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Square flux lines in YNi_2B_2C

M. Yethiraj
Solid State Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831-6393

D. McK. Paul and C. V. Tomy
Department of Physics, University of Warwick, Coventry, United Kingdom

J. R. Thompson
Solid State Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831-6393
and University of Tennessee, Knoxville, Tennessee 37996-1200
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Magnetic fields penetrate a type-II superconductor via quantized flux lines. Why these flux lines sometimes form square arrays, as in YNi_2B_2C with the field parallel to the *c* axis, rather than the expected hexagonal ordering, has long fascinated physicists and has eluded a simple explanation. Our latest measurements on YNi_2B_2C prove conclusively that the London penetration depth within the *a*-*b* plane is not isotropic. This anisotropy of the London depths implies that the cross section of an individual flux line has square symmetry, which makes a square lattice energetically favored over the more prosaic hexagonal ordering.
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Rare earth nickel borocarbides of the type RNi_2B_2C (R =rare earth) are of interest since those containing a magnetic rare earth ion exhibit[1,2] 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) exhibited fourfold symmetry. It was thought at first[3] 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 YNi_2B_2C proved[4] 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 in a hexagonal array since flux lines repel each other and a hexagonal lattice of these lines maximizes the distance between them, which can be reasonably assumed to be the lowest energy configuration. However, square lattices[5] of flux lines have been seen experimentally. The first such of these was in superconducting lead and most recently, square lattices are seen in the rare-earth nickel borocarbides 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 has no simple explanation. We show 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 thought. With a fourfold 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.

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 (SANS) 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 results in Bragg spots being located on an ellipse (as opposed to a circle in the isotropic case). 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 $\pm 2°$. Relative rotations were accurate to $\pm 0.25°$ where the field was applied at an angle to the *c* axis. Measurements were made at applied fields of 0.4 T and 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.

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 ^2B to reduce neutron absorption. Magnetic measurements[6] on a sample from the same growth run has shown that the material is extremely clean (the ratio of the critical current to the despairing current at 3 K was of the order of 3.3×10^{-6} at 0.4 T and 0.7×10^{-6} at 1.0 T, where measurements (reported here) were made. 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 and T_c (midpoint) of 14.5 K.

At the outset, the crystal was mounted (arbitrarily) and it was found that the vertical was approximately 22° from the *a* axis of the crystal. We saw that the flux lattice in the Y based

