Nonlocal Effects and Vortex Lattice Transitions in YNi_2B_2C

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High resolution, neutron small angle scattering studies have been performed to investigate the low field ($B < 250$ mT) region of the phase diagram for the vortex lattice in the superconducting state of YNi_2B_2C. The data present clear evidence for a vortex lattice reorientation transition from a state with the diagonal of the rhombic unit cell along a [110] direction to a [100] direction. Above this transition the lattice distorts under the influence of the applied field until the apex angle becomes constant at some higher field. For $\mathbf{B} \parallel \mathbf{c}$ a square lattice configuration is formed. These experiments confirm qualitatively many of the predictions of a general model based on nonlocal corrections to the London model as applied to this and similar materials. [S0031-9007(97)05235-6]

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The yttrium and rare earth nickel borocarbides present a useful series of compounds to examine the coexistence and interactions between magnetism and superconductivity. These intermetallic compounds, $R\mathrm{Ni_2B_2C}$, are tetragonal and are found to be superconducting for $R =$ Y, Dy, Ho, Er, Tm, and Lu with T_c's of up to 16 K and values of B_{c2} reaching 10 T [1–4]. These are type II superconductors with κ in the range 5–15, and the superconductivity is thought to be driven by a conventional, if strong, electron-phonon interaction [5]. The most interesting feature of these materials is that for those containing a magnetic rare earth ion, there generally exists a region of coexistence between superconductivity and magnetism. As it is possible to grow reasonably sized, high quality and purity crystals of these materials this permits a wide range of experimental techniques to investigate the correlations and competition between the two ground states. However, given the magnitude of the coherence length, $\xi_0 \sim 10$ nm, in these compounds it is essential to achieve an extensive understanding of nonlocal effects in the superconducting state for the nonmagnetic members of this series before assigning various effects to the influence of magnetic order or fluctuations on the superconducting state. The development of a new theoretical and quantitative model of the influence of nonlocal effects on the morphology of flux-line lattices in type II superconductors by Kogan *et al.* [6] is an important advance in understanding the mixed state. This model is still under development and its quantitative predictions require to be tested by measurements on a range of materials. This work provides one such test and finds good qualitative agreement with the predictions of the model.

In this Letter we present high resolution, neutron small angle diffraction measurements of the flux-line lattice in high quality, single crystals of YNi_2B_2C in the low field region ($B = 17.5$–240 mT). The data confirm the presence of a reorientation transition of the vortex lattice at a field of $B_1 \sim 112$ mT. The low field configuration has the flux lines arranged on a rhombic unit cell with the diagonal of the rhombus aligned along a [110] direction. Above the transition, the diagonal has switched to a [100] or [010] direction. On the low field side of the transition the lattice adapts to an increased field by decreasing the apex angle of the rhombus from 60° (a hexagonal lattice). After the transition the lattice increases its apex angle with increasing field until locking on to a fixed configuration (apex angle of 90° for $\mathbf{B} \parallel \mathbf{c}$) at a field of B_2. The transition at B_1 is most likely of first order, since there is no obvious route for the lattice to smoothly shift between these two configurations, while the higher field transition can be accommodated by smoothly varying the apex angle.

Neutron scattering has proved to be an extremely useful probe of the mixed state in type II superconductors. Such experiments provide a means to image directly the distribution of flux lines in the bulk of a superconductor. This is achieved by diffracting neutrons from the periodic distribution of magnetic induction in the superconducting state, under an applied magnetic field greater than B_{c1} but less than B_{c2}. There have been three previous studies of the vortex lattice [7–9] in borocarbides, using neutron diffraction techniques. Microscopic coexistence of superconductivity and ordered magnetism in $ErNi_2B_2C$ was inferred from the growth of disorder and the realignment of the vortex lattice away from the applied field [7]. A field-driven vortex lattice phase transition, B_2 in this Letter, has also been observed at ~50 mT in $ErNi_2B_2C$ [8]. Initial discussion on the role of anisotropy and disorder on the nonmagnetic analog YNi_2B_2C was presented in Ref. [9]. This last work was at considerably higher fields and lower resolution than the present study. Given the complexity found in the structure of the vortex lattice of YNi_2B_2C, it is essential to fully understand the properties of the vortex lattice in the nonmagnetic borocarbides before conclusions may be drawn about the influence of magnetic ordering on the mixed state properties of these compounds.

