Observation of vortex lattice melting in twinned YBa$_2$Cu$_3$O$_{7-x}$ using neutron small-angle scattering

C. M. Aegerter

Physik-Institut, Universität Zürich, CH-8057 Zürich, Switzerland

School of Physics and Space Research, University of Birmingham, Birmingham B15 2TT, United Kingdom

R. Cubitt

Institut Laue Langevin, Grenoble, France

S. L. Lee
School of Physics and Astronomy, University of St. Andrews, St. Andrews, Fife KY169SS, United Kingdom

D. McK. Paul

Department of Physics, University of Warwick, Coventry CV4 7AL, United Kingdom

M. Yethiraj and H. A. Mook

Solid State Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831-6033

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A neutron small-angle scattering study of the flux-line lattice in heavily twinned YBa$_2$Cu$_3$O$_{7-x}$ is presented. It is found that the diffraction signal disappears at temperatures well below $T_c$, associated with a melting of the flux lattice. The shape of the melting line is consistent with both a Lindemann criterion and the scaling expected for a vortex-glass transition with the superconducting parameters from the three-dimensional $XY$ model. The influence of twin planes on the structure of the vortex lattice and its melting is studied by applying the field at different angles to the $c$ axis. The results are compared with recent specific heat measurements on similar crystals. [S0163-1829(98)03022-7]

I. INTRODUCTION

High-temperature superconductors (HTS’s) are, in many respects, different from classic type-II superconductors. This is due to several factors, including the layered structure resulting in highly anisotropic superconducting behavior. Moreover, HTS’s have very short coherence lengths leading to extreme type-II behavior. The combination of these factors together with the high operating temperatures can produce exotic vortex phases such as the vortex-glass and the vortex-liquid phases. Over the past few years, these vortex phases have been intensively studied in various compounds, particularly in the model low- and high-anisotropy systems YBa$_2$Cu$_3$O$_{7-x}$ (YBCO) and Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (BSCCO). Until recently, the phenomenon of flux-line lattice (FLL) or vortex-glass melting in YBCO has only been observed by magnetization and transport measurements. This is because the vortex behavior is strongly influenced by the presence of twin-plane boundaries acting as strong, extended pinning sites and untwinned single crystals of sufficient size and quality have only recently become available. This is in contrast to BSCCO, in which there are no twin planes and where small-angle neutron scattering (SANS) and muon spin rotation (μSR) have both demonstrated the existence of a melting transition some years ago. Recently, a first-order melting transition has been observed using differential thermal analysis (DTA) on an untwinned crystal also used in magnetization and transport measurements. There have also been highly sensitive specific heat measurements on heavily twinned crystals of different purity. In the samples of a purity similar to those used in the present work, a jump in specific heat, consistent with a second-order phase transition, was found over a certain range of fields. For the very-high-purity crystals studied, a similar jump was found; however, on increasing the field a peak in specific heat, consistent with a first-order transition, could be observed. In this paper we present small-angle neutron scattering measurements on a heavily twinned YBCO crystal of a purity similar to those studied in Ref. 10, containing about 10% of nonsuperconducting inclusions (green phase).

In addition to the usual orientation of the field parallel to the $c$ direction, we applied the field at an angle of 51° to the $c$ axis and at 45° to the twin planes running along both a $\langle 110 \rangle$ and a $\langle 1\bar{1}0 \rangle$ direction (see Fig. 1). This is done in order to minimize the influence of the pinning to the twin planes on the vortex behavior. Such an arrangement has previously been shown to markedly influence the structure of the FLL. With the field parallel to the twin planes (and hence the $c$ direction) a fourfold symmetry has been observed previously, which has been attributed to the strong pinning effects of the twin planes combined with that of $ab$ anisotropy. Alternatively, such a morphology has been claimed to arise from $d$-wave effects. By applying the field
at angles to the twin planes greater than the depinning angle ($\sim 25^\circ$), a morphology with two triangular lattices, distorted by the superconducting $ac$ anisotropy was found. This change in the FLL structure can be clearly seen by comparing Figs. 2 and 3, which show typical neutron diffraction pictures for the two cases. For both orientations, we observe the disappearance of the diffraction signal at temperatures well below $T_c$. In this way we map the melting line over the field range 0.5–4.5 T. These results agree well with the irreversibility temperatures measured using a vibrating sample magnetometer on a small piece of the same crystal. Further discussion in Sec. III compares these results to those obtained in the recent specific heat measurements by Roulin et al.\textsuperscript{10}

