Neutron Scattering Study of the Flux Lattice in YNi_2B_2C

M. Yethiraj,[1] D. McK. Paul,[2] C. V. Tomy,[2] and E. M. Forgan[3]
[1]*Solid State Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831-6393*
[2]*Department of Physics, University of Warwick, Coventry, United Kingdom*
[3]*School of Physics and Space Research, University of Birmingham, Birmingham B15 2TT, United Kingdom*
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We observe a flux lattice with square symmetry in the superconductor YNi_2B_2C when the applied field is parallel to the *c* axis of the crystal. A square lattice observed previously by Yaron *et al.* in the isostructural magnetic analog $ErNi_2B_2C$ was attributed to the interaction between magnetism in that system and the flux lattice. Since the Y-based compound does not order magnetically, it is clear that the structure of the flux lattice is unrelated to magnetic order. We do not observe any change in the mosaic or direction of the flux lines with applied field; these phenomena may be related to magnetism. In YNi_2B_2C, the strength of the flux lattice signal was seen to fall off rapidly with applied field; the results imply considerable disorder in the arrangement of the flux lines at 2.5 T, but are not consistent with melting. We see a markedly anisotropic flux lattice when the applied field is at 60° to the *c* axis. In this case, the flux lattice is observed to be a distorted hexagon. [S0031-9007(97)03384-X]

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Shortly after rare earth nickel borocarbides of the type RNi_2B_2C (R = rare earth) were found [1] to be superconducting in 1994, it was found that some of these compounds containing a magnetic rare-earth ion exhibited coexistence [2,3] of magnetic order with superconductivity. The rare-earth ions order in the magnetic state; Ni ions do not have a moment. The interplay between superconductivity and magnetism in the rare-earth nickel borocarbides is a topic of considerable interest, since magnetism and superconductivity are typically competing interactions.

It is essential, however, to know the properties of the nonmagnetic compound to separate out those effects that are exclusive to the magnetic variants. In order to do this, we have studied YNi_2B_2C which does not order magnetically. This is the first flux line lattice study on the nonmagnetic rare-earth nickel borocarbide using small-angle neutron scattering (SANS). The intensity of the Bragg scattering from the ordered array of flux lines is inversely proportional to the fourth power of the London penetration depth in the material, making it possible to determine this quantity accurately.

Superconducting mass anisotropy results in Bragg spots being located on an elliptical rather than circular outline, which SANS can readily determine. The YNi_2B_2C system has been reported to be "remarkably isotropic" by torque magnetometry [4] measurements, while the Ho-based compound has been said [5] to have a mass anisotropy ($\gamma \approx 2$). We show that the flux lattice in the Y-based compound can be anisotropic when applied fields are not directed along principal crystal directions.

The neutron scattering measurements were carried out on the 30-m SANS facility at the High Flux Isotope Reactor at Oak Ridge National Laboratory. The *c* axis of the YNi_2B_2C crystal was initially aligned parallel to the field direction with an uncertainty of ±2°. Relative rotations were accurate to ±0.25° when the field was applied at an angle to the *c* axis. The divergence of the incident beam was set at 0.2° for field of 0.75 T or lower and 0.3° for fields between 1 and 2.5 T. The wavelength spread was less than 3%; the incident wavelength used was 4.75 Å. Measurements were made at fields from 0.4 to 2.5 T. The sample was a single crystal of YNi_2B_2C which was grown by a high temperature flux method using Ni_2B flux with isotopic ^{11}B to reduce neutron absorption. The crystal (of dimensions 3.4 mm × 3.7 mm × 0.6 mm thick) had a mosaic, determined by neutron diffraction, of less than 0.2°. The crystal had a T_c (onset) of 15.7 K.