In this experiment we have used the D22 instrument at the Institut Max von Laue–Paul Langevin, Grenoble with a mean incident wavelength of 1.55 nm and a 9% wavelength spread, collimation of 17.6 m and an area detector of 128 × 128 (7.5 mm) pixels at a distance of 18 m from the sample. An additional circular slit of 30 mm diameter was also placed before the incident collimation to better define the scattered beam at the detector and to equalize the horizontal and vertical divergences. This high resolution setup, which is required to resolve the Bragg reflections at low field, still provided sufficient neutron flux to permit a small crystal of YNi_2B_2C, $3.4 \times 3.7 \times 0.6$ mm^3, to be used in these experiments. The sample was cooled in a small-bore cryostat mounted between the pole pieces of an electromagnet with the applied field aligned parallel to the neutron beam. The magnetic field and the neutron beam were initially aligned by observing the diffraction signal from the flux-line lattice in niobium. For all measurements the vortex lattice was grown by cooling the sample through T_c with the required magnetic field already applied. At each temperature and applied field the sample was rocked through each Bragg reflection on the detector by rotating the cryostat and electromagnet together about the vertical axis to ensure that the diffraction spot position on the multidetector coincided with the Bragg condition. All the data presented in this Letter were taken with the crystal aligned with its [100] axis vertical and the applied field in the [010][001] plane.

The quality of the data obtained with this instrumental setup is excellent as can be seen from examining the diffraction images presented in Fig. 1. These figures were prepared by adding together a number of individual detector measurements, with a $T > T_c$ background subtracted, over a range of rocking angles to allow a complete picture of the diffraction pattern to be presented in a single image. Individual frames at the exact Bragg condition for each reflection were always used to extract the position of the spots for estimates of the lattice morphology and distortion. The apex angle of the real space rhombic unit cell was estimated by two independent means: (i) by directly calculating the angles between all the Bragg reflections visible on the detector and (ii) by determining the ratio of the magnitudes of the momentum transfer vector from each reflection. The anisotropy of this latter quantity was compared with that predicted from a calculation of the reciprocal lattice configuration for a range of apex angles at the appropriate orientation of the rhombic unit cell to the crystallographic axes. In all cases good agreement was found between these two methods of determining the apex angle. The apex angles as a function of applied magnetic

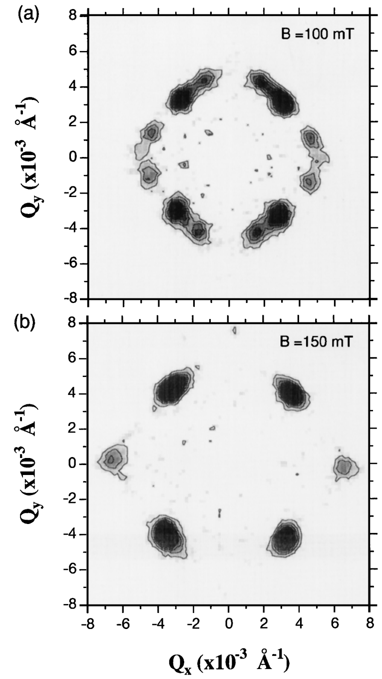

FIG. 1. Images of the diffraction pattern from the vortex lattice in YNi_2B_2C with a magnetic field of 100 and 150 mT applied at a 10° clockwise rotation of the c axis, about the vertical [100] axis, with respect to the applied field and the neutron beam. The splitting of the spots near the horizontal axis at low field has been removed at high field by the reorientation transition.

field are presented in Fig. 2 for **B** ∥ **c**, and for the crystal rotated clockwise by 10° and 30°, with respect to the applied field, about the vertical [100] axis. Most of the data were taken at 10° rotation since this gave the best definition for the Bragg reflections as it forced the vortex lattice to prefer a single domain configuration at high fields.

