Flux Lattice Symmetry in V_3Si: Nonlocal Effects in a High-κ Superconductor

M. Yethiraj,[1] D. K. Christen,[1] D. McK. Paul,[2] P. Miranovic,[3] and J. R. Thompson[1,4]
[1]*Solid State Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831-6393*
[2]*Department of Physics, University of Warwick, Coventry, United Kingdom*
[3]*Institute of Physics, University of Belgrade, Yugoslavia*
[4]*University of Tennessee, Knoxville, Tennessee 37996-1200*
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In the high-κ cubic superconductor V_3Si, phase transitions of the flux lattice structure occur as a function of applied field and temperature. With the field parallel to the fourfold [001] axis, the flux lattice transforms from triangular to square symmetry at approximately 1 T. With the field parallel to the twofold [110] axis, the lattice, which is a nearly perfect hexagonal array at the lowest fields, distorts as the field is increased; the rate of increase in this distortion changes abruptly at 1.3 T. As T_c is approached, the system tends towards a more isotropic hexagonal array of the flux lines. These transitions are largely but not completely in agreement with a recent theory of the effects on the flux line arrangements of nonlocal electrodynamics in the London limit. [S0031-9007(99)09351-5]

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Magnetic fields penetrate a type-II superconductor as quantized flux lines. Each flux line contains one quantum $(h/2e)$ of flux and an array of them creates a modulation of the magnetic field. Flux lines are generally expected to be arranged in a regular hexagonal lattice since their interaction is repulsive and a hexagonal lattice that maximizes the distance between them can reasonably be expected to be the lowest energy configuration. However, square lattices of flux lines have been seen experimentally. The flux line lattices (FLL) in the rare-earth nickel borocarbides have been quite interesting because they exhibited not just a square lattice [1,2] but phase transitions in the FLL symmetry [3,4] as the field was increased. It has recently been shown [5] that the square lattice arises from the fact that the cross section of a single flux line has a fourfold symmetric component at short distances from the core and hence the vortices stack in a square array when the distance between them becomes small enough. These symmetry changes have also been explained by theoretical calculations [6] of the effects on the flux line lattice symmetries of nonlocality in the London limit. The above mentioned studies which indicated a fourfold symmetry for the cross section of a single flux line in a material with no (in-plane) mass anisotropy implies a variation of the London penetration depth within the basal plane that is inconsistent with local London theory and points directly to the importance of nonlocal interactions.

In this Letter, we demonstrate that nonlocal electrodynamics plays an important role in clean, high-κ material [7] ($\kappa \approx 25$) V_3Si, which is a cubic superconductor. Nonlocality causes symmetry changes in the flux line lattice and its effects are encountered well outside the 40 Å range of the coherence length. This is quite contrary to what is generally considered to be the case that nonlocal electrodynamics is relevant only where the coherence length, ξ, of a superconductor is large, such as in niobium, where $\kappa \approx 1$. In the compound V_3Si, the effects of nonlocality were thought to be very minor.

Previously, Kogan and collaborators [8] applied their model of nonlocal effects in the London limit to existing small-angle neutron scattering (SANS) data [9] on the FLL in V_3Si, and predicted a number of changes in the vortex lattice symmetry in this compound as a function of field and temperature along various symmetry directions. Here, we report on the observation of the predicted phase transitions in the symmetry of the FLL in V_3Si, studied by SANS techniques.

The measurements were carried out on the 30-m SANS facility at the High Flux Isotope Reactor at Oak Ridge National Laboratory. The incident wavelength used was 4.75 Å. For all the measurements, the neutron beam was horizontal, and two different geometries for the applied field were used. In the horizontal field geometry, the field was nearly collinear with the neutron beam, except rotated (about the vertical axis) by the relatively small Bragg angles to satisfy the scattering condition. In the second geometry, the field was vertical and perpendicular to the neutron beam, the Bragg spots were in the horizontal plane of the detector, and the sample had to be rotated about the vertical axis by the angle between planes. (This would be a rotation of 60° in the case of a perfect hexagonal lattice to observe an equivalent symmetry-related reflection.) The horizontal field geometry has the advantage that all the Bragg spots can be seen simultaneously and symmetries of the lattice are readily apparent. In the vertical field geometry, the angles between basis vectors can be much more accurately determined and are related only to the direct rotation of the sample and not to experimental parameters, such as sample size, detector calibration, or resolution element. The flux line lattice was grown as the sample was cooled in a field.

