Evidence for a Magnetically Driven Superconducting Q Phase of CeCoIn_5

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(Received 30 November 2009; published 25 March 2010)

We have studied the magnetic order inside the superconducting phase of CeCoIn_5 for fields along the $[1\,0\,0]$ crystallographic direction using neutron diffraction. We find a spin-density wave order with an incommensurate modulation $\mathbf{Q} = (q, q, 1/2)$ and $q = 0.45(1)$, which within our experimental uncertainty is indistinguishable from the spin-density wave found for fields applied along $[1\ {-1}\,0]$. The magnetic order is thus modulated along the lines of nodes of the $d_{x^2-y^2}$ superconducting order parameter, suggesting that it is driven by the electron nesting along the superconducting line nodes. We postulate that the onset of magnetic order leads to reconstruction of the superconducting gap function and a magnetically induced pair density wave.

DOI: 10.1103/PhysRevLett.104.127001 PACS numbers: 74.70.Tx, 61.05.F−, 74.20.Mn

The interplay of superconductivity and magnetism is one of the most fascinating areas in condensed matter physics. Many unconventional superconductors feature strong magnetic fluctuations and complex ground states with either competing or coexisting magnetic and superconducting order. Because magnetic fluctuations are believed to be crucial for the formation of superconducting Cooper pairs in these materials [1,2], it is paramount to improve our understanding of how magnetism and superconductivity can interact.

One of the best known model materials for the study of magnetic superconductivity is CeCoIn_5. It is an ultraclean ambient-pressure superconductor which crystallizes in a tetragonal crystal structure [3]. It has a quasi-two-dimensional electronic structure in its normal phase [4,5], a d-wave superconducting gap function [6–8], and a magnetic spin resonance in the superconducting phase [9]. CeCoIn_5 shares all these properties with the widely investigated high-T_c cuprate superconductors. Superconductivity in CeCoIn_5 is Pauli-limited and is thus destroyed by a coupling of external magnetic fields to the spin of the Cooper pairs and not by orbital currents [10]. It may be significant that superconductivity in CeCoIn_5 occurs in the vicinity of a magnetic quantum critical point [11,12], which is associated with a divergence of an already strongly enhanced effective electronic mass [12].

Magnetism and superconductivity in CeCoIn_5 are closely intertwined, leading to a striking field-temperature (HT) phase diagram. The transition from the normal to the superconducting phase is first-order below $T_0 \simeq 1.1$ K for fields in the tetragonal basal plane [13]. There are two distinct superconducting phases: firstly the main phase where the superconducting gap is of $d_{x^2-y^2}$ symmetry [6–8], and secondly the Q phase, which exists only for high fields close to the upper critical field and $T < 0.3$ K [13–15]. The Q phase features static magnetic order in form of a spin-density wave (SDW) which is amplitude modulated by a wave vector that is incommensurate in the basal plane and doubles the unit cell along the tetragonal axis. The SDW does not exist in the normal phase [16,17], showing that magnetic order and superconductivity in CeCoIn_5 are directly coupled.

The origin of the field-induced SDW in CeCoIn_5 is not clear at present and has led to some speculation. Phenomenologically, the coupled nature of superconductivity and magnetic order leads to stringent symmetry constraints for superconductivity in the Q phase: we have suggested earlier that, in addition to the ambient d-wave superconducting component Δ_0, the SDW order $M_{\mathbf{Q}}$ is coupled to a pair density wave (PDW) $\Delta_{-\mathbf{Q}}$ [17] through a coupling term that is linear in $\Delta_0 M_{\mathbf{Q}} \Delta_{-\mathbf{Q}}$. It was shown that this coupling leads to a mixing of singlet and triplet superconductivity [18].

On a microscopic level, it has been suggested [19] that the rapidly modulated SDW is coupled to π-pairing superconductivity of odd parity and arises from the presence of a long-wavelength modulated PDW associated with the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) [20,21] mechanism. The presence of staggered π superconductivity in the presence of SDW order has also been proposed as a general feature for magnetic superconductors [22]. Yanase and Sigrist suggested that the SDW arises from a FFLO state due to Andreev bound states localized in the zeros of a Larkin-Ovchinnikov–type order parameter [21,23,24]. This latter scenario leads to additional Bragg peaks along the wave vector parallel to the field direction.

