In-Plane Anisotropy of the Interaction between Vortex and Twin Boundary in YBa₂Cu₃O₇ Single Crystals

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We report the temperature dependence of the magnetization hysteresis loops of high-quality $YBa_2Cu_3O_7$ single crystals, each with a single unique orientation of twin boundaries. We have observed magnetization anisotropy in the temperature range 6-77 K for various magnetic fields applied normal to the *c* axis and parallel (stronger pinning) or perpendicular (weaker pinning) to the twin-boundary orientation, thus causing an anisotropic critical current density J_c . Magnetization-time-relaxation and Meissner-fraction measurements also support the above observation.

PACS numbers: 74.60.Ge, 61.70.Ng, 74.70.Vy

The existence of twin boundaries in $RBa_2Cu_3O_x$ (R denotes rare-earth elements, except Ce, Tb, and Pr) provides a unique opportunity to study the flux-pinning mechanism in the layered high-temperature superconductors. While the general concept of the role of the twin boundaries in flux pinning has been addressed by comparing the magnetic measurements in the same crystal before and after detwinning,^{1,2} a detailed study on the interaction between vortex and twin boundaries is fundamentally important in understanding the basic pinning mechanism in the high- T_c superconductors. In a recent report, Kwok et al.³ used a YBa₂Cu₃O₇ (YBCO) single crystal with only one set of twin boundaries to study the dependence of the flux-flow-pinning strength on the orientation of the applied magnetic field with the twin planes at a temperature of 91.5 K. A remarkable decrease in resistivity was found when the field was rotated parallel to the twin planes, indicating a strong pinning effect in this vortex-twin-boundary (TB) interaction mode. In another paper, Swartzendruber et al.⁴ reported the magnetization measurements at 10 K on a very small YBCO single crystal with predominantly one variant of twin boundaries. The authors found that, in the case of the magnetic field perpendicular to the c axis, when the field is normal to the twin boundaries, the pinning force is measurably stronger than that of the case where the field is parallel to the twin boundaries. These opposite results on the nature of the anisotropic pinning of vortices by twin boundaries immediately called attention to a possible change in the sign of the energy for the vortex-twin-boundary interaction. In a later Comment to Kwok's work, Roitburd et al.⁵ claimed that a crossover in the sign of the interaction energy between flux vortices and twin boundaries was found at 70 K from their measurements. In a recent torque measurement, Gyorgy et al.⁶ found that when a field is applied perpendicular to the c axis and rotated with respect to the a-b axes, the torque increases roughly sixfold at 76 K when the field is within 2° of parallel to the twin boundaries. These peaklike features disappear at 27 K. The authors attributed this phenomenon to the increase in pinning strength from inherent defects.

In this Letter, we present our recent full magnetization-hysteresis measurements on high-quality crystals with only one set of twin boundaries. Our results show that the in-plane pinning energy in the parallel case is *always* larger than the perpendicular configuration. No evidence can be found for a change of sign in the vortextwin-boundary interaction between 6 and 77 K. Meissner-effect data and time-relaxation measurements also support the above observations.

High-quality YBCO single crystals with T_c between 93 and 94 K and transition widths between 1 and 2 K were grown in gold crucibles by a modified self-flux method based on previous work.^{7,8} It was observed that single crystals containing a large area of single twin boundaries are often found in locations where growth is undertaken more freely. Based on this observation, we developed a technique to control the nucleation so that many free-standing crystals can be grown. Crystals with a 1-2-mm²-large domain of single twin boundaries are not difficult to find. It is also noticed that the twin pattern on the surface of a crystal might be different from that obtained on inside layers, so care must be taken to select crystals with a one-to-one correspondence of twin structures on both faces. The photographs in Fig. 1 demonstrate this morphology for a typical crystal, where the one-to-one correspondence of the twin boundaries on both faces shows that the entire crystal contains only one set of twin boundaries. Our transmission-electronmicroscope observation showed that for a typical crystal of Fig. 1, the average twin-boundary separation is about 1300 Å. In order to utilize the extended Bean model⁹ to calculate the critical current density J_c , all samples were



FIG. 1. Morphology photographs of a YBCO crystal containing only one set of twin boundaries viewing from (a) side 1 and (b) side 2, respectively. Note the one-to-one correspondence of the twin boundaries at both sides.

cut into rectangular shapes with their edges either parallel or perpendicular to the twin boundaries. Two samples with the dimensions of $1 \times 1 \times 0.08$ and $1 \times 0.75 \times 0.05$ mm³, respectively, were used for measurements. The in-plane demagnetization factor for sample 1 is 0.06 (along the two edges). For sample 2, the in-plane demagnetization factors are 0.03 (along the longer edge) and 0.06 (along the shorter edge), respectively.

