uncovered $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ film and with an energy of 3 keV into the Ag covered film. According to the TRIM.SP simulations, muons with these energies come to rest in a mean depth of 135 and 18 nm below the $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ surface in case of the uncovered film and stop 55 nm in front of the superconductor film within the Ag layer in case of the Ag covered film. The displayed spectra are background corrected. In the original data an additional Gaussian distribution occurs at the external field value of 10.4 mT. This is due to a fraction of the muons (about 10%) stopping in an electrostatic lens placed in front of the sample used to focus the beam.

The line shapes show appreciable smearing relative to the theoretical line shapes displayed in Fig. 1(b). This is due to (a) range distribution of the muons, (b) deviations of the flux line lattice from its ideal configuration as a perfect triangular array (dislocations, microscopic disorder), (c) effects of nuclear dipolar fields, and (d) intrinsic resolution of the maximum entropy technique due to the time window selected and the statistics of the data. The range distribution of the muons (a) is taken into account by folding calculated field distributions $p(B)$ for different implantation depth z with the stopping distribution obtained from the TRIM.SP simulations. The resulting field distribution is then convoluted with a Gaussian to represent the effects of (b), (c), and (d). From the analysis of these line shapes we obtain $\lambda_{ab} = 145(5)$ nm

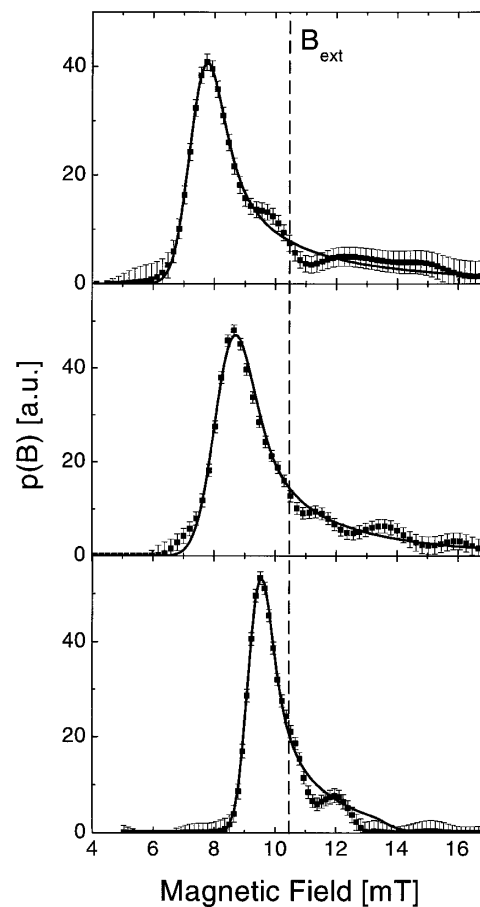


FIG. 2. The measured field distribution at $T = 20$ K and $B_{\text{ext}} = 10.4$ mT at distances of $z = 135$ and $z = 18$ nm below and $z = -55$ nm in front of the surface of the $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ film. As the flux lines emerge through the surface of the film both the cusp shift and the tail on the high field side are drastically reduced.

for the London penetration depth. This value of the penetration depth is in good agreement with the results from microwave transmission experiments [17] and mutual inductance techniques [18] on $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ thin films as well as muon spin rotation measurements on $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ single crystals [19].

The penetration depth can also be obtained from integral quantities such as the second moment of the field distribution or from the difference between the mean field B_0 and the most probable field B_{sad} of the field distribution [20]. In case of the limited statistics of our experiment the second quantity is more appropriate,

$$B_0 - B_{\text{sad}} \propto \frac{\Phi_0}{\lambda_{ab}^2}.$$

With the field applied perpendicular to the thin film above T_c , demagnetizing effects ensure that the mean field is equal to the applied field. Hence, the London penetration depth can thus be very reliably determined from the shift between the applied field and the cusp of the field distribution (so called cusp shift).

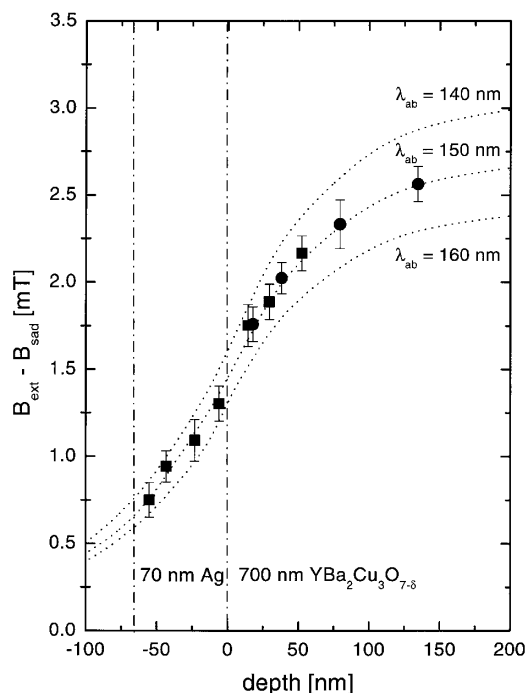


FIG. 3. Depth dependence of $B_{\text{ext}} - B_{\text{sad}}$ obtained at $T = 20$ K and $B_{\text{ext}} = 10.4$ mT. The dashed curves are predictions of the London model for different values of the London penetration depth λ_{ab} and $B_{\text{ext}} = 10.4$ mT. Excellent agreement between the experimental and calculated values is obtained for $\lambda_{\text{ab}} \approx 150$ nm.

In Fig. 3 we plot the measured depth dependence of the cusp shift $B_{\text{ext}} - B_{\text{sad}}$ and compare it to the results of calculations for three different values of the London penetration depth λ_{ab} of 140, 150, and 160 nm (dashed lines). One can see from Fig. 3 that λ_{ab} controls the magnitude of the cusp shift, but has very little effect on the depth dependence. Excellent agreement between the measured and calculated cusp shift is obtained for a penetration depth of about 150 nm, in good agreement with the value determined from the exact analysis of the line shape.

In summary, we have used for the first time low energy μ^+ as a microscopic probe to study the magnetic field distribution in the vortex state of a $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ film. By varying the energy of the muons we were able to measure the evolution of the field distribution as the flux lines emerge through the surface of the superconducting film. The results are in excellent agreement with calculations based on the London model and clearly demonstrate the potential of the LE- μ SR technique for investigations of the thin film properties with a depth resolution in the order of a few nanometers. We look forward to further measurements in samples such as multilayered high- T_c films, which offer the possibility to study the rich phenomenology of the vortex matter in a system with tunable anisotropy.

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