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The sample of V_3Si was a cylinder of diameter 8.3 mm and length 10.4 mm. The physical cylinder axis was determined to be the a axis of the crystal to within an uncertainty of 2°. The mosaic was less than 0.4°, the resolution with which the measurement was made. The sample had a dH_{c2}/dT slope of -2 T/K and a critical temperature of 16.3 K (midpoint), as characterized by SQUID magnetization measurements. The extrapolated $H_{c2}(0)$ as obtained from the expression $0.71(dH_{c2}/dT)T_c$ is approximately 22 T. With the Ginzburg-Landau expression $H_{c2} = \phi_0/2\pi\xi^2$, this corresponds to a coherence length $\xi_{\rm GL}(0) = 38$ Å. Just above T_c, the electrical resistivity ρ was 1.9×10^{-6} ohm cm (residual resistance ratio RRR = 32). From the product $\rho\ell = 0.64 \times 10^{-11}$ ohm cm^2 (obtained from analyzing the equilibrium magnetization data using Ginzburg-Landau-Abrikosov-Gor'kov (GLAG) expressions [10]), one then obtains the value $\ell = 320$ Å for the electronic mean free path. Thus $\ell \gg \xi$ and we conclude that the material is clean. Clean materials are a prerequisite for the nonlocal effects that have been observed. Although qualitative, another measure of cleanliness is the relatively weak vortex pinning, which gives a critical current density J_c of 1×10^4 A/cm^2 at 5 K in a field of 1 T. For comparison, the depairing current density is $J_0 = cH_c/3\sqrt{6}\,\lambda \approx 2 \times 10^8$ A/cm^2, corresponding to a ratio $J_c/J_0 \approx 10^{-4}$. The weak pinning and particularly the large mean free path imply that nonlocal effects may be pronounced in this material.

Of the predictions mentioned above, the first was that with the applied field parallel to the a axis at low temperature, there would be a transition from hexagonal to square symmetry at about 3 T. This hexagonal-square transition was observed, in fact, but at a field of 1 T, considerably smaller than predicted. Scattering patterns for the FLL at three different fields (0.75, 1.0, and 1.5 T) are shown in Fig. 1. Here, the horizontal and vertical axes are symmetry-equivalent 110 directions and the field is applied along the [001], an axis with fourfold symmetry. At low field, 0.75 T, the underlying fourfold symmetry of the crystalline lattice manifests itself in the existence of two triangular FLL domains, one of which is considerably larger than the other. Each of these domains is oriented such that the triangle base vector of each FLL is parallel to a crystallographic 110 direction. At 1 T, the intensity is split about equally between the triangular and square lattices and by 1.5 T, nearly all of the intensity is in the square lattice and a very small fraction of the intensity can be attributed to the triangular array. Because there is no mechanism by which this change could possibly occur in a continuous manner and there are no intermediate changes in structure, it would suggest that this transition is first order in nature. However, although the transition from triangular to square appears to happen in a single step, it does occur over an extended range of applied field. This could be influenced by a varied density of pinning centers or other inhomogeneities in the crystal.