With the field parallel to the *c* axis (long axis of the nuclear tetragonal cell) of the crystal, fourfold symmetry was observed for the flux lattice diffraction pattern (Fig. 1) for an applied field of 0.4 T. The first-order (10 and 01) spots were aligned along the 110 crystallographic axes. Unlike YBCO [6], however, the lattice had reasonably

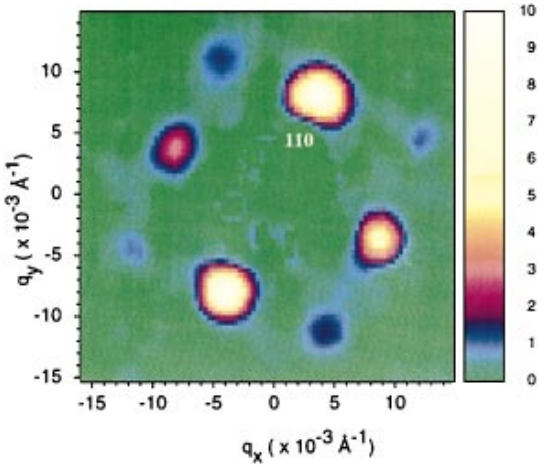

FIG. 1(color). A lattice with square symmetry is observed when the applied field (0.4 T) is parallel to the *c* axis of the crystal.

long-range translational order (which varies with applied field, as discussed later) so that second-order (11) peaks were also detectable. In this respect, it resembles the square flux lattice observed in Pb [7]. A square lattice is also seen in Nb [8] under certain conditions, when the field is parallel to the 100 direction. However, unlike Nb, the square lattice existed in the entire field range (0.4–2.5 T) studied. In Nb, the alignment mechanism is thought to be a fourfold symmetry of the minimum energy position for the nearest neighbor vortex, which is possible here. Square lattices are generally more common in low-κ materials. Clearly, the flux line lattice structure is completely unrelated to the magnetic order, contrary to the suggestion of Yaron *et al.* [9] based on the data for the magnetic Er analog.

Rocking curves about the vertical axis (perpendicular to the incident neutron and applied field directions) gave a width for the flux line crystal of $0.4° \pm 0.1°$. Although quite perfect, this is larger than the mosaic of the crystal. This width measures the combined effects of the straightness of the flux lines (η), and the length ℓ over which the flux lines are correlated. If the flux line is of finite length, its Fourier transform is an extended object, which gives rise to an observable angular width in a rocking curve. If the measured resulting rocking curve width is assumed to be due strictly to the finite length ℓ this length would be given by

$$\ell = d/\Delta\theta\,,$$

where d is the d spacing of the flux lattice and $\Delta\theta$ is the mosaic width. Our measured rocking curve width translates to a length ℓ of 9.2 μm over which the flux lines scatter coherently. However, since this mosaic is independent of the applied field, and hence the d spacing of the vortex lattice, it suggests that this width primarily reflects the straightness of the vortices; the correlation length would then be considerably longer. Pinning in this material is weak; the Bragg signal disappears when the field is turned off at 1.55 K subsequent to field cooling, in contrast to YBCO where 90% of the flux lattice signal is retained [6] when the field is turned off.

In $ErNi_2B_2C$, the flux lattice was seen to rotate away from the applied field, and its mosaic was seen to increase as the temperature was lowered. Here, in the nonmagnetic YNi_2B_2C, the vortex lattice direction coincided with the applied field to within 0.1°, and the mosaic was constant at all temperatures and fields in the ranges measured (1.55 to 7 K at 1 T and from 0.4 to 1.75 T at 1.55 K). The rotation of the flux lattice away from the field direction and change in mosaic are either properties of magnetically ordered systems or an artifact of misalignment of the c axis with the applied field. (The latter would imply a somewhat fortuitous near-exact alignment of the field and the c axis in our case; this angle was measured to be 1.6° in the other work.)

