High magnetic field studies of the Vortex Lattice structure in YBa$_2$Cu$_3$O$_7$

A.S. Cameron, J.S. White, A.T. Holmes, E. Blackburn, E.M. Forgan, R. Riyat, T. Loew, C.D. Dewhurst, and A. Erb

1 School of Physics and Astronomy, University of Birmingham, Edgbaston, Birmingham, B15 2TT, UK.
2 Laboratory for Neutron Scattering, Paul Scherrer Institut, CH 5232 Villigen, Switzerland.
3 Max Planck Institut für Festkörperforschung, D-70569 Stuttgart, Germany.
4 Institut Laue-Langevin, 6 rue Jules Horowitz, 38042 Grenoble, France.
5 Walther Meissner Institut, BAdW, D-85748 Garching, Germany.

(Dated: March 13, 2014)

We report on small angle neutron scattering measurements of the vortex lattice in twin-free YBa$_2$Cu$_3$O$_7$, extending the previously investigated maximum field of <11 T up to 16.7 T with the field applied parallel to the c axis. This is the first microscopic study of vortex matter in this region of the superconducting phase. We find the high field VL displays a rhombic structure, with a field-dependent coordination that passes through a square configuration, and which does not lock-in to a field-independent structure. The VL pinning reduces with increasing temperature, but is seen to affect the VL correlation length even above the irreversibility temperature of the lattice structure. At high field and temperature we observe a melting transition, which appears to be first order, with no detectable signal from a vortex liquid above the transition.

PACS numbers: 74.25.Wx, 74.72.Gh, 28.20.Cz

The high $T_c$ superconductor YBa$_2$Cu$_3$O$_7$ (YBCO$_7$) possesses an orthorhombic crystal lattice, with complete Cu-O chains running along the crystal b axis. This material is slightly over-doped, and pinning of vortex lines is greatly reduced by full oxygenation of the chains. It is believed that superconducting states within the Cu-O chains lead to a modification of the anisotropic d-wave superconducting gap in the ab plane. Anisotropies in both the Fermi surface and the order parameter are expected to distort the vortex lattice (VL) present in the mixed state away from the ideal triangular structure as the applied field and temperature are varied.

Here we extend the field range of previous small angle neutron scattering (SANS) studies from 10.8 T to 16.7 T, providing the first microscopic information about the behavior of the VL at such high fields in any superconductor. This can provide important clues about the evolution with field of the pairing state and the superfluid density. We investigate the structural evolution of the VL with field at both 2 K and at 60 K together with a temperature dependent study at 10 T and 16 T. Following this, we discuss the dependence of the spatial variation of magnetic field in the mixed state on both field and temperature. This is measured in terms of the VL lattice “form factor”, derived from the intensity of the SANS signal, which is expected to depend on the coherence length, $\xi$, of the Cooper pairs, the magnetic penetration depth $\lambda$, and the departure of the vortex lines from straightness, represented by a Debye-Waller factor. Over all our field range, we find that, on heating, the VL undergoes a melting transition to a vortex liquid. Our data complement existing thermodynamic measurements, while adding a unique insight into the VL structure and perfection near the transition.

The experimental work was carried out on the D22 cold neutron SANS instrument at the Institut Laue-Langevin in Grenoble, France. The sample was a mosaic of six single crystals of de-twinned YBa$_2$Cu$_3$O$_7$, prepared by the method of Ref. 3. The crystals, of total mass ~20 mg and $T_c$ ~89 K, were co-aligned on an aluminum plate. A cryomagnet of novel design applied a horizontal field along the crystal c axes. Full experimental details can be found in ref. 3.

Previous studies on de-twinned YBCO$_7$, taken in an applied field of up to 10.8 T for a range of temperatures, showed a series of first-order structural transitions from low field hexagonal structures to a high field rhombic phase. These transitions have been attributed to the increasing influence with field of non-local effects arising from the finite size and structure of the Cooper pairs. Within the rhombic phase, the vortex lattice structure was seen to evolve with increasing field towards a square configuration expected just above the field limit of 10.8 T. Extending these measurements, Fig. 1(a) shows the structural evolution of the VL versus field from 8 to 16.7 T, for two different temperatures, and panel (b) shows the evolution with temperature on warming from a base of 2 K for two different fields. The previously observed variation with field is seen to continue, with the structural evolution of the rhombic phase continuing until the highest available field, passing through a square configuration at around 12.5 T at base temperature.