\section*{II. EXPERIMENTAL DETAILS}

The experiments were carried out using the instrument D17 at the ILL, Grenoble, France. The neutron beam, collimated over a distance of 2.75 m with a wavelength $\lambda_n$ of typically 1.2 nm, was incident on the sample mounted in a horizontal-field cryomagnet capable of sample temperatures down to 1.5 K and a maximum field of 5 T. The scattered neutrons were detected by a position sensitive detector (approximately $0.8 \times 0.8$ m$^2$ total area with a pixel size of $\sim 10$ mm) and located at a distance from the sample that was varied between 2.88 and 3.43 m, depending on the applied field. The field was initially aligned with the neutron beam, to an accuracy of 0.1° both vertically and horizontally, by observing the diffraction pattern from the FLL in Nb. The sample was then aligned with the field either parallel or at $51^\circ$ to the crystallographic $c$ axis (see Fig. 1), by rotating the sample about the vertical axis.

The sample consisted of a large YBCO single crystal, of mass 7.8 g, grown using a melt processing technique. Measurements from an inductance coil mounted near the sample during the neutron experiments gave a value for $T_c$ of $\approx 92$ K. The small-angle neutron scattering data reported in this paper were obtained during three experimental periods at the D17 diffractometer. Data from the first experiment have been published separately\textsuperscript{15} and were obtained with the cryostat heater mounted on the sample stick. In the subsequent experiments the heating and thermometry were improved by placing the cryostat heater on the heat exchanger itself.
These changes of sample environment give rise to a small (approximately 1 K) discrepancy between the Pt resistance thermometer readings in the first experiment and those from later measurements. The critical temperature determined in situ in all cases, however, gives an independent calibration for the thermometry in the different setups. For this reason, the results presented below will always be given in terms of the reduced temperature $t = T/T_c$, with $T_c$ determined in each case by measuring the inductance at 1 kHz of a coil placed near the sample.

A brief consideration of small-angle scattering from a FLL is now given. Typical FLL plane spacings are ~50 nm (corresponding to a field of ~1 T). Thus the FLL can only be observed by using small angles and long wavelengths. In our experiments we used wavelengths between 1.2 and 1.5 nm, depending on the applied field. Using these typical values, we obtain scattering angles $2\theta = 2^\circ$ from the Bragg condition

$$\lambda_n = 2d_{hk}\sin(\theta),$$

where $d_{hk}$ is the layer spacing. Due to its magnetic moment ($\mu_n = -1.913\mu_N$) a neutron may interact not only with the nuclei in a sample, but also with magnetic inhomogeneities. This allows the observation of FLL’s in superconductors with small-angle neutron scattering.\(^{11,12}\) The intensity of scattered neutrons in a Bragg reflection, integrated over a small-angle neutron scattering.\(^{16,17}\) The intensity of scattered neutrons in a Bragg reflection, integrated over the rocking curve, then be calculated from the magnetic cross section to be

$$I_{hk} = \frac{\pi\mu_n^2\phi\lambda_n^2V}{8\Phi_0^2\tau_{hk}}|F(\tau_{hk})|^2,$$

where $\phi$ is the flux of incoming neutrons, $V$ is the sample volume, and $\Phi_0 = h/2e$ is the magnetic flux quantum. The magnetic form factor $F(\tau_{hk})$ is a measure of the $\tau_{hk}$ Fourier component of the spatial variation of the magnetic field in the sample and can be easily calculated for an extreme type-II superconductor in the London approximation. Due to their long penetration depths and short coherence lengths, HTS’s are extremely type II and one obtains

$$F(\tau_{hk}) = \frac{B}{1 + \lambda^2\tau_{hk}^2},$$

Here $\lambda$ is the (temperature-dependent) magnetic penetration depth and $B$ is the average induction. Thus, for fields greater than $B_{c1}$, corresponding to $\lambda \tau_{hk} > 1$, the scattered intensity depends on the penetration depth of the sample as $I \propto \lambda^{-4}$. Since the HTS’s have very long penetration depths ($\lambda_{ab} \approx 150$–200 nm at low temperature and even longer close to $T_c$) the scattered intensities are very weak compared to conventional type-II superconductors.