In order to understand the origin of magnetic order in the Q phase, we used high-field neutron diffraction to probe the magnetic order in the Q phase for magnetic fields along the crystallographic $[1\,0\,0]$ direction ($\mathbf{H} \parallel [1\,0\,0]$). This is a different field direction than in the previous experiment where $\mathbf{H} \parallel [1\ {-1}\,0]$. Originally, and for a long time since, the modulation wave vector $\mathbf{Q}_{\mathrm{FFLO}}$ in the FFLO state was often taken to be parallel to the applied magnetic field. Recently, several theoretical investigations explored the question of the direction of $\mathbf{Q}_{\mathrm{FFLO}}$ for d-wave superconductors [25]. In particular, for the case of the two-dimensional d-wave superconductor, without taking into account the orbital (vortex) effects, $\mathbf{Q}_{\mathrm{FFLO}}$ can switch between the nodal and antinodal directions of the superconducting order parameter. By measuring the properties of the magnetic order for different field directions in the basal plane, it is possible to distinguish if the field-induced SDW in CeCoIn$_5$ is pinned to the direction of the field or if it is pinned to the direction of the $d_{x^2-y^2}$ line nodes. Further, this experiment allows one to test some of the predictions of the FFLO-based theories.

The neutron experiments were performed using the two-axis diffractometer D23 at the ILL in Grenoble, France. The sample was a single crystal of about 50 mg, oriented with its $(0, k, l)$ reciprocal plane in the horizontal scattering plane. The lifting-arm detector of D23 permits neutron wave vector transfers in the field direction, giving thus access to out-of-plane wave vectors. A cryomagnet allowed the application of vertical magnetic fields $\mathbf{H} \parallel [1\,0\,0]$ up to $\mu_0 H = 12$ T. The sample was cooled down in a dilution insert. Checks of the magnetic field history inside the Q phase showed no observable effects. Measurements were performed using an incident neutron wavelength of $\lambda = 1.28$ Å obtained via the (002) Bragg reflection from a flat Cu monochromator.

Figure 1(a) shows the neutron diffraction intensity for wave vectors $(h, -h, 0.5)$, where h describes the wave vector transfer along the reciprocal $(1\ -1\,0)$ direction in reciprocal lattice units (r.l.u.). There is a well-defined diffraction peak centered at $h \sim 0.445$ for $\mu_0 H = 11.4$ T and $T = 60$ mK which is not present at $\mu_0 H = 12$ T, indicating that this incommensurate peak is of magnetic origin. Figure 1(b) shows the corresponding peak centered at $(0.54, -0.54, 0.5)$ as expected for magnetic order which is described by an ordering wave vector of $\mathbf{Q} = (q, -q, 0.5)$ with $q \sim 0.45(1)$.

We also looked for magnetic Bragg peaks at wave vector transfers $(0, -q, 0.5)$ and $(0.5 - q, -(0.5 + q), 0)$, but none were detected. The observed Bragg peaks are described by $\tau \pm \mathbf{Q}$, where τ is a reciprocal wave vector of the nuclear lattice. Bragg peaks with a large wave vector transfer are increasingly weaker and no incommensurate Bragg peaks were observed for $|(h, -h, l)| > 3.5\ \text{Å}^{-1}$. This reflects the wave vector dependence of the magnetic form factor of Ce^{3+}, which decreases with increasing wave vector, and thus provides further support for the magnetic origin of the incommensurate peaks. A refinement of 6 magnetic Bragg peak intensities shows that the magnetic structure is a SDW with a magnetic moment of $0.16(5)\mu_B$ along the c axis which is modulated with $\mathbf{Q} = (q, -q, 0.5)$ and $q \sim 0.45(1)$.

