Field Evolution of Coexisting Superconducting and Magnetic Orders in CeCoIn$_5$

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We present nuclear magnetic resonance (NMR) measurements on the three distinct In sites of CeCoIn$_5$ with a magnetic field applied in the [100] direction. We identify the microscopic nature of the long range magnetic order (LRO) stabilized at low temperatures in fields above 10.2 T while still in the superconducting (SC) state. We infer that the ordered moment is oriented along the $\hat{c}$ axis and map its field evolution. The study of the field dependence of the NMR shift for the different In sites indicates that the LRO likely coexists with a modulated SC phase, possibly that predicted by Fulde, Ferrell, Larkin, and Ovchinnikov. Furthermore, we discern a field region dominated by strong spin fluctuations where static LRO is absent and propose a revised phase diagram.

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The detrimental effect of an applied magnetic field on a superconductor has nourished the intuitive presumption of a *de facto* competition between magnetic and superconducting (SC) orders. However, it is now well established, both theoretically and experimentally, that not only can these orders coexist, but in some cases, they may even be essential for each other's stability [1]. Manifestations of coexistence span a rather wide range of materials including several cuprates, ferropnictides, and heavy fermion systems of which CeCoIn$_5$ is one of the most intriguing examples [2]. In the SC state of this compound application of a magnetic field (H_0) induces a long-range magnetic order (LRO), restricted to a narrow low-temperature (T) region of the phase diagram below the upper critical field H_{c2} [3,4]. What is more, this particular region of the phase diagram was initially identified as the first realization of the long-sought Fulde, Ferrell, Larkin, and Ovchinnikov (FFLO) state, a superconducting state with a nonzero pair momentum and a spatially modulated order parameter [5,6]. However, important questions regarding the true nature of the low-T high-H_0 SC phase, the details of the magnetic order and its field dependence, and the potential driving mechanisms of their coexistence remain unanswered [7]. Thus, CeCoIn$_5$ provides a strikingly rich ground to study the complex interplay between exotic SC and magnetism. Experimentally, nuclear magnetic resonance (NMR), as a microscopic probe sensitive to both magnetic and SC degrees of freedom, provides a powerful tool for the investigation of these puzzles.

In this Letter, detailed low-temperature NMR measurements on the three distinct In sites in CeCoIn$_5$ for $\mathbf{H}_0 \parallel [100]$ are presented. We establish that at $T \approx 70$ mK a phase with static magnetic LRO is stabilized for fields above ≈ 10.2 T in the SC state. We deduce that the LRO is an incommensurate spin density wave (IC SDW) with moments oriented along the $\hat{c}$ axis, independent of the in-plane $\mathbf{H}_0$ orientation. Further, the detailed field evolution of the moment is mapped. The study of the field dependence of the NMR shift implies that this IC SDW coexists with a novel SC state, characterized by an enhanced spin susceptibility [8]. Finally, we identify a new region in the H_0-T phase diagram, lying in between the low field SC (LFSC) and the IC-SDW states. This could be a FFLO phase without magnetic LRO.

High quality single crystals of CeCoIn$_5$, grown by a flux method, were placed in NMR radio frequency (rf) coils inside the mixing chamber of a dilution refrigerator so that $\mathbf{H}_0 \parallel [100]$. The rf coil was also used to determine the precise value of H_0 by performing ^{63}Cu NMR on its copper nuclei. The spectra were obtained, at each given value of H_0, from the sum of spin-echo Fourier transforms recorded at constant frequency intervals. Extremely weak rf excitation power [9] was used to discern the NMR signal from In sites sensitive to magnetism.

