

## Field Dependent Coherence Length in the Superclean, High- $\kappa$ Superconductor CeCoIn<sub>5</sub>

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(Received 8 April 2006; published 21 September 2006)

Using small-angle neutron scattering, we have studied the flux-line lattice (FLL) in the superclean, high- $\kappa$  superconductor CeCoIn<sub>5</sub>. The FLL undergoes a first-order symmetry and reorientation transition at  $\sim 0.55$  T at 50 mK. In addition, the FLL form factor in this material is found to be independent of the applied magnetic field, in striking contrast to the exponential decrease usually observed in superconductors. This result is consistent with a strongly field-dependent coherence length, proportional to the vortex separation.

DOI: [10.1103/PhysRevLett.97.127001](https://doi.org/10.1103/PhysRevLett.97.127001)

PACS numbers: 74.70.Tx, 61.12.Ex, 74.25.Op, 74.25.Qt

Since the discovery of heavy-fermion superconductivity in CeCoIn<sub>5</sub>, a plethora of interesting phenomena has been observed in this material. Among these are one of the highest critical temperatures ( $T_c = 2.3$  K) in any heavy-fermion superconductor [1],  $d$ -wave pairing symmetry [2,3], field- and pressure-induced quantum-critical points and non-Fermi liquid behavior [4–8], strong indications of the first realization of a nonuniform Fulde-Ferrell-Larkin-Ovchinnikov state [9–14], and suggestions of multiband or multiorder parameter superconductivity [15,16].

In addition, CeCoIn<sub>5</sub> is also found to represent an extreme case of a clean, high- $\kappa$  superconductor. The elastic electronic mean free path in this material ranges from  $l = 840$  Å at  $T_c$  [17], increasing exponentially as the temperature is decreased, and reaching values of 4 to 5  $\mu\text{m}$  at 400 mK [18,19]. Literature values for the penetration depth varies from 190 nm [18] to 280 nm [20]. Estimates of the orbital critical field based on  $dH_{c2}/dT|_{T_c}$  range from  $H_{\text{orb}}^{\parallel c}(0) = 13.2$  T [17] to 15.0 T [21], yielding an in-plane coherence length of  $\xi_{\text{orb}} = 47\text{--}50$  Å, and hence  $\kappa = 40\text{--}60$  and  $l/\xi \sim 10^3$  or larger at temperatures below a few hundred millidegrees Kelvin.

In many superconductors, detailed information about the nature of the superconducting state has been obtained from small-angle neutron scattering (SANS) studies of the flux-line lattice (FLL). Examples include symmetry transitions driven by nonlocal electrodynamics [22] or a superconducting gap anisotropy [23,24], and subtle changes in the fundamental length scales obtained from the FLL neutron reflectivity [25,26]. In this Letter we report on SANS studies of the FLL in CeCoIn<sub>5</sub>. In striking contrast to the exponential dependence usually observed, the FLL form factor in this material is found to be independent of the applied magnetic field. This result indicates a coherence length which decreases with increasing field, remaining proportional to the vortex separation over the entire measured field range.

SANS experiments were carried out at the D22 and D11 instruments at the Institut Laue-Langevin. The CeCoIn<sub>5</sub> single crystals used in the experiment were grown from excess indium flux and had a  $T_c = 2.3$  K and a  $H_{c2}(0) = 5.0$  T for fields applied parallel to the  $c$  axis [1]. The sample was composed of three individually aligned single crystals with thicknesses 0.13–0.22 mm mounted side by side. The total mass of the sample was 86 mg. The use of thin crystals was necessary, due to the strong neutron absorption by indium. Incident neutrons with a wavelength of  $\lambda_n = 4.5$  Å (D11) and 7 Å (D22) and a wavelength spread of  $\Delta\lambda_n/\lambda_n = 10\%$  were used, and the FLL diffraction pattern was collected by a position sensitive detector. For all measurements, the sample was field cooled to a base temperature of 40–50 mK in a dilution refrigerator insert, placed in a superconducting cryomagnet. Horizontal magnetic fields in the range 0.4–2.0 T were applied parallel to the crystalline  $c$  axis and the incoming neutrons. Background subtraction was performed using measurements following zero-field cooling.