According to microscopic calculations using Eilenberger theory [2], it is expected that at high fields the nearest neighbor vortex directions will lie along the nodal directions of the order parameter. Our observations of the VL structural evolution suggest a field-driven change in the nodal positions. At zero field, the nodes are ob-
served to lie away from the 45 degree configuration of a pure \(d_{x^2−y^2}\) order parameter. This is expected from an inherent \(s\)-wave contribution which must be present due to the orthorhombic crystal symmetry.

The role of the chains in the superconductivity of YBCO\(_7\) is illustrated by the value of the low field London penetration depth anisotropy \(\sim 1.28\) \([3, 6]\), which implies an anisotropy in \(n_s/m^*\) of \(>1.6\). This value seems to be higher than can be accounted for by plane superconductivity alone and the sign of the anisotropy implies an extra contribution to superfluid density, \(n_s\), from the chains. Furthermore, it is clear from the similar temperature dependence of the low field data for the \(a\) and \(b\) directions \([6]\) that the superconducting order parameter has nodes on both the plane and chain Fermi surfaces. This implies that the Josephson-like pair tunneling, rather than a single-particle proximity effect \([11, 12]\), is responsible for the chain superconductivity. In the data presented here, the anisotropy changes sign at high fields, which may be an indication of a modification of chain superconductivity, since we are at a small fraction of \(H_{c2}\) and the quasi 1-D chain superconductivity may be less robust than its plane counterpart.

The approach of applying non-local corrections to London theory has had some success in describing the VL of superconductors with a fourfold axis \([13, 14]\) and can be seen in a conventional high-\(\kappa\) material such as \(V_3\)Si \([15]\). Non-local effects (NLEs) can give extra anisotropy about a crystal axis, in addition to to effective mass anisotropy already included in London or Ginzburg-Landau theories. They can arise in the theory from two causes: either Fermi surface anisotropy, which was initially treated in combination with a constant superconducting energy gap \(\Delta\) \([13, 14]\), and/or from an anisotropic \(\Delta\) \([13, 14, 17]\). In \(d\)-wave materials, the effective coherence length \(\xi \propto 1/\Delta(k)\), such that non-local effects must always be important near nodes in the energy gap. However as shown by White et al \([6]\) for fields up to 11 T none of the NLE theories agree with the observed structural evolution of the VL in YBCO\(_7\), an observation which continues to hold for the present measurements.

Fig. 1(b) shows the evolution of the VL structure with temperature. We first consider the behavior near \(T_c\). Within non-local descriptions \([13, 16, 17]\), it is predicted that NLEs will be reduced on approaching \(T_c\). This suppression would bring the VL structure towards the hexagonal structure expected by local electrodynamics, which is in general agreement with the results in figure 1(b). The non-monotonic variation with temperature seen near 40-60 K is surprising; we believe this is intrinsic but since it occurs at a temperature where pinning effects are changing, we postpone discussion until these are considered.

Now we turn to the spatial variation of magnetic field within the VL. The local field within a vortex lattice is given as a sum over all its spatial Fourier components

\[ F(\mathbf{q}) \text{ at the various scattering vectors } \mathbf{q} \text{ of its reciprocal lattice. The magnitude of } F(\mathbf{q}) \text{ or form factor, is proportional to the square root of the integrated intensity of a VL Bragg reflection as detailed in the supplementary} \]
information in ref. [3]. This is obtained by summing the intensities obtained by rocking the vortex lattice through angles near the Bragg condition for diffraction of the neutron beam, producing a “rocking curve” - see inset of Fig. 2. The main figure shows the variation of VL form factor with applied field between 9 and 16 T at base temperature. To model the field dependence of the form factor, we have used the the local London model, extended to account for the $ab$ plane anisotropy of YBCO$_7$, and including a Gaussian cut-off term to account for the finite size of the vortex cores [4]:

$$F(q) = \frac{\langle B \rangle \exp(-c(q_x^2 \xi^2 + q_y^2 \lambda^2))}{q_x^2 \lambda_x^2 + q_y^2 \lambda_y^2}.$$  \hspace{1cm} (1)

Here, $\langle B \rangle$ is the average internal induction, $\lambda_i$ is the penetration depth arising from supercurrents flowing in direction $i$, $\xi_i$ is the coherence length along $i$, and $q_x, q_y$ are in-plane Cartesian components of the scattering vector, with $q_x$ parallel to $b^*$. The cut-off parameter $c$ accounts for the finite size of the vortex cores, and as discussed by White et al [4] a suitable value for $c$ in our field and temperature range is 0.44.

