Commensurability oscillations and smectic vortex phase transition in $YBa_2Cu_3O_y$ single crystals

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We have studied the oscillations of the magnetization induced by commensurability between intervortex and CuO plane distances in $YBa_2Cu_3O_y$ ($y=6.97\pm0.02$) single crystals using high angular resolution magnetometry. A sharp peak in the temperature dependence of the amplitude of oscillations was found at $T_f\sim60$ K. It is followed by other changes in the behavior of the amplitude and irreversible magnetization. We suggest that T_f reflects the vortex freezing transition from smectic to solid phase; the angular phase diagram is constructed. [S0163-1829(99)08517-3]

The layered crystal structure of high-T_c superconductors causes large anisotropy of these materials and brings a variety of specific features to their behavior. In combination with small coherence length, the layered structure induces intrinsic pinning of vortices between CuO layers.[1] For the vortex lattice the energy attains a minimum when the vortex spacing is commensurate with the distance between the layers. The theory of commensurable vortex structures was considered in Refs. 2 and 3. Oscillations of the magnetic moment induced by this commensurability effect have been observed experimentally.[4–6] These oscillations should be very sensitive to disorder in the vortex lattice, and probably represent the most delicate probe of the changes in the vortex arrangement.

Recently a theory[7] describing possible structures for vortex arrays closely parallel to the superconducting layers was proposed. It was suggested that three different vortex phases can be observed. In addition to the liquid and solid phases, a smectic vortex phase[8] is expected in the intermediate temperature range. In the latter, vortices are disordered within the layers, and are periodically arranged in the transverse direction, as in usual smectic liquid crystals. In this paper we study the commensurability oscillations in high quality $YBa_2Cu_3O_y$ single crystals, in order to reveal features related to the possible existence of smectic ordering of the vortices.

We have studied five flux-grown $YBa_2Cu_3O_y$ single crystals[9] that were long-time (300–700 h) annealed at 1 bar O_2, and had diamagnetic onset $T_c\approx91$ K and oxygen content $y=6.97\pm0.02$. These samples differ significantly in background impurities, so that their shielding currents vary by more than two orders in magnitude (from 80 A/cm^2 in sample CD to 4×10^4 A/cm^2 in sample MK at $B(\parallel c)=1$ T and $T=77$ K). Three samples VG, AZ, and CD were nearly monodomain as grown. Two others, WZ and MK, were detwinned using the uniaxial detwinning procedure.[10] The mosaic spread of four crystals was determined from (005) x-ray reflexes and did not exceed 0.02°. For the magnetic measurements we used a vector vibrating sample magnetometer (Ox-

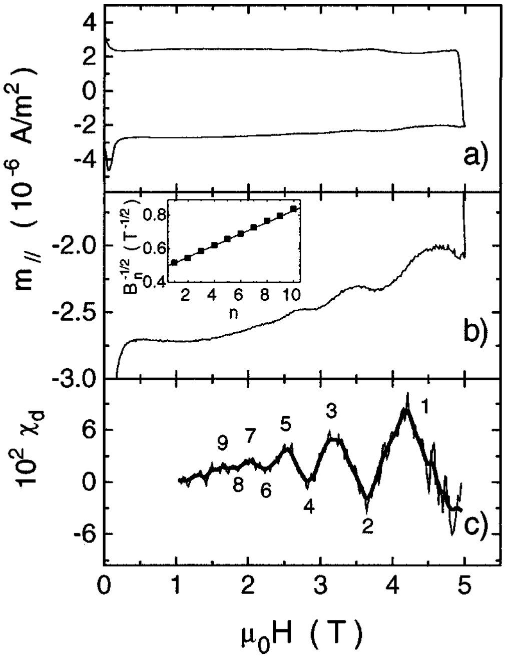

FIG. 1. (a) Magnetization loop of $YBa_2Cu_3O_y$ single crystal MK at 60 K with H accurately parallel to CuO planes. (b) Increasing-field leg on expanded scale. (c) differential susceptibility (thin line: direct differentiation; thick line: after preliminary smearing). Inset to Fig. 1(b) shows the extremum position vs number of extremum.

ford Instruments model 5[H]) equipped with two perpendicular pick-up coils. For the small angles ($<0.3°$) near the *ab* plane that we are interested in here, the m_{std} component parallel to the applied magnetic field gives the magnetization parallel to CuO planes $M_{\|}\approx m_{\mathrm{std}}/V$, and the transverse m_{ort} component corresponds to the magnetization along c axis $M_c \approx m_{\mathrm{ort}}/V$ (V is volume of the sample). The samples were rotated around the vertical axis with high angular resolution 0.01°, which was very important for our measurements; steps of 0.01° or 0.02° were used because of the sharp angular dependence of the commensurability oscillations.

