Temperature Dependence of the Flux Line Lattice Transition into Square Symmetry in Superconducting $LuNi_2B_2C$

M. R. Eskildsen,[1,*] A. B. Abrahamsen,[1] V. G. Kogan,[2] P. L. Gammel,[3]
K. Mortensen,[1] N. H. Andersen,[1] and P. C. Canfield[2]
[1]Risø National Laboratory, P.O. Box 49, DK-4000 Roskilde, Denmark
[2]Ames Laboratory and Department of Physics and Astronomy, Iowa State University, Ames, Iowa 50011
[3]Agere Systems, 700 Mountain Avenue, Murray Hill, New Jersey 07974
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We have investigated the temperature dependence of the **H** ∥ **c** flux line lattice structural phase transition from square to hexagonal symmetry, in the tetragonal superconductor $LuNi_2B_2C$ ($T_c = 16.6$ K). At temperatures below 10 K the transition onset field, $H_2(T)$, is only weakly temperature dependent. Above 10 K, $H_2(T)$ rises sharply, bending away from the upper critical field. This contradicts theoretical predictions of $H_2(T)$ merging with the upper critical field and suggests that just below the $H_{c2}(T)$ curve the flux line lattice might be hexagonal.

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Studies of the topology of the magnetic flux line lattice (FLL) in type-II superconductors have a long history. Early neutron scattering experiments on low-κ superconductors such as niobium revealed a multitude of different FLL symmetries and orientations, mainly determined by the symmetry of the atomic crystal structure in the plane perpendicular to the applied field [1]. This is not surprising since deviations from the hexagonal FLL characteristic of an isotropic superconductor, and the locking to the crystalline lattice are driven by the symmetry of the screening current plane and by nonlocal flux line interactions within a range determined by the coherence length ξ_0, and the electronic mean free path ℓ. Later, similar effects were also observed in the strong type-II superconductor V_3Si with $\kappa \approx 17$ [2], demonstrating that nonlocal effects can be equally important in high-κ materials.

Over the last couple of years, effects of nonlocality have been clearly observed in the borocarbide superconductors. The borocarbides are quaternary intermetallics with stochiometry RNi$_2$B$_2$C (R = Y, Gd–Lu) and a tetragonal unit cell ($I4/mmm$) [3]. These materials have attracted attention due to the coexistence of superconductivity (R = Y, Dy–Tm, Lu) and antiferromagnetic ordering (R = Gd–Tm). The borocarbides are strong type-II superconductors with Ginzburg-Landau (GL) parameter, $\kappa = 6$–15. The discovery of a square FLL in most of the **H** ∥ **c** phase diagram [4], which undergoes a smooth transformation into hexagonal symmetry at fields below 1 kOe [5], was the first observation of a purely field driven FLL symmetry transition.

Using nonlocal corrections to the London model and incorporating the symmetry of the screening current plane obtained from band structure calculations, one is able to calculate the FLL free energy and thereby to determine the stable FLL configuration in different fields in the borocarbides [6]. The model successfully describes the nature of the FLL square to hexagonal symmetry evolution in the borocarbides with **H** ∥ **c** as the applied field is reduced. Qualitatively, this can be understood as driven by the fourfold basal plane anisotropy which makes the vortex current paths "squarish" close to the core. At high densities this leads to a square FLL, whereas at low fields the system appears isotropic resulting in a hexagonal FLL [7]. The onset of the transition occurs as the field decreases, commencing at a critical field H_2, determined by the range of the nonlocal interactions [6]. Later studies of the FLL in a set of samples doped with various amounts of impurities to systematically change the nonlocality range showed a quantitative agreement between the predicted and measured onset of the transition [8,9]. The model also predicted a reorientation transition of the rhombic FLL at a field $H_1 \ll H_2$, which has been subsequently observed [10,11].

Whereas there is a growing experimental and theoretical understanding of the FLL geometry and transitions for fields well into the mixed state, there has been little attempt to understand what happens at fields just below the upper critical field. In this Letter we report on detailed measurements of the temperature dependence of the structural transition, $H_2(T)$, in $LuNi_2B_2C$ with the applied field parallel to the crystalline c axis, over a broad range of temperatures and applied fields. Below 10 K, $H_2(T)$ is essentially constant. Above 10 K, $H_2(T)$ rises sharply, eventually bending away from the $H_{c2}(T)$ curve, suggesting the existence of a hexagonal FLL in the region just below the upper critical field.

