Field Induced 3D to 2D Crossover of Shielding Current Path in $Bi_2Sr_2CaCu_2O_x$

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(Received 22 March 1993)

Magnetization curves of high quality $Bi_2Sr_2CaCu_2O_x$ single crystals in fields slightly misaligned with the *ab* plane show a peak at small fields (6 Oe at 5 K for misalignment angle of 6°). This is attributed to flux penetration into the Josephson coupling between the CuO_2 layers and a resultant three- to two-dimensional crossover of the shielding current path with the increase in the field. A surface barrier model for the interlayer flux penetration yields the anisotropy ratio γ of ≈ 700.

PACS numbers: 74.25.Ha, 74.60.Ge

The layered crystal structure of high T_c superconductors is considered to be responsible for their anisotropic properties. This is especially so for $Bi_2Sr_2CaCu_2O_x$ (Bi-2212) where the coherence length in the c direction ξ_c has been reported to be much shorter than the distance between the superconducting CuO_2 double layers [1]. Therefore, Bi-2212 has been modeled as a stack of superconducting sheets coupled by Josephson interaction [2]. This idea has been supported by several theoretical and experimental studies [3–5]. The interlayer weak coupling is believed to determine most of the superconducting properties. Nevertheless, there have been only a few reports on the direct detection of interlayer weak coupling [6].

Magnetization measurement is important not only to determine the basic superconducting parameters and their anisotropy, but also to obtain direct information on flux penetration into the weak-coupling regions. To date, however, the magnetization and other measurements show a significant discrepancy with the anisotropic properties of Bi-2212. The magnetic anisotropy ratio, $\gamma = \lambda_c/\lambda_{ab}$ [λ_{ab} and λ_c are the penetration depths for the screening currents parallel and perpendicular to the CuO_2 (*ab*) planes, respectively], has been found to be as large as 150 or more using magnetic torque measurements [4] or 710 from ac susceptibility [7]. On the other hand, several authors [8,9] have reported γ values of less than 10 from dc magnetometry. The discrepancy has not been resolved yet, and it is necessary to confirm the reliability of magnetization measurement.

In the present study, we have examined the anisotropy in magnetization of the Bi-2212 superconductor. For this purpose, high quality single crystals with the thickness of several hundred micrometers in the c direction have been prepared. The result has shown the existence of weak coupling in fields slightly misaligned with the *ab* plane. We attribute this to a three-dimensional (3D) to two-dimensional (2D) crossover of the shielding current path due to flux penetration along the CuO_2 interlayer regions through the surface barrier [10,11]. The γ is found to be ≈ 700, which is consistent with other reported measurements [4,7].

Bi-2212 single crystals were prepared by a traveling solvent floating zone method whose details are described elsewhere [12]. Two single crystal samples A and B with the dimensions of $a\times b\times c = 1.53\times 0.70\times 0.42$ mm^3 and $2.97\times 2.05\times 0.87$ mm^3, respectively, were selected from one batch. X-ray diffractometry and electron-probe microanalysis revealed that there were no other phases other than Bi-2212. To check the quality of the samples, the Meissner signal of sample A was measured for decreasing temperature in a field of 10 Oe perpendicular to the *ab* plane using a SQUID magnetometer (Quantum Design model MPMS2). The onset of the superconducting transition was 91 K, and the transition width was 2 K. The Meissner signal was as large as 90% of perfect diamagnetism, estimated from the sample volume and the demagnetization factor given below. Such a sharp transition and a large Meissner signal confirm the high quality of the samples. Magnetization measurements were performed using the SQUID magnetometer and a vibrating sample magnetometer (VSM; PAR model 4500). The former has more than 3 orders of magnitude better sensitivity than the latter, while the field direction can be more precisely controlled by the latter.

