

## Hexagonal and Square Flux Line Lattices in CeCoIn<sub>5</sub>

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Using small-angle neutron scattering, we have imaged the magnetic flux line lattice (FLL) in the *d*-wave heavy-fermion superconductor CeCoIn<sub>5</sub>. At low fields we find a hexagonal FLL. Around 0.6 T this undergoes what is most likely a first-order transition to square symmetry, with the nearest neighbors oriented along the gap node directions. This orientation of the square FLL is consistent with theoretical predictions based on the *d*-wave order parameter symmetry.

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Recently, a whole new family of heavy-fermion superconductors has been discovered. It includes pressure-induced superconductivity in CeRhIn<sub>5</sub> [1], ambient pressure superconductivity in CeIrIn<sub>5</sub> [2], and superconductivity with the highest known  $T_c = 2.3$  K for any heavy-fermion at ambient pressure in CeCoIn<sub>5</sub> [3]. The CeMIn<sub>5</sub> ( $M = \text{Rh, Ir, Co}$ ) family exhibits several similarities to other correlated electron superconductors such as high- $T_c$  cuprates and crystalline organic metals: their crystal structure consists of alternating units of CeIn<sub>3</sub> and MIn<sub>2</sub> stacked sequentially along the *c* axis [4,5], the superconducting state borders on a magnetically ordered phase giving rise to competition [1] or coexistence of magnetism and superconductivity [6,7]. Finally there is evidence from thermal conductivity measurements [8] and NMR [9] indicating that CeCoIn<sub>5</sub> is a *d*-wave superconductor, with line nodes along the [110] and [1 $\bar{1}$ 0] directions ( $d_{x^2-y^2}$ ). Theoretically, *d*-wave pairing is expected to stabilize a square flux line lattice (FLL) [10–13], which was indeed recently reported in the high- $T_c$  superconductor La<sub>1.83</sub>Sr<sub>0.17</sub>CuO<sub>4+ $\delta$</sub>  (LSCO) [14]. However, in the latter case the orientation was rotated 45° with respect to the theoretical predictions [10–13]. In addition, it is worth pointing out that studies of the FLL symmetry in LSCO [14] as well as in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$</sub>  (YBCO) [15–17] are susceptible to potential complications in interpretation due to the orthorhombic crystal structure, which leads to formation of twin planes capable of pinning the FLL. On the other hand, the crystal structure of CeCoIn<sub>5</sub> is tetragonal, which excludes twinning.

Here we report FLL imaging in CeCoIn<sub>5</sub>, obtained by small-angle neutron scattering (SANS). The FLL undergoes what appears to be a first-order, field-driven transi-

tion from a hexagonal to a square symmetry. A square FLL has not previously been observed in a heavy-fermion superconductor, and, furthermore, this is the first example of a square FLL in a *d*-wave superconductor oriented with the nearest neighbor directions parallel to the nodal directions of the gap.

The SANS experiment was carried out at the D11 small-angle neutron scattering diffractometer at the Institut Laue-Langevin, Grenoble, France. Single crystals of CeCoIn<sub>5</sub> were grown from an excess indium flux [3], and had a  $T_c = 2.3$  K and  $B_{c2}(0) = 5.0$  T parallel to the *c* axis. The sample was composed of four single crystals with thicknesses  $t = 0.16$ – $0.2$  mm mounted side by side, each of which was individually aligned. The rather thin samples were necessary, due to the strong absorption of low-energy neutrons by In. The total mass of the sample was 36 mg. Incident neutrons with wavelength  $\lambda_n = 0.6$  nm and a wavelength spread  $\Delta\lambda_n/\lambda_n = 10\%$  were used, and the FLL diffraction pattern was collected by a  $64 \times 64$  (1 cm<sup>2</sup>) position sensitive detector. For all measurements, the sample was field cooled to 50 mK in a dilution refrigerator insert, placed in a cryostat with a superconducting magnet. Magnetic fields in the range 0.3 to 2 T were applied parallel to the crystalline *c* axis, and background subtraction was performed using measurements following a zero-field cooling.

