Small-Angle Neutron Scattering Study of Flux Line Lattices in Twinned $YBa_2Cu_3O_7$

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(Received 8 September 1992)

A small-angle neutron study of vortex lattices in $YBa_2Cu_3O_7$ showed a diffraction pattern with square symmetry when the applied field was along the *c* axis with the four main spots aligned with the crystallographic {110} directions. With the field 30° from the *c* axis, a hexagonal lattice exists. At intermediate angles, a fraction of the flux lines follow the *c* axis and the two lattices coexist. The temperature dependence of the intensity deviates markedly from conventional predictions.

PACS numbers: 74.72.Bk, 61.12.Ex, 74.60.−w

In conventional type-II superconductors, when an applied field is above H_{c1} but below H_{c2}, flux lines penetrate the superconductor forming a 2D lattice. Small-angle neutron scattering (SANS) techniques have been used successfully [1,2] in the past to study these flux line lattices in traditional superconducting materials. Recently, similar experiments [3,4] have been carried out in high-T_c superconductors.

The properties of the flux lattice as a function of field and temperature are important both for a better understanding of the nature of the interaction causing pair formation in high-T_c superconductors and also for applications of the material in devices. $YBa_2Cu_3O_7$ has one of the strongest pinning characteristics of all the high-T_c superconductors and is of particular interest in applications since larger pinning leads to higher critical currents. The temperature (T) dependence of the London depth is vital information that can be measured using SANS. In the London model, the intensity of the Bragg peaks is proportional to the inverse fourth power of the London penetration depth (λ_L), and hence, this length can be accurately determined. The angular dependence of the scattering yields information about the long-range order and symmetry of the flux lattice, as well as the average straightness of the individual lines.

In this Letter we report on detailed measurements of the flux lattice in a large single-crystal sample of $YBa_2Cu_3O_7$ (YBCO). We made measurements with the field parallel to the *c* axis and with the field at an angle of up to 30° from the *c* axis. The T dependence of the intensity of the flux line scattering has been obtained much more accurately than in previous work. We also present the behavior of trapped flux as the temperature was varied.

The measurements were made on a large twinned single-crystal sample of YBCO that weighed 7.8 g. The mosaic width of the nuclear 006 Bragg peak was approximately 0.7°. The specimen had a superconducting transition temperature of 92.4 K with a width of 1 K. The sample contained an impurity phase that was about 15% of the total crystal volume. This phase is found in well separated regions of the crystal and is expected not to alter the behavior of the fluxoid lattice to any large extent. The experiment was carried out on the W. C. Koehler 30-m SANS instrument at the Oak Ridge National Laboratory. The neutron wavelength was 4.75 Å ($\delta\lambda/\lambda \approx 5\%$). The transmission of the single crystal was 70%. The instrumental resolution was roughly equal in horizontal and vertical directions in all cases.

The horizontal applied field, measured to be 0.8 T with a calibrated Hall probe, was along the incident beam direction. The sample and the magnet could be rotated together as a unit. In addition, the sample could be rotated independently of the magnet. The sample temperature was stabilized in a Displex (closed-cycle helium) refrigerator, with a resistive heater to maintain any temperature in the range 10–110 K for the extended period of the experiment. The absolute accuracy in the measured temperature was ± 0.5 K with a stability of ± 0.1 K. The sample was then aligned *in situ* by measuring the angular position of the 004 nuclear Bragg reflection on both sides of the incident beam to minimize the error in alignment. The alignment was good to within 0.2°; it was verified by strongly angle-dependent metallurgical small-angle scattering observed when the *c* axis was exactly 90° from the direct beam.

The signal from the flux lines was smaller than the small-angle scattering from the sample and was observed as a difference between the signal with and without flux lines in the sample. The flux lattice signal was observed after cooling through T_c in the applied field. Background runs were taken with the field on, above the transition temperature at 100 K. Some difference in scattering (between 100 K and low temperatures) was present and could not be avoided, but this grew smaller as the temperature differential was decreased; hence, the errors were small as the signal from the flux lines decreased.

The sample was aligned with the *c* axis in the horizontal plane and the (100) about 22° from the horizontal. The flux lattice was observed both for the high-symmetry geometry with field parallel to the c axis, as well as for an orientation where the applied field was up to 30° in the horizontal plane from the crystallographic c direction. Because neither the (100) nor the (110) was in the horizontal plane, the rotation was not about a high-symmetry crystallographic axis.