As Fig. 1 clearly shows, the primary differences between low and high field data is the splitting of the spots, at B_1, to produce two equally populated domains at low field. Each of these domains contributes a pair of the diagonal spots in Fig. 1(a). In particular, the spots which are on the horizontal axis at high field [Fig. 1(b)] rotate away from that axis. This is the clearest evidence for a reorientation of the vortex lattice. This rather abrupt change in morphology for the vortex lattice can be explained by proposing that the diagonal of the real space rhombic unit cell rotates from a [100] to approximately a [110] direction as the field is decreased. In addition, the pattern of angles between the Bragg reflections and the associated lengths of the Bragg momentum transfer vectors can be understood only if the apex angle for the rhombic unit cell becomes less than 60°, in the field region near 100 mT. We have chosen to show our data at 100 mT in Fig. 1, since at this particular field there remains a slight trace of Bragg intensity on the horizontal axis and indeed a complete analysis of all our data at 10° from the c axis,

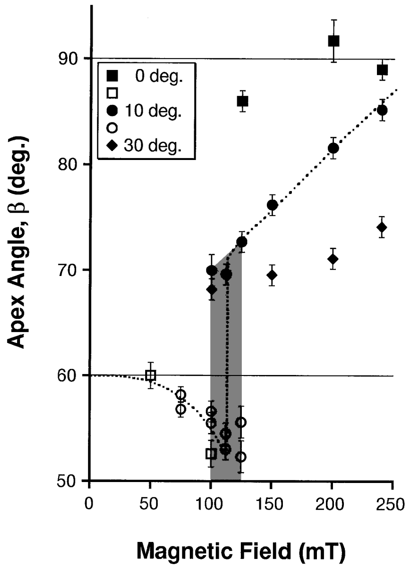

FIG. 2. The derived apex angle (β) for the rhombic unit cell as a function of magnetic field at three orientations of the applied field to the c axis. Open symbols, on the lower branch, correspond to an alignment of the diagonal of the unit cell near to [110], while closed symbols, upper branch, refer to a [100] orientation.

Fig. 2, confirms that there is a region of coexistence between the two phases over the range 100–125 mT. In passing through the coexistence regime there is a rapid increase in the proportion of one phase to the other. Such coexistence is probably associated with the first order nature of this transition. At fields greater than 125 mT the distortion of the vortex lattice continues with increasing field and tends towards a "square" configuration but never reaches this value, nor becomes constant, in our restricted range of applied field, up to 240 mT. Although we currently have limited data taken with the field parallel to the c axis and at 30° off the c axis, a comparison of these measurements with the 10° off c information is of interest. For $\mathbf{B} \parallel \mathbf{c}$ the low field data is in good agreement with that taken at 10° from c and again suggests a rhombic unit cell with an apex angle which initially decreases from 60° with increasing field. At 125 mT the vortex lattice has reoriented to align with the [100] axis and the apex angle is 86° ± 2°. At higher fields an apex angle of 90° is estimated within our experimental accuracy and a square vortex lattice has formed. These data suggest that a field independent square configuration forms between 125 and 200 mT for $\mathbf{B} \parallel \mathbf{c}$, B_2 in the notation of Kogan *et al.* [6]. B_2 is probably closer to the lower bound given the shape of the apex angle vs field curves in that work. Since there is no evidence for such a stabilization with B 10° off c, in our measurement range, this datum implies that the B_2 transition is very sensitive to orientation while the B_1 reorientation transition is much less so. At 30° off c, the cell diagonal is aligned along the [100] direction, and the apex angle is markedly reduced and has only a small field dependence. Extrapolation of the available data points also suggests that the low field apex angle would not be 60°, which is reasonable given the anisotropy of this material. The B_1 transition must also have moved to lower fields or no longer be present to account for these observations.

A simple, qualitative argument for the sense of the change in apex angle for the two presumed orientations of the vortex lattice is presented in Fig. 3. Assuming that a hexagonal lattice will tend toward a high field configuration which is square with the nearest neighbor direction along [110], this figure shows how at constant field the vortices would move to achieve this aim. If the diagonal of the rhombic cell is aligned along [010] then the apex angle will increase, alignment along [$\bar{1}$10] would produce the opposite result.

In the work of Kogan *et al.* [6], nonlocal corrections are applied to the London model of a superconductor in the mixed state and the free energy calculated for various alignments of the vortex lattice as a function of the apex angle. The minima in free energy were mapped to produce a phase diagram for the apex angle and configuration of the vortex lattice as a function of applied field for $\mathbf{B} \parallel \mathbf{c}$ and $\mathbf{B} \parallel \mathbf{a}$. The required parameters, to make this model quantitative, such as averages of products of Fermi velocities were obtained from a tight binding fit to a first principles linearized plane wave calculation of the band structure of $LuNi_2B_2C$ and a scaling factor ($C = 0.363$ for $\lambda = 71$ nm in the clean limit), adjusted to a value of $C = 0.221$ to position the "square to triangle" transition near 50 mT. For $\mathbf{B} \parallel \mathbf{c}$ Kogan *et al.* predict that initially, at low field, the diagonal of the rhombic unit cell is directed along a [110] direction and the apex angle decreases as the field is