In Fig. 1 we show FLL diffraction patterns for applied fields of 0.3, 0.6, and 2 T. The images were constructed by summing a number of measurements at different angular positions, in order to satisfy the Bragg condition for the different peaks. A clear evolution of the FLL symmetry and orientation is evident. At the lowest field, 12 peaks are observed evenly distributed on a circle in reciprocal space as shown in Fig. 1(a). This corresponds to two

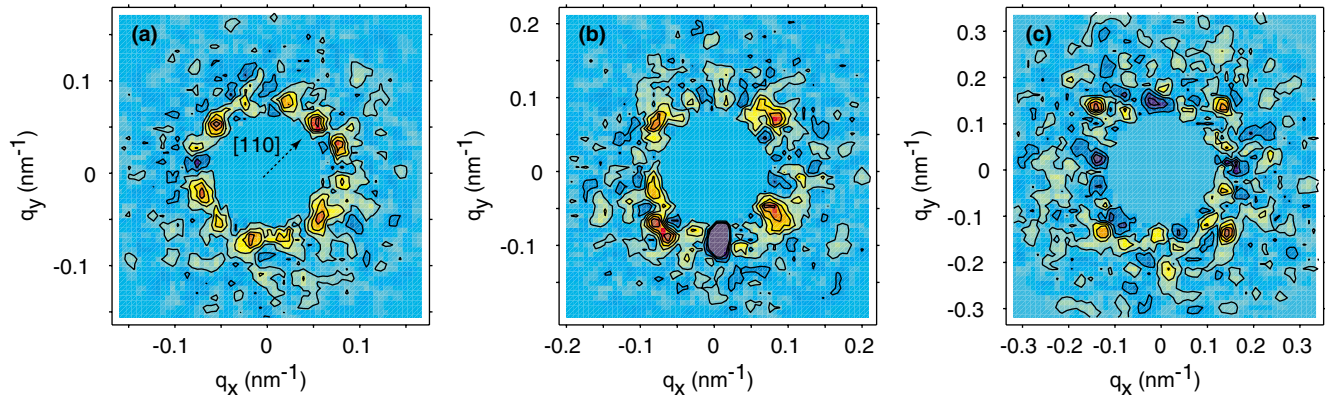


FIG. 1 (color). FLL diffraction patterns for CeCoIn<sub>5</sub> obtained at 50 mK and applied fields of 0.3 T (a), 0.6 T (b), and 2 T (c), after subtraction of background measurement. In addition the data are smoothed by a  $3 \times 3$  box car average, and the center of the image is masked off. The arrow in panel (a) shows the orientation of the crystalline axes.

hexagonal domains oriented along the  $[110]$  or the  $[\bar{1}10]$  directions. For a hexagonal FLL oriented with respect to an underlying square crystal symmetry, the existence of two degenerate domain orientations having equal population is expected. As the field is increased to 0.6 T, a primarily square FLL is found [Fig. 1(b)], with the majority of the scattered intensity concentrated in four irregularly shaped peaks. Again, the FLL is oriented with the nearest neighbor direction along  $[110]$  and  $[\bar{1}10]$ . The square FLL remains stable up to the highest measured field of 2 T [Fig. 1(c)], where the diffraction pattern now shows four distinct Bragg peaks. The symmetry and orientation of the FLL is shown schematically in real space in Fig. 2, and compared to the symmetry of the superconducting gap. While preserving the nearest neighbor direction along  $[110]$ , the transition from hexagonal to square symmetry can, in principle, be continuous. However, at intermediate fields this would result in diffraction patterns with contributions from four sheared hexagonal lattice orientations, analogous to what was previously observed in YBCO [15,16]. Such a distortion of the hexagonal FLL was not observed, and we speculate that the transition from square to hexagonal symmetry is therefore most likely discontinuous, i.e., of first order. On the other hand, we cannot exclude that the transition to square symmetry is preceded by a weak rhombic distortion of the hexagonal FLL, similar to what was ob-

served in the borocarbides [18]. However, such a distortion would not alter the order of the transition from being first order. Finally, a coexistence of domains having, respectively, square and hexagonal symmetry is expected and usually observed in a narrow field range around a first-order transition [19]. We expect this to be the reason for the slightly disordered square diffraction pattern seen in Fig. 1(b), and hence take the corresponding applied field of 0.6 T to be at or close to the transition field. The most likely first-order FLL symmetry transition, and the orientation of the square FLL along the nodes of the superconducting gap, are the main results of this Letter, and will be addressed in further detail below.