The temperature dependence of the intensity for an applied field of 1 T is shown in Fig. 2. For these data,

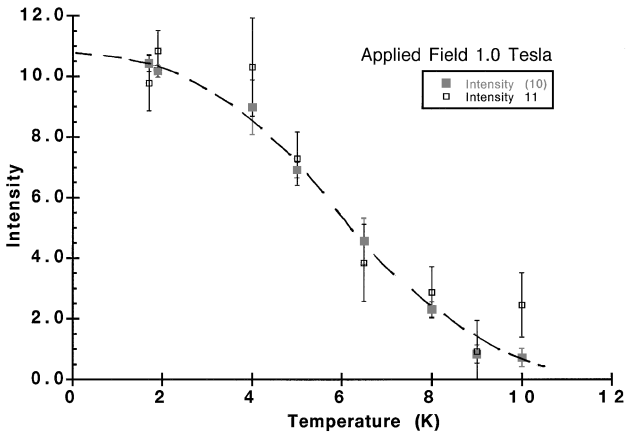

FIG. 2. Temperature dependence of the Bragg intensity for 1.0 T (filled squares) with the field parallel to the c axis. The intensity of the (scaled) second-order peak at 1 T is indicated by open squares. $T_c = 15.7$ K.

it is seen that the temperature (T) dependence of the second-ordered peak (scaled to allow direct comparison) is identical to that of the first-order reflection, further evidence that the mosaic does not change with T for the following reason. The T-dependence data were taken at a set angular position, where both first- and second-order peaks could be observed simultaneously, such that the 10 peak was exactly on the Bragg condition. If the mosaic changed, the T dependence of the 11 peak would not mirror that of the 10 peak.

The London penetration depth λ_L is obtained [10] from the Bragg intensity I integrated over the mosaic using the relation,

$$\frac{I}{I_0 V} = \frac{2\pi}{q}\left(\frac{\gamma}{2}\right)^2 \lambda_n^2 \left(\frac{1}{\phi_0}\right)^2 \left(\frac{B}{(1 + q^2\lambda_L^2)}\, e^{-2\pi^2 B\xi^2/\phi_0}\right)^2,$$

where V is the sample volume, γ is the neutron gyromagnetic ratio, B the applied field, ξ is the coherence length, and λ_n is the neutron wavelength. The term involving ξ approximately allows for finite core size (and agrees to within 30% of exact numerical solutions [11] of the Ginzburg-Landau equation for $B < 0.5B_{c2}$). In addition, even quite large differences in the form factor do not greatly change the effective x/d because it is the square root of an exponent. For instance, a change in the form factor by a factor of 3 would only make a ratio of 10. Using this equation at 0.4 T [with $H_{c2}(0) = 5.5$ T [12]], the London depth is deduced to be 1100 ± 80 Å, which is considerably larger than that for $ErNi_2B_2C$ from SANS measurements [9].

Even after correcting for geometrical $(1/q)$ and core effects, the intensity drops rather quickly as the field is increased [Fig. 3(a)]. We assume this field dependence to be due to a static Debye-Waller-like factor [13], which depends on x, the root mean square displacement of the flux line from its mean position, and on the average spacing of the vortices, d. The resulting x/d