For studying both the magnetic-field and the temperature dependences of the vortex-twin-boundary interaction, we measured the hysteresis loops of the two samples at different temperatures from 6 to 77 K with the magnetic field parallel and perpendicular to the twin boundaries. Magnetic measurements were conducted on a commercial SQUID magnetometer¹⁰ with the magnetic field varying from 0 G up to 5.5 T. In the magnetization-hysteresis measurements, 5 min waiting time was used after setting a new field, so that the magnetization inside the sample had ample time to relax to a relatively stabilized value. Figure 2(a) presents two magnetization-hysteresis loops of sample 1 at 6 K for the magnetic field parallel and perpendicular to the twin boundaries, respectively. The remarkable size difference between the two loops clearly demonstrates that when $H > H_{c1}$ $(H_{c1} \approx 200 \text{ Oe at } 6 \text{ K})$ the pinning force in the case of the magnetic field parallel to the twin boundaries is significantly stronger than that of the perpendicular case. The same results were obtained at different temperatures. Figure 2(b) presents some examples of the field dependence of the magnetizations of sample 1 for a field range between 0 and 250 Oe for the two interaction modes. Over the full temperature range, our results show that the parallel mode has stronger pinning than the perpendicular mode at lower fields. For comparing the pinning effects at higher field, the critical current density J_c at 1 T for each temperature was calculated by using the extended Bean model.⁹ Figure 3 presents the calculated J_c for the two samples in a magnetic field of 1



FIG. 2. (a) Magnetization-hysteresis loops obtained from sample 1 at 6 K for a magnetic field parallel to the twin boundaries and perpendicular to the boundaries, respectively. (b) Lower-field behavior of magnetization of sample 1 for two orientations. Here, effective magnetic field $H_{\text{eff}} = H_a - D4\pi M$, with H_a the applied field and D = 0.06.

T. In the temperature range between 6 and 77 K, the fact that the magnetization critical current density J_c in the parallel case is always larger than that in the perpendicular case again rules out the possibility of a sign change in the vortex-twin-boundary interaction. The ratio of $J_c^{\parallel}/J_c^{\perp}$ for sample 1 in the temperature range 6-77 K changes from an average value of 1.6 at lower temperatures to a maximum value of 3.5 at 77 K (see Fig. 3). For sample 2, the ratio remains approximately constant, with an average value of 1.65 between 6 and 70 K (see inset of Fig. 3). Since J_c is proportional to the pinning potential energy U_0 , the ratio $J_c^{\parallel}/J_c^{\perp}$ should be the same as $U_0^{\parallel}/U_0^{\perp}$. In order to verify this point, we conducted a shielding-magnetization time-relaxation experiment on sample 1 at different temperatures and magnetic fields. Figure 4 presents an example obtained at 30 K in a 1-T magnetic field for $H \parallel TB$ and $H \perp TB$, respectively (TB denotes the twin-boundary direction). The observed faster decay of magnetization with time in the perpendicular mode compared to that of the parallel configuration



FIG. 3. Temperature dependences of the magnetization critical current density J_c of sample 1 at 1 T for a magnetic field parallel and perpendicular to the twin boundaries, respectively. Inset: Results from sample 2.

again confirms that the pinning force in the former case is weaker than in the latter case. The pinning potential U_0 can be obtained from $U_0 = k_B T (M_0 - M_{eq}) d \ln(t) / d \ln(t)$ dM, where k_B is the Boltzmann constant, M_0 is the initial magnetization taken at t = 400 sec [where the linear behavior of $dM/d\ln(t)$ starts], and M_{eq} is the final stable value which can be replaced approximately by the Meissner magnetization at the same temperature and magnetic field. The pinning potentials U_0 obtained from Fig. 4 for the parallel and perpendicular cases are $U_0^{\parallel} = 0.06$ eV and $U_0^{\perp} = 0.04$ eV, respectively. The difference in pinning potential energy for these two vortex-twin-boundary interaction modes is 0.02 eV, a significant value relative to the size of U_0 . We have a ratio of $U_0^{\parallel}/U_0^{\perp} = 1.5$, which is in good agreement with the value obtained from Fig. 3.

