

# Nonlocal effects in the superconductor $\text{YNi}_2^{11}\text{B}_2\text{C}$

M. Yethiraj<sup>a,\*</sup>, H. Kawano<sup>a,b</sup>, D.McK. Paul<sup>c</sup>, C.V. Tomy<sup>c</sup>, H. Takeya<sup>d</sup>, H. Yoshizawa<sup>e</sup>

<sup>a</sup>*Solid State Division, Oak Ridge National Laboratory, Oak Ridge, TN, 37831, USA*

<sup>b</sup>*The Institute of Physical and Chemical Research (RIKEN), Wako, Saitama 351-0198, Japan*

<sup>c</sup>*Department of Physics, University of Warwick, Coventry CV4 7AL, UK*

<sup>d</sup>*National Research Institute for Metals, Sengen 1-2-1, Tsukuba, Ibaraki 305, Japan*

<sup>e</sup>*Neutron Scattering Laboratory, ISSP, University of Tokyo, Tokai, Ibaraki 319-1106, Japan*

## Abstract

Flux lines repel each other, thus a hexagonal lattice which maximizes the distance between them is generally the lowest energy configuration. In the rare-earth nickel borocarbide superconductor  $\text{YNi}_2\text{B}_2\text{C}$ , square lattices of flux lines have been observed when the field is parallel to the  $c$ -axis. Using the  $c$ -axis penetration depth as a gauge, we find that the cross-section of an individual flux line has a four-fold symmetry, which makes square lattices energetically favored over the more prosaic hexagonal ordering. The dependence of the penetration depth within the basal plane is discussed. © 1999 Elsevier Science Ltd. All rights reserved.

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## 1. Introduction

Rare-earth nickel borocarbitides of the type  $\text{RNi}_2\text{B}_2\text{C}$  ( $R =$  rare-earth) are of interest since those containing a magnetic rare-earth ion exhibit the coexistence of magnetic order with superconductivity. The flux line lattices in these materials (when the applied field is parallel to the tetragonal  $c$ -axis of the crystal) exhibit four-fold symmetry. It was thought at first that the square symmetry of the flux lattice observed in these materials was related to the presence of long range magnetic order. The same square symmetry occurring in the nonmagnetic  $\text{YNi}_2\text{B}_2\text{C}$  proved that this symmetry was not related to the magnetism. It is known from diffraction that Ni does not have an ordered moment in these compounds and that only the rare-earth ions order in the magnetic state and Y is nonmagnetic. The square symmetry of the flux lattice seemed to be an intrinsic property of clean borocarbide superconductors.

Flux lines in a superconductor are generally expected to be [1] in a hexagonal array since flux lines repel each other and a hexagonal lattice of these lines, which maximizes the

distance between them, can be reasonably assumed to be the lowest energy configuration. However, square lattices of flux lines have been seen experimentally. The first observation of this was in superconducting lead [2] and more recently, square lattices are seen in the rare-earth nickel borocarbitides [3,4] when the applied field is parallel to the  $c$ -axis of the crystal. The driving mechanism for the square symmetry has been loosely attributed to Fermi surface features and pinning by the same or even the interplay of flux lines with magnetic order. However, the question of why flux lattices are square had no simple explanation. We have shown [5] conclusively that square arrays originate from an in-plane anisotropy of the penetration depths which makes the cross-section of a single flux line square, rather than circular (or oval) as generally assumed. With a four-fold symmetry of the flux line, the square lattice now becomes energetically favorable at an intervortex distances comparable to the London penetration depth, where the vortices interact strongly. We also studied the variation of the London penetration depth within the basal plane to further elucidate the features caused by the details of the electronic structure. The origins of the variation of the penetration depth are discussed.

It is not completely unexpected that the four-fold symmetry of the structure of the compound is not reflected

\* Corresponding author. Tel.: + 1-423-576-6069; fax: + 1-423-574-6268.

*E-mail address:* yethirajm@ornl.gov (M. Yethiraj)

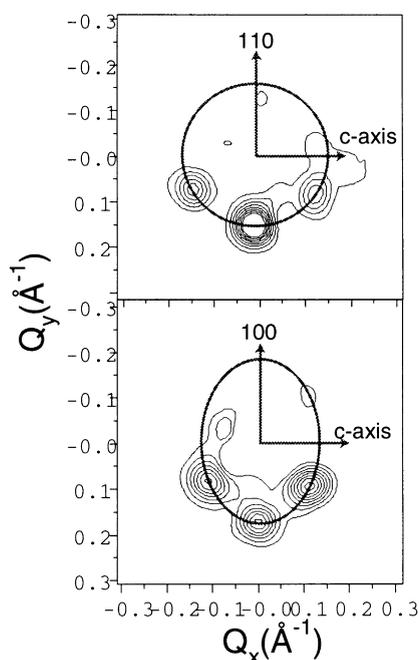


Fig. 1. The flux lattice with the  $c$ -axis horizontal in both cases, but with (a) 110 vertical and (b) 100 vertical. The ellipse through the first order peaks are a measure of the anisotropy of penetration depths.

