Evidence for Three Fluctuation Channels in the Spin Resonance of the Unconventional Superconductor $CeCoIn_5$

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Polarized inelastic neutron scattering under a magnetic field is used to get a microscopic insight into the spin resonance of the heavy fermion superconductor $CeCoIn_5$. The resonance line shape is found to depend on the neutron polarization: Some of the spectral weight is common to the two polarization channels while the remaining part is distributed equally between them. This is evidence for the spin resonance being a degenerate mode with three fluctuation channels: A Zeeman split contribution and an additional longitudinal mode.

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The spin resonance is a ubiquitous magnetic excitation that appears in many unconventional superconductors: cuprates, iron pnictides, and chalcogenides and heavy fermion systems. This mode is observed in inelastic neutron scattering (INS) experiments below the superconducting transition temperature, T_c. It appears at the wave vector for which the superconducting gap, Δ, changes sign and at an energy $\Omega_{\mathrm{res}}/2\Delta \approx 0.64$ [1]. While there is no consensus on the origin of such an excitation and its relevance to the pairing mechanism, it provides important information about the superconducting state. The spin exciton model describes the resonance as an $S = 1$ collective state [2] while an alternate view considers the resonance as a magnon in a disordered Néel state [3]. It is therefore important to get a more microscopic insight into the spin resonance and in particular to investigate the multiplicity of this excited state. The application of a magnetic field may lift its degeneracy and by polarized neutron scattering technique the nature of the corresponding fluctuations can be analyzed. Among unconventional superconductors, heavy fermion systems are particularly suited to the observation of strong effects under magnetic field because of their intrinsically low characteristic energy scales. Resonance peaks are reported so far for three heavy fermion systems, UPd_2Al_3 [4,5], $CeCu_2Si_2$ [6], and $CeCoIn_5$ [7].

$CeCoIn_5$ has the highest superconducting transition temperature among Ce heavy fermion compounds ($T_c = 2.3$ K) [8,9]. It crystallizes in the tetragonal space group P4/mmm, the superconducting gap symmetry is established to be the singlet $d_{x^2-y^2}$ state, and the Fermi surface shows nesting features for the wave vector $\mathbf{k} = (1/2, 1/2, 1/2)$. The spin resonance occurs at this wave vector $\mathbf{k}$ at an energy of $\Omega_{\mathrm{res}} = 0.6$ meV [7]. Under magnetic field, early investigations report that the resonance peak energy decreases [10,11]. One of the most intriguing properties of $CeCoIn_5$ is the occurrence of magnetic field induced incommensurate magnetic order [12,13]. Strikingly, this order disappears above $H_{c2} = 11.7$ T indicating a collaborative effect between the magnetic ordering and the superconductivity. In the present letter, we show, using polarized INS, that the spin resonance splits under magnetic field and that the magnetic response is composed of three elements: two Zeeman split peaks, that have a chiral nature, and an additional nonchiral contribution that appears at the same energy as the lowest of the Zeeman split peaks.

The experiment was performed on the cold neutron three-axis spectrometer IN14 at ILL, Grenoble. The sample is the same as that used in previous works [10,11]. It was put in a dilution insert inside a 3.8 T horizontal field magnet with the [1, −1, 0] axis vertical. The scattering plane was defined by [1,1,0] and [0,0,1]. The initial neutron beam was polarized with a bender placed after the pyrolitic graphite monochromator (polarization $\mathbf{P_0}$ with an experimental magnitude of 0.92; see Supplemental Material I [14]) and the polarization of the scattered beam was not analyzed (double focusing pyrolitic graphite analyzer). The spectrometer was setup in W configuration with fixed $k_f = 1.2$ Å^{-1}. The energy resolution given by the full width at half maximum (FWHM) of the incoherent signal was 0.1 meV. A Mezei flipper and a Be filter were placed before the magnet. The magnetic field defines the direction of $\mathbf{P_0}$ and is applied parallel to [1,1,1]. Hence, for $\mathbf{Q} = \mathbf{k} = (1/2, 1/2, 1/2)$, we have $\mathbf{H}//\mathbf{Q}//\mathbf{P_0}$. The measured magnetic intensity for a given momentum $\mathbf{Q}$ and energy transfer E is for such an experimental setup (without taking into account instrumental corrections):