Due to these difficulties, intrinsic in neutron scattering studies of HTS’s, long counting times were required. Furthermore, there is very strong background scattering from extended defects in the sample. To obtain clear scattering images from the FLL in the sample, we subtracted a background measurement taken above the transition temperature $T_c$. Due to thermal contractions of the cryostat and sample stick, this background was numerically shifted by a fraction of a detector pixel in order to obtain good subtractions near the beam stop. Typical counting times were 1 h each for a foreground and a background.

**III. RESULTS AND DISCUSSION**

A typical diffraction pattern for the field parallel to the $c$ axis is shown in Fig. 2, at low temperatures and a field of 2 T. There is a distinct fourfold symmetry visible in the scattering. This should not, however, be interpreted as the observation of a square lattice. As pointed out earlier,\(^{13,14}\) measurements performed on similar crystals using higher resolution were able to distinguish 24 diffraction peaks corresponding to four different orientations of a hexagonal lattice. All of these orientations, corresponding to different domains in the sample, have strong diffraction peaks at 45° to the horizontal, consistent with strong pinning of FLL planes to twin boundaries. The different FLL orientations with the same strong 45° spots correspond to different orientations of the crystallographic $a$ and $b$ axes due to twinning.\(^{11,12}\) The slight distortion from an ideal hexagonal lattice for the different orientations can then be understood as due to the small $ab$ anisotropy present in YBCO. It can thus be seen that the main features of the FLL in the case where the field is parallel to the $c$ axis is determined by the twin boundaries.

In the case of an inclined field (at 51° to the $c$ axis), the situation is drastically changed. This is shown in Fig. 3, where a typical diffraction pattern at low temperatures and a field of 1.5 T is shown. The figure does not show a full diffraction pattern, as the scattering angles of the farthest spots differ by more than the width of the rocking curve. Therefore, the left-hand side of the pattern was deliberately illuminated by rocking close to the Bragg condition for spots on that side. One can clearly see six Bragg peaks on the left half, showing a distorted hexagonal symmetry. Taking the right-hand side of the pattern into account, this results in 12 spots corresponding to two orientations of a hexagonal lattice distorted by $ac$ anisotropy. Assuming that the heavy twinning in our sample results in an effectively isotropic $ab$ plane, this distortion can be parametrized by the ratio of the minor ($\alpha$) and major ($\beta$) axes of the ellipse on which Bragg spots are situated. This should scale with length as

$$\left(\frac{\alpha}{\beta}\right)^2 = \cos(\vartheta)^2 + 1/2\gamma^2\sin(\vartheta)^2,$$

where $\vartheta$ is the angle between the field and the $c$ axis and $\gamma = \lambda_c/\lambda_{ab}$ parametrizes the anisotropy. For an angle of 51°, this scaling function is still determined almost entirely by the cosine term. To obtain an estimate of the $ac$ anisotropy, we performed $\mu$SR measurements with the field both parallel and perpendicular to the $c$ direction.\(^{15}\) This gives an anisotropy of $\gamma = 4.3(2)$, in agreement with earlier determinations by SANS of 4.5(2).\(^{14}\) This parameter will be used again in the discussion of the melting line.

The temperature dependence of the scattered neutron intensity, summed over all diffraction peaks, is shown for applied fields of 1 and 4 T in Fig. 4. The temperature well below $T_c$, at which the intensity falls to zero, is interpreted as the melting of the FLL.\(^4\) In both cases the field was at 51° to the $c$ axis. There are several interesting features in this figure. First, the scattered intensity is zero above the
melting temperature. As is the case in BSCCO, this is most probably due to the entanglement of the vortices (disorder along their length) above the melting temperature, which reduces the scattered intensity to zero. The observation of entangled (or decoupled) vortex lines has already been reported from measurements applying a current at the top of a sample, while recording the voltage difference on the bottom. Above the irreversibility line, the voltage measured on the melting temperature is different for the low-field data from 14 .