The FLL in $LuNi_2B_2C$ ($T_c = 16.6$ K) was studied using the small angle neutron scattering spectrometer on the cold neutron beam line at the Risø National Laboratory DR3 research reactor. The sample used was a 1 g single crystal, grown from a high temperature flux [3] using isotopically enriched ^{11}B to enhance the neutron transmission. Incident neutrons with wavelengths in the range $\lambda_n = 6$–15.6 Å and a bandwidth $\Delta\lambda_n/\lambda_n = 24\%$ were used, and the FLL diffraction pattern was collected by a position sensitive

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detector at the end of a 6 m evacuated tank. Magnetic fields in the range 1.5 to 10 kOe were applied parallel to the crystalline c axis and to the incoming neutrons. In order to keep the resolution of the spectrometer unchanged, the neutron wavelength was varied in concert with the applied field to keep the scattering angle, $2\theta = q_{10}\lambda_n/2\pi$, constant. Here q_{10} is the FLL scattering vector shown in Fig. 1. The measurements were performed at temperatures between 2 and 20 K, following a constant field cooling procedure. A background signal obtained above T_c was subtracted from the data.

In Fig. 1 we show three different FLL diffraction patterns obtained in $LuNi_2B_2C$. In quasi-2D structures such as that of the FLL, the real space flux line configuration and the reciprocal space diffraction pattern are related by a simple 90° rotation and relabeling of the axes. The diffraction pattern therefore directly shows the FLL symmetry and orientation, with the caveat that scattering from differently oriented domains is superposed. For the square FLL there is no orientational degeneracy with respect to the atomic lattice, due to the tetragonal crystal structure of the borocarbides, as reflected in the top panel of Fig. 1. Here both the FLL $(1,0)$ and $(1,1)$ reflections are clearly visible, and resolution limited both in the radial and the azimuthal direction. The square FLL is aligned with the scattering vector q_{10} along the $[110]$ crystal direction.

However, as soon as the FLL is distorted away from a square symmetry there is a twofold degeneracy, since orientations with the long rhombic unit cell diagonal along either the a or the b axis have the same energy. Experimentally this will first manifest itself as an azimuthal broadening and, when the distortion increases as a splitting of the $(1,0)$ reflections, with the two peaks each belonging to a different domain orientation [5]. The magnitude of the splitting will eventually reach 30° if the FLL is transformed to a perfect hexagonal symmetry. The azimuthal broadening is evident in the middle panel of Fig. 1, and in the bottom panel the $(1,0)$ reflections are clearly split. The effect of the transition is also reflected in the square FLL $(1,1)$ reflections. Ideally a radial splitting should be observed, with one peak moving towards shorter q and gaining intensity and the other moving to longer q and fading, to become, respectively, hexagonal $(1,0)$ and $(1,1)$ reflections belonging to different domain orientations. However, due to the very rapid decrease in the scattered intensity with increasing q, one observes only a radial broadening and a relative increase in the intensity.

In general, the azimuthal splitting was only directly observable when above $\sim 10°$, comparable to experimental resolution. To determine the transition onset field $H_2(T)$, we analyzed the azimuthal intensity distribution in each diffraction pattern at a scattering vector $q = q_{10}$, where $q_{10} = \alpha \times 2\pi(B/\phi_0)^{1/2}$, $\phi_0 = hc/2e$ is the flux quantum, and α is a constant which depends on the FLL

