Lock-In Oscillations in Magnetic Hysteresis Curves of YBa$_2$Cu$_3$O$_{7-x}$ Single Crystals

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We report the first direct experimental evidence of intrinsic pinning in hysteresis loops of untwinned YBa$_2$Cu$_3$O$_{7-x}$ single crystals, for magnetic fields perpendicular to the $c$ axis. We have observed oscillations in the $M-H$ loops that are well explained when one considers the layered structure of this compound. The oscillations are found to be periodic in $H_c^{1/2}$ and are induced by a succession of transitions between states for which the vortex lattice is commensurate with the crystal lattice periodicity in the direction perpendicular to the CuO$_2$ planes. The following anisotropy parameters were extracted from the experimental results: $\Gamma_{ab} = (m_c/m_b)^{1/2} = 6.3$, $\Gamma_{ac} = (m_c/m_a)^{1/2} = 5.6$, and $\Gamma_{ab} = (m_a/m_b)^{1/2} = 1.13$.

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It is well established that the anisotropy in high-$T_c$ superconducting oxides plays an important role in their physical properties. However, many of these properties cannot be accounted for by a simple anisotropic 3D description because the layered structure of these materials confers some characteristic features on them. These characteristics are not specific to the cuprate superconductors; interest in this topic originated in the early seventies through the study of conventional layered materials [1]. The layered structure leads to interesting physical properties. For example, a 3D to 2D transition [2] is expected at a temperature $T^*$, where the coherence length along the $c$ axis becomes smaller than the spacing between the CuO$_2$ planes (the superconducting layers). Also, when a magnetic field $H > H_{c1}$ is applied along the layers, it is energetically favorable at low temperatures for a vortex to lie between two CuO$_2$ planes. Feiberg and Villard [2] have shown that for an applied magnetic field making an angle $\theta$ with the $a$-$b$ plane, the vortices lock in to the $a$-$b$ plane if $\theta < \theta_c$ (the critical angle). Experimentally, an anomaly in torque measurements on an untwinned YBa$_2$Cu$_3$O$_{7-x}$ (YBCO) single crystal has been observed by Farrell et al. [3] and attributed to lock-in by Bulaevskii [4]. A similar anomaly was reported [5,6] for Tl$_2$Ba$_2$CaCu$_2$O$_8$ and Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ single crystals. In this paper, we present the first clear evidence of intrinsic pinning by the layered structure of high-$T_c$ superconductors observed in hysteresis loops of untwinned YBCO single crystals recorded for magnetic fields parallel to CuO$_2$ planes. We will also deduce accurately the anisotropy parameters $\Gamma_{ij} = (m_i/m_j)^{1/2}$, where $m_i$ and $m_j$ are the effective masses defined such that the subscripts refer to the principal axes along which screening currents flow.

The crystals investigated were grown as described in Ref. [7] by a conventional self-flux method using yttria stabilized zirconia crucibles. They were oxygenated for 6 d at 500°C in an oxygen atmosphere. Detwinning was achieved by applying a uniaxial pressure of $\sim$ 50 MPa at 550°C in air for 15 min. Detwinned crystals were then reoxygenated for 1 d at 500°C in oxygen atmosphere, providing an overall oxygen content [8] $6.9 < x < 6.92$. Polarized optical microscopy revealed a surface fraction of misaligned domains of less than 2%. The crystal whose data are reported here has a $T_c = 93.8$ K and $\Delta T_c = 0.2$ K, and has a size of 1.45 x 1.2 x 0.125 mm$^3$. The width of the transition, $\Delta T_c$, is the temperature range over which the zero field cooled magnetization in a field of 0.1 mT varies from 10% to 90%. The thickness has been estimated using the mass and the theoretical density of 6.8 g/cm$^3$. Magnetic hysteresis measurements were carried out on a 12 T vibrating sample magnetometer (Oxford Instruments, model VSM-3001). Samples were first zero field cooled at the desired temperature, and then subjected to a magnetic field ($H_a$) which was swept at a rate of 5 mT/s.

Figure 1 shows magnetic hysteresis loops at $T = 60$ K, for applied magnetic fields up to 12 T making an angle $\theta$ with the $b$ axis. The curves in Fig. 1(a) correspond to two measurements in which the crystal was each time placed with the $b$ axis parallel to $H_a$ (with the accuracy of our setup $\theta_1, \theta_2 < 1^\circ$). For the curve presented in Fig. 1(b) the crystal was placed under a slight angle $\theta_3 = 2^\circ \pm 0.5^\circ$. Similar results have been obtained for magnetization isotherms with $H_a$ along the $a$ axis. The oscillations observed in the magnetization are reported for the first time. The period of the oscillations increases with increasing magnetic field. We will show that the response is periodic in $H_a^{-1/2}$. An important observation is that the positions of the maxima are temperature and orientation independent, indicating pinning caused by a matching of the vortex lattice constant to a regular pinning structure. From the magnetization data presented in Fig. 1 it is clear that there exists a critical angle of the order of 1° above which the oscillations disappear. This demonstrates that the oscillatory pinning behavior has its origin in the lock-in of vortices between the layers.