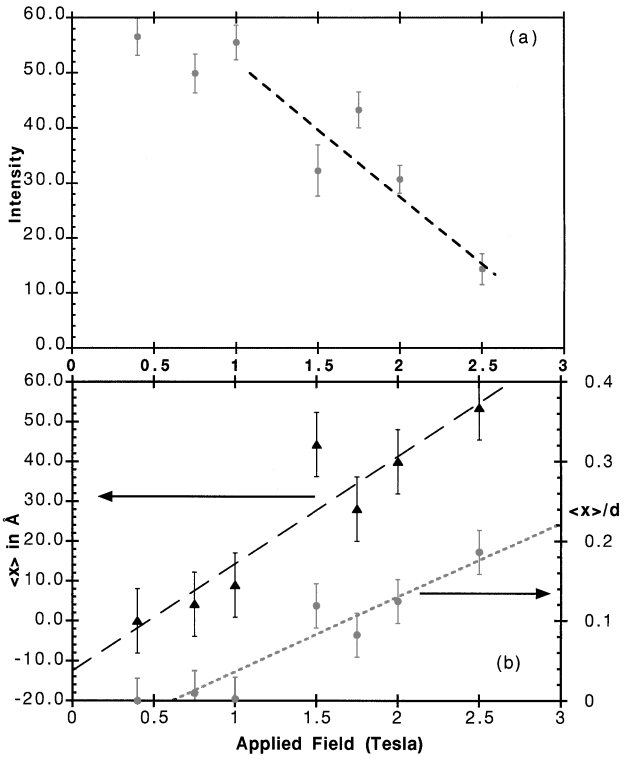

FIG. 3. (a) Field dependence of the intensity and (b) the actual change in the displacement; x and x/d were obtained from these intensities (see text). Lines are intended as a guide to the eye.

and x [Fig. 3(b)] are seen to increase monotonically with applied field. By analyzing the ratio of 10 to 11 intensities from rocking curves, the absolute magnitude of x/d at 0.4 T was obtained. The disorder for other fields was determined from the corrected first-order intensities. At 2.5 T, x/d approaches 18%. However, spots indicating long-range directional order can still be seen, hence an average lattice still exists. Only local displacements about the mean position are implied. The monotonic increase x/d with applied field is mirrored by an increase in the value of x.

A criterion for a glass could be established where static fluctuations of 25% or greater are present. By this measure, the glassy phase is not reached in our range of fields. Hence, our data disagree with the recent suggestion [14] that the flux lattice in YNi_2B_2C forms a glassy phase at fields of 2 T. The relative intensities of first-order to second-order peaks at 0.4 T imply little or no (zero $\pm 3\%$) disorder. Hence, we can conclude that, at 0.4 T, we have a well-formed lattice. Further, since we can identify no sharp changes in behavior as a function of field, it is reasonable to say that it remains a lattice up to 2.5 T. In short, there is no lattice-glass transition, although the lattice becomes progressively more disordered as the field is increased. This increase in disorder probably reflects a relative decrease of the shear modulus of the flux line lattice as H_{c2} is approached. Our observations provide a microscopic description of what has been seen in bulk measurements, independent of the semantics of where the lattice-glass boundary lies or even of its existence.

In addition, a liquid peak should be centered at a significantly different q from that of a solid. The intensity, $I(q)$, for a two-dimensional liquid is given by the integral over the pair correlation function multiplied by $J_0(qr)$. Assuming the pair correlation function has a well-defined peak at the nearest neighbor distance r, then the peak in $I(q)$ corresponds to the first maximum in $J_0(qr)$, which occurs [15] for $qr = 7.01$. Hence the q of the liquid peak is given by $7.01/r$ which is 12% (and measurably) larger [16] than $2\pi/r$ for a square lattice (but $\approx 3\%$ smaller than the q of a triangular lattice, which we do not have, with the same d spacing). The intensity as a function of q for peaks on the Bragg condition for applied fields of 2.5 T (Fig. 4) is inconsistent with scattering from a liquid or glass. In addition, liquids, polycrystalline materials, and glassy systems will produce a ring of scattering rather than distinct Bragg spots. Exceptions are where a hexatic or tetratic phase exists where the directional order is long range but with sort-range translational order. However, the existence of higher-order spots is not consistent with a hexatic phase. The observation of the suggested melting of the lattice at temperatures closer to T_c was not attempted in this study.

Unexpectedly, although all measurements report little or no anisotropy in YNi_2B_2C, an obviously distorted hexagonal lattice was observed when the c axis was 60° from the (0.4 T) applied field direction (Fig. 5). The rotation was carried out about an axis that was 21° from the a (or equivalently, b) axis of the crystal in the a-b plane. The resulting hexagonal lattice had an orientation locked to the square spot that was closer to the