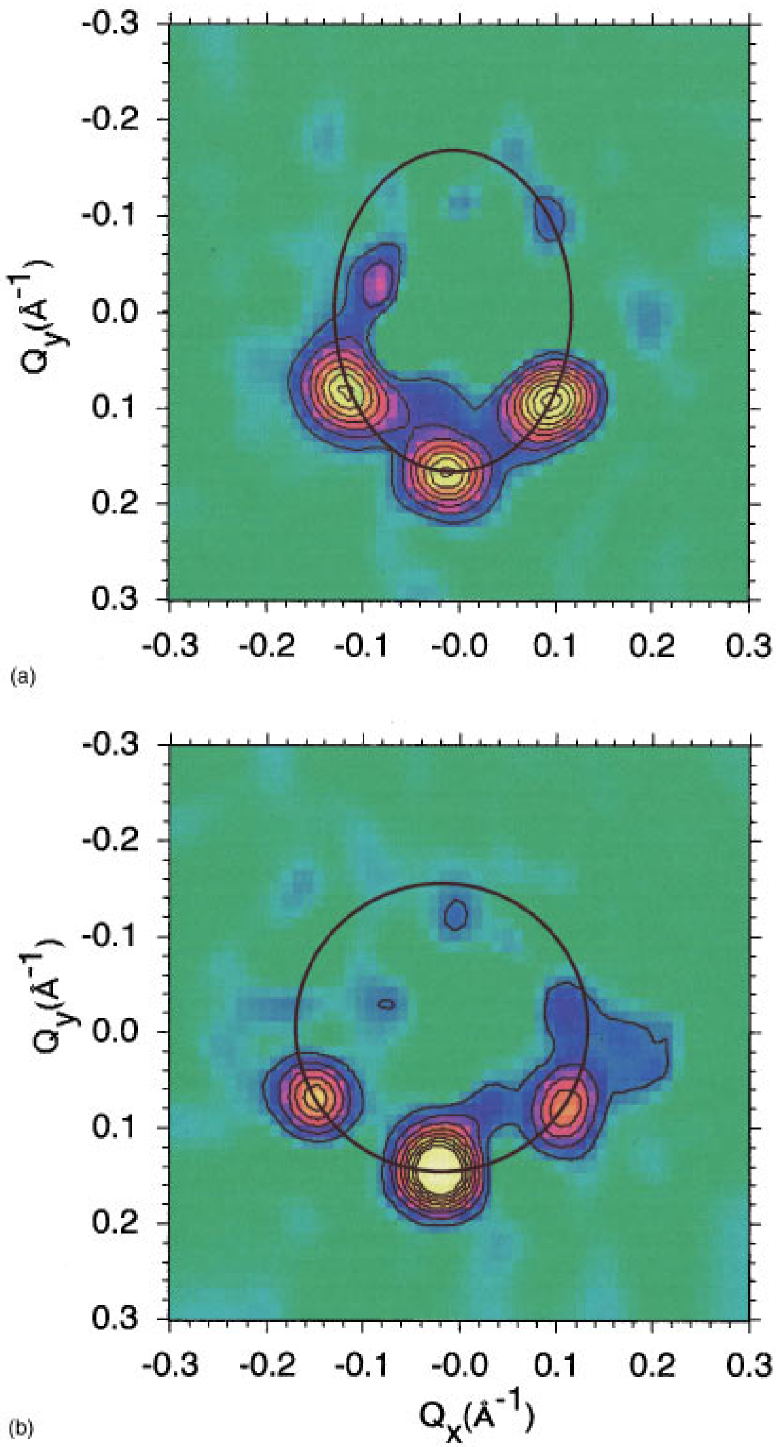

FIG. 1. (Color) The flux lattice with the *c* axis horizontal in both cases, but with (a) 100 vertical and (b) 110 vertical. The ellipse through the first order peaks are indicative of the anisotropy of penetration depths.

compound is rather complex. With $B \| c$, the flux lattice was square, as had also been seen in $ErNi_2B_2C$ earlier[3] by Yaron *et al.* As the angle between the *c* axis and the field was increased, the lattice became hexagonal.[7] For these data, the rotation of the field was performed about the vertical axis. When the applied field was at 45° 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.[8] 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° to *c*, the eccentricity of the ellipse was smaller than when the field was at 45° 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.

While anisotropy was evident, we saw from an analysis of the structural details that, in this orientation, the anisotropy was small when the field was applied 90° from the *c* axis. That indicated that the principal directions seemed to exhibit rather similar London penetration depths. However, in order

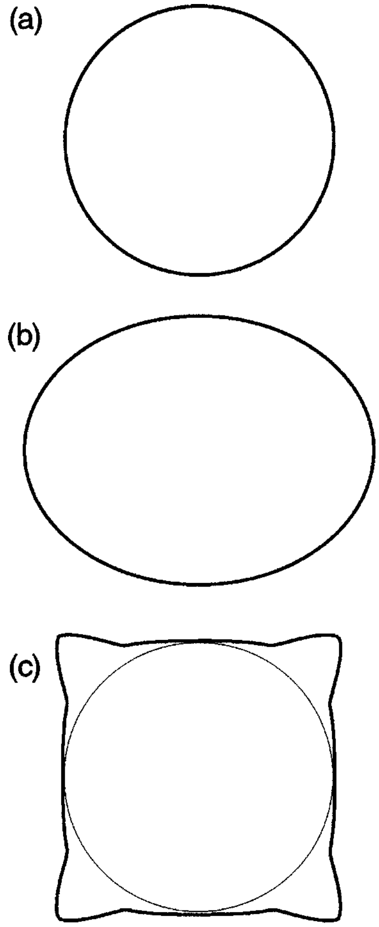

FIG. 2. A schematic of a constant local field contour of a single flux line at a distance comparable to the London penetration depth in the (a) isotropic case, (b) local with superconducting mass anisotropy not equal to 1, and (c) isotropic case with nonlocal effects as is the case for YNi_2B_2C.

to explain the nonmonotonic variation of the eccentricity of the ellipse that the spots lay on, it was our thesis that the London depth must vary within the basal plane.

