Magnetic Field Splitting of the Spin Resonance in $CeCoIn_5$

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Neutron scattering in strong magnetic fields is used to show the spin resonance in superconducting $CeCoIn_5$ ($T_c = 2.3$ K) is a doublet. The underdamped resonance ($\hbar\Gamma = 0.069 \pm 0.019$ meV) Zeeman splits into two modes at $E_{\pm} = \hbar\Omega_0 \pm \alpha\mu_B\mu_0 H$ with $\alpha = 0.96 \pm 0.05$. A linear extrapolation of the lower peak reaches zero energy at 11.2 ± 0.5 T, near the critical field for the incommensurate "Q phase." Kenzelmann *et al.* [Science **321**, 1652 (2008)] This, taken with the integrated weight and polarization of the low-energy mode (E_-), indicates that the Q phase can be interpreted as a Bose condensate of spin excitons.

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The presence of an underdamped resonance peak in the neutron scattering response has proven to be a strong indication of unconventional superconductivity [1–3]. Spin resonances have been reported in a series of heavy fermion-, cuprate-, and iron-based superconductors and have been associated with the gap function undergoing a change in sign $(\Delta(\mathbf{q} + \mathbf{Q_0}) = -\Delta(\mathbf{q}))$ [4]. Therefore, neutron scattering can be used to probe the electronic superconducting gap symmetry.

It is to be expected that applied magnetic fields, which suppress the superconducting order parameter, should have a strong effect on the spin resonance. Such effects have been difficult to pursue in the cuprates and iron based superconductors where chemical doping is required and resonance energies are high [5]. $CeCoIn_5$ is, however, particularly well suited owing to the stoichiometric nature of the compound and the accessible field and energy scales.

$CeCoIn_5$ displays an unconventional superconducting phase at ambient pressures and at temperatures below 2.3 K with a gap characterized by d-wave symmetry [6–9]. The structure is layered tetragonal with magnetic Ce^{3+} ions in Ce-In(1) planes stacked along the c-axis and separated by a Co-In(2) network [10]. While the Fermi surface is characterized by three dimensional sheets (Refs. [11–13]), the superconductivity reflects the underlying lamellar structure with a critical field of ~12 T within the a-b plane and ~5 T for fields along c [14–17]. Neutron scattering (Ref. [18]) shows the normal state has overdamped magnetic excitations peaked near $\mathbf{Q}_0 = (1/2, 1/2, 1/2)$ indicative of antiferromagnetic interactions between Ce^{3+} ions both within the a-b plane and along c. The commensurate magnetic spin response differs from nonsuperconducting, though metallic, $CeRhIn_5$ which displays a magnetic Bragg peak at the incommensurate point $\mathbf{Q} = (1/2, 1/2, 0.297)$ with a spiral magnetic structure [19]. On entering the superconducting phase in $CeCoIn_5$, an underdamped resonance peak at $\hbar\Omega_0 = 0.60$ meV develops gathering spectral weight from low energies. These results indicate strong coupling between f-electron d-wave superconductivity and magnetism. A similar result and analysis has been applied to the heavy fermion superconductor $CeCu_2Si_2$ where a spin resonance has also been observed in the superconducting phase [20].

While no magnetic Bragg peak was found at zero fields in $CeCoIn_5$, incommensurate order with $\mathbf{Q} = (0.45, 0.45, 0.5)$ was observed for fields within the a-b plane in a narrow field range below H_{c2} [1,21–23]. This, so called Q phase appears to be directly linked to superconductivity as it vanishes abruptly for magnetic fields above H_{c2}.

The underlying structure of the resonance peak has been a matter of considerable theoretical interest. One means of probing this is through high field spectroscopy which may lift any degeneracy of the resonance mode. Here we demonstrate that the spin resonance in $CeCoIn_5$ is a doublet and the lower branch may represent the soft mode of the Q phase order.