For $\mathbf{H}_0 \parallel [100]$, there are three inequivalent In sites. The axially symmetric In(1) is located in the center of the tetragonal Ce planes, while In(2_{ac}) and In(2_{bc}) sites correspond to In atoms located on the lateral faces (parallel and perpendicular to the applied field, respectively) of the unit cell [10]. In Fig. 1 the H_0 evolution of the In(2_{ac}) and In(2_{bc}) spectra at $T \sim 70$ mK is plotted. Lowering the field below $\sim$11.7 T establishes magnetic LRO. The LRO is evident in the fact that the In(2_{ac}) line broadens into a spectrum with two extrema or peaks with finite signal weight in between them. Such spectra are characteristic of IC LRO along one spatial dimension [11]. Furthermore, at $H_0 = 11.67$ T, both the broad In(2_{ac}) and the sharp normal state spectra are observed. This reflects the coexistence of normal and IC phases in the vicinity of the phase transition confirming its first order character. From independent NMR measurements [12] and the tuning resonance of the tank circuit, we establish that this transition to the IC-LRO state coincides precisely with the transition from the normal to the SC state. For $9.2 \lesssim H_0 \lesssim 10.2$ T,

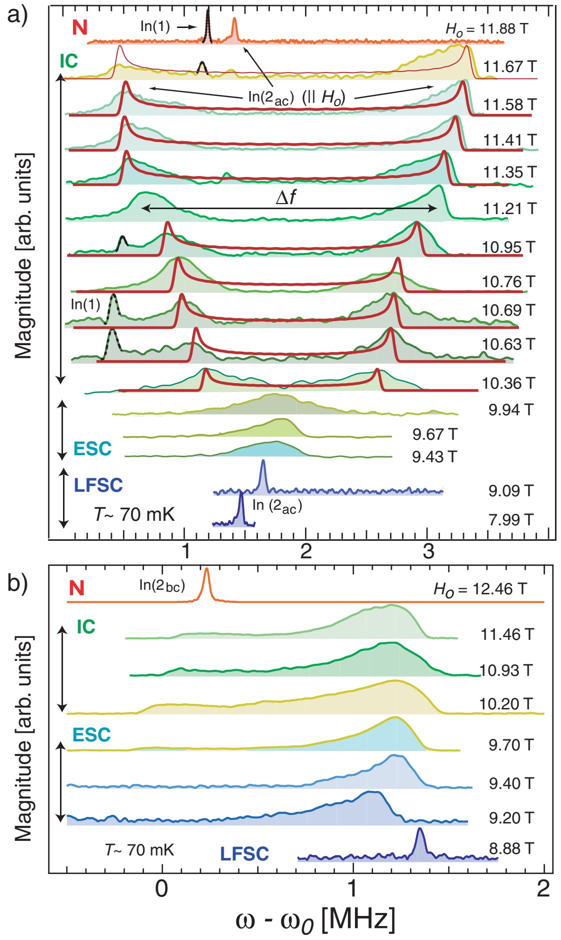

FIG. 1 (color online). NMR spectra of (a) In(1), In(2_{ac}) and (b) In(2_{bc}) at $T = 70$ mK for various $\mathbf{H}_0 \parallel \hat{a}$. The frequency scale is defined by subtracting ω_0, the zero NMR shift frequency. N denotes the normal phase, IC the LRO phase, ESC the state with strong fluctuations, and LFSC the Abrikosov SC state. Solid lines in (a) are simulated spectra for the IC-SDW order described in the text.

the spectra of all In sites consist of a single peak; i.e., no signature of the IC state is observed. However, these spectra remain significantly broader than the ones for $H_0 \lesssim 9.2$ T, where the linewidth of all sites can be adequately described by the spatial distribution of magnetic fields resulting from the vortex lattice [12].