$$I(\boldsymbol{Q}, E) \propto \int_{-\infty}^{+\infty} (\langle \boldsymbol{M}^{\perp}_{-Q} \boldsymbol{M}^{\perp}_{Q}(t)\rangle - i\boldsymbol{P}_0 \langle \boldsymbol{M}^{\perp}_{-Q} \times \boldsymbol{M}^{\perp}_{Q}(t)\rangle) dt e^{-i(E/\hbar)t}, \quad (1)$$

where $\boldsymbol{M}_Q^{\perp}(t)$ is the Fourier transform of the sample magnetization perpendicular to **Q** probed by INS and $\langle \ldots \rangle$ is the quantum statistical expectation value. The first term of Eq. (1) is the usual correlation function and the second term is the chiral correlation function. It is the antisymmetric part of the cross-correlation function between two orthogonal components of $\boldsymbol{M}_{\perp Q}$. A nonzero chiral correlation function indicates breaking both time reversal and parity reversal symmetry. It can arise from an intrinsic axial vector in the system like the Dzayloshinskii-Moriya interaction, from the presence of electron spin currents or from the application of a magnetic field [15].

The search for a chiral contribution in a spin superconducting resonance is motivated by the fact that in other spin singlet ground-state compounds, it was shown that the chirality enhances or suppresses different contributions to the magnetic excitation spectrum [16] (see Supplemental Material II [14]). In the following, we label I^{+0} the intensity of Eq. (1) for $P_0 = 1$ and I^{-0} for $P_0 = -1$. Figure 1(a) shows the magnetic excitation spectrum obtained at $\mathbf{Q} = (1/2, 1/2, 1/2)$ at 50 mK and 1 T. A single (nonsplit) resonance peak is obtained for both polarization channels at 0.57(1) meV in agreement with previous unpolarized data for the same field direction [11]. The background measured away from the resonance peak at $\mathbf{Q} = (0.38, 0.38, 0.9)$ shows no polarization dependence and we averaged the data over the two polarization channels. Figure 1(b) shows the same constant **Q** scan performed at a higher field of 2 T. Here, the energy dependence of the signal depends on the different initial polarizations. For I^{+0}, most of the spectral weight is located around *a single energy* $E_1 = 0.5$ meV (blue arrow) while for I^{-0}, spectral weight is present *at two distinct energies* $E_1 = 0.5$ and $E_2 = 0.7$ meV (red arrow). Such a polarization dependence of the magnetic excitation spectrum is confirmed by constant energy scans performed at E_1 and E_2, shown in Figs. 1(d) and 1(e), respectively. Finally Fig. 1(c) shows the data above T_c at the same magnetic field of 2 T where the spin resonance is replaced by a quasielastic signal independent of the polarization.

Figures 2(a) and 2(b) show the difference between the spectra collected at $\mathbf{Q} = (1/2, 1/2, 1/2)$ at 2 T and 50 mK and the background fit shown in Fig. 1. To go further, we focus on the chiral part of the scattering which is shown in Fig. 2(c) as the difference of the raw data $I^{-0} - I^{+0}$. From this figure, we can see that the chiral part has similar spectral weight, but with opposite sign, above and below an energy $\Omega_0 = 0.60(2)$ meV for which $I^{-0} = I^{+0}$. Since this symmetry of the chiral intensity is not reflected in the total signal shown in Fig. 1(b), we propose as the simplest possible analysis that the measured signal is composed of two components: a contribution with a chiral character which has a similar spectral weight on both sides of Ω_0 and a contribution without chiral character that is responsible for the asymmetry of the total signal between the two polarization channels. The data analysis follows in a straightforward way. The resolution effects are not taken into account since the energy width of the nondispersive modes is found to be twice the FWHM of the incoherent signal. Each mode i is described by a Lorentzian function $I_{\Omega_i}(E) = A_i/((E - \Omega_i)^2 + \Gamma_i^2)$ (see Supplemental Material III [14]) and the fit is made simultaneously for the two polarization channels with the following coupled equations (using the experimental value $|P_0| = 0.92$):