To test if the London penetration depth was indeed anisotropic with the *a-b* plane, we performed the following measurements. We used the *c*-axis penetration depth as the invariant gauge to measure the ratio of the penetration depths between the 100 (*a* axis) and 110 directions. To do this, the sample was first mounted with the *a* axis vertical and with the *c* 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 [see Fig. 1(a)]. Next, we realigned the sample such that the (110) axis was vertical, but with the *c* axis still in the scattering plane. Here, we measure the ratio between the London penetration depths along the *c* axis and that in the 110 direction [see Fig. 1(b)]. Given that the *c*-axis London depth is the same in both cases, we can get the ratio between the penetration depths along the (100) and (110) directions. The data for these measurements are somewhat less well determined than for $B\|c$, since the sample was viewed edge-on and the extent of the incident beam was effectively the thickness of the sample, which was only 0.4 mm. Only the ''lower'' half of the reflections were in Bragg condition for these measurements, since the magnetic was tilted ''down'' to improve the signal for these reflections. The flux crystal was quite perfect with a mosaic of approximately 0.4°. The data were obtained at an applied field of 1.0 Tesla.

The geometry for the two cases and the respective data sets are shown in Fig. 1. From the eccentricity of the ellipse that the spots lie on, in each case, we find that

$$\lambda_{110}/\lambda_c = 0.96 \pm 0.03 \quad \text{and}$$

$$\lambda_{100}/\lambda_c = 1.39 \pm 0.035.$$

Hence, the ratio: $\lambda_{100}/\lambda_{110} = 1.45 \pm 0.05$. This ratio of penetration depths implies that the cross section of an individual flux line has fourfold symmetry.

Our finding is not inconsistent with our previous measurement and with torque magnetometry[9] where it was found that the penetration depths in the basal plane and along the *c* axis were virtually identical. Since our data show that anisotropy exists between different directions in the basal plane, the comparison of ''basal plane'' penetration depths to the *c*-axis values is relatively meaningless; this anisotropy will depend on the exact orientation within the basal plane to which the *c*-axis value is being compared. In effect, it appears that nonlocal effects are very significant in this compound. In fact, even when the external applied field is directed along a principal symmetry direction, it appears there is considerable anisotropy in the plane perpendicular to it.

A cursory analysis of these data make it apparent that nonlocal effects must be involved to explain this behavior for the following reasons. In the local picture, $\mathbf{j} = 4\pi \boldsymbol{M}^{-1}\mathbf{A}$ where **j** is the current, **A** is the vector potential and **M** the mass tensor. In an isotropic system, the diagonal element of the mass tensor are all the same whereas in an anisotropic system, the superconducting mass anisotropy gives rise to a mass tensor that has different diagonal terms corresponding to the three major symmetry directions. However, in either scenario, the penetration depth must vary monotonically between the symmetry directions. That is, it is not possible for λ_{110} to be larger than λ_{100}. (By symmetry, $\lambda_{010} = \lambda_{100}$.) However, the finite extent (ξ) of a Cooper pair gives rise to nonlocality, which in Fourier space has been written[10] as follows: $j(q) = Q(q)A(q)$. Here, $Q(q)$is the q-dependent kernel that modifies the effect of the vector potential on the current. More specifically, the current $j(r)$ at r can have contribution from the vector potential within a coherence length around the point r. This has immediate implications on the symmetry of the flux lattice. The q-dependent kernel causes the flux lattice structure to be field-dependent.

The physical argument of the field dependence of the flux lattice symmetry is quite simple. The data show that in the borocarbides with $B\|c$, the cross section of a single flux line has square symmetry. This is schematically shown in Fig. 2 and compared with the expected cross sections for an isotropic material and for a system having superconducting mass anisotropy, but without nonlocal effects. At low fields, where the distance between flux line is large, the effects of the square flux line are not relevant[11] and the lattice formed is hexagonal. However, when the distance between the flux lines becomes comparable to the London penetration depth, the minimum energy configuration is reached in a square rather than triangular arrangement of the flux lines. The measured London penetration depth (from SANS measurements) for $B\|c$ is 1100 Å, which is the value along the 110 since the reflections are aligned along that direction. From the in-plane anisotropy determined here, $\lambda_{100}=1600$ Å. An intervortex distance of 1600 Å is achieved at an applied field of approximately 800 Oe (for a square lattice, 920 Oe for a triangular one). This is approximately the field value at which the hexagonal-square transition is observed[12,13] for $B\|c$ in this material. The distance between vortices at the same applied field is smaller in a hexagonal lattice than in a square one by about 7%. However, if the repulsion of two vortices is smaller in a particular direction because of a smaller penetration depth, there may be numerous fields where a rearrangement of the lattice results in a lower energy.