The temperature dependence of the FLL signal below the melting temperature is different for the low-field data from the high-field data. We note that this temperature dependence will arise from two causes: (i) the variation of the superconducting penetration depth, which for a perfect FLL would give a temperature dependence of intensity according to Eqs. (2) and (3), $I(T) \propto \lambda^4(T)$, and (ii) the variation of the FLL order as thermal destruction of the flux line structure increases with temperature. Clearly, the second term is the cause of the zero intensity as the flux lattice melts, but the first term will contribute to the temperature dependence below $T_m$, so we consider its contribution. The Ginzburg-Landau mean field picture gives a variation of the penetration depth as $\lambda(T) = \lambda(0)(1 - t)^{1/2}$. If we assume that $\lambda$ is little affected by our applied fields, which are much less than the mean field $H_{c2}$ at the melting temperature, then this gives $I(t) = I_0(1-t)^2$. However, it appears from microwave measurements at zero field that critical fluctuations renormalize the temperature dependence of the penetration depth to $\lambda(T) = \lambda(0)(1-t)^{1/3}$; this is the dependence expected in the three-dimensional (3D) $XY$ model and gives $I(t) = I_0(1-t)^{5/3}$. Whatever the functional dependence of $\lambda(T)$, unless it tends to infinity at the melting temperature, it will not affect the temperature dependence very close to $T_m$. This will be controlled by the FLL order. Hence it is of some importance to note that within our experimental error, the intensity falls to zero continuously at $T_m$, implying that the transition is not of first order. At 4 T the FLL intensity falls to zero approximately linearly with temperature, while the falloff is much more gradual at 1 T. This can be quantified by fitting a power law of the form $I(t) = I_0(1-t)^n$, where $n$ parametrizes the shape of the temperature dependence and is $\sim 1$ at 4 T and $\sim 1.5$ at 1 T. If we regard the scattered intensity as the square of a phenomenological FLL order parameter, then at 4 T the intensity surprisingly follows the temperature dependence expected for an order parameter in mean field theory. At low fields, however, this is not the case. Since we do not know of a detailed theory for these results, we confine ourselves to reporting the rest of our data in terms of the power law $I\propto(1-t/t_m)^n$ that best describes the variation of the intensity below $T_m$. The results are summarized by Fig. 5, where we see a general trend to smoother transitions at low fields.

It is interesting to compare this field dependence with that of the jump observed in specific heat (Fig. 11 in Ref. 10). For their sample with $x = 0.03$, which is very similar to the one
studied here, the step shows a comparable qualitative dependence. \( \Delta C \) in that sample is almost field independent at fields above 5 T and then decreases below fields \( \sim 3 \) T, until the jump cannot be resolved at fields lower than 1.5 T. This corresponds to our observation of the field dependence of the exponent \( n \), being field independent above \( \sim 4 \) T and increasing at lower fields. Viewing the scattered intensity as a FLL order parameter as discussed above, an increasing exponent \( n \) corresponds to a smaller change in the slope of the order parameter at the melting transition. Thus a bigger exponent \( n \) corresponds to a smaller jump in specific heat, consistent with the observation.

As already mentioned, the melting transitions that we observe are apparently continuous. Such a behavior is consistent with a second-order phase transition, as observed in similar crystals by Roulin et al., using specific heat measurements on single crystals, where DTA measurements demonstrate the existence of a first-order transition. Similar results are also found in specific heat measurements on ultrahigh-purity twinned YBCO crystals, where a peak in specific heat was found at intermediate fields. Moreover, in SANS experiments on a large untwinned crystal (with very few remaining twin planes), show a much sharper transition than those presented here at high fields. Although no clear jump in neutron intensity, as expected for a first-order transition, could be found in that sample, the data are not inconsistent with a smeared first-order transition.

A second-order transition, as observed in this and an other work, has been predicted for the melting of a vortex glass. The glass transition line is predicted to scale as 

\[
B_G \propto (1 - t)^{4/3}
\]

in the limit where the 3D \( XY \) model is applicable. However, as we observe a diffraction pattern the supposed vortex glass has to be what is called a "weak glass" or "Bragg glass." It is thus rather surprising that such weak glassiness may turn the first-order transition expected for an ideal lattice into a second-order transition. The different available data seem to indicate that not only the twin planes, but also the degree of purity may be important in modifying the order of the transition.

In a twinned crystal containing no green phase, Junod et al. observed a melting line of first or second order depending on the applied field. Similarly, DTA measurements on untwinned crystals find no latent heat at the melting transition in low fields \( (B < 0.5 \) T), indicating that there is either no transition or a second-order transition at low fields. Moreover, the first-order (high-field) and second-order (low-field) transitions found in Ref. 9 lie on the same phase line, indicating a single process.