somewhat in the superconducting properties as well and that critical fields and vortex lattice structures in these superconductors are direction dependent. However, many properties contradict a simple mass tensor description of anisotropy. For example, peculiarities in the behavior of Nb has been attributed to the anisotropy of its Fermi surface, which has been treated theoretically through nonlocal electrodynamics. This provides the natural explanation for coupling between crystal lattice and vortex lattice and a mechanism where the details of the Fermi surface, and not simply the crystal symmetry, affects the superconducting properties. Observation of square vortex lattices and first order transitions at  $H_{c1}$  are common for low- $\kappa$  cubic superconductors. However, it is widely accepted that nonlocality effects are significant only for low- $\kappa$  superconductors. We show that, even in high- $\kappa$  clean superconductors, the effects of nonlocal electrodynamics should not be disregarded.

## 2. Experimental

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  $\text{YNi}_2\text{B}_2\text{C}$  crystal was initially aligned parallel to the field direction with an uncertainty of  $\pm 2^\circ$ . Relative rotations were accurate to  $\pm 0.25^\circ$  when the field was applied at an angle to the

$c$ -axis. Measurements were made at applied fields of order 1.0 T at an incident wavelength of 4.75 Å. For all the measurements, the neutron beam was horizontal and the applied magnetic field was collinear with the incident neutron direction, except rotated by the Bragg angle to satisfy the scattering condition. This is the geometry most commonly used today.

The two single crystal samples were used; each was grown by a different technique. Both utilized isotopic  $^{11}\text{B}$  to reduce neutron absorption. The smaller of the two single crystals of  $\text{YNi}_2\text{B}_2\text{C}$  which was grown by a high temperature flux method using  $\text{Ni}_2\text{B}$  flux. Magnetic measurements on a sample from the same growth run has shown that the material is extremely clean (the ratio of the critical current to the depairing current at 3 K was of the order of  $3.3 \times 10^{-6}$  at 0.4 T and  $0.7 \times 10^{-6}$  at 1.0 T. The crystal (of dimensions 3.4 mm  $\times$  3.7 mm  $\times$  0.6 mm thick) had a mosaic, determined by neutron diffraction, of less than  $0.2^\circ$ . The crystal had a  $T_c$  (midpoint) of 14.5 K. The larger sample, grown by techniques detailed elsewhere [6], had a  $T_c$  (midpoint) of 14.2 K width of 0.5 K as characterized by magnetization measurements.

Each flux line contains one quantum ( $h/2e$ ) of flux and an array of them creates a modulation of magnetic fields. The neutron, which has a magnetic moment, interacts with this array giving rise to diffraction peaks from the flux lattice. Small angle neutron scattering is an ideal probe of the structural details of flux line lattices in the bulk. Further, any anisotropy of the penetration depth is readily apparent as this implies that flux lines have elliptical cross-sections and this results in Bragg spots being located on an ellipse (as opposed to a circle in the isotropic case).

At the outset, the crystal was mounted (arbitrarily) and it was found that the vertical was approximately  $22^\circ$  from the  $a$ -axis of the crystal. We saw that the flux lattice in the  $Y$ -based compound is rather complex. With  $B \parallel c$ , the flux lattice was square, as had also been seen in  $\text{ErNi}_2\text{B}_2\text{C}$  earlier by Yaron et al. As the angle between the  $c$ -axis and the field was increased, the lattice became hexagonal. For these data, the rotation of the field was performed about the vertical axis. When the applied field was at  $45^\circ$  to the  $c$ -axis, the lattice observed showed that the spots lie on an ellipse (eccentricity = 1.2) instead of on a circle. The eccentricity of the ellipse is a measure of the anisotropy between the penetration depths along different directions in the scattering plane. For the case of effective mass anisotropy, the eccentricity of the ellipse varies monotonically as the applied field is rotated from one symmetry direction to the other. When the applied field was at  $60^\circ$  to  $c$ , the eccentricity of the ellipse was smaller than when the field was at  $45^\circ$  to the  $c$ -axis, hence the lattice was somewhat more isotropic. This was a very curious result as it implied that there were quite large variations of the effective penetration depth in a manner that was not consistent with a simple uniaxial superconducting mass anisotropy.