The anisotropic pinning potential energy U_0 for the parallel and perpendicular vortex-twin-boundary interaction modes could result in different Meissner fractions. To avoid the irregular-shape effect, sample 1, a square platelet, was used to measure the low-field susceptibilities. For each orientation ($H \parallel TB$ and $H \perp TB$), the sample was first cooled in a zero field down to 6 K, and the shielding effect was measured in a 5-Oe field as the sample was warmed to a temperature above T_c . In both cases, we obtained $\chi_{ZF} = -1/4\pi$, indicating a full diamagnetic shielding effect. Meissner data were taken under a field-cooling condition (5 Oe) from a temperature above T_c down to 6 K. Figure 5 presents the Meissner fractions relative to the shielding effects for the two different orientations. Again, the fact that the Meissner fraction for $H \parallel TB$ is smaller than that for $H \perp TB$ provides evidence of stronger pinning effects existing in the parallel mode at a temperature near T_c , which is consistent with the results obtained by transport measurements.³



FIG. 4. Magnetization-time-relaxation curves of sample 1 at 30 K and 1 T for a magnetic field parallel and perpendicular to the twin boundaries, respectively.

It is believed that the difference in the vortex-twinboundary interaction between the parallel and perpendicular modes is associated with the local microstructure related to the twin boundaries. An electron microscope study on twin boundaries¹¹ showed that the boundary region has a structure different from the bulk orthorhombic structure. The value of $\Delta a/a$ ($\Delta a = b - a$) measured by electromicrodiffraction at the boundary region is much smaller than that of the bulk value. This implies that at the boundary the tetragonal structure might be a stable phase. A recent field-ion-microscopic image study on the twin structure confirms the depletion of oxygen concentration across the boundary.¹² Hence, elastic strain exists in the region between the two crystallographic structures. The energy of vortex-twin-boundary interaction depends on the orientation of flux lines with



FIG. 5. Meissner fraction of sample 1 at 5 Oe for a magnetic field parallel and perpendicular to the twin boundaries, respectively.

respect to the distribution of the stress field. This will generally result in anisotropic pinning effects. The relative distribution of the stress field should be independent of temperature for $T < T_c$. This is consistent with our observation of no sign change in the vortex-twinboundary interaction. We believe that the different results among Swartzendruber et al.,⁴ Roitburd et al.,⁵ and this work arise mainly from the samples. First, their sample contains a substantial fraction of crossing twin boundaries [see Fig. 1(e) of Ref. 4]. To our knowledge, the existence of crossing twin boundaries would greatly change the pinning behavior. This can be evidenced by their higher ratio of J_c for $H \parallel c$ and $H \perp c$ (~62 at 10 K and H=0). In our case, this J_c anisotropy is as low as 15 at 10 K and H=0. Kes and Van den Berg¹³ have pointed out that when HIIc, the pinning force of a crossing-twin-boundary structure is much stronger than that of a single set of twin boundaries. Our experiments confirm this point. Second, the volume of their sample $(0.12 \times 0.135 \times 0.12 \text{ mm}^3)$ is almost 40 times smaller than our samples. This significant reduction in size presents major difficulties in sample orientation with respect to the applied field. Since this kind of measurement is quite sensitive to the accuracy of the orientation, caution must be taken when comparing the results obtained from different samples.

In summary, magnetization-hysteresis loops have been measured for two YBCO crystals, each with a single, unique orientation of twin boundaries, for the parallel and perpendicular interaction modes respectively. By comparing the low-field magnetization and the magnetization critical current density J_c in a field of 1 T, it is found that the pinning force in the parallel mode is *always* stronger than that of the perpendicular mode over the entire temperature range of 6-77 K. Magnetization-time-relaxation measurements at 1 T and 30 K support the above conclusion, where the difference in pinning potential between the two modes is 0.02 eV. This observation can also be extended to temperatures near T_c , which was confirmed by our Meissner-effect measurements.

This work was performed under the auspices of the U.S. Department of Energy for Lawrence Livermore National Laboratory under Contract No. W-7405-ENG-48.

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(a) (b) FIG. 1. Morphology photographs of a YBCO crystal containing only one set of twin boundaries viewing from (a) side 1 and (b) side 2, respectively. Note the one-to-one correspondence of the twin boundaries at both sides.