The results are based upon experiments performed on four cold neutron spectrometers. The sample consisted of ~300 crystals aligned such that Bragg reflections of the form (HHL) defined the horizontal scattering plane [18]. High resolution measurements in a vertical magnetic field aligned along the $[1\bar{1}0]$ direction perpendicular to the scattering plane, were performed on the OSIRIS spectrometer (ISIS, UK) with a fixed $E_f = 1.84$ meV. By rotating the sample through ~15 positions spaced 0.5°

apart, a map in momentum and energy was constructed from which constant Q spectra near the commensurate $(1/2, 1/2, 1/2)$ position were extracted. Triple-axis measurements with vertical fields (aligned along the $[1\bar{1}0]$ direction) were also performed at SPINS and MACS (NIST, USA) with $E_f = 3.7$ meV and 3.5 meV, respectively. Horizontal field measurements, with the magnetic field aligned within the scattering plane, were taken at FLEX (Helmholtz Zentrum Berlin) where the field was within the (HHL) plane rotated 30° from [001] to improve access for the incident and scattered beams. For the horizontal field data discussed in this Letter, we list the component of field projected along the c axis.

The effect of magnetic fields, close to the upper critical field, on the spin resonance is summarized in Fig. 1. Panels (a) and (b) show results for fields along $[1\bar{1}0]$, where $H_{c2} = 12$ T. For 0 T, we reproduce our previous results (Ref. [18]), while panel (b) shows that at 11 T a resonance is no longer observed. Panel (c) demonstrates the resonance peak remains visible under the more constrained condition imposed by the horizontal field configuration on FLEX at 2 K. For modest fields along [001] near $H_{c2} = 5$ T [panel (d)], the resonance is suppressed, presumably replaced by the over damped fluctuations reported at similar fields by NMR [24]. We infer that the resonance peak is associated with superconductivity.

Figure 2 illustrates the response of the spin resonance to intermediate fields in the superconducting phase well below H_{c2}. The constant-Q scans are formulated by integrating around $H = [0.45, 0.55]$ and $L = [0.45, 0.55]$ on the OSIRIS indirect geometry spectrometer. The scans were performed at $T = 0.1$ K, well below the transition to superconductivity ($T_c = 2.3$ K), and the vertical field was applied along the $[1\bar{1}0]$ axis with the sample aligned in the (HHL) scattering plane. The resolution at the elastic line on OSIRIS is 0.025 meV (full-width at half maximum) and increases to 0.026 meV at 0.5 meV [25]. The resolution function is illustrated by the solid curve in panel a) centered at 0.63 meV [25]. A background derived from a similar scan at 10 K has been subtracted. The solid lines are fits to damped harmonic oscillator response functions convolved with the measured elastic resolution function. While previous measurements on SPINS found the resonance peak width to be largely defined by the energy resolution of the spectrometer, panel a) shows that the zero field resonance does have a finite lifetime with

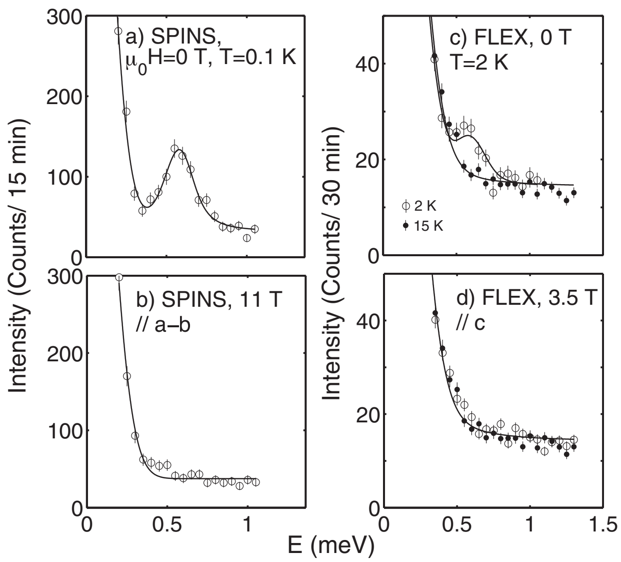

FIG. 1. Panels (a) and (b) illustrate the $\mathbf{Q} = (1/2, 1/2, 1/2)$ resonance in zero field ($T = 0.1$ K) and 11 T (near H_{c2}) along the $[1\bar{1}0]$ direction. Panels (c)–(d) are acquired with the field applied along the c axis. The curves are fits to a simple harmonic oscillator and a background originating from incoherent elastic scattering. The temperature independent peak at $E = 0$ meV results from background associated with strong nuclear incoherent scattering and is not magnetic in origin.