It can be shown easily in the case of a mass anisotropy

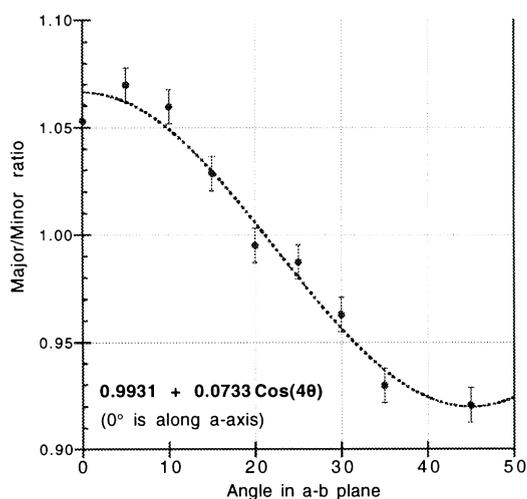


Fig. 2. The ratio of the major to the minor axis of the ellipse is the ratio of penetration depths between the  $c$ -axis and the particular direction in the basal plane perpendicular to it. The line through the data is a fit of the function  $a + 2b\cos(4\theta)$ ; the fit parameters are as indicated on graph. the value of  $a - b$  represents the total anisotropy between the  $c$ -axis and the 110; including both mass anisotropy and nonlocality considerations.

(where the variations of the penetration depth between the major axes are monotonic) that the ratio of the major to minor axis of the ellipse that the Bragg diffracted spots lie on is the same as the ratio of the penetration depths along those directions. Keeping this in mind, we decided to use the  $c$ -axis penetration depth as a gauge and study the variation of the in-plane London depth by comparing it to the  $c$ -axis value. Here, it should be mentioned that while this treatment is sound in the mass anisotropy case, there may be problems

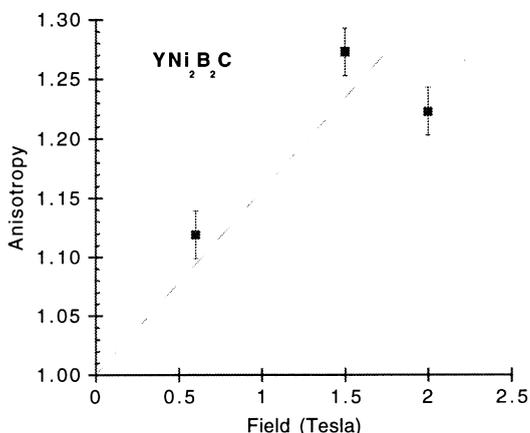


Fig. 3. The ratio of the penetration depths between the 100 and 110 increases as the applied field is increased. This ratio in the first sample was  $1.45 \pm 0.05$  at an applied field of 1.0 T, which is larger than in the bigger sample up to 2.0 T, the highest field at which we measured the flux lattice.

if it is applied to a case where there are unusual variations of the London depth between the two directions. However, since the lattices with the  $c$ -axis in-plane are always either hexagonal or distorted hexagonal and never square, we assume that this is not the case in a plane which has the  $c$ -axis in it and that unusual variations of the penetration depth occur only within the basal plane since that is the only geometry in which square flux lattices are observed.

We then used the  $c$ -axis penetration depth to measure the ratio of the penetration depths between the 100 ( $a$ -axis) and 110 directions in the basal plane. To do this, the sample was first mounted with the  $c$ -axis vertical and with the  $a$ -axis in the plane of scattering. The applied field was along the  $b$ -axis of the crystal (which is equivalent by symmetry to the  $a$ -axis). In this way, we could measure the ratio of the penetration depth between the  $a$ -axis and the  $c$ -axis. The scattering observed here (Fig. 1(b)) shows that there is a 40% anisotropy in this case (the penetration depth along the 100 is larger). Then, the sample was rotated such that the 100 and the  $c$ -axis were in the plane of scattering. In this instance, the two penetration depths were different by only 6% (Fig. 1(a)). This confirmed that the penetration depths in the  $(hk0)$  plane were, in fact, anisotropic and different by 45% between the 100 and 110 directions.