Setting aside the nature of the transition, we may still obtain predictions as to the position of the melting line in a \( B-T \) phase diagram using a Lindemann approach. The Lindemann criterion for the melting of a lattice involves calculating the thermal fluctuations of the lattice. When these fluctuations reach a certain fraction of the lattice constant, the phase transition is thought to occur. This leads to a criterion for melting

\[
\langle u^2(T) \rangle = c_L^2 a_0^2,
\]

where \( c_L = 0.1-0.2 \) is the Lindemann number and \( a_0 \sim (\Phi_0/B)^{1/2} \) is the intervortex distance. Calculating the thermal fluctuations using the (dispersive) elastic constants of a FLL then leads to the expression

\[
B_m(T) = \frac{\Phi_0^4}{1.5 \pi k_B T \lambda(T)^2}.
\]

where \( k_B \) is Boltzmann’s constant. Using the temperature dependence of the penetration depth in the 3D \( XY \) model presented earlier, this gives a similar temperature dependence of the melting temperature to that predicted for the vortex-glass transition [see Eq. (5)]. For temperatures close to \( T_c \), where the 3D \( XY \) model is applicable and the \( 1/T \) in Eq. (7) may be regarded as constant, the two predictions are exactly the same and cannot be distinguished. At higher fields, however, where \( 1/T \) becomes important and mean field theory might be applicable, the two theories predict different melting lines. Such fields are, however, beyond our range of measurement.

Figure 6 shows the melting temperatures, as determined from the point above which the scattered intensity disappears, for both orientations studied. The line through the data
is a fit to $B_m = B_0(1-t)^{4/3}$, corresponding to Eq. (7). According to the anticipated angular scaling for the melting line,

$$B_m(\theta) = B_m(0)/[\cos^2(\theta) + 1/\gamma^2 \sin^2(\theta)]^{1/2},$$

the prefactors in the two orientations are expected to differ by a factor of $B_0(0)/B_0(51) = 0.66$ (assuming $\gamma = 4.2$; see above). From the fitted values $B_0(0) = 100(10)$ T and $B_0(51) = 135(10)$ T, we obtain 0.74(9), in fair agreement with the prediction. Taking into account the values for the anisotropy and the penetration depth as determined by $\mu$SR,18 we can then determine the Lindemann number to be $c_L = 0.15$, also in reasonable agreement with theoretical expectations. It should be noted, however, that Kwok et al. find an angular dependence of the melting temperature showing a cusp around $\theta = 0$. In contrast to these transport measurements however, SANS and specific heat measurements10 find a clear melting transition for fields parallel to the c axis. The anisotropies determined from the melting line and the upper critical field $B_{c_2}$ parallel and perpendicular to the c axis. The anisotropies determined from the melting line are consistently smaller than those determined from $B_{c_2}$ but as in the case presented here the experimental uncertainty is too large for a quantitative discussion of such a small effect. It is possible that in the presence of twin plane pinning, SANS and specific heat measurements give a better indication of melting than transport measurements because the former tell us about equilibrium thermodynamic properties of the system. Considering that the structure of the FLL is mainly determined by the twin planes (see Figs. 2 and 3) it is surprising, however, that the twin planes do not have a stronger influence on the melting temperatures.

IV. CONCLUSIONS

In conclusion, we have presented SANS measurements of the melting of the FLL in heavily twinned YBCO. The data are consistent with a recent specific heat study on crystals with similar twinning and purity.10 The findings are consistent with a second-order phase transition, at a phase boundary as predicted by a Lindemann criterion. The observed melting line also shows the field and temperature dependence expected for a vortex-glass melting line as predicted from the 3D $XY$ model. With the limited range of fields studied we are not able to distinguish between the two theoretical predictions. The effect of the twin-plane boundaries and the superconducting anisotropy on both the melting line and the FLL structure were studied by applying the field parallel to the c direction and at an angle of $51^\circ$. It was found that the lattice structure is strongly influenced, with the diffraction pattern showing a fourfold symmetry (but not a fourfold lattice) for the field parallel to c, whereas two distorted hexagonal lattices are found for the field at $51^\circ$. In contrast, the melting line is only very slightly affected by the presence of the twin planes, consistent with the findings of specific heat measurements.10

Finally, we find unexpected behavior in the temperature dependence of the scattered intensity close to the melting temperature. The temperature dependence of the scattered intensity, as parametrized by the exponent $n$, depends on the applied field, falling more rapidly (decreasing $n$) with increasing field. This behavior is reminiscent of that observed by heat capacity measurements on a similar sample,10 where the jump in heat capacity at the melting transition rises with field. However, in both these cases, the results reflect the temperature dependence of not only the FLL structure, but also the underlying superconducting parameters, such as the penetration depth and the coherence length.

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