Observation of a Triangular to Square Flux Lattice Phase Transition in YBa$_2$Cu$_3$O$_7$

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We have used the technique of small-angle neutron scattering to observe magnetic flux lines directly in an YBa$_2$Cu$_3$O$_7$ single crystal at fields higher than previously reported. For field directions close to perpendicular to the CuO$_2$ planes, we find that the flux lattice structure changes smoothly from a distorted triangular co-ordination to nearly perfectly square as the magnetic induction approaches 11 T. The orientation of the square flux lattice is as expected from recent $d$-wave theories, but is 45° from that recently observed in La$_{1.83}$Sr$_{0.17}$CuO$_{4+\delta}$.

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The technique of small-angle neutron scattering (SANS) from flux lines has a long and honorable record in measuring the properties of flux lines in superconductors. However, it continues to bring new dividends, especially in unconventional superconductors, since important information about the nature of the superconducting state is often revealed by the flux line lattice (FLL) structure, for example $^1$$^1$, $^2$$^2$, $^3$$^3$. The diffraction pattern not only reveals the co-ordination and perfection of the FLL, and its correlation with the crystal lattice, but also the absolute intensity may be used to determine the actual spatial variation of the magnetic field within the mixed state and the values of the coherence length and penetration depth $^1$$^1$, $^2$$^2$, $^3$$^3$. In the simplest approximation, flux lines would order in a regular triangular FLL; however, anisotropy of the Fermi surface or of the superconducting order parameter can cause distortions of the triangular lattice or transitions to other structures. The simplest situation in a high-$\kappa$ material is anisotropy of the magnetic penetration depth associated with effective mass anisotropy $^3$$^3$; for example, the anisotropy in the $ab$ plane of YBa$_2$Cu$_3$O$_{7-\delta}$ (YBCO) leads at low values of field to a corresponding distortion of triangular FLLs $^3$$^3$. At the lower values of $\kappa$ in borocarbides, “non-local” effects are expected and observed $^4$$^4$, $^5$$^5$, $^6$$^6$, to give a variety of FLL distortions and transitions. If the superconducting order parameter has a different symmetry from that of the crystal, this can again be revealed via its effects on the FLL structure, for instance in the $p$-wave superconductor Sr$_2$RuO$_4$ $^7$$^7$, $^8$$^8$. In general in $d$-wave superconductors $^9$$^9$, $^{10}$$^{10}$, there is expected to be a tendency towards a square FLL as the field is increased and the anisotropic flux line cores overlap. This may be the cause of the FLL phase transition recently observed in overdoped La$_{1.83}$Sr$_{0.17}$CuO$_{4+\delta}$ (LSCO) at the comparatively low field of 0.4 T $^3$$^3$. According to $^{12}$$^{12}$, the FLL nearest-neighbour directions should lie along the directions of the nodes of the order parameter, which would be at 45° to the Cu-O bonds in the superconducting layers. This is not, however, the orientation of the square FLL observed in LSCO $^2$$^2$. The orientation may instead be controlled by band structure effects $^{13}$$^{13}$, even if the symmetry of the FLL is controlled by $d$-wave effects. It has been suggested that an peak effect in magnetisation measurements on overdoped YBCO may be a signature of a continuous triangular-to-square FLL transition in this material at high fields $^{14}$$^{14}$. However, others have suggested that there is a glass transition in this region $^{15}$$^{15}$. Only by direct measurements may such suggestions be tested and the correlation between FLL and crystal lattice (or superconducting order parameter) determined.