We now proceed to the analysis of the nature of the IC LRO phase to elucidate the field evolution of its magnetic moment and its ordering wave vector ($\mathbf{Q}$). The NMR spectrum reflects $\mathcal{P}(H_{\rm int}^{\parallel})$, the probability distribution of the internal magnetic field at the nuclear site projected along the $\mathbf{H}_0$ direction, denoted by $\hat{z}$. In the LRO phase, the component of the internal hyperfine field parallel to $\mathbf{H}_0$, at an In site, is given by $H_{\rm int}^{\parallel} = \hat{z} \cdot \sum_{\langle i \rangle} \mathbb{A}_i \cdot \boldsymbol{\mu}_i$, where $\mathbb{A}_i$ is the symmetric 3×3 hyperfine coupling tensor with the ith nearest-neighbor Ce atom and $\boldsymbol{\mu}_i$ is its magnetic moment. The exact form of the hyperfine tensor is derived in Ref. [13]. Our data and the fact that certain off-diagonal elements of $\mathbb{A}$ [A_{ab}, A_{ac}, and A_{bc} for In(1), In(2_{ac}), and In(2_{bc}), respectively] are nonzero allows us to place stringent constraints on the possible nature of the magnetic LRO. Specifically, the form of $\mathbf{Q}$ and $\boldsymbol{\mu}_i$ should be such that the In(2_{ac}) spectrum [Fig. 1(a)] broadens into a double peak structure, while no such broadening of the In(1) [12] and In(2_{bc}) [Fig. 1(b)] lines is induced. In principle, such spectra can be effectively described by an appropriate sinusoidal variation of $H_{\rm int}^{\parallel}$, that is, of the moment itself. For an IC-SDW state, this spatial variation of the moment can be written as $\boldsymbol{\mu}_i(\mathbf{r}) = \boldsymbol{\mu}_0 \cos(\mathbf{Q} \cdot \mathbf{r} + \phi_0)$ where $\mathbf{Q}$ is the IC wave vector, $\mathbf{r}$ defines the lattice coordinates of the ith Ce moment, and ϕ_0 is an arbitrary phase. The separation between the two extrema of the In(2_{ac}) spectrum is $\Delta f = \gamma(H_{\rm int}^{\parallel\,\max} - H_{\rm int}^{\parallel\,\min})$, where γ is the nuclear gyromagnetic ratio. Thus, Δf depends on a product of hyperfine tensor components, the moment amplitude μ_0, and some trigonometric function dictated by $\mathbf{Q}$.

A priori, the magnetic structure with the ordered moments $\boldsymbol{\mu}_i \parallel \mathbf{H}_0 \parallel \hat{a}$ and $\mathbf{Q} = \mathbf{Q}_{\rm AF} + \boldsymbol{\delta}$, where $\mathbf{Q}_{\rm AF} = (0.5, 0.5, 0.5)$ and incommensuration $\boldsymbol{\delta} = (\delta, 0, 0)$, can satisfy the requirements imposed by our data, as was suggested in [3]. In this case, Δf for the In(2_{ac}) peaks is equal to $4\gamma A_{aa}\mu_0 \sin(\delta\pi)$, with $A_{aa} \approx A^{LT}_{{\rm In}(2_{ac})}/2 \approx -0.6$ T$/\mu_B$ [10,13]. However, reproducing the observed spectra requires either a moment μ_0 that is 3 times larger than that found by neutrons for $\mathbf{H}_0 \parallel [1\bar{1}0]$ [4] or an excessive value of $\delta \simeq 0.21$. It is thus unlikely that such a structure with $\boldsymbol{\mu}_i \| \hat{a}$ is stabilized in the IC phase.