Figure 1 shows FLL diffraction patterns obtained at three different applied fields. Each image is a sum of the scattering from the FLL, as the sample is rotated and tilted in order to satisfy the Bragg condition for the different reflections. At fields below 0.55 T, 12 ( $2 \times 6$ ) reflections are observed as shown in Fig. 1(a), corresponding to two nearly hexagonal domains with Bragg peaks aligned along the  $\langle 110 \rangle$  directions. As the field is increased above  $\sim 0.55$  T the FLL undergoes a first-order transition to a rhombic symmetry as shown in Figs. 1(b) and 1(c). Again two rhombic FLL domain orientations are observed, indicated by the 8 ( $2 \times 4$ ) Bragg peaks. As evident from the decreasing peak splitting in the rhombic phase, the FLL gradually evolves towards a square symmetry as the field is increased. These results are in agreement with our earlier studies [27].

The evolution of the symmetry transition, quantified by the FLL opening angle  $\beta$ , is summarized in Fig. 2.

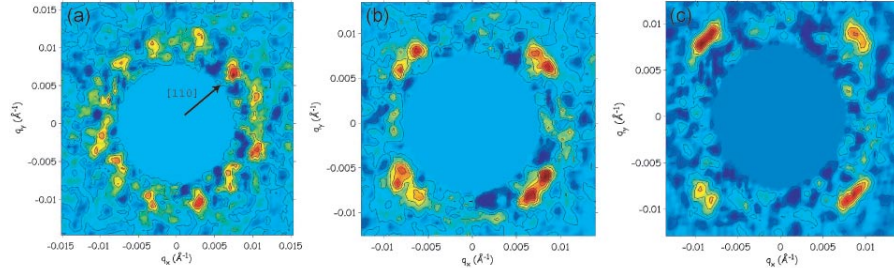


FIG. 1 (color). FLL diffraction patterns for CeCoIn<sub>5</sub> with applied fields 0.5 T (a), 0.55 T (b), and 0.75 T (c), after background subtraction. The data are smoothed and the center of the image is masked off. The crystalline *a* axis is vertical.

Although it was not possible to reliably fit and extract a FLL split angle above 0.85 T, high-resolution measurements up to 1.0 T still showed a weak rhombic distortion. A linear extrapolation of the opening angle to  $\beta = 90^\circ$  yields a transition field  $H_2 = 1.1$  T above which a perfectly square FLL is realized. In addition, scattering vectors belonging to the same domain were used to obtain the internal field by the relation  $|\mathbf{q}_1 \times \mathbf{q}_2| = (2\pi)^2 B / \phi_o$ , where  $\phi_o = 20.7 \times 10^4 \text{ T}\text{\AA}^2$  is the flux quantum. This yielded  $dB/d(\mu_o H) = 0.992 \pm 0.012$ , leading us to set  $B = \mu_o H$  in the remainder of this work.

A square FLL can either be stabilized by a gap anisotropy as observed, e.g., in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> [24], or by nonlocal electrodynamics coupled with a Fermi surface anisotropy as seen in the borocarbide superconductors [22,28]. In the case of CeCoIn<sub>5</sub> the orientation of the gap nodes has been subject to controversy [2,3,16], although recent theoretical work aimed at resolving this issue concluded that the pairing symmetry in this material is  $d_{x^2-y^2}$  [29]. As we have previously reported, such an orientation of the gap nodes is consistent with the high-field square FLL being stabilized by *d*-wave pairing [27]. On the other hand, an extrapolation to  $H = 0$  of the high-field opening angle in Fig. 2 yields  $\beta \approx 60^\circ$ , as expected if the FLL symmetry is determined by nonlocal effects [28].