The initial magnetization curve of sample A at 20 K measured by SQUID is shown in Fig. 1. The angle θ between the *ab* plane and the applied field H is 90°. The absolute value of magnetization $4\pi M$ increases linearly with the increase in the applied field in the low field region. It deviates from the linearity at $H_{d\perp} \approx 160$ Oe. Assuming perfect diamagnetism in the linear region, one

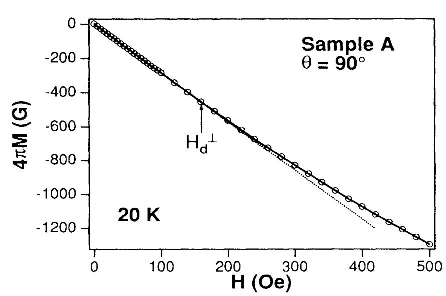

FIG. 1. Initial magnetization curve of sample A for $\theta = 90°$ at 20 K.

can regard the slope of -2.82 as $-1/(1-N_c)$ where N_c is the demagnetization factor for fields parallel to the c axis. From this we can estimate N_c as ≈ 0.65 and N_{ab}, the demagnetization factor in fields parallel to the ab plane, as $\approx (1-N_c)/2 \approx 0.18$. The effective field $H_{d\perp}/(1-N_c)$ does not give the lower critical field $H_{c1,c}$ itself but the upper bound of $H_{c1,c}$, because the pinning effect may shift the flux penetration field higher.

For $\theta=6°$, in a slight misalignment between H and the ab plane, the initial magnetization curve for sample A (Fig. 2) shows a quite different feature from that in Fig. 1; it shows a peak around $H_0 \approx 10$ Oe (note that H_0 is not the exact peak position but an upper bound of it, because the increment of the field is as large as 10 Oe. A more precise peak position, H_p, is given later). In fields higher than H_0, the magnetization is linear with H, and then deviates from the linearity at a field $H_d \approx 1500$ Oe. However, the slope $4\pi\chi^*$, defined as $4\pi\Delta M/\Delta H$ in the field range of $H_0 \ll H < H_d$, is much smaller than that predicted by perfect diamagnetism.

Such behavior shows that there are two kinds of flux penetration around H_0 and H_d; magnetization in fields below and above H_0 may be dominated by different kinds of shielding currents. Similar behavior has been observed in polycrystalline samples, in which flux starts to penetrate along grain boundaries which act as weak coupling between the grains, in a field much smaller than the lower critical field of the superconducting grains. However, there are no grain boundaries in the Bi-2212 single crystal, and this weak-coupling-like behavior disappears in fields perpendicular to the ab plane. Hence, this seems to be due not to the granularity in the crystal, but to an intrinsic property of Bi-2212.

First, we discuss the magnetization behavior in fields above H_0. Previous torque measurements [4] have suggested that only the transverse component of H to the ab plane, H_T, contributes to the magnetization for Bi-2212 (see the lower inset in Fig. 3). Under this assumption, if H_T is smaller than $H_{d\perp}$, the Meissner signal of $4\pi M_T = -H_T/(1-N_c)$ will be induced perpendicular to the layers. Then the observed magnetization of the magnetometer, $4\pi M_{\rm obs}$, will be the component parallel to the applied field, $4\pi M_{\rm obs} = 4\pi M_T \sin\theta$. Using $H_T = H\sin\theta$ and dividing $4\pi M_{\rm obs}$ by H, we can obtain an expression of the susceptibility as