A better candidate for describing our data is an IC SDW with ordered moments perpendicular to the plane, i.e., $\boldsymbol{\mu}_i \parallel \hat{c} \perp \mathbf{H}_0$ as in [4], and $\boldsymbol{\delta}$ in the plane. We cannot uniquely determine the direction of $\boldsymbol{\delta}$, since the relevant $\mathcal{P}(H_{\rm int}^{\parallel})$ is nearly the same for $\boldsymbol{\delta} \parallel \hat{a}$, $\boldsymbol{\delta} \parallel \hat{b}$, or $\boldsymbol{\delta} \parallel [110]$. However, the case of $\boldsymbol{\delta} \parallel \hat{b} \perp \mathbf{H}_0$ appears to be the most plausible one, since it would agree with both the experimental findings for $\mathbf{H}_0 \parallel [1\bar{1}0]$ [4] and the theoretical prediction of a $\boldsymbol{\delta} \perp \mathbf{H}_0$ for coexisting IC-SDW and FFLO order parameters [14]. In this case, Δf is proportional to $4\gamma A_{ac}\mu_0$, where A_{ac} is the off-diagonal hyperfine tensor component, whose value is not known from other independent measurements. Assuming that at 11 T $\mu_0 = 0.15\mu_B$ and $|\boldsymbol{\delta}| = 0.085$ (the same magnitude as in [4]), we find that a reasonable value of $A_{ac} \approx 0.38$ T$/\mu_B$ is required to fully account for our data. The calculated distributions $\mathcal{P}(H_{\rm int}^{\parallel})$, convolved with the underlying vortex lattice line shape [12], are depicted as the solid lines in Fig. 1(a). Based on this result, the magnetic LRO for $\mathbf{H}_0 \parallel \hat{a}$ is most likely an IC SDW with the ordered moment perpendicular to the plane. Hence, in conjunction with the neutron findings [4], we infer that the direction of the ordered moments is independent of the in-plane orientation of H_0.

Next, we extract the field evolution of the magnitude of the magnetic moment μ_0 from Δf of the In(2_{ac}) depicted in Fig. 1(a). To do so, we fit the data to $\mathcal{P}(H_{\rm int}^{\parallel})$ with μ_0 as a fitting parameter and assuming that $|\boldsymbol{\delta}| = 0.085$ is H_0

independent, as experimentally found [4] and theoretically predicted [14,15], and that the hyperfine component $A_{ac} \approx 0.38\ \mathrm{T}/\mu_B$. The deduced field evolution of μ_0 is shown in Fig. 2. The moment is zero outside the IC phase below $H_0 \approx 10.2$ T. It increases by a factor of ~2.5 as the field changes from ≈ 10.3 to 11.67 T, where it reaches its maximum value of $\approx 0.2\mu_B$.

In order to better understand the perplexing relationship between the coexisting SC and IC-SDW orders, we next discuss the field dependence of the NMR shift of In(2_{ac}) and compare it to that of In(1) [12]. This allows us to distinguish between the contribution from the low energy local density of states (LDOS), characteristic of the SC state, which affects both sites and that from the localized magnetic moments, affecting predominantly In(2_{ac}). Our previous analysis of the In(1) shift has revealed that a novel SC state, possibly a FFLO state, is stabilized for $H_0 = H^* \gtrsim 10$ T via a second order phase transition [12]. We were, however, unable to exclude the existence of a magnetic order in this novel SC phase. In Fig. 3, the field dependence of the shift, determined by diagonalizing the full nuclear spin Hamiltonian, is plotted. In the LFSC phase, the DOS increases with H_0 due to excess Zeeman and Doppler-shifted nodal quasiparticles [12], leading to an increasing spin susceptibility (χ_s). The In(1) and In(2_{ac}) shifts ($K \propto A\chi_s$) exhibit reversed H_0 evolution. That is, they scale with their respective hyperfine coupling constants, which are nearly equal but have opposite signs [10], with that for In(2_{ac}) being negative. The same is also true for the relative shift change between the normal and LFSC state for both sites.

For $H^* \le H_0 \le H_{c2}$, where the IC-SDW LRO is established, we plot the In(2_{ac}) shift of its first moment and of the two extrema of its broadened line shape. The difference of the shift of the two extrema strongly varies with the field reflecting the increase of Δf with increasing H_0. Further, the In(2_{ac}) shift associated with its first moment has essentially the same field dependence as that reported for In(1), once the difference in their hyperfine couplings is taken into account. Thus, independently of the local magnetism, both In sites sense an additional common local field, due to a spin polarization along $\mathbf{H}_0$.