The measurements on the first sample were made difficult by the fact that it was only 0.5 mm thick and as the geometry required that for the measurements of this sample must be viewed with in the narrow dimension facing the incident neutrons. For a more detailed measurement of how the penetration depth varied within the basal plane, we used the larger sample. We began again with the  $a$ - $c$  plane perpendicular to the field. For each successive measurement, we rotated the sample about the vertical  $c$ -axis and probed the London depth in-plane in  $5^\circ$  increments from  $0^\circ$  (i.e.  $a$ -axis) to  $45^\circ$  (110 axis). Given that the  $c$ -axis London depth is the same in all these cases, we obtain the variation of the penetration depths in the basal plane. The full angular dependence data, obtained at an applied field of 0.6 T, are shown in Fig. 2. The values of the anisotropy at the two end points (i.e. 100 and 110 directions) as a function of applied field is shown in Fig. 3. It is observed that the anisotropy increases with field up to 2 T, the highest field measured. ( $H_{c2} = 4.5$  T).

### 3. Conclusion

In the local London model, mass anisotropy allows the penetration depth to vary within a plane. However, the variation can only be monotonic and the symmetry directions represent the upper and lower limits of the London penetration depth in the plane. In  $\text{YNi}_2\text{B}_2\text{C}$ , we see that there is no mass anisotropy difference within the  $a$ - $b$  plane, since the  $a$  and  $b$  axes are equivalent. Here, the difference in penetration depths occurs along the 110 directions. This directly implies the influence of nonlocal

electrodynamics, that is, the effects due to the finite size of the core of the flux line. Theoretical work of nonlocality [7] in the London limit has explained quite well the transition from hexagonal to square in the symmetry of the flux lattice with increasing field ( $B \parallel c$ -axis) and deduce an anisotropic in-plane London penetration depth. Extensions [8] of this work, presented elsewhere, are consistent with the results presented here. The good agreement between the theory and experiment suggests that the nonlocal electrodynamics in the London limit is the mechanism responsible for determining the vortex lattice structure in the borocarbide superconductors.

The structure of the flux line lattice is always consistent with that of the underlying crystalline lattice and presumably its associated Fermi surface. The penetration depth also has the same symmetry. Note that the penetration depth is a measure of the  $1/e$  distance of the decay of an external applied field that is less than  $H_{c1}$  and the half width at half maximum of the field modulation of a single flux line in the mixed state. The theoretical construct uses the penetration depth in a manner that makes it invariant as a function of rotation about the applied field. The “penetration depth” in the theory is the in-plane average of the physical value. In the local London model, the theoretical and experimental definitions coincide and both values become identical.

It is clear from measurements in this and other materials, that quite small coherence lengths can contribute to nonlocal effects that are considerable. Naturally, effects of nonlocality such as the anisotropy of the penetration depths should become smaller as the mean free path decreases. In the present measurement, the smaller sample showed an anisotropy between the  $a$ -axis and the 110 direction of about 45% at 1 T. The larger sample was not measured at 1 T, but at 1.5 T showed a smaller 27% anisotropy in-plane. This may be due to a smaller mean free path in the larger sample, but we have not yet made quantitative comparisons to verify this thesis. Although they suggest that the anisotropy may have saturated at 2 T, the data are far from clear and further measurements are necessary.

The nonlocal correction to the field modulation is in addition to any anisotropy that may be caused by any mass anisotropy that may be present. It is impossible to say from a single measurement at the lowest temperature what the anisotropy is attributable to. The apparent anisotropy varies, depending to which direction in the basal plane the  $c$ -axis value is being compared; this could yield quite dramatically different values of the “mass anisotropy”. The best way would be to measure this at temperatures rather close to  $T_c$ , where mass anisotropy would dominate.

The in-plane variation of the London penetration depth follows a  $\cos(4\theta)$  functional form, consistent with the above mentioned theory. This kind of variation of the upper critical field has also been observed in borocarbide [9] superconductors, as well as in high- $T_c$  [10] materials, although this behavior has been attributed to a d-wave character in the high- $T_c$  case.

It would be extremely interesting to see if the anisotropy of the coherence length begins to affect the flux lattice symmetry, rather than that of the penetration depth. Presumably, a large penetration depth would be associated with a small coherence length. In that case, the field distribution of a single flux line would once more appear more or less isotropic at some distance between the coherence length and the penetration depth from the center of the core. This may once more give rise to hexagonal symmetry. Unfortunately, at higher fields, even with the large sample, the signal dropped off too rapidly to be observable.

Finally, the drop off in the signal intensity is presumed to be to its proximity to the upper critical field, for which the only solution is to have higher neutron flux with which to measure the diffracted signal. The problem of insufficient neutron flux should be alleviated considerably after the planned installation of a cold source at the HFIR, where the flux is expected to increase by a factor of more than two orders of magnitude compared to the present small-angle scattering instrument.

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