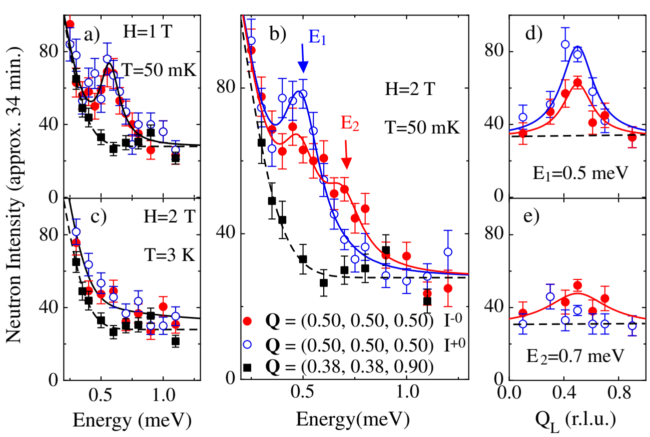

FIG. 1 (color online). Magnetic excitation spectra measured for initial polarization $P_0 = 1$ (open circles) and $P_0 = -1$ (full circles). The left and central panels show constant **Q** scans performed at $\mathbf{Q} = (1/2, 1/2, 1/2)$ for (a) $T = 50$ mK and $H = 1$ T (b) $T = 50$ mK and $H = 2$ T and (c) $T = 3$ K, and $H = 2$ T. The background (squares) is measured at $\mathbf{Q} = (0.38, 0.38, 0.9)$ and averaged over the two polarizations. The right panels show constant energy scan performed at $T = 50$ mK and for $H = 2$ T for (d) 0.5 meV and (e) 0.7 meV. Those scans are obtained by rocking the sample, the label Q_L is given for indication. All lines are fits to the data as explained in the text.

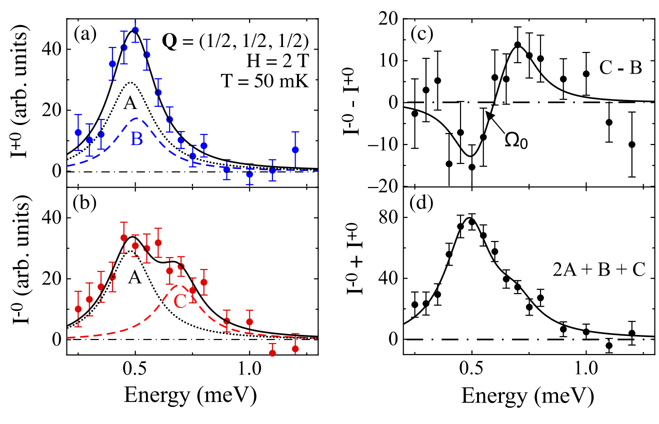

FIG. 2 (color online). Excitation spectra measured at $T = 50$ mK and $H = 2$ T (a) and (b) show the data obtained with the background fit subtracted. Panel (c) shows $I^{-0} - I^{+0}$ and panel (d) shows the sum $I^{+0} + I^{-0}$. The lines are fits as indicated in the text.

$$I^{+0} = I_{\Omega_0} + (1/2)((1 + |P_0|)I_{\Omega_{res}-\delta} + (1 - |P_0|)I_{\Omega_{res}+\delta}), \quad (2)$$

$$I^{-0} = I_{\Omega_0} + (1/2)((1 - |P_0|)I_{\Omega_{res}-\delta} + (1 + |P_0|)I_{\Omega_{res}+\delta}), \quad (3)$$

where I_{Ω_0} corresponds to the contribution without chiral character (contribution A) and $I_{\Omega_{res}-\delta}$ and $I_{\Omega_{res}+\delta}$ correspond to the magnetic field split contributions with a chiral character (contributions B and C).