$$4\pi\chi_{\rm obs} = -\sin^2\theta/(1-N_c)\,. \tag{1}$$

Figure 3 shows the field angle dependence of $4\pi\chi^*$, normalized by the value at $\theta=90°$ to eliminate the demagnetizing factor, for sample B measured by VSM at 4.2 K. The origin of θ has been determined as the minimum point of the normalized $4\pi\chi^*$. It is noteworthy that the $4\pi\chi^*/4\pi\chi^*(90°)$ versus $\sin^2\theta$ relationship shows excellent linearity as shown in the inset of Fig. 3, as predicted by Eq. (1). This indicates that $4\pi\chi^*$ is equivalent to $4\pi\chi_{\rm obs}$, and that one can estimate θ by measuring $4\pi\chi^*$. In fact, $\theta=6°$ in Fig. 2 has been derived from Eq. (1) using the $4\pi\chi^*$ value of -3.23×10^{-2} and the normalization factor $4\pi\chi^*(90°)$ of 2.82 from Fig. 1. In addition, the deviation from the linearity at H_d in Fig. 2 should indicate that $H_d\sin\theta$ becomes $H_{d\perp}$. Using the values of $H_d=1500$ Oe and $\theta=6°$ for Fig. 2, we can obtain $H_d \sin\theta \approx 160$ Oe, which is consistent with $H_{d\perp}$ in Fig. 1. These results suggest that the magnetization for $H \gg H_0$ is dominated by the 2D in-plane ($\parallel ab$) shielding current which is induced by H_T, and $4\pi\chi^* H$ is the resultant magnetization for $H < H_d$.

Therefore, magnetization in fields below H_0 is considered to be a superposition of the contributions from the in-plane and the out-of-plane ($\perp ab$) currents. In fields around H_0, such a 3D shielding current path changes to a 2D path due to the penetration of longitudinal flux into the interlayer regions. This 3D to 2D crossover field ($\approx H_0$) may be proportional to $1/\cos\theta$ while H_d is given as $H_{d\perp}/\sin\theta$. It means that the crossover field exceeds H_d at a certain angle θ_c. Using $H_{d\perp} \approx 160$ Oe and $H_0(\theta=0°) \approx 10$ Oe, $\theta_c = \tan^{-1}[H_{d\perp}/H_p(\theta=0°)]$ is estimated as $\approx 86°$. Therefore, in the vicinity of $\theta=90°$, flux will not penetrate into the sample in fields below H_d, i.e., $H_{d\perp}/\sin\theta$. This explains the magnetization curve without the weak-coupling-like behavior shown in Fig. 1.

For high T_c superconductors, a change in the dimensionality of the system has been suggested [5,13]: ξ_c in-

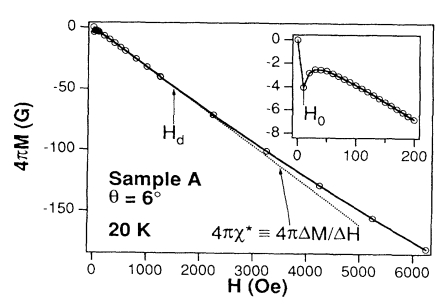

FIG. 2. Initial magnetization curve of sample A for $\theta=6°$ at 20 K. Inset shows an expansion of the low field region.

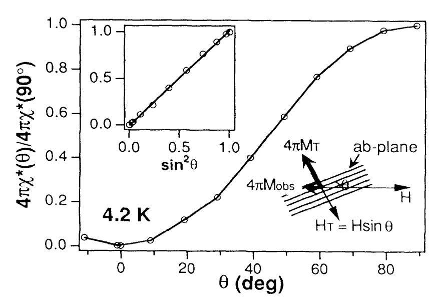

FIG. 3. Angle dependence of normalized $4\pi\chi^*$ of sample B at 4.2 K. The lower inset shows a scheme of a stack of superconducting sheets in an applied field.

creases with increasing temperature, and if ξ_c exceeds the CuO_2 interlayer distance d, an anisotropic 3D picture becomes a better description than the Josephson coupled quasi-2D layered system. For Bi-2212, the crossover temperature has been reported to be higher than $(T_c - 1)$ K [13] which exceeds our experimental range. Therefore, the dimensional crossover of the shielding current path illustrated by the present results takes place within the quasi-2D regime.

In the special case of $\theta = 0°$, as predicted by Eq. (1), $4\pi\chi^*$ will become zero: We will observe magnetization by the longitudinal fields only, and obtain information on the flux penetration into the interlayer regions around H_0. However, it is difficult to adjust θ exactly to zero because of the instrumental limitation. Hence, we have subtracted $4\pi\chi^* H$ from the data taken at a small θ to extract the contribution of H_L, the longitudinal component of H.