Figure 1 shows the commensurability oscillations for the MK sample as an example of the behavior common for the other crystals. Because of their small amplitude and the monotonic background, a quantitative analysis of the oscillation directly from the magnetic moment is rather ambiguous; instead we used the differential susceptibility $\chi_d = dM/dH$. It was calculated numerically from the preliminary smoothed experimental curves. The degree of smoothing was always chosen to avoid distortion of the oscillations. For χ_d the oscillatory part is dominant and the quantitative analysis becomes robust. Moreover, we have found that the oscillations in $\chi_d(H)$ have clearly pronounced triangular shape [Fig. 1(c)]; this gives a clear indication of the transition between different commensurable vortex states at the cusps of $\chi_d(H)$.

Commensurability implies that an integer number k of intervortex spacings a_c in the direction normal to the layers equals an integer number m of the interlayer periods d. The vortex energy is minimized for $k=1$. Thus the corresponding commensurability condition is

$$a_c = \left(\frac{\sqrt{3}\Phi_0}{2\Gamma B} \right)^{1/2} = md,$$

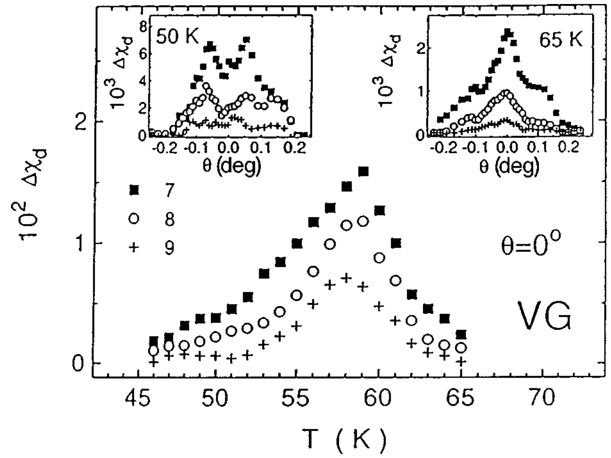

FIG. 2. Temperature dependence of the oscillation amplitude of the differential susceptibility of $YBa_2Cu_3O_y$ single crystal VG. The applied magnetic field is parallel to the CuO planes. Commensurability numbers $m=7$, 8, and 9 correspond to magnetic fields $\mu_0 H = 3.8$, 3.0, and 2.4 T, respectively. Insets: angular dependence of the amplitude above (right) and below (left) T_f.

where $\Gamma^2 = m_c/m_{ab}$ is the effective-mass anisotropy. The small anisotropy between a and b directions will be ignored here.

As a function of magnetic field, commensurability is periodic in $B^{-1/2}$ scale, with period $\Delta B^{-1/2} = d(2\Gamma/\sqrt{3}\Phi_0)^{1/2}$. Our experiments do show the periodicity in $B^{-1/2}(n)$ scale [inset in Fig. 1(b)]. For all our crystals of high oxygen content, as well as for the samples studied in Refs. 4–6, the period falls within quite a narrow range of $\Delta B^{-1/2} = (6.6 \pm 0.3) \times 10^{-2}\ \mathrm{T}^{-1/2}$ and, as expected, is practically independent of the field direction and temperature.

Using the expression resulting from the commensurability condition

$$\Gamma = \frac{\sqrt{2}}{2} \Phi_0 \left(\frac{\Delta B^{-1/2}}{d} \right)^2,$$

and the crystal lattice constant $d = 11.7$ Å,[11] one can find the effective mass anisotropy $\Gamma^2 = (30 \pm 5)$; this value agrees well with other measurements (e.g., Ref. 12). Note[11] that the unit cell contains two CuO layers spaced by ~3.4 Å, these bilayers are separated by the distance ~8.3 Å. The consistency of the above estimates shows that the spatial period of the intrinsic pinning potential coincides with the crystal lattice constant. This implies that there is just a single minimum energy well for a vortex within the unit cell in the middle of the bilayer spacing.