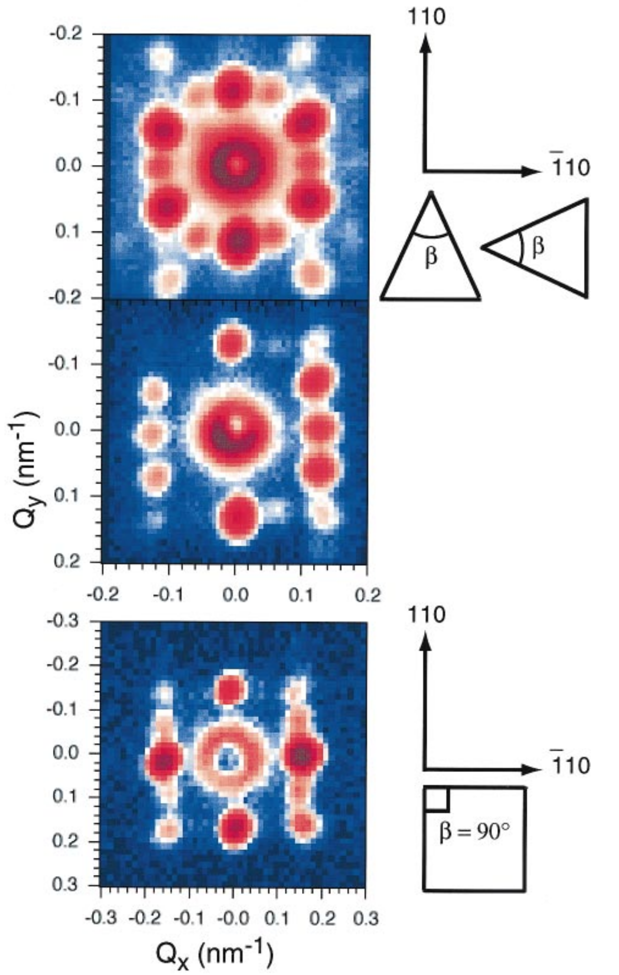

FIG. 1(color). The scattering pattern from the FLL at 0.75, 1.0, and 1.5 T shows the structural transition in the flux line lattice. The real space arrangement and its relationship to the crystal orientation is indicated alongside. The scattering pattern at 1.0 T is a superposition of the triangular and square lattices. The sample temperature was 1.8 K. The 0.75 and 1.0 T data are taken at a single angular setting. The 1.5 T data are summed over a rocking curve.

The stable structure near T_c, where the lattice first forms (since the sample is field cooled), could be different than that at low temperatures. Pinning could cause the structure stabilized near T_c to persist. However, the narrow transition $(\Delta T_c(10\%-90\%) \approx 0.2$ K) indicates relatively high compositional homogeneity, and nearly reversible magnetization curves are observed near $T_c(H)$.

Although the FLL appears to have sixfold symmetry at lower fields ($B \parallel [001]$), the lattice here is not perfectly hexagonal. That is, the apex angle β, the angle between the two Bragg peaks, indicated in Fig. 1 is not exactly 60°. The angle β extrapolates to 60° at zero field and it is smaller than 60° for low values of the applied field. The apex angle decreases with increasing field until the transition occurs to the square symmetry (where β is 90°) (see Fig. 1). The measurement of the lattice perfection in the triangular phase was resolution limited. However, for the square FLL, the mosaic spread [11] (vertical field geometry) was quite broad (2° FWHM).

In the geometry where the applied field is parallel to the $[\bar{1}10]$, a nearly perfect hexagonal lattice is seen at the lowest field ($\beta = 62°$ at 0.5 T) in agreement with previous [9] measurements. Here, only one domain exists and the primary Bragg reflections are aligned once again to a crystallographic [110] direction. The existence of a single domain is consistent with the twofold crystal symmetry in this geometry, since the [110] and [001], which are perpendicular to each other, are not equivalent directions. With an increasing field, the lattice is seen to be distorted from the perfect hexagonal. As a function of the field, the variation of the apex angle, β, for this geometry is shown in Fig. 2. It is seen that β increases with the applied field; however, the field dependence changes abruptly at 1.3 T, approximately the same field at which the hex-square phase transition occurs for $B \parallel [001]$. Beyond 1.3 T, the increase in β appears to remain linear up to 5 T, the highest field measured. The FLL at all fields was perfect and the mosaic was limited by the resolution for $B \parallel [\bar{1}10]$.

The theoretical prediction for the FLL in this geometry ($B \parallel [\bar{1}10]$) is the same as that for $B \parallel [001]$. That is, a hexagonal to square lattice transition is expected at the same field as that in the other geometry. However, it should be noted that the FLL symmetry in the lowest order theoretical construct (fourfold) is higher than the actual crystallographic (twofold) symmetry.