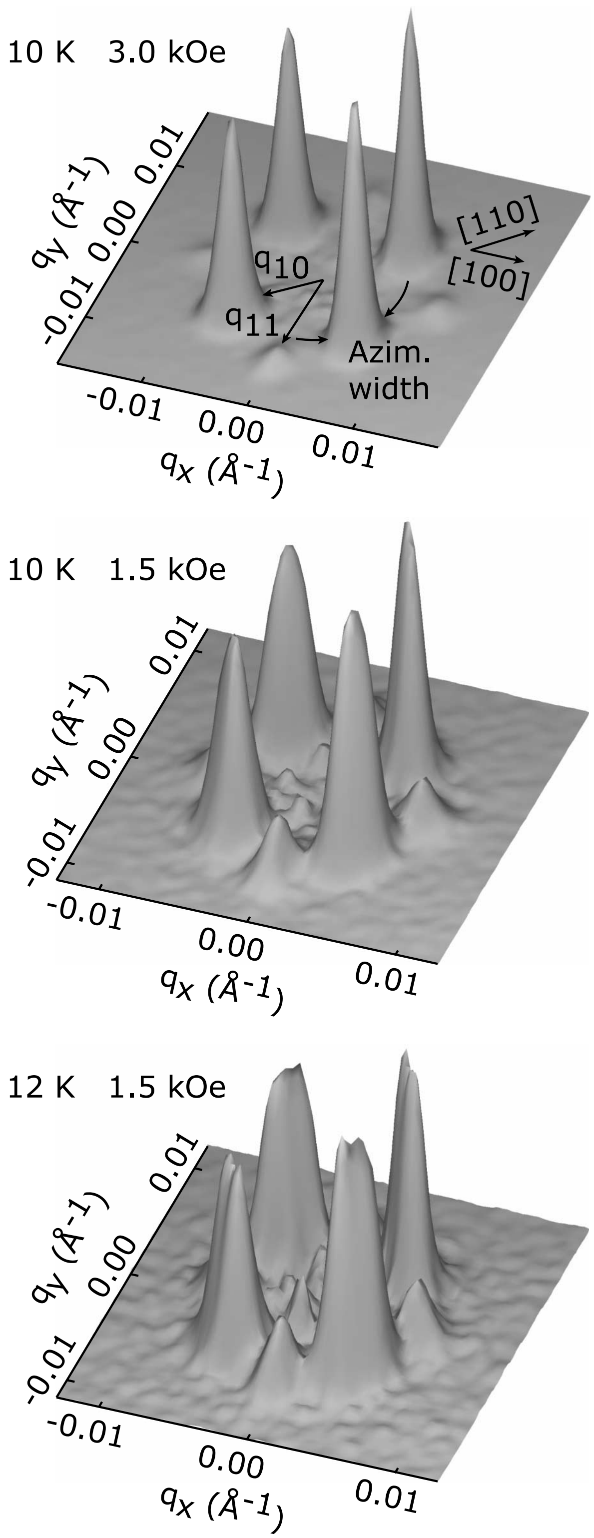

FIG. 1. FLL diffraction patterns from $LuNi_2B_2C$ obtained at temperatures of 10 and 12 K and fields of 1.5 and 3 kOe. The orientation relative to the crystallographic axes, and the FLL $(1,0)$-peak azimuthal width is shown in the top panel. Each diffraction pattern is a sum of four measurements with the sample oriented to satisfy the Bragg condition for each of the main peaks/groups of peaks, and subtracted by background measurements taken at 20 K. The neutron wavelengths used were, respectively, $\lambda_n = 15.6$ Å (1.5 kOe) and 11.0 Å (3 kOe). The scattering close to $q = 0$ is an artifact of imperfect background subtraction.

symmetry: With a FLL apex angle β as shown in Fig. 3, $\alpha = (\sin\beta)^{-1/2}$, and hence $\alpha = 1$ for the square case ($\beta = 90°$) and $(2/\sqrt{3})^{1/2} = 1.075$ for the hexagonal FLL ($\beta = 60°$ or $120°$). In the top panel of Fig. 2 we show the temperature dependence of the azimuthal width for $H = 3$ kOe, obtained by fitting to a single Gaussian. A clear broadening is seen for $T > 10$ K. The low-T width of $13.8° \pm 0.2°$ is a direct measure of the azimuthal resolution, which was kept constant throughout the experiment. To determine the azimuthal splitting, the intensity distribution at each temperature and field was then fitted to two Gaussians with a width fixed of 13.8°, as demonstrated in the top panel inset. In the bottom panel of Fig. 2 the azimuthal splitting is plotted versus temperature for a number of fields between 1.5 and 6 kOe. At low temperatures the splitting goes to $0° \pm 2°$ as the field is increased. As T is raised the splitting increases smoothly for $H \leq 2$ kOe and abruptly above a certain threshold for $H \geq 3$ kOe. The threshold is determined by $H_2(T)$. In addition, one notes a crossing of the curves for 4.5 and 6 kOe, which means that $H_2(T)$ is a multivalued function of T.