There are two likely origins for this local field for $H_0 \ge H^*$. One is a canting of the transverse staggered magnetization of the localized moments along $\mathbf{H}_0$. To test this hypothesis one needs a quantitative description of canting in an IC phase, which is missing. Thus, we calculate the effect of several crude models of canting on the NMR observables. We find that our data are effectively reproduced by considering an appropriate fixed value of $\mu_{\hat{z}}$ [$\sim\mu_0 \sin(5°)$] on all lattice sites, regardless of $|\boldsymbol{\mu}_i(\mathbf{r})|$. Nevertheless, it is not clear how this type of canted moment could be induced in an IC-SDW phase.

Alternatively, the additional common field could originate from the enhanced LDOS of the spin polarized quasiparticles in the nodal planes of the FFLO state [8]. In that case, since in a FFLO state LDOS varies on a length scale that significantly exceeds the spacing between two In sites, its contribution to the shift of the two distinct In sites should be equivalent as observed. This spatial modulation of the LDOS can also contribute to asymmetric spectral broadening, which for different In sites should scale as their respective hyperfine couplings. For In(1), this LDOS modulation gives rise to a tail on the high frequency side of the line [9,12], while for In(2_{ac}) the tail should be on the low frequency side, due to the sign difference of their hyperfine coupling. This is indeed observed for In(2_{ac}) spectra, where only the low frequency side is essentially broadened beyond IC-SDW line shape as shown in Fig. 1(a).

Next, we consider the intriguing low field limit of the IC-LRO phase. The low-T shift data provide evidence of a continuous phase transition at ≈10.2 T, as shown in Fig. 3. However, significant broadening of In(2_{ac}) and In(2_{bc}) spectra, as compared to the ones in the LFSC phase, onsets for $H_0 \gtrsim 9.2$ T, as evident in Fig. 1. Strikingly, 9.2 T is precisely the field at which the amplitude of the IC SDW

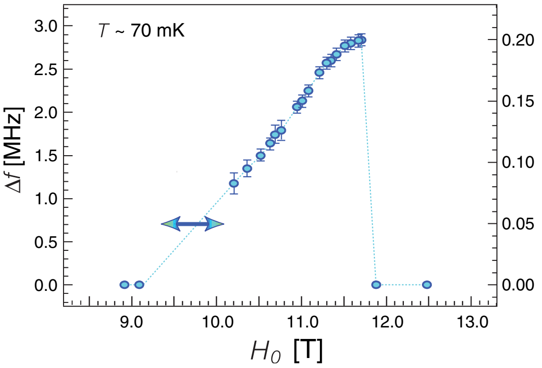

FIG. 2 (color online). Magnetic moment as a function of H_0 deduced from Δf at $T \approx 70$ mK. The double arrow denotes the region of H_0 within which LRO is absent but spectra remain much broader than in the LFSC state.

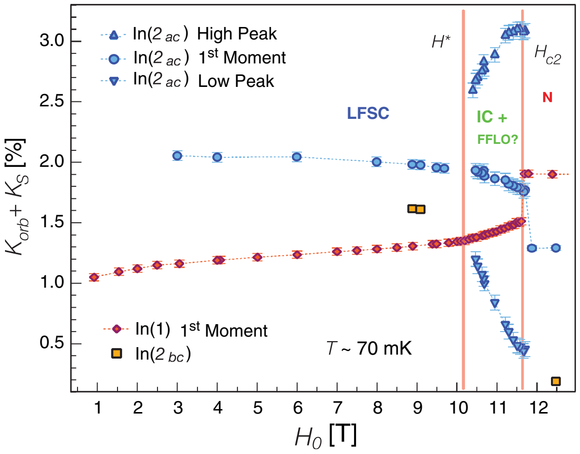

FIG. 3 (color online). NMR shift (spin part, K_s, and constant orbital part, K_{orb} [10]) of In(1), In(2_{ac}), and In(2_{bc}) at $T \approx 70$ mK as a function of $\mathbf{H}_0 \parallel \hat{a}$. The solid lines indicate the values of H^* and H_{c2}.