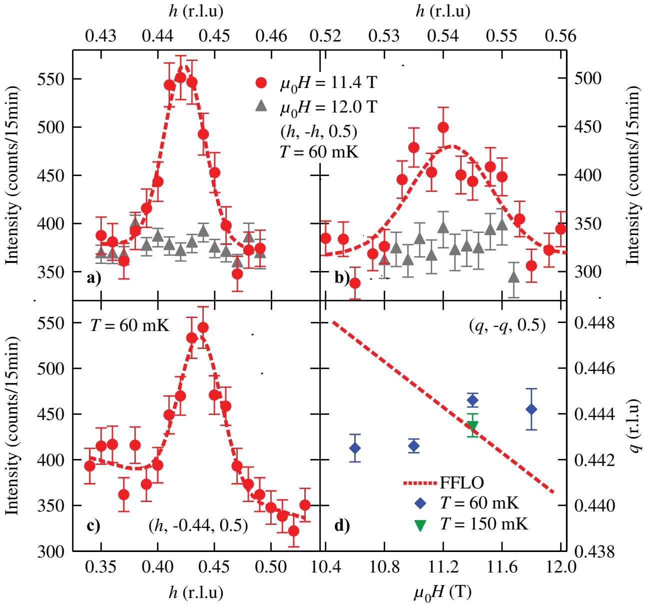

FIG. 1 (color online). (a) Neutron diffraction intensity for wave vectors $(h, -h, 0.5)$, centered around $h \sim 0.445$, for two magnetic fields $\mathbf{H} \parallel [1\,0\,0]$ at $T = 60$ mK, showing the magnetic diffraction peak of the field-induced magnetic order in the superconducting phase of CeCoIn$_5$. (b) Intensity of the magnetic Bragg peak for wave vectors $(h, -h, 0.5)$, centered around $h \sim 0.541$, for $\mu_0 H = 11.4$ T and $\mu_0 H = 12$ T at $T = 60$ mK. (c) Tilt scan of the magnetic Bragg peak along $(h, -0.44, 0.5)$, showing that the magnetic order is long range along the flux line direction. (d) Magnetic field dependence of the incommensuration $h = q$, for which the peak was observed. The solid line represents the expected field dependence of q predicted by Miyake [19] with the Fermi velocity adjusted to $v_F = 2.6 \times 10^3$ m/s for the best agreement with the data.

The SDW for $\mathbf{H} \parallel [1\,0\,0]$ has thus identical symmetry to that detected for $\mathbf{H} \parallel [1\ {-1}\,0]$, and even the incommensuration q is very similar in both cases [17]. This demonstrates that the field-induced SDW in CeCoIn$_5$ depends only weakly, if at all, on the magnetic field direction in the basal plane and on the direction of the vortex flux lines. Further, the ordered magnetic moment is similar to the one previously observed for $\mathbf{H} \parallel [1\ {-1}\,0]$ [17].

The magnetic peak width measured by rotating the sample around the field direction is resolution limited and we estimate that the correlation length of the magnetic order in the crystallographic $(0, k, l)$ plane is larger than 3000 Å. This is much larger than the diameter of vortex cores, which is of the order of the coherence length $\xi_0 \sim 100$ Å, showing that the SDW is not limited to the vortex cores. The lifting-arm detector of D23 also allowed us to determine a lower limit for the correlation length of the SDW along the field direction. Figure 1(c) shows the wave vector dependence, $(h, -0.44, 0.5)$, of the Bragg intensity along the magnetic field direction, showing a well-defined magnetic Bragg peak broadened by the much looser vertical resolution. We find that the lower limit for the correlation length along the field direction is 250 Å, indicating that the SDW is long-range ordered in three dimensions.

Figure 1(d) shows the field dependence of the incommensuration q, hinting at a possible small variation of q with the strength of the applied field, which, however, is only a fraction of the width of the diffraction peak. We can compare our data to the field dependence of q predicted by the model by Miyake [19] where the magnetic order is driven by a FFLO-type PDW. Our data are clearly incompatible with the field dependence predicted by this model. Further, Fig. 1(c) clearly shows the absence of additional magnetic Bragg peaks apart from the peak at $(q, -q, 0.5)$, which appears to be incompatible with FFLO driven Andreev bound states as the origin for the SDW [24]. These two independent observations indicate that the magnetic order is not related to an FFLO-type PDW.

Figure 2 shows the peak intensity as a function of field for $T = 60$ mK and $T = 250$ mK, respectively, and as a function of temperature at $\mu_0 H = 11.6$ T. These measurements were performed by scanning the temperature or field while keeping the diffractometer at the magnetic Bragg peak position. Such an approach is justified because any movement of the peak position $h = q$ with field or temperature is much smaller than the width of the diffraction peak. The field dependence shows a gradual onset at low field with a fast decrease at $\mathbf{H}_{c2}$, similar to what was observed for fields parallel to $[1\ -1\,0]$. As a function of temperature, the intensity also drops suddenly, because at $\mu_0 H = 11.6$ T a temperature scan crosses the first-order HT phase boundary into the normal phase. These measurements thus show that the SDW disappears simultaneously with superconductivity.