Our experiments were performed on the SANS-I instrument at SINC, PSI, Switzerland. Cold neutrons (8 Å to 14 Å, with a FWHM wavelength spread of 10 %) were collimated over distances from 4.5 m to 15 m, depending on the field and hence $q$-range required. The diffracted neutrons were registered on a 128 $\times$ 128 $\times$ 7.5 mm$^2$ multidetector, which was similarly adjustable in distance from the sample. The undiffracted main beam was intercepted by a cadmium beamstop. A magnetic field of up to 11 T applied approximately parallel to the neutron beam, was provided by a cryomagnet with a field uniformity of 0.2 % over a 1 cm sphere. A variable temperature insert containing He heat exchange gas allowed sample temperatures from 1.5 K to 300 K. The sample was a 40 mg low-twin-density high-purity single crystal of YBCO grown in a BaZrO$_3$ crucible $^{17}$$^{17}$ and oxygenated close to O$_7$ by high-pressure oxygen treatment in order to reduce pinning by oxygen vacancies in the Cu-O chains $^{18}$$^{18}$. It was therefore overdoped and had a $T_c$ of 86 K. It was initially mounted with its $c$-axis parallel to the field direction. In order to satisfy the Bragg condition for each diffraction spot in turn and hence establish the FLL structure, the cryomagnet and sample together could be rotated or tilted to bring the FLL Bragg planes to the appropriate small angles ($\sim 1^\circ$) to the incident neutron beam.

In Fig. 1 is shown the FLL diffraction pattern obtained at the low field of 1 T. The most obvious feature of this pattern is its fourfold symmetry which reflects the average fourfold symmetry of the twinned or-
the orthorhombic structure of our YBCO sample. However, the FLL structure itself has triangular coordination, and the symmetry of Fig. 1 arises from four orientations of distorted triangular FLLs, present in different domains in the sample, as was first observed by Keimer et al. [10]. The diffraction spots arising from these four triangular lattices are represented in Fig. 2. It appears that the distortion of the individual lattices arises mainly from the $a/b$ anisotropy present in each orthorhombic domain in the crystal [21, 22]. This interpretation was confirmed by measurements on an untwinned sample [8] which show diffraction spots distributed around an ellipse aligned with the $a$ and $b$ axes. The ratio of the principal axes of the ellipse should represent the anisotropy of the London penetration depth for $B_{c1} \ll B \ll B_{c2}$ [2]. The value we observe for the anisotropy ratio, $\gamma_{ab}$, in our sample is 1.28(1), whereas many estimates of this quantity are rather larger [22]. However comparable values to ours were obtained by measurements on a separate untwinned sample using neutrons [8] and muons and torque magnetometry [22]. Our results are also corroborated by recent surface-sensitive measurements using a novel atomic-beam magnetic-resonance technique [24]. It seems likely that the precise value of $\gamma_{ab}$ depends on the degree of perfection of the Cu-O chains along the $b$ direction [22]. The orientation of the triangular FLLs has been ascribed to pinning of a pair of spots, and hence planes of flux lines, to the twin planes [21, 22]. However, results reported later in this Letter also support the existence of a correlation between the nearest-neighbour FLL directions and the directions of zeroes of the $d$-wave order parameter.

In Fig. 3, we show diffraction patterns taken at higher fields. The data taken at 7 T show a distortion of the triangular FLLs so that some of the weaker spots are closer to the strong spots, and others have moved towards the diagonals. There is clearly another source of distortion than pure $a/b$ anisotropy. Finally at 11 T, the FLL has become almost exactly square, with the weak corner spots now playing the role of second order $\{1,1\}$ spots of a square FLL instead of first order spots from a distorted triangular FLL. In order to investigate this steady change in the FLL structure with field, we rotated the crystal about the vertical axis in Fig. 3, so that the field was $5^\circ$ from the $c$-axis. This was done in order to break the degeneracy between those FLL structures giving strong vertical diffraction spots (Fig. 2(a) and (c)), and those giving strong horizontal spots (Fig. 2(b) and (d)). Within anisotropic London theory, this small angle of rotation should make a negligible change to the FLL distortion. As shown in Fig. 4., we found that at high fields the FLL structures giving horizontal spots were suppressed and instead only the structures depicted in Fig. 2(a) and (c).
were observed. The advantage of this arrangement is that the pairs of spots near the horizontal axis in Fig. 4 could be observed easily, without being overlaid by the strong ones on the axis. This allowed us to measure accurately the spot positions and hence the FLL distortion. Nevertheless, at high field, this pair of spots overlaps, but by assuming that the spot size is independent of field, we may estimate the angle between them even when they overlap. Further measurements of spot positions allow us to give a complete description of the FLL distortions versus field in terms of the angles between the FLL reciprocal lattice vectors. The results of this analysis are shown in Fig. 5. It is clear that the low field structure progressively changes with increase of field, although in our available field range the FLL never exactly reaches a perfectly square shape. This may partly be because the phase transition is at the extreme of our available field range, but is also clearly a result of the orthorhombic structure of YBCO. As depicted in the inset to Fig. 5, the $a/b$ anisotropy of each domain must, on symmetry grounds, distort a square lattice to a rectangular one, causing a slight splitting of the “square” spots from a twinned crystal such as ours.