We now turn to measurements of the FLL form factor, which are the main results of this Letter. Figure 3(a) shows the FLL reflectivity obtained from the integrated intensity

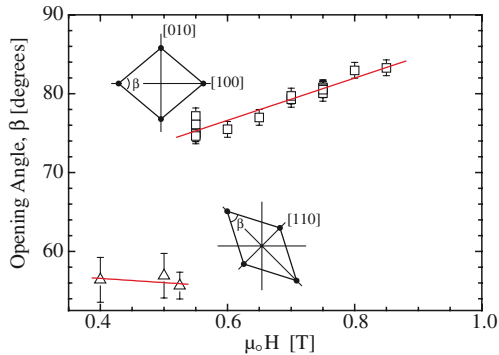


FIG. 2 (color online). Field dependence of the FLL opening angle,  $\beta$ . The insets show the real space orientation of the FLL unit cell in the low- and high-field configurations.

of the Bragg peaks, as the sample is rotated through the diffraction condition. The reflectivity is given by

$$R = \frac{2\pi\gamma^2\lambda_n^2 t}{16\phi_o^2 q} |F(q)|^2, \quad (1)$$

where  $\gamma = 1.91$  is the neutron gyromagnetic ratio,  $\lambda_n$  is the neutron wavelength,  $t$  is the sample thickness, and  $q$  is the scattering vector [30]. The form factor,  $F(q)$ , is the Fourier transform of the magnetic-field profile around a vortex and depends on both the penetration depth and the coherence length. Figure 3(b) shows the FLL form factor determined from the reflectivity. Here we have used the

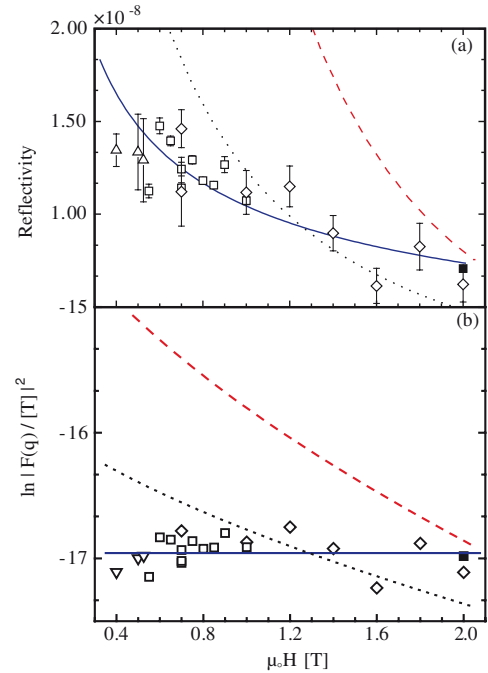


FIG. 3 (color online). Field dependence of the reflectivity (a) and form factor (b) for the (1,0) FLL Bragg reflections. Triangles correspond to a hexagonal FLL and the squares and diamonds to a rhombic FLL. The solid square is from previous work [27]. The dashed and dotted lines are calculated using Eqs. (1) and (2) with the following values for the coherence length and the penetration depth:  $\xi = 81 \text{ \AA}$ ,  $\lambda = 2350 \text{ \AA}$  (dashed line);  $\xi = 50 \text{ \AA}$ ,  $\lambda = 3750 \text{ \AA}$  (dotted line). The solid line corresponds to a constant form factor,  $F = 2.08 \times 10^{-4} \text{ T}$ .

measured opening angle,  $\beta(H)$ , in Fig. 2 to determine the magnitude of the scattering vector, and hence compensate for effects due to the FLL symmetry transition. In the high-field rhombic FLL phase  $q_{sq} = q_o/\sqrt{\sin\beta}$ , where  $q_o = 2\pi\sqrt{B/\phi_o}$ . In the low-field distorted hexagonal phase, the 4 Bragg peaks aligned along the  $\langle 110 \rangle$  directions have  $q_{hex1} = q_o\sqrt{2(1 - \cos\beta)}/\sin\beta$ , while for the remaining 8 peaks  $q_{hex2} = q_o/\sqrt{\sin\beta}$ .

The field-independent form factor observed for CeCoIn<sub>5</sub> is in striking contrast to the exponential decrease observed in other superconductors. However, it is important to note that if the form factor was truly independent of  $q$ , this would imply an unphysical situation with a diverging magnetic field at the vortex center and a coherence length equal to zero. To reconcile this apparent paradox, it is necessary to assume a field dependence of either the superconducting penetration depth, the coherence length, or both.