We explain these observed features in the magnetic hysteresis in terms of states of the vortex lattice which are commensurate with the layered structure of the cu-
prate superconductors. The anisotropic London model, which assumes a homogeneous material, predicts a hexagonal vortex lattice compressed along the crystal c axis with a field independent ratio $a/l = 2\Gamma/\sqrt{3}$, where $\Gamma$ is the anisotropy parameter and $a$ and $l$ are the vortex lattice spacings perpendicular and parallel to the c axis, respectively (see Fig. 2). In the cuprate superconductors, the CuO$_2$ planes are responsible for superconductivity and there will be a modulation of the superconducting order parameter along the c axis. The vortex lattice with the $a/l$ ratio calculated from the anisotropic London model (and therefore with minimum vortex-vortex interaction energy) will be commensurate with this modulation for certain applied magnetic field values, $H_n$. At $H_n$, the vortex lattice spacing in the direction of the c axis fulfills the condition $l = nd$, where $d$ is the c-axis crystal lattice constant and $n = 1, 2, 3, \ldots$. The ratio $a/l$ will change for intermediate fields, $H_n \neq H_{n+1}$. For $H_n$ near a given $H_n$, changes in vortex density in response to a magnetic field change will be accommodated by motion of the vortices in the $a$-$b$ plane (decreasing or increasing the vortex spacing $a$ for increasing or decreasing fields, respectively). Pinning centers in the regions between the CuO$_2$ planes are less effective and hence the effective pinning force for these displacements is small. However, this process alone cannot lead to another commensurate state. For field values in the intermediate region between two consecutive commensurate states, $H_n$ and $H_{n+1}$, changes in $l$ must occur. This crossing of the CuO$_2$ planes by vortices is generally thought to be accomplished by the nucleation and motion of pairs of kinks and antikinks [2].

These kinks consist of a vortex disk in the CuO$_2$ plane. Motion of these vortex disks is impeded by the relatively stronger pinning interactions in the CuO$_2$ planes. From this we expect that the resulting effective pinning force, and hence the magnetic hysteresis width, to be larger for the intermediate magnetic field regions. This is indeed what we observe: The magnetic hysteresis width shows lock-in oscillations which are periodic in $H_n^{-1/2}$. Bulgakii and Clem [9] have calculated the equilibrium structure of the vortex lattice for magnetic fields parallel to the planes using the Lawrence-Doniach theory [10]. They have found a field dependence of the ratio $a/l$ which supports our explanation of the magnetic behavior.

In Fig. 3 we have presented the magnetization curves for increasing field as a function of $(\mu_0H_a)^{-1/2}$, at $T = 30, 40, 50$, and 60 K. The magnetic field has been applied parallel to the a axis for the data in Fig. 3(a) and parallel to the b axis in Fig. 3(b). In order to see the oscillations clearly, we have subtracted a linear field dependent background from the original data and shifted the curves along the vertical axis. This procedure does not, of course, affect the periodicity of the signal. For both sets of measurements the curves are periodic with an identical period at all temperatures. The positions of the maxima are temperature independent except for $T \geq 60$ K where a slight shift towards lower fields is observed. The amplitude of the oscillations increases with increasing magnetic field. At low magnetic fields the vortex-vortex interactions are weaker and the energy differences between commensurate and incommensurate states are smaller. We argue that this decreases the amplitude of the oscillations. From Fig. 3 we can see that the observed period for $H_a \parallel a$ and $H_a \parallel b$ are slightly different, indicating a difference in the anisotropy parameters $\Gamma_{ab}$ and $\Gamma_{ca}$.

In the anisotropic London model, for magnetic fields parallel to either the a axis or the b axis, the vortex spacing along the c axis is given by [9,11]

$$l = (\sqrt{3}\phi_0/2\Gamma)^{1/2}B_a^{-1/2},$$

(1)

where $\phi_0$ is the flux quantum and $B_a = \mu_0H_a$. The periodicity in Fig. 3 corresponds to transitions between commensurate states at $H_a = H_n$ in which case Eq. (1) gives the correct vortex spacing. When the applied magnetic
The magnetic hysteresis curves for $T \geq 60$ K display some characteristics not observed at lower temperatures. As mentioned earlier the oscillations are slightly shifted towards lower fields. These hysteresis curves also exhibit additional peaks at high fields (see Fig. 3). A possible explanation is that the lock-in of different regular vortex structures which are not minimal energy solutions in the anisotropic London model causes these additional features.

We have also performed similar magnetic hysteresis measurements ($H_a$ or c) on microtwinned YBCO crystals. However, in these measurements we have not observed any oscillations. This absence of oscillations can be explained by the following arguments. First, the periodicities along the a and b directions differ, and second, as the anisotropy factor is dependent upon the oxygen concentration $[14]$, the wavelength of the oscillations is dependent on it as well. In microtwinned samples, the oxygen concentration is less homogeneous because of the disorder introduced by twinning planes. Both these effects will cause an averaging out of the hysteresis oscillations.

In summary, we have reported direct experimental evidence of lock-in pinning in hysteresis loops of untwinned YBCO single crystals, in longitudinal geometry. Values of the in plane as well as the out of plane anisotropy factors have been obtained from the experimental data and are found to be in agreement with those reported in the literature using other experimental techniques. Our observation of the lock-in oscillations in the magnetic properties opens up a new method to study the vortex lattice by using the layered crystal structure as a ruler against which we can determine accurately the vortex spacing in the c direction.

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