Figure 4 shows extracted magnetization curves for sample A in fields below 50 Oe at several temperatures. Since this field range is much smaller than H_d, we can safely assume that Fig. 4 shows the contribution from H_L only. We now regard H_L as H because the correction factor of cos6° is close to unity. The peak position H_p is 6 Oe at 5 K, and this monotonically decreases with the increase in temperature as shown in the inset. In fields below H_p, magnetization almost obeys perfect diamagnetism shown as a dashed line in Fig. 4, although there is a slight deviation, presumably due to the surface roughness described later. Nevertheless, the main portion of flux penetration into the interlayer regions seems to begin at H_p, since the magnetization is rapidly depressed with the increase of H beyond H_p.

The existence of hysteresis between ascending and descending field branches suggests that there should be some kind of pinning effect on the flux parallel to the plane. Nevertheless, the hysteresis shows some strange features which are not expected in the conventional bulk flux pinning picture. In the increasing portion of the field, the magnetization curve is depressed with the increase in temperature. On the contrary, in the decreasing portion of the field, magnetization is almost temperature independent and close to zero.

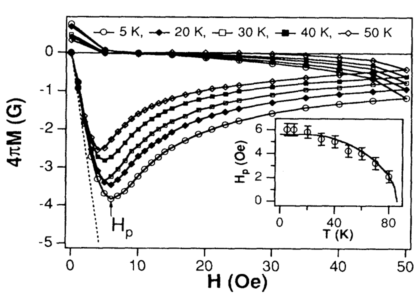

FIG. 4. Minor hysteresis loops of sample A at several temperatures for a small θ. Contribution from H_T has been subtracted (see text). Inset shows a plot of H_p vs temperature.

Such asymmetry can be attributed to the effect of the Beam-Livingston type surface barrier [10]. In the field descending process, interaction with the surface shielding current pushes the flux lines inside the sample. This is the origin of the surface barrier preventing flux lines from escaping. Campbell and Evetts [11] have argued that there will always be such a barrier unless the flux density inside the sample exceeds that outside the sample, because the surface current is a consequence of the difference between them. The surface current becomes zero at the early stages of the decreasing portion of the field, since the current from the flux lines, trapped inside the sample by the surface barrier, cancels the Meissner current which decreases with the applied field. If the field is further decreased, then flux lines leave the sample to stabilize the overall surface current at zero. For a sample without bulk pinning, magnetization is proportional to the surface current. Therefore, magnetization will become nearly zero in the descending branch of the hysteresis loop.

A necessary condition for the surface barrier to be effective is that the surface roughness is less than the vortex current dimension, which is equivalent to the penetration depth. In the Josephson coupled layered superconductors, it should be replaced by the Josephson length γd (≈ 1 μm for Bi-2212 according to the following estimation) for vortices parallel to the *ab* plane [6,14]. Sample A satisfies the condition since the surface roughness has been estimated as less than 1 μm by optical microscopy. There are, however, some irregular regions with roughness of more than several micrometers. This may result in a partial depression of the surface barrier, and some deviation from the ideal situation. It qualitatively describes the slight deviation from the perfect diamagnetism below H_p in Fig. 4.

The possibility of effective surface barriers in fields perpendicular to the *ab* plane has been reported for Bi-2212 [15] and $YBa_2Cu_3O_x$ [16] single crystals which have shown the same features in their magnetization curves. Chikumoto *et al.* [15] and Konczykowski *et al.* [16] have suggested the crossover from the bulk pinning regime to the surface barrier regime as a function of temperature; at high temperatures, the bulk pinning vanishes and the surface barrier becomes dominant. For the present measurements, the characteristic feature in magnetization curves is observed even at as low a temperature as 5 K. This indicates weak bulk pinning for Bi-2212 in this field direction over a wide range of temperatures. Actually, once the flux parallel to the *ab* plane penetrates into the sample, intrinsic pinning [3] may act on flux motion perpendicular to the *ab* plane. There seems, however, to be no flux pinning mechanism on the flux motion parallel to the plane. This may result in the apparent zero bulk pinning in this field direction [17]. Therefore, the surface barrier acting on the flux motion parallel to the *ab* plane is considered to be responsible for the bulk magnetization behavior.