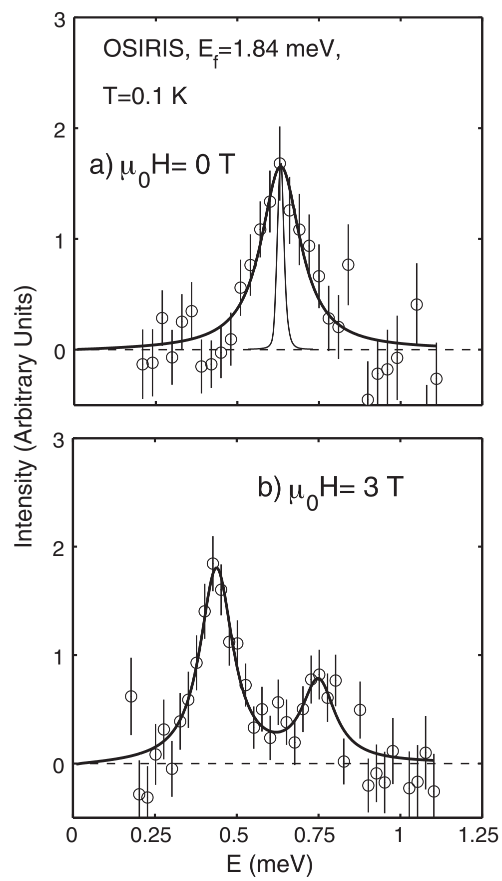

FIG. 2. (a) A high resolution scan through the spin resonance at zero applied field. The solid curve centered at 0.6 meV illustrates the resolution function on OSIRIS with a full width of 0.025 meV. The resolution is derived from the elastic line and it was assumed not to change significantly between 0 and 0.5 meV (Ref. [25]). (b) Demonstration of the splitting of the resonance into two peaks under an applied field of 3 T. A 10 K background was subtracted from the scans.

$\hbar\Gamma = 0.069 \pm 0.019$ meV (half width at half maximum) after correcting for resolution. This was confirmed through additional measurements on SPINS described in the Supplemental Material [26].

Figure 2(b) shows the same scan but in an applied vertical field of 3 T along the $[1\bar{1}0]$ direction at 0.1 K. The single peak observed in panel (a) at zero field is seen to be split into two peaks and this demonstrates that the resonance peak in $CeCoIn_5$ is a doublet. The intensity ratio between the two peaks is 0.41 ± 0.11 at 3 T. The width of the two peaks are equal, to within experimental error, and fitted to be $\Gamma = 0.056 \pm 0.008$ meV (half width). The solid curve is a fit to two damped harmonic oscillator line shapes convolved with the resolution function. We have put an upper bound of 5% of the zero field intensity that could reside in a putative central and field independent peak, which would be associated with a triplet. This upper bound is confirmed by measurements performed at intermediate fields as described in the Supplemental Material [26].

For measurements over a broader range of fields, we use the coarser resolution and higher intensity of the cold neutron triple-axis spectrometer SPINS. Figure 3 illustrates the evolution of the resonance peak as a function of fields ranging over 2–5 T. Panel (a) is a contour plot of the 11 T background corrected intensity as a function of magnetic field. The data show an intense lower peak which softens with field and an upper peak which diminishes in intensity. Constant Q scans are summarized in panels (b)–(e) where the solid lines are fits to a linear combination of two damped harmonic oscillators of equal width. The data at 2 T [panel (b)] show a broadening of the resonance which persists to 3 T and is consistent with Fig. 2 with the larger resolution width of 0.15 meV. At 4 T, [panel (d)] a distinct splitting can be resolved and two peaks are observed. The intensity ratio at 4 T is 0.39 ± 0.1, consistent with the 3 T OSIRIS data illustrated in Fig. 2. At 5 T, the two peaks are displaced and the intensity at the central position nearly approaches the background measured at 11 T. These results are consistent with a previous cold triple-axis study (Ref. [27]), which tracked the softening of the lower peak with field, but did not observe the upper peak of the doublet shifted to higher energies.