SANS FLL imaging in CeCoIn<sub>5</sub> is complicated by two factors, illustrated by the diffraction patterns in Fig. 1 and the 2 T rocking curve shown in Fig. 3. The most limiting factor is the long superconducting penetration depth in this material, which results in a very small field modulation and hence low scattered intensity. This inevitably leads to imperfect background subtractions, which is

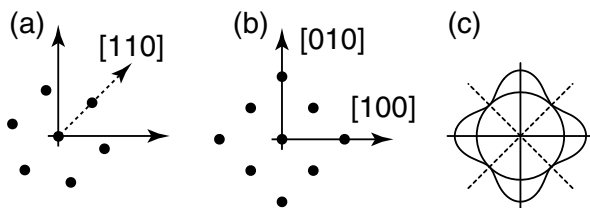


FIG. 2. Symmetry and orientation of the hexagonal (a) and square (b) FLL. The orientation of the nodes in the superconducting gap is shown in (c).

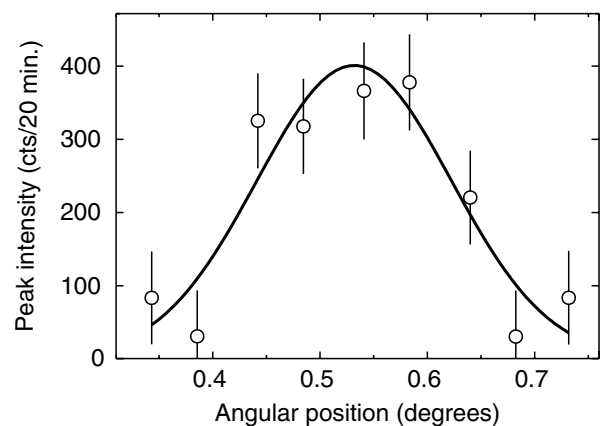


FIG. 3. Rocking curve for the CeCoIn<sub>5</sub> (1, 0) FLL reflection at 50 mK and 2 T. The curve is a fit to a Gaussian.

seen as negative scattered intensity in the diffraction patterns in Fig. 1 (dark blue regions). Figure 3 shows the intensity of a FLL Bragg reflection, as the cryostat is gradually tilted and rotated in such a way that the scattering vector cuts the Ewald sphere at a right angle. The integrated reflectivity is given by [20]

$$R = \frac{2\pi\gamma^2\lambda_n^2 t}{16\phi_0^2 q} |h(q)|^2,$$

where  $\gamma = 1.91$  is the neutron gyromagnetic ratio,  $t$  is the sample thickness,  $\phi_0 = h/2e = 2067 \text{ T nm}^2$  is the flux quantum, and  $q = 2\pi\sqrt{(B/\phi_0)} = 0.1955 \text{ nm}^{-1}$  is the calculated scattering vector for a square FLL and a field of 2 T. This expectation and the measured value of  $q = 0.19 \text{ nm}^{-1}$  agree within 3%. The flux line form factor  $h(q)$  for a square lattice is given by [21]

$$h(q) = \frac{\phi_0}{(2\pi\lambda)^2} e^{-\pi B/B_{c2}},$$

where the exponential factor represents the so-called core correction. Fitting the rocking curve to a Gaussian and using the area under the curve as the integrated reflectivity, together with the upper critical field  $B_{c2}(0) = 5.0 \text{ T}$  [3], we obtain  $\lambda = 247 \pm 10 \text{ nm}$ . This falls inside the range of values  $\lambda_0 = 190\text{--}281 \text{ nm}$  reported in the literature [22–24]. Calculating the coherence length from the upper critical field,  $\xi = \sqrt{(\phi_0/2\pi B_{c2})} = 8.1 \text{ nm}$ , we estimate the Ginzburg-Landau parameter  $\kappa = \lambda/\xi \approx 30$ , making CeCoIn<sub>5</sub> a strongly type-II superconductor.