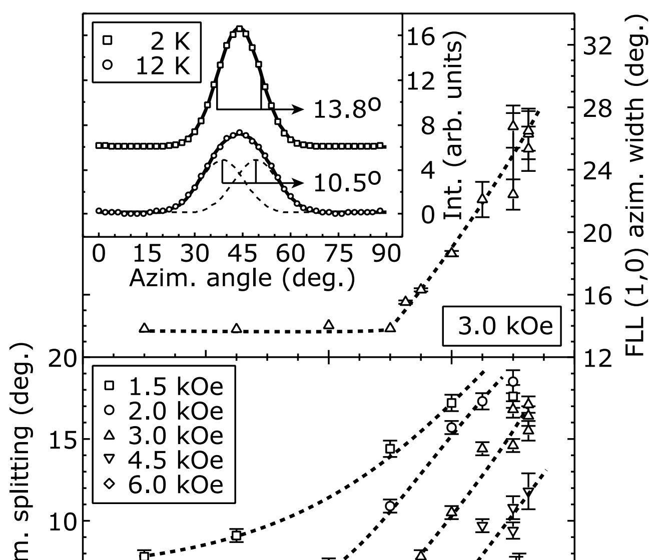

FIG. 2. Top panel: The FLL (1,0)-reflection azimuthal width obtained by fitting a single Gaussian to the azimuthal intensity distribution, as a function of temperature at an applied field of 3 kOe. Inset: FLL azimuthal intensity distribution folded into one quadrant, at 3 kOe and 2 and 12 K. The 2 K data (offset for clarity) are well fitted by a single Gaussian of width 13.8° equal to the experimental resolution. The 12 K data are fitted by two equal weight Gaussians with the same width, yielding a splitting of $10.5° \pm 0.4°$. Bottom panel: Azimuthal splitting versus temperature for applied fields between 1.5 and 6 kOe. The error bars include both the error on the direct fit of the splitting and the uncertainty in the experimental resolution. The dashed lines are guides to the eye.

Figure 3 is a contour plot of the azimuthal splitting in a (H,T)-phase diagram. Here we have used the 3° contour, well above the zero-splitting error of 2°, as the criterion for the transition onset field. Below 10 K, $H_2(T)$ is nearly constant, equal to 2.25 kOe. This is somewhat higher than the literature values [5], but as shown by Gammel *et al.* [9], H_2 depends strongly on the sample purity as this affects the range of the nonlocal flux line interactions. Above 10 K, $H_2(T)$ increases sharply, bending away from $H_{c2}(T)$ and thereby avoiding to cross the upper critical field. From this it is clear that the square FLL is not stable at $H_{c2}(T)$. The strong temperature dependence contradicts predictions based on extended GL theory [12,13], where $H_2(T)$ intercepts $H_{c2}(T)$ at a finite field. An extrapolation of our measurements suggests that in the vicinity of $H_{c2}(T)$, the FLL may become hexagonal.

Theoretically the nonlocal contribution to the vortex-vortex interaction weakens when the temperature rises, and $H_2(T)$ should somewhat increase. This relatively weak increase is essentially captured in the London approach, where the order parameter $|\Delta|$ is taken as a constant in space [14]. Moreover, $|\Delta|$ is assumed equal to the zero-field uniform value $\Delta_0(T)$ which determines the condensation energy $F_N - F_S$ of uniform superconductors ($\propto\Delta_0^2$ at low temperatures and $\propto\Delta_0^4$ near T_c). Being constant in space and field independent, the condensation energy so defined is commonly disregarded in calculations of the equilibrium FLL. Within this scheme, $|\Delta|$ affects only the value of the penetration depth, $\lambda \propto 1/|\Delta|$. The model is good for flux line spacings large relative to the