We plot the peak positions [Fig. 4(a)] and intensities [Fig. 4(b)] as a function of magnetic field applied along the $[1\bar{1}0]$ direction. The solid lines are fits to $E_{\pm} = \hbar\Omega_0 \pm \alpha\mu_B\mu_0 H$ as expected for a Zeeman split doublet. The slope $\alpha = 0.96 \pm 0.05$ results in a net and field dependent splitting between the E_+ and E_- modes of $2\alpha\mu_0\mu_B H = (1.92 \pm 0.10)\mu_0\mu_B H$. This slope may be compared with the Lande factor of 0.83 for a free Ce^{3+} ion and $g_{\parallel} = 1.95$ calculated from a crystal field analysis with an in-plane magnetic field as discussed in the supplementary information [26,28]. This comparison illustrates that the spectral

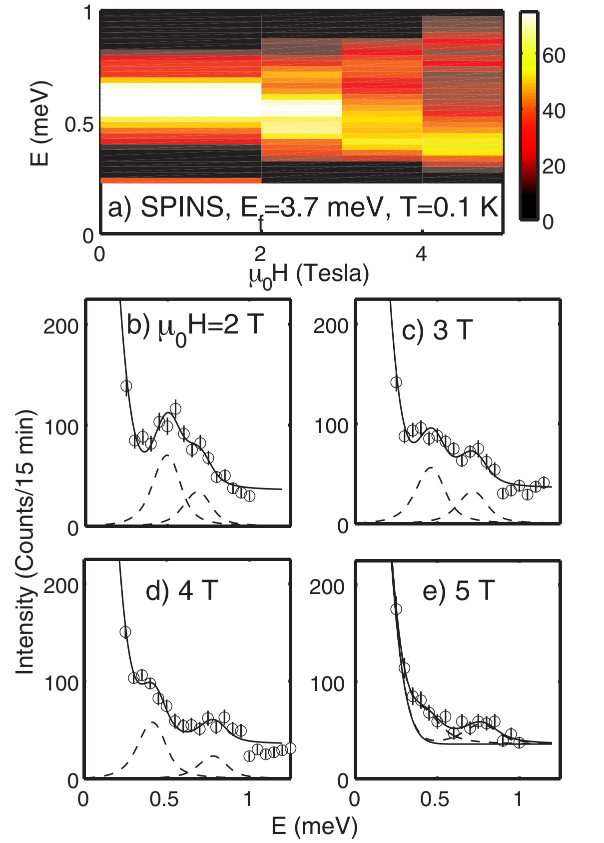

FIG. 3 (color online). The magnetic field dependence of the resonance peak at 0.1 K taken on SPINS. (a) background corrected intensity as a function of magnetic field. (b)–(e) The solid lines are fits to two damped harmonic oscillators and the dashed lines indicate the individual fits. An overall background fixed to the 11 T scan has also been subtracted and is represented by the solid line in (e) for the 5 T data.

weight in the resonance may originate from localized $4f$ electrons associated with the Ce^{3+} ions in a tetragonal crystal field. The dashed line is the calculated energy position from Ref. [29] normalizing the y axis energy scale to match the zero field resonance (0.6 meV) energy and the horizontal axis to agree with the onset of magnetic order in the Q phase (taken to be 10.6 T). The intensity of the two peaks is displayed in Fig. 4(b). The E_+ peak shows a consistent trend to decreasing intensity at larger fields while the E_- peak intensity is constant within error. The dashed line is 1/2 of the zero field resonance spectral weight.

Predictions for the field splitting of the resonance in the cuprates (in the context of $Pr_{0.88}LaCe_{0.12}CuO_{4-\delta}$) have suggested a splitting into three peaks reflecting an excitation from a singlet ground state to a triplet excited state [30] The central field independent peak is longitudinally polarized while the field dependent peaks are transverse. In this theoretical study, the intensity of the two field dependent peaks was predicted to be equal. However, in the close