The second complicating factor is the narrow rocking curve, necessitating a very precise alignment in order to obtain scattering. The fit to the data in Fig. 3 yields a width of  $0.21^\circ$  FWHM comparable to the experimental resolution estimated to be  $0.15^\circ$  FWHM. In principle, the rocking curve width can be used to determine the longitudinal correlation length or straightness of the flux lines, but with the width being close to the experimental resolution this should rather be taken as a lower bound. We find  $\Delta q_L \leq 0.21^\circ (\pi/180^\circ) q = 7 \times 10^{-4} \text{ nm}^{-1}$ , and hence  $\xi_L = 2/\Delta q_L \geq 3 \text{ }\mu\text{m}$ . This is a large value corresponding to  $\sim 100$  flux line spacings, and indicates very weak pinning in this material.

We now return to the discussion of the symmetry and orientation of the FLL. In an ideal isotropic type-II superconductor this will be hexagonal [25,26]. However, if one evaluates the free energy difference between the hexagonal and square symmetry, this is found to be only about 2% [26]. A relatively weak anisotropy is therefore capable of changing this delicate balance, leading to a distorted hexagonal or a square FLL. A number of theoretical studies have addressed the effect of  $d$ -wave pairing on the structure and orientation of the FLL. As the field is increased or temperature decreased, they consistently find that a square FLL is stabilized, oriented with the nearest neighbor direction *along* the direction of the gap

nodes [10–13]. Determining the orientation of the hexagonal FLL is more difficult, since the energy difference between the two configurations aligned  $45^\circ$  apart is very small [11,13]. Ichioka *et al.* [13] conclude that both the square and the hexagonal FLL are oriented with the nearest neighbors along the node direction, with a first-order transition separating the two symmetries. The transition field is predicted to be  $0.15 \times B_{c2}$ , which in the case of CeCoIn<sub>5</sub> corresponds to 0.75 T at  $T = 0 \text{ K}$ . This is in agreement with our results concerning the orientation of the FLL as well as the nature of transition. Furthermore, their prediction of the transition field is in good agreement with our estimate of  $\sim 0.6 \text{ T}$ .

In principle, there is another mechanism that could be responsible for the FLL symmetry transition: a four-fold Fermi surface anisotropy combined with nonlocal electrodynamics due to the finite coherence length. Theoretically, this was studied extensively by Kogan and co-workers who used nonlocal corrections to the London model to calculate the FLL free energy and thereby determine the stable configuration as a function of the flux line density [27–29]. This is the driving force behind the transition between a low-field (distorted) hexagonal and a high-field square FLL seen in the rare-earth nickel borocarbides [18] as well as in V<sub>3</sub>Si [30]. In the case of CeCoIn<sub>5</sub> such an analysis has not yet been carried out. Band structure calculations have been performed on the isostructural compound CeIrIn<sub>5</sub> [5], and (partially) confirmed on CeCoIn<sub>5</sub> by measurements of de Haas–van Alphen oscillations [31]. The calculations show a Fermi surface with at least one sheet having a four-fold anisotropy [5]. However, this warps between two orientations  $45^\circ$  apart, and at present it is not clear what implication this has on the FLL.

In summary, we have studied the symmetry and orientation of the magnetic flux line lattice in the  $d$ -wave superconductor CeCoIn<sub>5</sub> using small-angle neutron scattering. At low fields a hexagonal FLL was found, which undergoes which is most likely a first-order transition to square symmetry around 0.6 T. Though the possibility of a Fermi surface anisotropy combined with nonlocal effects cannot be ruled out as the determining factor, our measurements agree well with the predictions for a pairing-symmetry driven transition. In particular, the nature of the transition, the field at which it occurs, and above all the orientation of the square FLL with the nearest neighbors aligned parallel to the node directions, are all consistent with being driven by the  $d$ -wave symmetry of the order parameter.

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*Note added.*—We have been informed (private communication by T. Sakakibara and K. Machida) that the gap nodes in CeCoIn<sub>5</sub> are probably oriented along the [100] and [010] directions ( $d_{xy}$ ), in contradiction with what was reported in Ref. [8]. While this does not affect the experimental results presented in this Letter it will, if confirmed, change some of the conclusions.

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