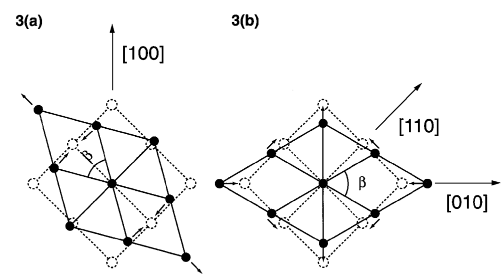

FIG. 3. A representation of how a hexagonal lattice, solid lines and symbols, would distort, at constant unit cell area, to match a square lattice, dotted lines and symbols, aligned along a [110] direction, depending on the initial alignment of the hexagonal lattice. (a) Orientation of rhombic unit cell diagonal along [$\bar{1}$10]; (b) rhombic unit cell diagonal along [010]. Note that in (a) a decrease in β makes [110] the nearest neighbor direction, while in (b) distortion towards a square lattice corresponds to an increase in β, as observed and predicted by Kogan *et al.* [6]. It is also clear from this diagram that it is possible to smoothly distort (b) to form a square lattice; hence a second order transition is possible at B_2, while it is not possible to simply shift the orientation from (a) to (b) suggesting a first order transition at B_1.

increased. Eventually, when $B = B_1$, this configuration becomes unstable and the lattice flips to lie with its diagonal along a [100] direction and the apex angle is $>60°$. This is thought to be a first order transition. The apex angle now increases until at $B_2 = 50$ mT a second order transition occurs where the apex angle locks on to 90°. This is the stable configuration for all fields $B > B_2$. For $\mathbf{B} \parallel \mathbf{a}$, the high field state is a rhombic cell with an apex angle of 81°, $B_2 = 130$ mT and there is no evidence for a B_1 transition. The low field configuration extrapolates to an apex angle of 53°.

In general, the predictions and conclusions from the theoretical study are in excellent qualitative agreement with the experimental data presented in this Letter. Differences between experiment and theory exist in the positions of the transitions. Our estimate for B_1 is an order of magnitude larger than that predicted by the model and B_2 approximately a factor of 3 higher. This may be due to the use of the band structure for the Lu compound in the calculations, suggesting a high sensitivity to the composition for such transitions (a further source in the differences between experiment and theory may be the neglect of any superconducting gap anisotropy in the theoretical model). These discrepancies may be reduced when full band structure calculations become available for YNi_2B_2C. It would be of particular interest to see such calculations extended to other orientations to compare more directly with current and future neutron measurements of the phase diagram for this and similar materials.

This work has demonstrated, for the first time, the presence of a first order reorientation transition with a marked increase in apex angle for the rhombic unit cell of the vortex lattice in YNi_2B_2C. In addition, it is suggested that there is considerable anisotropy in the position of the lock-in transition at higher fields, with much less anisotropy being associated with the reorientation transition. The experimental data provide strong evidence for the validity of a general model based on nonlocal corrections to the London model and should prompt further studies along these lines. Such investigations stress the importance of understanding, in detail, the properties of the vortex lattice of the nonmagnetic superconducting borocarbides before it will be possible to extract the influence of magnetic order on such properties.

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[1] R. Nagarajan *et al.*, Phys. Rev. Lett. **72**, 274 (1994).
[2] R. J. Cava *et al.*, Nature (London) **367**, 252 (1994).
[3] B. K. Cho *et al.*, Phys. Rev. B **52**, 3684 (1995).
[4] C. V. Tomy *et al.*, Physica (Amsterdam) **248C**, 349 (1995).
[5] W. E. Pickett and D. J. Singh, J. Supercond. **8**, 425 (1995).
[6] V. G. Kogan *et al.*, Phys. Rev. B **55**, R8693 (1997).
[7] U. Yaron *et al.*, Nature (London) **382**, 236 (1996).
[8] M. R. Eskildsen *et al.*, Phys. Rev. Lett. **78**, 1968 (1997).
[9] M. Yethiraj *et al.*, Phys. Rev. Lett. **78**, 4849 (1997).