Several models exist for the magnetic-field profile around the vortices. In the following we have based the analysis on the form factor obtained by Clem [31]:

$$F(q) = B \frac{gK_1(g)}{1 + \lambda^2 q^2}, \quad g = \sqrt{2}\xi(q^2 + \lambda^{-2})^{1/2}, \quad (2)$$

valid for  $\kappa \gg 1$ . It is worth pointing out that while this form factor was obtained for an isotropic superconductor, numerical simulations found no significant difference between superconductors with  $s$ - or  $d$ -wave pairing [32].

The dashed lines in Figs. 3(a) and 3(b) show the reflectivity and form factor calculated from Eqs. (1) and (2) using an average literature value for the penetration depth  $\lambda = 2350 \text{ \AA}$  and  $\xi_{c2} = \sqrt{\phi_o/2\pi H_{c2}} = 81 \text{ \AA}$ . As already discussed, these calculated values are not consistent with the experimental data. A more appropriate value for the coherence length is obtained from the orbital critical field,  $\xi_{orb} = \sqrt{\phi_o/2\pi H_{orb}} \approx 50 \text{ \AA}$  [17,21]. However, while this does correspond to a smaller slope of  $|F(q)|^2$ , it also increases the discrepancy between the measured and calculated values of  $|F(q)|^2$  since, for a given  $\xi$  and  $q$  (or  $H$ ), the magnitude of the form factor is determined by  $\lambda^2$ . Using the penetration depth as a fitting parameter, one obtains the dotted lines in Fig. 3 for  $\lambda = 3750 \text{ \AA}$ . Although this provides a better agreement with the data, such a large value of  $\lambda$  is not consistent with reports in the literature [18,20]. In contrast, a perfect fit to the data is obtained with a constant value of the form factor,  $F = 2.08 \times 10^{-4} \text{ T}$ , and correspondingly a reflectivity  $\propto 1/q$ , as shown by the solid lines in Fig. 3.

It is important to note that no significant disordering of the FLL is observed. Except for systematic differences between the two beam lines, the FLL rocking curve widths remain essentially constant throughout the measured field range. On D22 we find rocking curve widths going from  $0.14^\circ \pm 0.02^\circ$  FWHM at 0.55 T to  $0.16^\circ \pm 0.01^\circ$  FWHM at 1 T, comparable to the experimental resolution ( $\sim 0.08^\circ$

FWHM). Below the reorientation transition a slightly higher value of  $0.20^\circ \pm 0.04^\circ$  FWHM is observed. On D11 FLL rocking curve widths decrease from  $0.26^\circ \pm 0.03^\circ$  FWHM at 0.7 T to  $0.19^\circ \pm 0.01^\circ$  FWHM at 2 T. Such narrow rocking curve widths indicate a very well ordered FLL with a longitudinal correlation length in the micron range, consistent with weak pinning due to the high cleanliness of CeCoIn<sub>5</sub>. Furthermore, FLL disorder above a certain threshold has been shown to lead to a decrease in the scattered intensity, exceeding the usual exponential field dependence of the form factor [33]. We can therefore exclude FLL disordering as the cause for the field independence of the form factor in CeCoIn<sub>5</sub>.

A constant form factor could in principle be due to a decrease of the penetration depth with increasing field, caused by either an increasing superfluid density or a nonuniform spin magnetization contributing to the magnetic flux carried by each vortex. We do not consider the first possibility realistic. Furthermore, while CeCoIn<sub>5</sub> is indeed paramagnetic [21], spin polarization effects will lead to an enhancement of the form factor in contrast to the strong suppression observed in this material [34,35]. We therefore conclude that while paramagnetic effects may contribute, they are not the dominating mechanism behind the field-independent form factor. In the following we therefore restrict our analysis to consider only a field-dependent coherence length.