With the applied field and the c axis along the incident beam, a diffraction pattern with square symmetry was observed in the difference between 11 and 100 K data, shown in Fig. 1(a). All spots could be seen simultaneously due to the large mosaic of the flux line lattice, discussed later. The four intense spots arise by diffraction from flux lattice planes parallel to the orthorhombic {110} planes of the crystal. Since this is the direction along which twin plane defects are known to exist, it is probable that pinning due to these defects gives rise to the square pattern. The two orthogonal sets of twin planes give rise to four spots 90° apart. Scattering with square symmetry (referred to as square spots, for brevity) has been observed previously in SANS measurements of the flux lines in YBCO with field parallel to the c axis [3,4]. Less intense features were also seen 45° from the main spots. The origin of these secondary features is not clear, but they are possibly due to defects in the flux lattice or due to additional pinning along the {100} directions. The simplest structure that corresponds to the observations is two domains of 1D arrangements constrained by the orthogonal sets of twin planes. The morphology of the lattice is most likely to be very dependent on the density of twin plane defects and the results reported here are on a strongly twinned sample.

With the field along the c axis, the square spots have been interpreted in the past as being due to two triangular lattices that are rotated 90° from each other. Each of these would give rise to six Bragg spots; hence, twelve peaks 30° apart would arise from two triangular lattices. Of the twelve, four would be strong due to pinning effects; the rest would be somewhat smeared. However, the evidence suggests that this is not the case in the present sample. The intensity between the spots is low and its mosaic spread is only slightly larger than that of the main square spots. A Gaussian fit to the sector average of one of the spots (on the Bragg condition) indicates flux lattice intensity centered at $q = 0.0124 \pm 0.002$ Å^{-1}. The q calculated from the applied field is 0.0124 Å^{-1} for a square lattice and 0.0133 Å^{-1} for a triangular lattice. The fit obtained with the center constrained to 0.0133 Å^{-1} is considerably worse. It is unlikely that the lattice is triangular. The measured $\Delta q/q$ of the lattice is $(40 \pm 5)\%$. After accounting for instrumental resolution contributions, the intrinsic $\Delta d/d$ is determined [5] to be approximately $(10 \pm 15)\%$. Hence, our present measurement is insufficient to discriminate between a vortex lattice and a vortex glass.

Decoration experiments [6] in YBCO show a triangular lattice with long-range order. The flux line lattice spacing in these studies is much larger (~ 1 μm) since the applied field is much smaller. The square symmetry we observe is almost definitely due to pinning effects on a small length scale that is not significant in a decoration experiment. It is possible that a change from triangular to square morphology occurs at an intermediate field. Another decoration experiment [7] in YBCO in untwinned regions has suggested pinning by defects other than twin boundaries. Although this is possible, we see that for the **B**$\|$**c** geometry, at least the alignment of the

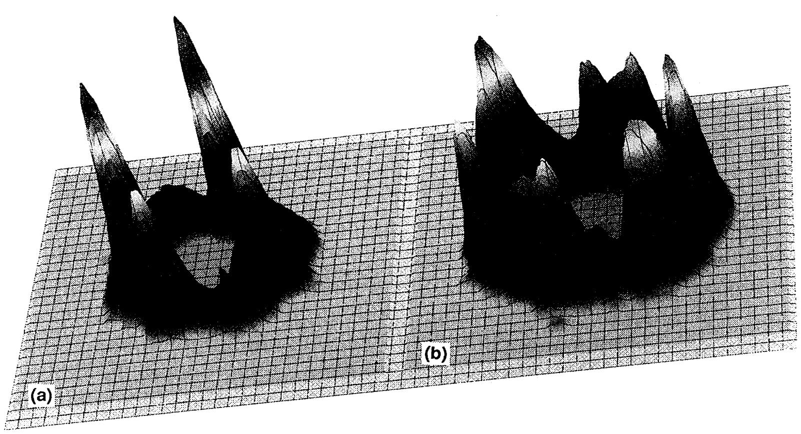

FIG. 1. (a) Diffraction pattern by the flux line lattice with an 0.8 T field applied parallel to the c axis has fourfold symmetry. The flux line lattice peaks are aligned along the {110} direction. [Sample-detector distance (SDD) = 15.8 m; 22×15 mm aperture at source and 8×14 mm aperture at sample]. (b) A distorted hexagonal lattice is formed when the applied field is 30° from the crystallographic c axis. (SDD = 19.0 m; 17 mm diam and 8 mm diam circular apertures.) Note that the intensities of the hexagonal spots have been scaled up relative to the square spots.

flux lattice is controlled by twin planes. When the field is turned off at low temperatures in a field-cooled sample, the scattered intensity remains at $(94\pm2)\%$ of the field-on value. This indicates strong pinning.