Although the oscillation period is temperature independent, the oscillation amplitude varies rapidly, with an unexpected sharp maximum (Fig. 2). This behavior, which is common to all the samples studied, is the central theme of the discussion and interpretation that follow. The peak position T_f (which we will refer to as a freezing temperature) shifts slowly to higher temperatures with the decrease of the commensurability order m, or correspondingly with the increase of the magnetic field. For fixed m, the values of T_f in different samples are similar. For example, for $m=7$ (corresponding to magnetic field $(4.0 \pm 0.4)T$ depending on the intrinsic parameters of different samples) T_f is 59, 66, 58,

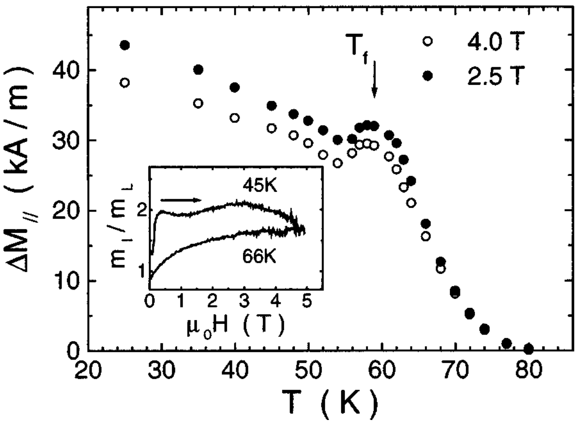

FIG. 3. Temperature dependence of the irreversible magnetization $\Delta M_{\parallel}$ of the $YBa_2Cu_3O_y$ single crystal VG. Inset: field dependence of the ratio of irreversible magnetic moments for H parallel to the long (m_L) and short (m_l) transverse sides of the sample. The arrow indicates the ratio between transverse sizes of the sample.

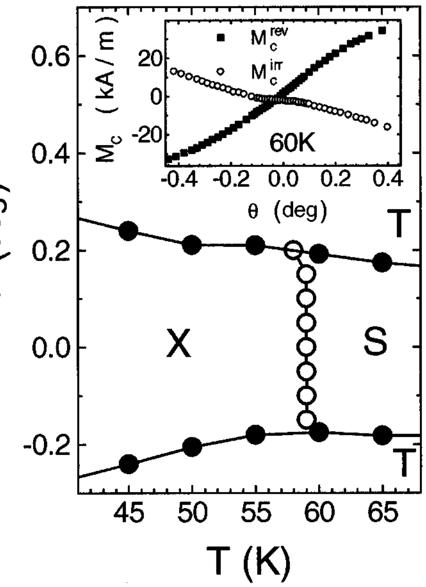

FIG. 4. Phase diagram for $YBa_2Cu_3O_y$ single crystal VG, as a function of temperature and angle θ between the field direction and the CuO planes for $m=7$. Open circles: position of the peak in the temperature dependence of the oscillation amplitude. Closed circles: vanishing of oscillations. Inset: angular dependences of the reversible and irreversible parts of M_c.

63, and 59 K for samples VG, AZ, WM, CD, and MK, respectively. Although there are small differences in T_f (which might be attributed to the difference in intrinsic and pinning parameters) the overall consistency of the values obtained is an indication of the thermodynamic nature of the peak anomaly, particularly given the very different levels of background impurities.

The angular dependencies of the oscillation amplitude differ above and below T_f. At high temperatures (right inset to Fig. 2) the amplitude exhibits a continuous decrease when the magnetic field tilts from the CuO plane. Below T_f two sharp maxima are observed on either side of the CuO plane direction (left inset in Fig. 2). In both cases the angle ($\sim 0.2°$) at which oscillations vanish is rather magnetic field and temperature independent.