In an alternate picture, when nonlocal effects are small, an individual flux line may have a reasonably circular cross section even at quite small distances from its core. In this case, going to higher fields (and approaching H_{c2}), which is expected to cause nonlocal effects to become smaller will mean that a hexagonal lattice can be expected at higher fields than measured here.

The penetration depth referred to here is the physical ($1/e$) length for penetration of an applied field (lower than H_{c1}), which is also the physical half-width at half-height of the field modulation for a single flux line in the mixed state. In the formulation of the theory[8] of nonlocal effects, the terminology used is somewhat different. The authors define a quantity referred to as the ''penetration depth'' which is an invariant as a function of rotation about the applied field direction. In this theoretical construct, the kernel averages over all of the Fermi surface in this plane. Hence their ''penetration depth'' is equivalent to the average penetration depth in this plane perpendicular to the external applied field rather than the actual penetration depth in any given direction. This is strictly a difference in terminology and no difference is implied in the physics.

We note that there has been much discussion about the symmetry of the flux lattices in high T_c superconductors and the possibility that a symmetry other than rhombohedral[14] may be indicative of *d*-wave superconductivity. We caution first that the symmetry of the flux lattice is not a good test of *d*-wave versus *s*-wave theses. Further, since nonlocal effects are temperature and field dependent, this could possibly give rise to ''anomalous'' temperature and field dependence of the order parameter as well.

It is unclear at this time whether the anisotropy of the penetration depths is directly related to the local shape of the Fermi surface. If it is, such experiments could be used to probe the Fermi surface in superconductors. In addition, it is well known[15,16] that, in these rare-earth nickel borocarbides, a transverse $(h,0,0)$ phonon mode softens as a function of temperature. We think it would be exciting to try to correlate the nonlocal effects on the flux lattice with the effects of field on the softening. Such experiments are planned.

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[2] A. I. Goldman *et al.*, Phys. Rev. B **51**, 678 (1995); J. Zarestky *et al.*, *ibid.* **51**, 681 (1995).

[3] U. Yaron *et al.*, Nature (London) **382**, 236 (1996).

[4] M. Yethiraj *et al.*, Phys. Rev. Lett. **78**, 4849 (1997).

[5] B. Obst, *Anisotropy Effects in Superconductors* (Plenum, New York, 1977), p. 139.

[6] K. J. Song and J. R. Thompson (unpublished).

[7] The distinction between a ''distorted square'' and a ''distorted hexagon'' is often not meaningful in strict symmetry terms. We use ''distorted hexagon'' loosely to describe the lattice when there are clearly six spots with comparable intensities, but the angle between the basis vectors is not 60°.

[8] The eccentricity measures directly the ratio of penetration depths along the major and minor axis of the ellipse.

[9] E. Johnston-Halperin *et al.*, Phys. Rev. B **51**, 12 852 (1995).

[10] V. Kogan *et al.*, Phys. Rev. B **55**, R8693 (1997).

[11] The (anisotropic) nonlocal terms fall off much faster than the local terms. See, for example, Appendix B in V. G. Kogan *et al.*, Phys. Rev. B **54**, 12 386 (1996). Hence, at large distances from the core, the effects from the local London penetration depth predominate.

[12] D. McK. Paul *et al.*, Phys. Rev. Lett. **80**, 1517 (1998).

[13] M. Eskildsen *et al.*, Phys. Rev. Lett. **78**, 1968 (1997).

[14] All the data on YBCO that we have seen can be explained with one-dimensional or hexagonal structures coupled with a superconducting mass anisotropy which is different in the three principal directions. No exotic structures are suggested by the data. However, discussion of distortions of the rhombohedral structure abound.

[15] H. Kawano *et al.*, Phys. Rev. Lett. **77**, 4628 (1996).

[16] C. Stassis *et al.*, Phys. Rev. B **55**, R8678 (1997).