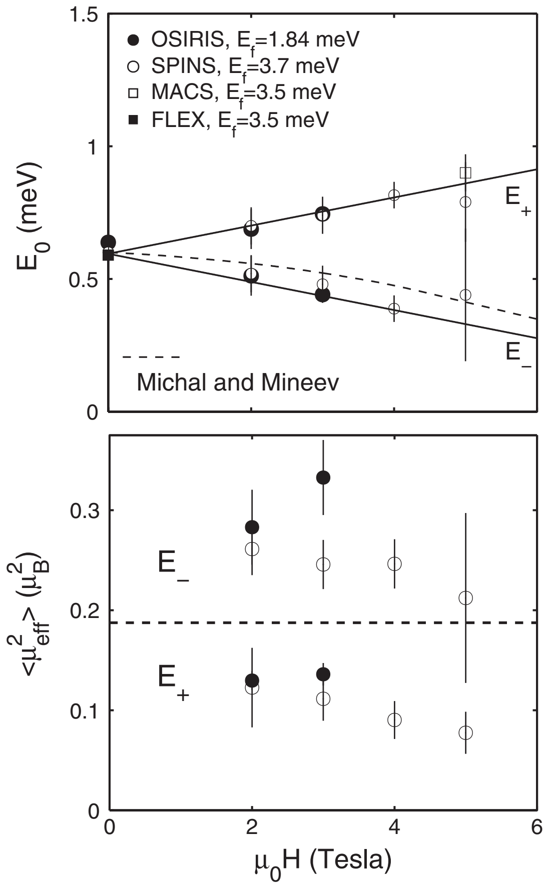

FIG. 4. (a) The peak positions of the field spit resonance as a function of applied field within the *a*-*b* plane. The solid lines are fits to $E_{\pm} = \hbar\omega_0 \pm \frac{1}{2} g\mu_B\mu_0 H$ with $g = 1.92 \pm 0.10$. The dashed line is the theory described in Ref. [29]. (b) The integrated intensity of the upper split peak in absolute units. The dashed line is 1/2 the integrated intensity of the zero field resonance peak.

proximity of a particle-hole continuum the upper mode was predicted to weaken.

The splitting of the $CeCoIn_5$ spin resonance is different because only two peaks splitting with the Zeeman energy are observed. Below 4 T, the total integrated spectral weight encompassed by $E_{\pm}$ is conserved to within 20% of the zero field value of $0.37 \pm 0.05\mu_B^2$. However, the upper peak (E_+) intensity weakens beyond 4 T [Fig. 4(b)]. This may indicate the resonance is located near a particle hole continuum causing a loss of intensity in E_+ as this mode is driven into the continuum. Such a scenario was suggested in Ref. [31], though other theories have been proposed (Ref. [32]). Missing spectral weight in a spin excitation also occurs in the cuprates at high energies and has been suggested to result from the close proximity of a continuum related to the pseudogap [33,34].

The results for $CeCoIn_5$ differ from excitations observed in dimer quantum magnets (namely $TlCuCl_3$ and PHCC) where the ground state is a singlet and the first excited state is a triplet [35,36]. The $\Delta S_z = \pm 1$ modes have equal intensity and the $\Delta S_z = 0$ mode (the central peak) is the strongest. In $CeCoIn_5$, only two field dependent peaks with differing intensity are observed and no intensity is measurable in the central component, which is normally the strongest in insulating quantum magnets. Therefore, we cannot interpret the resonance as an excitation from a singlet ground state to an excited triplet.

Any description of the resonance ($\hbar\Omega_0 = 0.60$ meV) must reconcile the experimental facts that the resonance is a doublet, the total zero field spectral weight is $\sim 0.37\mu_B^2$, and that a polarization analysis (based upon L scans using unpolarized neutrons) indicates that the fluctuations are polarized along the c axis corresponding to J_z matrix elements (see supplementary information). One way of understanding this is to consider excitations from a superconducting d-wave condensate ($|\psi\rangle$) to an excited state that can be described as a condensate with a localized $4f$ spin ($|\psi, \pm\rangle$). This exciton (Ref. [29]) state lies at an energy ($\hbar\Omega_0$) and is a doublet on account of the $4f$ crystal field environment. Based on the zero field results, these two states are connected by J_z but not by $J_{\pm}$, which presumably reflects a characteristic of the condensate. In this picture the effect of the applied field would be to split the doublet into two peaks $E_{\pm} = \hbar\Omega_0 \pm \frac{1}{2} g\mu_B\mu_0 H$, with g being the Lande factor for the localized $4f$ crystal field doublet. This is consistent with the observation of two peaks and the similarity of the experimental $g \equiv 2\alpha = 1.92 \pm 0.10$ to the value of 1.95, derived from a crystal field analysis.

Extrapolating the lower energy position in Fig. 4(a) to $E = 0$ correspondingly suggests a quantum critical point at 11.2 ± 0.5 T, close to the field where the Q phase is observed [1]. The spectral weight of the low-energy mode [Fig. 4(b)] is similar to the $0.16\mu_B$ ordered moment reported for the Q phase [21] and the moments are aligned along c as are the spin fluctuations associated with the zero field resonance. Therefore, it appears the lower peak of the split doublet is the soft mode of the Q phase which in turn may be interpreted as a Bose condensate of $|\psi, \pm\rangle$ excitons.

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