From the rocking curve, the measured mosaic width of the square flux lattice was $2.0°\pm0.3°$, which is large compared to widths of vortex lattices in conventional superconductors. This mosaic is a measure of the straightness of the flux lines along the applied field direction or a measure of the length over which they remain straight. If it is assumed that the mosaic is entirely due to finite sample size (i.e., flux line length), this length must be at least 1.5 μm. This is a lower limit since the measured rocking width convolutes this length effect with any meandering of the flux lines away from the c axis. A single flux line sees a square grid of defects since twin plane defects do not span the length of the crystal long the c axis. The mosaic may represent a degree of zigzag of the flux lines as they adjust to pin themselves along defects as much as they can. Additional experiments on lattice morphology are in progress. There is no change, within the estimated errors, in the rocking curve width of the square spots between 11 and 70 K.

When the applied field was at an angle to the c axis, a substantial fraction of the flux lines were parallel to the c direction of the crystal rather than along the external field direction. As the angle between the field and the c axis is increased above 5°, two sets of diffraction spots are observed. A square arrangement along the c axis coexists with a hexagonal arrangement along the applied field direction. These measurements were made on trapped flux (i.e., with the field off). At 30° from the c axis, the square spots were suppressed and a distorted hexagonal lattice was seen [Fig. 1(b)]. In contrast to the square lattice, the tangential spread of these diffracted spots was marked and much larger than the instrumental spread. The full width at half maximum of the Bragg peak in the tangential direction, a measure of the degree of disorder in the plane perpendicular to the length of the flux lines, was 33° ($\pm4°$) for the distorted hexagonal lattice and was resolution limited for the square lattice. The existence of spots is indicative of long-range orientational order in the system. If the structure had only short-range order, the 2D data would be spread out into a ring of scattering at the expected q instead of the observed spots, since a unique direction would not be maintained from one end of the sample to the other. Within experimental error, the mosaic width (which measures the disorder along the length of the flux lines) of the hexagonal lattice was the same as that of the square lattice.

The T dependence of the flux lattice intensity is most unusual. Unlike any standard theoretical predictions for the order parameter, the intensity falls very rapidly for $T<T_c/3$ as the temperature is increased, as shown for both the square and hexagonal spots in Fig. 2. One is reminded that the sample has a very sharp transition ($T_c=92.4\pm1$ K) and has been characterized by lattice

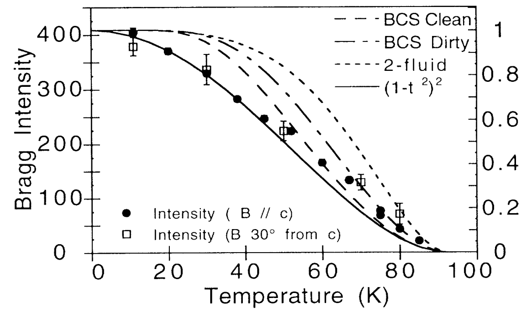

FIG. 2. T dependence of the intensity of the square spots (averaged over all four reflections) and of the hexagonal spots (averaged over six reflections and scaled) are indicated by open and closed symbols, respectively. The dashed lines represent theoretical predictions from BCS and two-fluid models. The solid line is given by $(1-t^2)^2$.

parameter measurements as being very uniform; hence the T dependence of the intensity is not due to variations in oxygen stoichiometry. We found that the mosaic width of the square spots at 11 and 70 K remained the same, and thus broadening the mosaic is not the cause of the observed drop in intensity. The observed T dependence is also consistent with that seen [3,4] (with much larger error bars) on other samples at other fields.

Since the T dependence of the hexagonal spots (applied field 30° from the c axis) is the same as for the square spots, it appears that this behavior of the order parameter is intrinsic. As shown in Fig. 2, BCS s-wave [8] and two-fluid models predict a much slower drop in intensity. The T dependence of both the triangular and the square pattern intensities is approximated better, although not exactly, by $(1-t^2)^2$, where t is the reduced temperature. The observed long-range order does not disappear until at least 85 K, and there is no change except in the intensity of the spots. The scattering does not deteriorate into a ring. The temperature regime studied here is ≤ 85 K and thus below the irreversibility line for this sample. Recent microwave and muon-spin-rotation (μSR) work [9] on single-crystal samples with $T_c=93$ K shows a $1-t^2$ and a $1-t^{2.37}$ dependence of λ_L^{-2} by the two techniques. However, earlier μSR results [10] on ceramic samples showed agreement with the two-fluid model.