In Figs. 2(a) and 2(b), the black dotted line shows the contribution without chiral character and the contribution which has a chiral character is shown by the dashed blue curve for $P_0 = 1$ and dashed red curve for $P_0 = -1$, both having the same spectral weight, as inferred from the raw data shown in Fig. 2(c). At 1 T, the measurements do not exhibit any difference between the two polarization channels, we therefore fit the data by a single mode Ω_0. The following parameters were found: $\Omega_0(1\text{ T}) = 0.57(1)$ meV, $\Gamma_0(1\text{ T}) = 0.11(2)$ meV, and $A_0(1\text{ T}) = 4.6(4)$ arb. units in each channel. As indicated above, the FWHM of the peak, $2\Gamma_0$, is of about twice that of the incoherent signal. At 2 T, all the energy widths were fixed to 0.11 meV at first. The nonchiral part has an energy $\Omega_0(2\text{ T}) = 0.47(2)$ meV and an intensity $A_0(2\text{ T}) = 3.3(4)$ arb. units in each channel. The chiral part is characterized by an averaged energy $\Omega_{res}(2\text{ T}) = 0.60(2)$ meV and a total splitting $2\delta = 0.16(4)$ meV. The respective peak intensities are $A_{\Omega_{res}-\delta}(2\text{ T}) = 2.3(3)$ arb. units and $A_{\Omega_{res}+\delta}(2\text{ T}) = 2.2(5)$ arb. units. When the energy widths were unfixed, similar parameters were obtained and the fitted values for the relaxation rates are equal within the error bars to previously fixed value. The splitting 2δ between contributions B and C corresponds to a magnetic moment of $\mu = 0.69\mu_B$ for a linear Zeeman splitting. [Note that $\Omega_0(2\text{ T})$ corresponds to the zero field resonance energy, $\Omega_{res}(0\text{ T})$.] For completeness, the sum $I^{+0} + I^{-0}$ is shown in Fig. 2(d); it is consistent with the unpolarized data obtained previously [10,11]. The broad nature of the signal observed in the previous unpolarized INS experiments arises from the degeneracy of the mode, which is lifted under magnetic field and resolved here in different channels using polarized INS. A very recent work using *high resolution* unpolarized INS also reports the splitting of the peak under field and comparison with the present study will be made below [17]. In Fig. 3, the magnetic field dependence of the resonance energy is summarized, also showing previous unpolarized INS data.

The present polarized INS study shows a splitting of the magnetic excitation under a magnetic field of 2 T. Since this splitting is not seen at 1 T, it is ascribed to the Zeeman effect. Some insight on the effect of a magnetic field on the resonance excitation in a superconductor is given by theoretical studies carried out for the $S = 1$ exciton model. If only the orbital effect is taken into account, the resonance

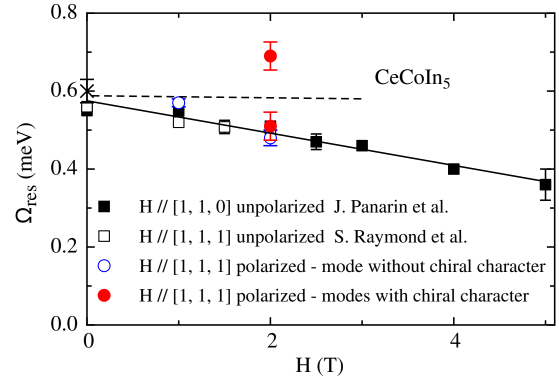

FIG. 3 (color online). Magnetic field dependence of the resonance energy. The full (empty) squares are obtained by unpolarized INS data with magnetic field along [1,1,0] [10] ([1,1,1] [11]). The full circles are the modes with chiral character and the empty circles are the nonchiral contribution. The cross at $H = 0$ T is obtained by Stock *et al.* [7]. Lines are guides for the eyes.

peak shifts to lower energy without any splitting and the line shape broadens [18]. When the effect on the spin part of the Cooper pairs is taken into account (the Pauli effect), a splitting is expected as initially theoretically proposed for electron doped cuprates [19]. This behavior is confirmed by a recent theoretical study dedicated to $CeCoIn_5$, a prime candidate for resonance splitting due to its strong Pauli limited nature. In this study, which considers Ising symmetry for simplicity, the spin resonance splits into two branches and the lower branch is considered to be the soft mode of the field induced magnetic ordering [20,21].

Beyond establishing a splitting under magnetic field, our measurements exhibit an asymmetry of scattering between the two polarization channels. This is in contrast to the results of the seminal work performed on spin triplet excitations of the nearly isotropic spin ladder compound $Sr_{14}Cu_{24}O_{41}$ using the same experimental setup [16]. In that case, the magnetic field splits the triplet into three components labeled by the quantum number S_z. The $S_z = 0$ component is not measurable with this experimental setup. The $S_z = -1$ ($S_z = 1$) mode of the triplet is observed at the energies $\Omega - \mu H$ ($\Omega + \mu H$) in the polarization channels I^{+0} (I^{-0}). Hence, the modes at different energies $\Omega \pm \mu H$ contribute separately to different polarization channels.