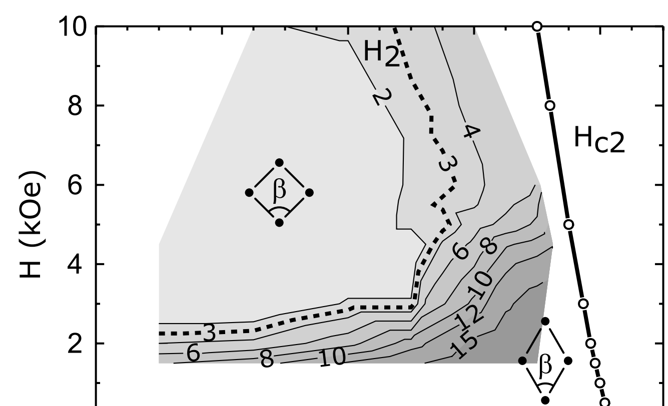

FIG. 3. (H,T)-phase diagram showing constant azimuthal splitting contours. The dashed line corresponds to a splitting of 3° and is taken as the FLL symmetry transition onset field $H_2(T)$. Approaching $H_{c2}(T)$ the scattered intensity vanishes and the shaded area shows the range of our measurements. A schematic illustration of the FLL symmetry and apex angle is shown for the square (center) and hexagonal case (low H, high T).

core size $\xi(T)$, i.e., in small fields, and for temperatures well below T_c.

However, with increasing density of vortices at a fixed T, the spatial average $\langle|\Delta|\rangle$ decreases thus causing an overall increase of the system energy [15]. The $\langle|\Delta|\rangle$ suppression depends on the ratio $a/\xi(T)$, with a being the intervortex spacing. Therefore, at a given flux line density, B/ϕ_0, the system energy can be reduced if the intervortex distance is maximized. This can be interpreted as an extra (to the London interaction) repulsion of vortices. This repulsion is isotropic for cubic materials or for tetragonal crystals with $\mathbf{H} \parallel \mathbf{c}$ and will lead to a deviation from square symmetry and eventually to a hexagonal FLL (or triangular with the structure determined only by the ratio ξ_a/ξ_b for $\mathbf{H} \parallel \mathbf{c}$). We speculate that this effect should become dominant on approaching $H_{c2}(T)$ where $|\Delta|$ is strongly suppressed, in accord with the data presented in this Letter.

In addition, we have earlier reported that in $TmNi_2B_2C$ at low temperatures the FLL is hexagonal near $H_{c2}(T)$. With decreasing field, the FLL transforms first to a rhombic structure followed by the square [16]. As H is further reduced, the FLL should go through the same evolution in inverse order, confirmed by low field decoration data [11]. This material is antiferromagnetic below 1.5 K, and it is possible that the magnetic ordering has a detrimental effect on the nonlocality range in fields near $H_{c2}(T)$. If so, both the antiferromagnetism and the order parameter suppression would have similar effects on the FLL, which may explain why the domain of "reverse evolution" is so broad in $TmNi_2B_2C$. Finally, also in agreement with our hypothesis are data on the FLL in V_3Si that show how a distorted triangular lattice which is seen at $T < 5$ K and $H = 10$ kOe evolves toward hexagonal when T approaches $T_c(H)$ [17].

Within the microscopic theory applied to the mixed state, there is no artificial separation of the electromagnetic (London) intervortex forces and those due to the varying order parameter. This problem, however, has never been addressed for the FLL [18]. The GL approach does provide such a complete description, but it is valid only near the zero-field T_{c0}. The accuracy of results obtained within GL for $T < T_{c0}$ is difficult to control. Moreover, the very existence of the GL expansion in powers of the order parameter gradients away from T_{c0} is questionable. The coherence length $\xi(T)$ does not diverge as $T \to T_c(H)$; as a consequence the gradients of Δ are not necessarily small, and the convergence of the GL series is difficult (if not impossible) to monitor. In this respect, calculations based upon keeping certain quartic terms in the GL expansion while discarding others [12,13] can only be justified by a microscopic theory, which is still to be done.

In conclusion, we have in this Letter reported measurement of the temperature dependence of the FLL square to hexagonal symmetry transition in $LuNi_2B_2C$. Our data show that the transition field $H_2(T)$ bends away from the upper critical field line $H_{c2}(T)$. This suggests that while approaching the $H_{c2}(T)$ curve, the FLL is driven towards hexagonal symmetry. We propose a possible qualitative argument for why this might be the case. This poses new questions and challenges concerning the phase diagram of the flux line solid.

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*Present address: DPMC, Université de Genève, 24 Quai E.-Ansermet, CH-1211 Genève 4, Switzerland.
Email address: morten.eskildsen@physics.unige.ch

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