Using the values for $\xi$ and $\lambda$ found in previous neutron scattering experiments [4], with $\lambda_a = 138\text{nm}$, $\lambda_b = 107\text{nm}$, $\xi_a = 3.04\text{nm}$ and $\xi_b = 3.54\text{nm}$, labeled as the ‘Low Field’ model in Fig. 2 we find that the form factor above 10 T is much larger than the extrapolation from low-field data. Taking smaller values of $\xi_a = 2.60\text{nm}$ and $\xi_b = 3.03\text{nm}$ with $\lambda$ unchanged provides the ‘High Field’ line in Fig. 2 however, this departs from the lower field data, and we suspect that even smaller values of $\xi$ would be appropriate if we could go to higher fields. This highlights our finding that no physically-reasonable constant values of the parameters can be found to fit the full field-range of our data.

We note that the previously fitted values for $\xi_a$ and $\xi_b$ seemed to be too large, since the upper critical field estimated from them is around 30 T, which is too low for YBCO$_7$. The larger values probably reflect frozen-in disorder in the vortices giving a static Debye-Waller (DW) factor, reducing the scattered intensity from the VL. A mean square deviation $\langle u^2 \rangle$ from straightness would contribute an additional term $\exp(-q^2 \langle u^2 \rangle / 4)$ to Eqn. (1), which has the effect of increasing the Gaussian cut-off term and simulating a larger vortex core size. We deduce that the effects of disorder in our high-field range are relatively smaller, and that the actual values of $\xi$ are smaller than those obtained from the lower-field data.

![FIG. 2. (Color online) The variation with field of the VL form factor for the 1st order diffraction spots (shown in Fig. 1(c)) at a temperature of 2 K. Integrated intensities were obtained by fitting the data to a Lorentzian line-shape. The solid red line shows the extended London model (ELM) prediction (Eqn. (2)), using parameter values obtained by fitting earlier data taken below 11 T [6], whilst the solid blue line shows a prediction, discussed in the text, from the ELM using smaller values of $\xi$.](image)

![FIG. 3. (Color online) (a) The variation with temperature of the VL form factor at 10 and 16 T, with data taken on warming after cooling by the OFC procedure to 2 K. The integrated intensity was calculated by fitting the rocking curves to a Lorentzian line-shape. (b) rocking curve widths vs. temperature, compared with the resolution width of the instrument (solid line).](image)
3D XY variation. The melting line of the data for a range of fields plotted as a fraction of the equilibrium, followed by VL melting when the displacement from the increasing thermal excursions of the VL from vortex lines away from straightness even though the VL as a whole can move. In the temperature region between these, the 10 T and 16 T data exhibit differing behaviors, with the rocking curve width at 10 T decreasing towards \( T_c \), whilst at 16 T the width increases before also narrowing on the approach to \( T_c \). In the same temperature region, the opening angle of the VL structure in fig. 1(b) has a peak. Further measurements, not reported here, show that the non-monotonic variation of the opening angle occurs both on warming and cooling, indicating that this is not due to annealing the VL on warming. We speculate that temperature/field dependent changes in the nodes, particularly in the chain order parameter, may be influencing the VL structure and pinning.

The temperature dependence of the form factor in Fig. 3, particularly at 10 T, may also represent such intrinsic effects, although we cannot rule out the effects of changes in a static DW factor. However, well below the irreversibility temperature both the VL structure and its lattice perfection remain fixed; hence the temperature dependence of \( F \) should reflect intrinsic properties of the sample. It has been suggested that the weak temperature dependence of \( F(q) \) seen at low temperatures is due to a stronger non-local response in large fields \([6, 20]\). Following the non-local model employed by White et al to fit data from YBCO at lower fields, we find that using the parameters for \( \xi \) and \( \lambda \) from fig. 2 provides a fit to the data at base temperature, and allowing these parameters to vary provides a good fit up to around 40 K. Above this region we expect that the static DW factor would reduce as the VL becomes able to move.