In Fig. 4, we show the coherence length obtained by varying  $\xi$  in Eq. (2) to achieve the measured form factor at each field. The coherence length is found to follow a  $1/\sqrt{H}$  behavior. While different models for  $F(q)$  will provide slightly different values for the coherence length, the qualitative behavior will remain unchanged. Figure 4 also shows the extracted coherence length as a function of intervortex spacing,  $a$ . Within the experimental error shown by the scatter in the data, the coherence length is found to increase linearly with the intervortex spacing with a slope  $d\xi/da = (0.55 \pm 0.02)/\sqrt{2\pi}$ , while at all times

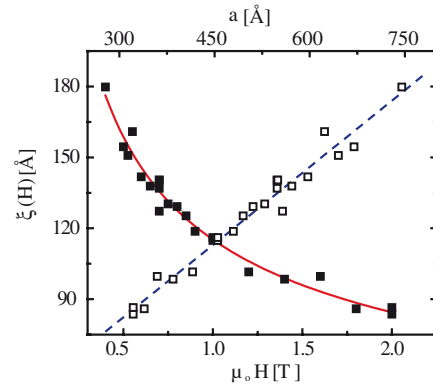


FIG. 4 (color online). Field dependence of the coherence length,  $\xi(H)$ , plotted versus applied field (solid symbols), and vortex separation (open symbols). The solid and dashed lines are fits to the data described in the text.

satisfying  $\xi < a$ . Extrapolating to a vortex separation of 125 Å (corresponding to an orbital critical field  $H_{\text{orb}} = 13.2$  T) yields  $\xi = 43.0 \pm 4.6$  Å, in reasonable agreement with  $\xi_{\text{orb}} = 50$  Å.

A field-dependent vortex core size determined from muon spin rotation experiments have been reported by Sonier for a wide range of low- and high-temperature superconductors with varying Ginzburg-Landau parameters [36]. While these reports are in qualitative agreement with the measurements presented here for CeCoIn<sub>5</sub>, SANS measurements on the compounds in Ref. [36] have not shown an anomalous field dependence of the FLL form factor. Consequently, CeCoIn<sub>5</sub> presents the first case of a field-dependent coherence length observed by SANS. We believe that the field-dependent coherence length is so prominently observed in CeCoIn<sub>5</sub> due to the combination of a large  $\kappa$  and the very high cleanliness of this material.

Additional evidence for the unusual magnetic-field response of CeCoIn<sub>5</sub> has been observed in the quasiparticle mean free path, as extracted from measurements of the Hall angle [37]. Specifically, the mean free path is found to decrease as the applied field increases, being roughly equal to the vortex separation for the range of fields covered by our SANS measurements. This leads us to speculate that a connection exists between  $\xi$  and  $l$  beyond the simple Pippard model.

Recently, a field-dependent coherence length in superclean, high- $\kappa$  superconductors was predicted theoretically [38]. While the underlying model, based on assumptions of weak-coupling  $s$ -wave superconductivity and a simple Fermi surface topology (sphere or cylinder), is strictly speaking not applicable to CeCoIn<sub>5</sub>, it is for the lack of a better alternative still interesting to compare the theoretical results to our measurements. In particular, the model predicts the coherence length to be proportional to  $1/\sqrt{H}$  and correspondingly depending linearly on the vortex separation although with a slope which is roughly half our measured value [38]. This suggests that a field-dependent coherence length is a universal property of very clean superconductors. Such a notion is supported by calculations showing a shrinking vortex core size in a charged Bose liquid with increasing density [39].

In summary, we have presented SANS measurements of the FLL in CeCoIn<sub>5</sub>. The measurements show a highly anomalous field-independent FLL form factor, indicating a strong reduction of the superconducting coherence length with increasing field. More work, experimental as well as theoretical, is needed to further explore this behavior and provide an understanding of the underlying microscopic details.

We are grateful to N. Jenkins for assistance with the SANS measurements and to K. Machida for valuable discussions. M. R. E. acknowledges support by the Alfred P. Sloan Foundation, and B. W. H. acknowledges support by the NCCR Nanoscale Science. Part of this work was carried out at Brookhaven National Laboratory, which is

operated for the U.S. Department of Energy by Brookhaven Science Associates (No. DE-Ac02-98CH10886).

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