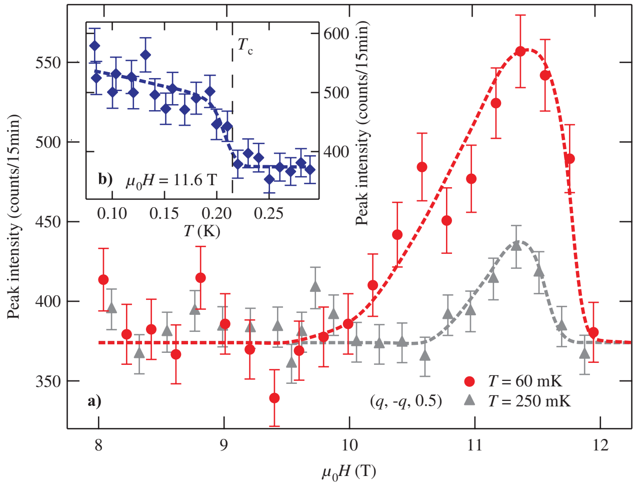

FIG. 2 (color online). (a) Field dependence for $\mathbf{H} \parallel [1\,0\,0]$ of the peak intensity of the magnetic Bragg peak at wave vector $(q, -q, 0.5)$ at $T = 60$ mK and $T = 250$ mK, respectively. (b) Temperature dependence of the magnetic Bragg peak intensity at wave vector $(q, -q, 0.5)$ at $\mu_0 H = 11.6\,\mathrm{T} \parallel [100]$, revealing a sudden drop at T_c. The dashed lines are guides to the eye.

Figure 3 shows the HT phase diagram for $\mathbf{H} \parallel [1\,0\,0]$. The phase boundary at H_{c2} was determined from the midpoint field of the first-order collapse of the magnetic diffraction intensity, while the low-field onset of the intensity was determined by inspection of the field dependence of the peak intensity. The neutron data are in good agreement with the phase boundaries of the Q phase determined from macroscopic measurements [12]. In addition, we were able to extend the phase line to lower temperatures. The comparison shows that the magnetic order only exists in the Q phase and in the superconducting phase of CeCoIn$_5$. The HT phase diagram for $\mathbf{H} \parallel [1\,0\,0]$ confirms that superconductivity is an essential component for the existence of magnetic order in CeCoIn$_5$.

Our experiment clearly shows that the SDW wave vector does not depend on the magnetic field direction in the basal plane. This should severely restrict the symmetry of the PDW that is allowed to couple to the SDW [18]. From a microscopic point of view, it may be significant that the ordering wave vector $\mathbf{Q}$ is along the direction in reciprocal space where line nodes are present for $d_{x^2-y^2}$ superconductivity. Our results are thus consistent with $d_{x^2-y^2}$-wave superconductivity in CeCoIn$_5$, where a SDW arises through electron nesting from low-energy states along the $[1\,1\,l]$ reciprocal direction.

Our results are not explained by the available theories where the SDW is driven by a FFLO-type pair density wave. The observed difference between experiment and

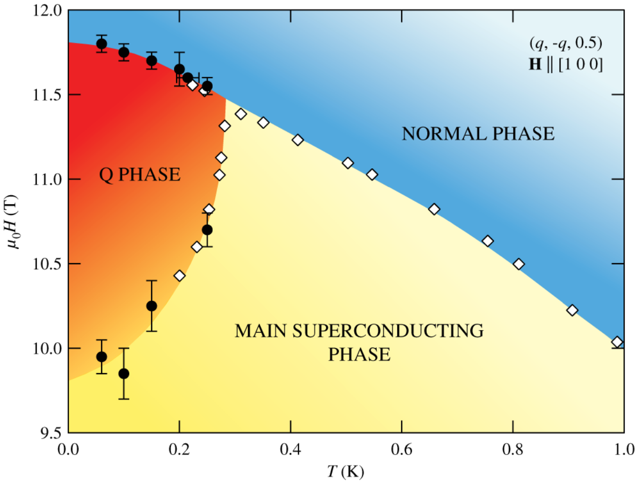

FIG. 3 (color online). HT phase diagram of CeCoIn$_5$ for $\mathbf{H} \parallel [1\,0\,0]$, showing the phase boundaries of the Q phase as determined from the present experiment as full circles (compared to the phase boundaries determined with specific heat measurements [12]). Magnetic order was only observed in the superconducting part of the phase diagram. The upper phase boundary H_{c2} for $T < T_0$ indicates a first-order transition where magnetic order suddenly collapses simultaneously with superconductivity.