The T dependence of the intensity due to the trapped flux in a field-cooled sample when the field is turned off at low temperature is an indication of the strength of the pinning. For the high-symmetry case of **B**$\parallel$**c**, the ratio of intensity with field off to field on as a function of temperature is shown in Fig. 3. The intensity due to trapped flux up to 50 K is 94% ($\pm2\%$) of the field-on value. Above 50 K, the trapped flux was sharply lower. However, on the time scale of our measurement (3 h), the signal at each temperature did not vary with time, even at 75 K. The hexagonal lattice was also strongly pinned at 11 K, but no measurement was made of the T dependence of

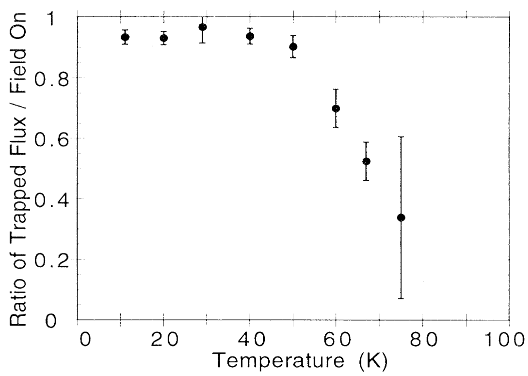

FIG. 3. T dependence of flux pinning. Ratio of intensities of (square) Bragg spots with field off to field on. (Before measurements, taken with increasing temperature, the sample was always field cooled to base temperature.)

the pinning in this geometry. The sample was aligned such that the (110) direction was ~23° from the rotation axis; hence twin planes should not be directly involved in the pinning. The fact that square spots occur only when the flux lines are along the c axis and that the (10) axis of the flux lattice coincides with the (110) crystal axis suggest that the square symmetry is due to the twin-plane defects. However, the fact that the hexagonal lattice (field 30° from c) trapped flux almost as efficiently as the square lattice implies additional pinning mechanisms.

The value of the London penetration depth can be estimated from the absolute integrated intensity of the spots. If we take half the superconducting volume of the sample to be in each of the 1D domains, the penetration depth in the basal plane (λ_{ab}) was 1625 ± 50 Å. If the scattering was indeed due to a square lattice rather than two sets of pinned lines, then a larger value of 1850 Å is obtained for the London depth. (If a square lattice is assumed, a correction is made to account for 15% of the intensity being between the strong spots.) The $T=0$ value will be somewhat smaller since the lowest temperature of our work was ~11 K where the intensity had not saturated. Our measurement is a lower limit on the intensity, hence an upper limit on the London depth. The absolute intensity calibration (accuracy $= \pm 5\%$) was obtained by comparison with a sample of Al with voids whose scattering has been well characterized [11].

Irrespective of the structure we assume, the value of λ_{ab} obtained is significantly larger than μSR [10] and other [12] estimates of ~1400 Å. If the lattice had a random static distortion together with long-range order, this would lower the intensity in the spot without broadening the peaks. This type of local disorder would also decrease the intensity of higher-order peaks.

In conclusion, we observe in $YBa_2Cu_3O_7$ a flux line lattice that is affected by directional pinning. We see that the variation of the order parameter with temperature is contrary to conventional s-wave predictions. The intrinsic nature of the T dependence of the flux lattice intensity is confirmed by measurements at two different geometries; experiments at different applied fields can further verify this.

We are grateful to Brian Chakoumakos for sample annealing and characterization, to Amit Goyal for microscopy studies on the sample, to Bill Hamilton and John Hayter for SANS software modifications, and to Steve Lee for useful discussions. This work was supported in part by the Division of Materials Sciences, U.S. DOE under Contract No. DE-AC05-84OR21400 with Martin Marietta Energy Systems, Inc.

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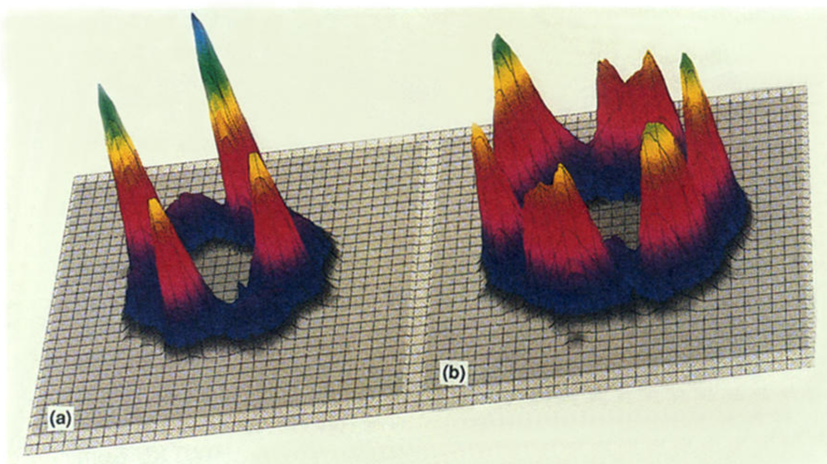

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