As the temperature is increased, the apex angle tends towards 60°. The apex angles for any given field are stable up to approximately 7 K (Fig. 3). Above this temperature, β systematically decreases with temperature. There is some unusual dependence of β at 1 T, near the field where the transitions occur in the FLL as a function of field. At 1 T, the nearest neighbor distance between the flux line centers is approximately 450 Å in a square arrangement (or 480 Å for a triangular configuration). This is not a particularly suggestive length scale; it would appear that the features in the anisotropy are strong at this distance from the vortex core. It has been postulated that the magnetic field falloff has a short range anisotropic part and a more slowly varying isotropic part. In this case, at low fields when the vortices are far from each other, the interaction potential is nearly isotropic. But at larger fields, the vortices are closer to each other and the anisotropy becomes more evident. This is certainly the trend that is observed; the hexagonal lattice is more strongly distorted as the field is increased.

The intensity of the flux lattice Bragg peak with $B \parallel [001]$ is considerably lower (by a factor of more than 3) than that with the field parallel to the $[\bar{1}10]$. This is not entirely unreasonable for the following reason: The lattice in the first instance ($B \parallel [001]$) consists of two lattices related by symmetry to each other by a rotation of 90°. Even though one of these lattices is much less populated than the other, the average size of a "perfect" domain in the more prevalent direction is most likely smaller than in the case where a single lattice exists. Since the intensity of a Bragg peak [12] is proportional to the square of the number of (coherent) scatterers, the Bragg peak for B parallel to $[\bar{1}10]$, with its single lattice orientation, is likely to have a higher intensity in the Bragg peak, all other factors remaining equal. However, given this inequity in the intensities and the fact that there exists a symmetry transition in the middle of the range, the temperature dependence of the intensities cannot be analyzed in a simple manner. More information is required, particularly for higher order reflections, in order to adequately correct the intensities and elicit the "true" dependence of the order parameter.

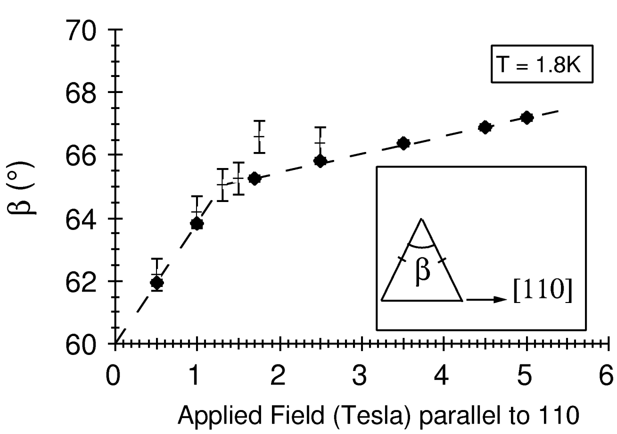

FIG. 2. The apex angle, β, varies as a function field and shows an abrupt change in slope at 1.3 T. Note that the data acquired in the vertical field geometry (filled diamonds) are considerably more precise than those obtained in the horizontal field geometry (crosses).

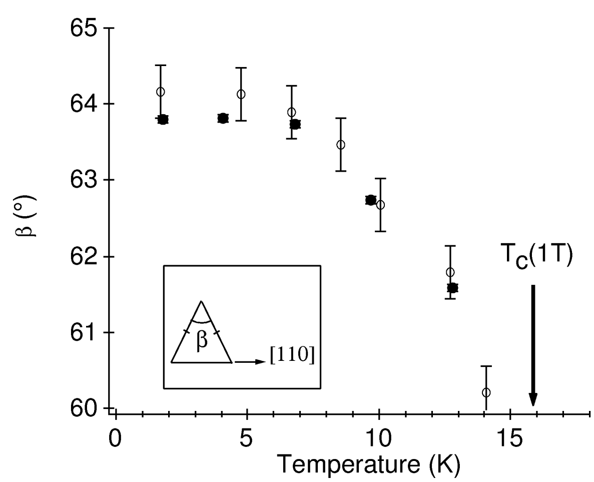

FIG. 3. The temperature dependence of the apex angle, β, at an applied field of 1 T. $T_c(1\ \text{T}) = 15.8$ K (vertical field: filled circles; horizontal field: open circles).