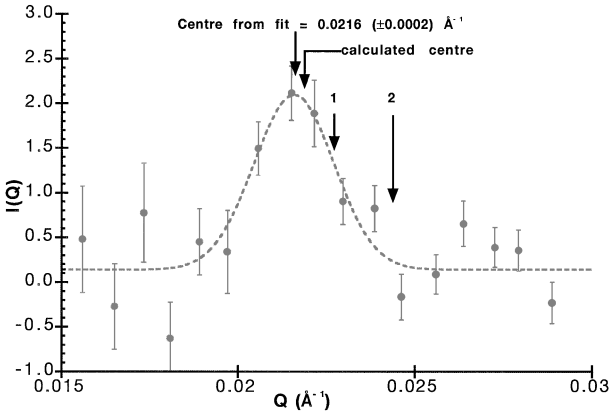

FIG. 4. Radial q average of the Bragg spot (on Bragg condition) for an applied field of 2.5 T. The peak is centered at the calculated position for a square lattice, within the errors. The position for a liquid peak with triangular coordination is marked "1" and a liquid with square coordination is marked "2."

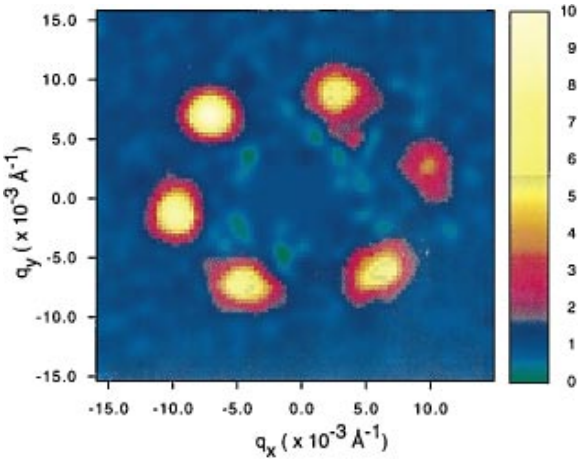

FIG. 5(color). A distorted hexagonal lattice is observed when the applied field is 60° from the *c* axis of the crystal. The vertical axis (rotation axis) is in the *a*-*b* plane and is 21° from the *a* axis.

vertical rotation axis. Apparently, the same orientation mechanism as for $B \parallel c$ is still at play here. The mosaic of this lattice ($0.6 \pm 0.1°$) is slightly larger than for *B* parallel to *c*. The anisotropy from this lattice can be ascertained from the eccentricity of the ellipse on which the Bragg spots lie. However, even these preliminary results indicate that the anisotropy cannot be described as an effective mass anisotropy [17].

This study clearly demonstrates the complexity to be found in the flux line lattice structure of this nonmagnetic member of the $R\mathrm{Ni}_2\mathrm{B}_2\mathrm{C}$ series of compounds. It is evident that a good understanding of the properties of this material in the superconducting state and further measurements on this particular compound are required before any conclusions can be drawn about the role of the coexistence of magnetic order on the vortex lattice in the mixed state.

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[11] E. H. Brandt (private communication).
[12] J. R. Thompson (private communication).
[13] The Debye-Waller factor for a Bragg reflection in a 2D system is given by $\exp(-q^2\mathbf{x}^2)$, where $\mathbf{x}$ is the component of the root mean square displacement in the direction of $\mathbf{q}$, the reciprocal lattice vector. Since $q = |\mathbf{q}| = 2\pi/d$, the Debye-Waller factor can be expressed as $\exp[-4\pi^2(\mathbf{x}/d)^2]$, where d is the d spacing.
[14] M. Mum *et al.*, Phys. Rev. Lett. **76**, 2790 (1996).
[15] $J_0'(qr) = -J_1(qr)$, thus the first maximum in the scattering occurs at the second zero of $J_1(qr)$. For the case of 3D system, see J. M. Schulz, *Diffraction for Materials Scientist* (Prentice-Hall, Englewood Cliffs, NJ, 1982) Sect. 2.4, p. 43.
[16] The exact q of the liquid peak depends on the average coordination number, which is four for a square arrangement and six for a triangular arrangement. For both these arrangements, the liquid q is bigger than that for a square lattice.
[17] M. Yethiraj, D. McK. Paul, C. V. Tomy, and E. M. Forgan, (to be published).