In Fig. 3, the form factor at 16 T shows a sudden drop close to 70 K. A more detailed scan between 60 and 80 K at 16.7 T is shown in Fig. 4(a), together with a prediction for the variation from the 3D XY model for superfluid density given as a solid line \([21]\). This model can be used to give the VL form factor as:

\[
F \propto \left(1 - \frac{T}{T_c(\xi)}\right)^n,
\]

with \( T(\xi) \) taken from work by Junod et al \([22]\), and \( n = 0.66 \). It can be seen in Fig. 4(a) that the form factor data drops well below the model prediction. The falloff will begin as a (true) Debye-Waller effect arising from the increasing thermal excursions of the VL from equilibrium, followed by VL melting when the displacements become large enough \([23]\). In Fig. 4(b) are shown the data for a range of fields plotted as a fraction of the 3D XY variation. The melting line \( B_m(T) \) derived from these results is very close to that found by heat capacity measurements \([24]\) of the lattice-liquid transition. It has been seen that a transition point between first and second order melting is dependent on pinning disorder and oxygen content \([2, 24, 25]\). First-order melting is expected in our case, because macroscopic measurements on YBCO show that the first order transition continues up to \( \sim 30 \) T \([26]\).

The intensity of Bragg reflections is predicted to drop sharply to near zero upon melting of the VL due to the loss of crystalline order, which would smear the sharp spots into a ring, which we estimate would reduce the signal at the reflection by an order of magnitude. However, even after counting for an extended period, no signal from the vortex liquid was detected above the transition. Should the rocking curve width of the vortex liquid remain similar to that of the vortex solid - and we note that the FWHM of the VL remained narrow during the melting transition - then we can estimate the Linde mann number required for a Debye-Waller factor to reduce the scattering from the vortex liquid to the point...
where we could no longer observe it. From the relation \( \langle u^2 \rangle = c_L^2 a^2 \), where \( \langle u^2 \rangle \) is the aforementioned mean-square deviation from straightness, \( c_L \) is the Lindemann number and \( a \) is the spacing between Bragg planes, we find \( c_L \) to be at least \( \sim 0.25 \), which is well within predicted values . Whilst we expect a first order transition to be sharp, we see that these data do not appear to have a sharp fall to zero at the melting transition, although this may be obscured by the initial fall due to the Debye-Waller factor, and we note that this may reflect a slight spread in \( T_c \) of our sample .

In conclusion, we report in this paper the microscopic investigation of the structure of vortex matter in a high-\( T_c \) superconductor in a new high-field range. We have observed the structural evolution of the VL rhombic phase in YBCO, with no observation of a field independent structure. These measurements suggest a field-driven change in the superconducting states associated with the CuO chain layers. The evolution of the VL form factor with field away from the low-field London+core model indicates that the VL will remain measurable by SANS well above our maximum field, allowing for measurements to be extended further when equipment permits. Further, the deviation of both the field and temperature dependence of the VL form factor from the expected behaviour in the extended London model, combined with the unusual variation in VL structure with increasing temperature, suggests that a static Debye-Waller factor is present in the low field and temperature data. This effect appears reduced both at high field and temperature, and suggests that estimates of the coherence length obtained from lower field data are too long . Observations of the VL melting transition are consistent with a first order lattice to liquid transition, with the intensity scattered from the VL falling sharply across the transition to an unmeasurable value. In summary, our study provides a further perspective on vortex matter under extreme conditions.

We acknowledge funding from the UK EPSRC, support from the ILL, where the measurements were performed, and the assistance of S. Mühlbauer and A. Helm at SANS-I of FRM-II with additional measurements not reported here.