We do not find in $CeCoIn_5$ an equivalent total enhancement or suppression of intensity for the energies $\Omega \pm \mu H$ in each polarization channel. We introduce an additional mode (contribution A) in the total magnetic scattering to describe this fact. In a classical description of spin motion, the contribution with chiral character corresponds to spin precession perpendicular to the field while the contribution without chiral character is understood as longitudinal fluctuations. (They are polarized along an axis that must have a component perpendicular to the field given our experimental setup.)

Our polarized INS data are consistent with the very high resolution unpolarized INS data recently reported by Stock *et al.* for $H//[1,1,0]$ [17] with regards to the magnitudes of the Zeeman splitting and the intensities. However, our work establishes that the single low energy mode at energy $\Omega - \mu H$ seen by Stock *et al.* is in fact composed of two parts with different character. One of these two parts is chiral and has the same intensity as its counterpart mode at energy $\Omega + \mu H$. There is therefore no need to invoke any damping effect related to the electron-hole continuum to explain the intensity of the mode at $\Omega + \mu H$.

The present experiment was performed for $H//[1,1,1]$, as in Ref. [11], while all other INS investigations are performed for $H//[1,1,0]$ [10,17]. In our discussion we neglect the 23° angle between these two directions owing to the fact that the previous INS experiments performed for both field directions show similar results [10,11]. Bulk measurements also indicate a smooth behavior when the field is tilted off the tetragonal basal plane except for the occurrence of the field induced ordered phase [22]. The very good agreement between the unpolarized high resolution INS data for $H//[1,1,0]$ and the polarized INS data for $H//[1,1,1]$ *a posteriori* shows that the effect of a 23° difference in field direction is negligible within the achieved statistics.

Finally, we comment on the striking fact that the lower branch of the split mode (red point in Fig. 3) has a similar energy to that of the longitudinal-like contribution (blue point in Fig. 3). This observation casts some doubt on the interpretation of the data using spectroscopic g factors of localized $4f$ electrons in their crystal field environment, although this interpretation describes the data surprisingly well, if the low energy peak is considered as a single object [11,17]. A recent calculation shows that the appearance of a resonance peak in any given spin fluctuation channel (longitudinal, transverse) is very sensitive to the anisotropy of both the g factors and the quasiparticle interactions, leading to many possibilities for the excitation spectrum under magnetic field [23]. An important step will be to find a set of parameters describing the overall INS experimental results within such a model.

Several experiments have been devoted to the study of anisotropy of the spin resonance in unconventional superconductors using polarized INS at $H = 0$ T. A global picture is missing due to contrasting results. In the iron-based superconductors, the spin resonance is slightly anisotropic with a larger in-plane component in $FeSe_{0.5}Te_{0.5}$ [24] whereas for $BaFe_{1.9}Ni_{0.1}As_2$ [25] only a planar component is observed. In the cuprates, a study on $YBa_2Cu_3O_{6.9}$ reveals spin anisotropy below the resonance energy with a dominant in plane component while the resonance peak has isotropic spin fluctuations [26]. Together with our results on $CeCoIn_5$, the common trend for all these superconductors is that the fluctuations associated with the resonance peak span cases ranging from XY to isotropic symmetries but not Ising-like. This could be related to the fact that all these compounds achieve a singlet superconducting state.

In conclusion, we have seen by polarized INS experiments under magnetic field, a Zeeman splitting of the superconducting spin resonance of $CeCoIn_5$, a feature related to its strong Pauli limited nature. This splitting involves only one part of the magnetic response which has a chiral nature. The remaining spectral weight consists of a contribution whose energy is close to the lower mode of the Zeeman split contribution. The spin resonance in $CeCoIn_5$ is thus a composite excitation which contains, under magnetic field, three excitation channels involving both precessional and longitudinal modes. This work puts strong constraints on further theoretical descriptions of the spin resonance in $CeCoIn_5$.

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