For a Josephson coupled layered superconductor with an interlayer spacing of d and the anisotropic parameter $\gamma=\lambda_c/\lambda_{ab}$, the Josephson length γd along the layers and d across them can be regarded as an effective core size of a vortex parallel to the layers [14]. Then one can apply the result of de Gennes [18] to write down the Gibbs potential G for a vortex at a distance of x from a surface perpendicular to the layers, and the distance of x as

$$G=\frac{\phi_0}{4\pi}\left[H\exp\left(-\frac{x}{\lambda_c}\right)-\frac{\phi_0}{4\pi\lambda_{ab}\lambda_c}K_0\left(\frac{2x}{\lambda_c}\right)+H_{c1}-H\right], \tag{2}$$

where ϕ_0 is the flux quantum and K_0 is a modified Bessel function of the second kind. Then, under the condition of $\partial G/\partial x=0$ and $x=\gamma d$, an expression of the field for the beginning of flux penetration along the layers through the surface barrier is obtained as [19]

$$H_{\text{pen}}\approx\phi_0/4\pi\lambda_{ab}\gamma d\,. \tag{3}$$

If λ_{ab} and λ_c have the same temperature dependence, then γ is a constant and λ_{ab} is the only temperature dependent parameter in Eq. (3). Therefore, if H_p corresponds to H_{pen}, the temperature dependence of H_p should be the same as that of $1/\lambda_{ab}$. For Bi-2212, the London local limit [20] describes well the temperature dependence of the penetration depth λ [21]. In fact, $1/\lambda$ deduced from this limit depends ón temperature as a solid line in the inset of Fig. 4, which is consistent with the data. It suggests the validity of the surface barrier model for the present result.

From Eq. (3) we can derive several important parameters. Using the values of $\phi_0=2\times10^{-7}$ G cm^2, $\lambda_{ab}=3000$ Å [20], and $H_{\text{pen}}=H_p=6$ Oe for Eq. (3), a γ value of ≈ 700 is obtained. This is consistent with other measurements as torque [4] and ac susceptibility [7], and suggests the validity of the surface barrier model. The lower critical field parallel to the ab plane is expressed as $H_{c1,ab}=H_{c1,c}/\gamma$. As the upper bound of $H_{c1,c}$(20 K) is given as $H_{d\perp}/(1-N_c)=460$ Oe from Fig. 1, one can estimate $H_{c1,ab}$(20 K) as 0.7 Oe or less. This is much smaller than the previously reported value of ≈ 60 Oe at the same temperature [8].

Finally, some remarks should be made about the inconsistency in the anisotropy with the previous magnetization measurements [8,9]. As platelet single crystals with a thickness of up to several tens of micrometers in the c direction were used in previous works, misalignment between the ab plane and the applied field is crucial. Krusin-Elbaum *et al.* [8] have pointed out that a slight misalignment of less than 2° results in a 100% increase of the apparent signal, which is presumably due to the large demagnetizing effect for H_T. It may mask the contribution from the out-of-plane currents, and then the correct anisotropic nature of magnetization.

In conclusion, magnetization measurements on high quality Bi-2212 single crystals have shown the existence of anisotropic weak coupling for the first time. It has been attributed to flux penetration into the CuO_2 interlayer regions and a resultant 3D to 2D crossover of the shielding current path with the increase of the field; diamagnetism dominated by the bulk shielding current changes to that by in-plane current only. The shape of the hysteresis loops around the crossover field indicates the existence of an effective surface barrier which sustains the out-of-plane current in the low field region. Based on a model of the surface barrier in a layered superconductor, a large γ value of ≈ 700 has been obtained.

The authors would like to thank M. Suenaga, M. Murakami,and H. Kubota for their helpful discussions, D. N. Matthews for a critical reading of this manuscript, and S. Tanaka for his continuous encouragement. This work was supported in part by NEDO.

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