Further support for a phase transition at T_f and an evidence to the nature comes from the temperature dependence of the magnetization hysteresis $M_{\parallel}$. The irreversible magnetic moment is dominated by the nonoscillating part, and therefore probes a different physical process. As will be discussed below, $M_{\parallel}$ is related to the current j_c^c along the c axis, which induces vortex motion parallel to CuO planes. $M_{\parallel}$ also shows a maximum at T_f (Fig. 3), supporting further a change in the nature of the vortex system at this point. Outside a very narrow angular region of about $\pm 0.2°$, the maximum in $M_{\parallel}$ as well as the oscillations disappear. At low temperatures the current $j_c^c \sim \Delta M_{\parallel}$ has a rather weak temperature dependence; however, above T_f it decreases very fast and vanishes significantly below either T_c or the expected[13] three-dimensional (3D) melting temperature $T_m \approx 90$ K. Such behavior is consistent with the melting of a vortex solid to a smectic structure. In fact, several theories[8,14] consider the smectic phase as a combination of solid in c direction with liquid in CuO plane. Bearing in mind the existence of fluctuations and background pinning we can tentatively relate fast decrease of $\Delta M_{\parallel}(T)$ to a second-order[7] melting that results in free vortex shear parallel to CuO layers. The presence of oscillations shows good vortex order in the c direction above T_f.

Let us briefly discuss the relation of the irreversible magnetic moment to the currents in the sample. According to the anisotropic Bean model,[15] Δm is determined by two different shielding currents. One (j_c^c) flows in c direction and another ($j_c^{\parallel}$) is parallel to CuO planes. These two components can be extracted by nondestructive measurements for a rectangular sample at $H \| ab$: two measurements of $\Delta M_{\parallel}$ with H directed parallel to the long and to the short sides of the sample suffice. For the dominant vortex penetration in the c direction, the magnetization $\Delta M_{\parallel} \approx j_c^{\parallel} t$ (t is sample thickness) should be equal for both field directions. For vortex penetration parallel to the CuO plane, $\Delta M_{\parallel}$ is dominated by the j_c^c current and is determined by $\Delta M_{\parallel} \approx j_c^c L/2$ where L is the crystal size in the direction perpendicular to the applied magnetic field.[15]

The only elongated sample studied was VG. As can be seen from the inset in Fig. 3 the ratio of the magnetizations for different directions of the magnetic field exceeds unity significantly, and at large H, correlates well with the ratio of the sample sides. This result means that the measured $\Delta M_{\parallel}$ is dominated by vortex motion parallel to CuO planes.

Using the high angular resolution measurements of the commensurability oscillations, we are able to construct the vortex phase diagram shown in Fig. 4. The boundary between the low (X) and high (S) temperature phases is determined by the position T_f of the peak in the oscillation amplitude. The behavior of $\Delta M_{\parallel}(T)$ and comparison with the theory[7] lead us to relate the lower temperature phase X to the 3D vortex solid phase (presumably vortex glass phase due to pointlike disorder) and phase S to a 2D smectic phase. Within 1 K the S-X boundary is temperature independent in accordance with the theory.[7] The boundaries to the high angle phase (T) are the angles φ^* at which the oscillation amplitude vanishes. Note also that in this angular range M_c^{irr} is very small, and M_c^{rev} is almost linear. We suggest therefore[6,16] that φ^* marks the boundary between the low angle locked state and a tilted vortex structure.

Theory also predicts a melting of the smectic phase to a vortex liquid at high temperatures. Resistive measurements[17,7] support a second-order transition to a liquid at $T_M \sim 90$ K but because of the vanishing pinning we cannot see this transition in our measurements. We should stress that our results give only indirect evidence of the transition to the smectic phase and the final confirmation requires structural measurements.

The decrease of oscillation amplitude in the solid phase with decreasing temperature can be connected with the rapid increase of pinning by pointlike disorder[18] which suppress order in the vortex structure. According to theory,[7] the smectic order should be insensitive to pointlike defects, hence thermal disorder and reduced pinning become the dominant factors leading to a decreasing oscillation amplitude with increasing temperature.

Summarizing, we have studied commensurability oscillations in $YBa_2Cu_3O_y$ ($y = 6.97 \pm 0.02$) single crystals. A sharp peak anomaly in the temperature dependence of the oscillation amplitude was found at $T_f \approx 60$ K. It is followed by other changes in the behavior of the amplitude and irreversible magnetization. Our analysis suggests that T_f reflects a freezing transition of a smectic vortex phase to a solid. These phases exist only within a very narrow ($\pm 0.2°$) angular interval of the direction of the magnetic field near the CuO planes.

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