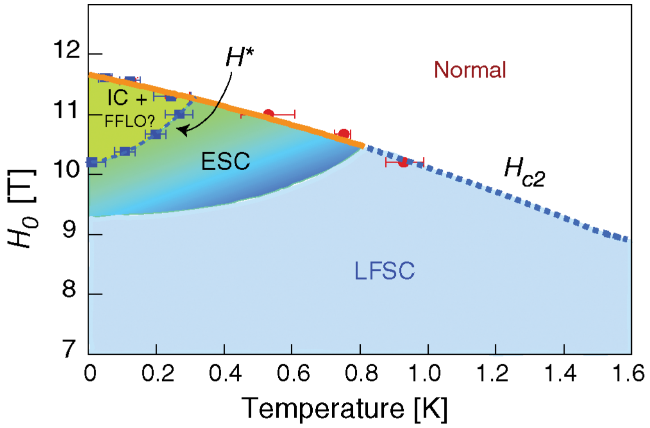

FIG. 4 (color online). A sketch of an alternative phase diagram of CeCoIn$_5$ (data points adopted from Ref. [9]). The solid line and the dashed line indicate first and second order phase transitions, respectively.

should vanish if one extrapolates from its field dependence in Fig. 2. For $9.2 \lesssim H_0 \lesssim 10.2$ T, the In(2_{ac}) signal is weak due to the rapid loss of spin coherence, caused by strong field fluctuations. Thus, although the static LRO is absent, strong antiferromagnetic (AFM) fluctuations are still present. These fluctuations can be responsible for the apparent collapse, evident in the disappearance of the double-horned broad In(2_{ac}) line, of the LRO in this field region. Additionally, the observed extra static line broadening, as compared to the LFSC state, in the presence of these fluctuations, can only emerge from highly enhanced χ_s around defects [16], such as nodal planes in a FFLO state. Thus, this field range possibly corresponds to a FFLO state in the presence of strong AFM fluctuations, as predicted in [14]. This is all more likely since the width of the low-T spectra of all three In sites in the same field range is comparable to that observed in the higher T ($T > T^*$), high-H_0 ($H_0 > H^*$) SC phase outside the limits of the LRO state [3,9].

The second order phase transition previously identified by the In(1) shift can be, then, the transition to the IC LRO state. As recently shown, the IC magnetism can arise in the FFLO state in a d-wave SC as a consequence of the formation of Andreev bound states near the zeros of the FFLO order parameter [14]. It is a large LDOS in the bound states that triggers the formation of the IC LRO [17]. At finite T the LRO phase is stabilized only when a sufficient number of the nodal planes containing the bound states is induced by H_0 [14,18]. Since these same states contribute to the shift, it is likely that the transition identified by it is indeed the one to the IC-LRO phase. Alternatively, if the visible increase in the In(1) shift as compared to that in the LFSC phase is attributed to canting, an evident increase should also onset only after LRO is established as observed.

Based on these observations and consistent with the discussion in the previous paragraphs, we postulate that a phase with strong AFM fluctuations [referred to as “exotic” SC (ESC)] develops for $9.2 \lesssim H_0 \lesssim 10.2$ T at low T, while the second order phase transition previously reported [6,12,19] at $H^* \approx 10.2$ T marks the transition to the state with well-established static IC-SDW order. Such a two-step phase transition from LFSC to FFLO (assuming it exists in ESC) and then to the IC state was theoretically predicted when spin fluctuations are considered in Ref. [14]. In light of our NMR results, we propose the revised phase diagram sketched in Fig. 4.

In conclusion, our comprehensive low-T NMR data provide a clear picture of the field evolution of the IC magnetism, and the magnitude of the magnetic moment, confined within the high-H_0 low-T phase of CeCoIn$_5$. Our analysis of the field dependence of the NMR shift on different In sites indicates that the IC-SDW order likely coexists with a FFLO state. Finally, we identify a novel phase, in the field regime in between the LFSC and IC-SDW states, which is likely a true FFLO phase in the presence of strong AFM fluctuations.

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