theory indicates that either the SDW in CeCoIn_5 is not FFLO driven or that some of the assumptions in the theories are not correct. For example, Yanase and Sigrist [24] assumed that $\mathbf{Q}_{\mathrm{FFLO}} \parallel \mathbf{H}$, and predictions for different modulation vectors would be very useful.

We suggest that the SDW may have its origin in the local magnetism inside the vortex cores observed using small-angle neutron scattering and NMR [16,26]. This enhanced vortex core magnetism may be due to the confined nature of the normal vortex region, possibly leading to a different electronic structure and to higher density of low-energy excitations that can mediate interactions. Long-range SDW order would occur spontaneously at high enough field where the vortex cores are separated by only 130–150 Å and thus almost overlap, leading to a strong tendency towards the formation of a SDW at low temperatures. No correlated magnetism would be observed in the normal phase of the HT phase diagram where superconducting vortices are absent.

In the proposed scenario, the Q phase is magnetically driven, through a gain in magnetic energy when the SDW is formed. The ordering of a SDW through electronic nesting at a particular wave vector would drive a reconstruction of the electronic structure and lead to a short-wavelength PDW $\Delta_{-\mathbf{Q}}$ with opposite momentum to the SDW. $\Delta_{-\mathbf{Q}}$ appears as the result of SDW magnetic order, and thus corresponds to a magnetically driven spin pairing channel with finite momentum Cooper pairs. This mechanism should lead to a much less field-dependent incommensuration q than for a FFLO driven scenario, which is consistent with the experiment. The interaction between the localized spins in the Q phase may be similar to the proposed interaction that leads to the spin resonance at zero field, where the spin resonance appears as a superconducting feedback on magnetic excitations [27].

Open remains the question whether the field-induced magnetism in CeCoIn_5 is a special case or whether it is present in a wide range of superconductors. Evidence for field-induced magnetism has also been observed in high-T_c superconductors with evidence for a complex interaction between SDW and superconductivity [28].

Finally, we point out that for $\mathbf{H} \parallel [1\,0\,0]$, there should be two different equally populated domains of coupled SDW-PDW ordering, described by $(q, q, 0.5)$ and $(q, -q, 0.5)$, respectively, which lie at the same angle to the magnetic field. For $\mathbf{H} \parallel [1\ {-1}\,0]$, the field is at a closer angle to the $(q, -q, 0.5)$ wave vector, and one of the two domains may be favored. Through switching of the SDW domains using magnetic fields, it may thus be possible to switch the PDW domains magnetically. For $\mathbf{H} \parallel [0\,0\,1]$, it can be expected that the SDW is unstable due to a spin-flop transition, so magnetic order may be a spiral and the PDW may be chiral.

In summary, our study shows that the symmetry of the field-induced magnetic order in CeCoIn_5 for $\mathbf{H} \parallel [1\,0\,0]$ is identical to that observed for $\mathbf{H} \parallel [1\ {-1}\,0]$, thus strongly suggesting that the magnetic structure does not depend on the magnetic field direction in the basal plane. Our results are not explained by two theories where the Q phase is FFLO driven. Instead, we propose that the SDW arises from electron nesting along the line nodes of the $d_{x^2-y^2}$ gap function, inducing a novel spin pairing channel with finite momentum Cooper pairs.

We thank M. Sigrist, Y. Yanase, and D. Agterberg for discussions and acknowledge the technical services of ILL. This work was supported by the Swiss NSF (Contract No. 200021-122054) and by the Swiss NCCR program MaNEP. Work at Los Alamos was performed under the auspices of the U.S. Department of Energy and supported in part by the Los Alamos Laboratory Directed Research and Development program. A. D. B. received support from the Natural Sciences and Engineering Research Council of Canada (Canada), Fonds Québécois de la Recherche sur la Nature et les Technologies (Québec), and the Canada Research Chair Foundation.

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