[1] J. R. Kirtley et al., Nature (Physics) 2, 190 (2006).
[2] M. Roulin, A. Junod, A. Erb, and E. Walker, Phys. Rev. Lett. 80, 1722 (1998).
[3] J. S. White, V. Hinkov, R. W. Heslop, R. J. Lycett, E. M. Forgan, C. Bowell, S. Strässle, A. B. Abrahamson, M. Laver, C. D. Dewhurst, J. Kohlbrecher, J. L. Gavilano, J. Mesot, B. Keimer, and A. Erb, Phys. Rev. Lett. 102, 097001 (2009).
[4] A. T. Holmes, G. R. Walsh, E. Blackburn, E. M. Forgan, and M. Savey-Bennett, Rev. Sci. Inst. 83 (2012).
[5] See supplementary material at [URL] for experimental and analysis details.
[6] J. S. White, R. W. Heslop, A. T. Holmes, E. M. Forgan, V. Hinkov, N. Egottenmeyer, J. L. Gavilano, M. Laver, C. D. Dewhurst, R. Cubitt, and A. Erb, Phys. Rev. B 84, 104519 (2011).
[7] M. Ichioka, A. Hasegawa, and K. Machida, Phys. Rev. B 59, 8902 (1999).
[8] H. J. Smilde, A. A. Golubov, Ariando, G. Rijnders, J. M. Dekkers, S. Harkema, D. H. A. Blank, H. Rogalla, and H. Hilgenkamp, Phys. Rev. Lett. 95, 257001 (2005).
[9] T. Xiang and J. M. Wheatley, Phys. Rev. Lett. 76, 134 (1996).
[10] K. Zhang, D. A. Bonn, S. Kamal, R. Liang, D. J. Baar, W. N. Hardy, D. Basov, and T. Timusk, Phys. Rev. Lett. 73, 2484 (1994).
[11] W. A. Atkinson and J. P. Carbotte, Phys. Rev. B 52, 10601 (1995).
[12] J. S. White, C. J. Bowell, A. S. Cameron, R. W. Heslop, J. Mesot, J. L. Gavilano, S. Strässle, L. Mächler, R. Khasanov, C. D. Dewhurst, J. Karpinski, and E. M. Forgan, Phys. Rev. B 89, 024501 (2014).
[13] V. G. Kogan, M. Bullock, B. Harmon, P. Miranović, L. Dobrosavljević-Grujić, P. L. Gammel, and D. J. Bishop, Phys. Rev. B 55, R8693 (1997).
[14] V. G. Kogan, P. Miranović, L. Dobrosavljević-Grujić, W. E. Pickett, and D. K. Christen, Phys. Rev. Lett. 79, 741 (1997).
[15] M. Yethiraj, D. K. Christen, D. McK Paul, P. Miranović, and J. R. Thompson, Phys. Rev. Lett. 82, 5112 (1999).
[16] M. Franz, I. Affleck, and M. H. S. Amin, Phys. Rev. Lett. 79, 1555 (1997).
[17] K. M. Suzuki, K. Inoue, P. Miranović, M. Ichioka, and K. Machida, J. Phys. Soc. Jpn. 79, 013702 (2010).
[18] R. Cubitt, E. M. Forgan, D. McK Paul, S. L. Lee, J. S. Abell, H. Mook, and P. A. Timmins, Physica B 180, 377 (1992).
[19] U. Yaron, P. L. Gammel, D. A. Huse, R. N. Kleiman, C. S. Oglesby, E. Bucher, B. Batlogg, D. J. Bishop, K. Mortensen, K. Clausen, C. A. Bolle, and F. DeLaCruz, Phys. Rev. Lett. 73, 2748 (1994).
[20] M. H. S. Amin, M. Franz, and I. Affleck, Phys. Rev. Lett. 84, 5864 (2000).
[21] S. Kamal, D. A. Bonn, N. Goldenfeld, P. J. Hirschfeld, R. Liang, and W. N. Hardy, Phys. Rev. Lett. 73, 1845 (1994).
[22] A. Junod, M. Roulin, J.-Y. Genoud, B. Revaz, A. Erb, and E. Walker, Physica C: Superconductivity 275, 245 (1997).
[23] J. Kierfeld and V. Vinokur, Phys. Rev. B 69, 024501 (2004).
[24] R. Khasanov, S. Strässle, D. Di Castro, T. Masui, S. Miyasaka, S. Tajima, A. Bussmann-Holder, and H. Keller, Phys. Rev. Lett. 99, 237601 (2007).
[25] T. Nishizaki and N. Kobayashi, Superconductor Science and Technology 13, 1 (2000).
[26] K. Shibata, T. Nishizaki, T. Sasaki, and N. Kobayashi, Phys. Rev. B 66, 214518 (2002).