In the local London model, the local supercurrent density, j, is proportional to the vector potential, a, at the corresponding point in space. In the nonlocal generalization, the current density becomes k dependent with $j(k) = Q(k)a(k)$, where Q is k dependent kernel. This implies that the current j at a point r is proportional to an average over a surrounding volume of radius $\approx \xi_{BCS}$. Within the first order terms of the nonlocal correction (in the theoretical treatment), the magnetic field distribution has fourfold symmetry for any field direction, except along the [111] direction where it is isotropic (since the anisotropy parameter in this case is zero). Since the true symmetry for $B \parallel [111]$ is sixfold rather than isotropic, the correct symmetry is reproduced in the model only for the field parallel to the 001 direction. The first nonlocal correction term with its fourfold symmetry will stabilize a square FLL in high fields for any field direction other than the [111]. On the other hand, a square FLL is experimentally observed only for the field along the fourfold (001) axis. It is obvious from the data that at high fields, the full k dependence of the expression [6,8] becomes important and just the first order term is insufficient. Accounting for the higher order terms in the nonlocal correction has proved to be cumbersome, but it was readily apparent that the next term in the expansion additionally breaks the symmetry such that it is twofold for $B \parallel [\bar{1}10]$ and sixfold symmetry for $B \parallel [111]$, while remaining 90° invariant along the 001 direction. Thus, the formal incorporation of the next higher order correction term would make the symmetry of the field coincide with the symmetry of the underlying lattice.

In conclusion, it is seen that nonlocal effects are important even in high-κ cubic materials. Here, since mass anisotropy is absent, all distortions of the FLL may be considered as variations of the penetration depth due to the effects of the finite size of the core. Where the anisotropy of the intervortex interaction is felt strongly enough to precipitate the triangular-square transformation, the distance between vortices is about 450 Å, which is more than 10 times the coherence length.

It is evident from present and previous measurements that the FLL and crystal lattice symmetry are highly correlated. The FLL shape is dependent on the field direction and always reflects the symmetry of the crystal plane normal to the field. The other distinctive feature is that the hexagonal FLL at low inductions distorts as the field increases and abruptly becomes square for certain field directions. This is in contradiction with the prediction of the London model for cubic superconductors. Within the simple local London model, vortices repel each other with force that depends on distance only; the interaction is isotropic, which should result in a hexagonal rather than square lattice. Presumably, in fields much smaller than H_{c2}, the vortex-vortex interaction is mainly due to the Lorentz force and is given by a single vortex field distribution. The experimental results indicate that there is a region around the vortex core where magnetic field distribution is anisotropic (even in cubic crystals) and that the anisotropy depends on the vortex orientation within the crystal. Finally, since the field falloff from a vortex is direction dependent, this will result in an anisotropic suppression of the gap as well. This may well effect the predictions of the FLL symmetry, since the theoretical construct assumes a naive picture where the gap is isotropic and the core has no structure.

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[1] M. Yethiraj, D. McK. Paul, C. V. Tomy, and E. M. Forgan, Phys. Rev. Lett. **78**, 4849 (1997).
[2] U. Yaron *et al.,* Nature (London) **382**, 236 (1996).
[3] M. R. Eskildsen *et al.,* Phys. Rev. Lett. **78**, 1968 (1997).
[4] D. McK. Paul *et al.,* Phys. Rev. Lett. **80**, 1517 (1998).
[5] M. Yethiraj, D. McK. Paul, C. V. Tomy, and J. R. Thompson, Phys. Rev. B **58**, R14 767 (1998).
[6] V. G. Kogan *et al.,* Phys. Rev. B **55**, R8693 (1997).
[7] The coherence length is approximately 40 Å and the penetration depth is 1000 Å.
[8] V. G. Kogan *et al.,* Phys. Rev. Lett. **79**, 741 (1997).
[9] D. K. Christen *et al.,* Physica (Amsterdam) **135B**, 369 (1985). These measurements were all performed at fields less than 1 T; hence the structural transition was not observed.
[10] BCS and GLAG relations are conveniently tabulated in the appendix of T. P. Orlando, E. J. McNiff, Jr., S. Foner, and M. R. Beasley, Phys. Rev. B **19**, 4545 (1979).
[11] The rocking curve width in the vertical field geometry measures the perfection of the two-dimensional lattice over the whole sample, i.e., the mosaic of the flux line crystal. This is not a measure of the straightness or length of the flux lines, as it is in the horizontal field geometry.
[12] C. G. Darwin, Philos. Mag. **27**, 315 (1914).