We have further investigated the temperature-dependence of the FLL distortion shown in Fig. 5. We find that with increasing temperature, the FLL structure changes more towards triangular. Thus the boundary between square and triangular phases must curve up in field as temperature is increased. We would expect this if the triangular to square transition is due to $d$-wave effects, as the nature of the pairing becomes less important as $k_B T$ becomes comparable with the magnitude of the gap. The shape of the phase boundary is similar to that seen in an overdoped sample by macroscopic measurements, but not the same as that proposed in ref. Unlike LSCO, the orientation of the FLL that we observe is aligned as expected from $d$-wave theories. It may be argued that twin planes, which are present in LSCO and YBCO are controlling the FLL orientation. To rule this out, measurements were also taken with the field at an angle to both sets of twin planes in our sample and the shape and orientation of the FLL was essentially unchanged. One should also note that the predicted difference in free energy between the two orientations of a
that one pair of spots is tied to the $\alpha \alpha$ hexagonal lattice would have order to resolve this angle more clearly (see text). A regular lines are predictions of anisotropic London theory $[7]$ for $\alpha \kappa$ do not believe that in a large-

square pattern (exaggerated for clarity), expected in the two 

tion and probable direction of $\gamma$. We square FLL is much larger than that between any trian-

gonic and the lower energy square orientation $[12]$. We also note that a similar correlation between FLL orienta-

tion is as expected from an isotropic $d$-wave theory $[12, 13]$, unlike that in LSCO $[2]$. It seems very 

likely from our measurements that at high fields, the FLL orientation, particularly when the FLL becomes square, is caused by the correlation between FLL planes and the directions of zeroes of the $d$-wave order parameter. It would clearly be of great interest to repeat these investiga-

gations in an unwinned crystal, where the effects of any pinning by twin planes would be completely removed, and the effect of the $a/b$ anisotropy on the “square” FLL would be crystal-clear. It appears that further investiga-

tions of these phenomena will allow stringent tests of the-

ories of the order parameter in the mixed state of high-$T_c$ materials as a function of angle of field and doping.

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ligen, Switzerland.

FIG. 5: Field-variation at 5 K of two of the angles between reciprocal lattice vectors, $\alpha$ and $\beta$, depicted in Fig. 2. (Errors are comparable with the marker size.) All data were obtained with $\mathbf{B}$ parallel to the crystal $c$-axis except the data for $\alpha$ at fields greater than 6 T, which were taken with $\mathbf{B}$ at $5^\circ$ to $c$ in order to resolve this angle more clearly (see text). A regular hexagonal lattice would have $\alpha = \beta = 60^\circ$, and an exactly square one $\alpha = 90^\circ$ and $\beta = 45^\circ$. Also marked by horizontal 

lines are predictions of anisotropic London theory $[7]$ for $\alpha$ and $\beta$, using a basal plane anisotropy, $\gamma_{ab} = 1.28$, and assuming that one pair of spots is tied to the $\{110\}$ directions. In the inset is shown the orthorhombic distortion from an exactly square pattern (exaggerated for clarity), expected in the two orthorhombic domains present in our crystal.

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