

Spin Resonance in the d -Wave Superconductor CeCoIn₅

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Neutron scattering is used to probe antiferromagnetic spin fluctuations in the d -wave heavy fermion superconductor CeCoIn₅ ($T_c = 2.3$ K). Superconductivity develops from a state with slow ($\hbar\Gamma = 0.3 \pm 0.15$ meV) commensurate [$\mathbf{Q}_0 = (1/2, 1/2, 1/2)$] antiferromagnetic spin fluctuations and nearly isotropic spin correlations. The characteristic wave vector in CeCoIn₅ is the same as CeIn₃ but differs from the incommensurate wave vector measured in antiferromagnetically ordered CeRhIn₅. A sharp spin resonance ($\hbar\Gamma < 0.07$ meV) at $\hbar\omega = 0.60 \pm 0.03$ meV develops in the superconducting state removing spectral weight from low-energy transfers. The presence of a resonance peak is indicative of strong coupling between f -electron magnetism and superconductivity and consistent with a d -wave gap order parameter satisfying $\Delta(\mathbf{q} + \mathbf{Q}_0) = -\Delta(\mathbf{q})$.

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While magnetism and s -wave superconductivity are thought to be incompatible, spin fluctuations may actually mediate superconductivity with other order parameter symmetries such as a d -wave gap symmetry [1]. A possible indication of this comes from the suggested observation of a spin resonance in the superconducting state of various high temperature superconductors. Intricacies of the cuprates, including the electronic inhomogeneity and the complex phase diagrams, complicates the interpretation so it is of interest to examine related phenomena in chemically distinct materials which are homogeneous and lack electronic and structural disorder. In this Letter we report the observation of a spin resonance in the CeCoIn₅ superconductor, a rare earth, and chemically homogeneous d -wave superconductor.

CeCoIn₅ is a superconductor with the highest recorded transition temperature thus far ($T_c = 2.3$ K) for a heavy fermion material [2]. The compound is part of the CeTIn₅ ($T = \text{Rh, Ir, and Co}$) family of intermetallics featuring both antiferromagnetism (AFM) and superconductivity (SC). The tetragonal HoCoGa₅ type crystal structure of these systems is built from alternating layers of CeIn₃ and TIn₂ stacked along the [001] direction. The presence of two-dimensional planes of magnetic Ce³⁺ ions links these materials to the SC cuprates and other materials where AFM and SC coexist [3]. The quasi two-dimensional nature is reflected in de Haas and van Alphen measurements which show that the Fermi surface includes nearly cylindrical surfaces and the largely two-dimensional nature of the electronic properties has been suggested as a contributing factor to the high superconducting transition temperature [4]. This contrasts with AFM ordered CeRhIn₅ where the Fermi surface exhibits a stronger three dimensional character [5,6]. The detected cyclotron masses of 5–87 m_0 are extremely large with the $4f$ electrons contributing greatly to the Fermi surface [7]. While node locations

within the basal plane remain under investigation [8,9], it is clear that the SC state has d -wave symmetry.

The experiments were conducted on the SPINS cold neutron spectrometer at the NIST Center for Neutron Research. A variable vertically focusing graphite (002) monochromator and a horizontally focused graphite (002) analyzer subtending a solid angle of 0.021 sr with 11° horizontal acceptance were used. Energy transfer, $\hbar\omega = E_i - E_f$, was defined by fixing the final energy to 3.7 meV and varying the incident energy. Cooled Be and BeO filters were placed before and after the sample, respectively. The sample consisted of ~ 300 CeCoIn₅ crystals with a total mass of 5 grams coaligned on a series of aluminum plates so reflections of the form (HHL) lay within the horizontal scattering plane. The room temperature lattice constants were measured to be $a = b = 4.60$ Å, and $c = 7.51$ Å. The crystals were secured to the mount using Fomblin-Y mechanical pump oil and covered by a thin aluminum plate. The combined mosaic spread was measured to be $\sim 3^\circ$ and $\sim 5^\circ$ for rotations around the ($1\bar{1}0$) and (001) directions, respectively. The magnetic intensity was normalized to absolute units using the integrated intensity from the (110), (002), and (111) Bragg peaks. The formula and notation used for the cross sections are the same as those described elsewhere [10].

The magnetic excitation spectrum at $\mathbf{Q} = (\frac{1}{2} \frac{1}{2} \frac{1}{2})$ in the normal state ($T = 3$ K) and in the SC state ($T = 1.3$ K) is plotted in Fig. 1. The normal state spectrum is featureless over the 1 meV energy range probed. The dashed line through these data show a Lorentzian response function, $\chi''(\mathbf{Q}, \omega) = \chi'_0 \Gamma \omega / (\Gamma^2 + \omega^2)$, with a relaxation rate $\hbar\Gamma = 0.3 \pm 0.15$ meV. This contrasts with the excitation spectrum in the SC state which displays a sharp peak for $\hbar\omega_0 = 0.60 \pm 0.03$ meV with a relaxation rate $\hbar\Gamma < 0.07$ meV.

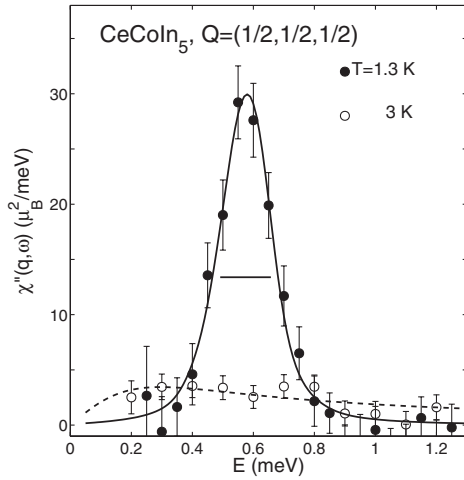


FIG. 1. The imaginary part of the dynamic susceptibility at $\mathbf{Q} = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ is plotted in the normal (3 K) and in the superconducting (1.35 K) states. A background taken at $\mathbf{Q} = (0.3, 0.3, 0.5)$ and $\mathbf{Q} = (0.7, 0.7, 0.5)$ was subtracted. The horizontal bar is the resolution width.

The momentum dependence of the magnetic neutron scattering cross section is plotted in Fig. 2. The left-hand panel shows scans along the $(HH\frac{1}{2})$ direction in the normal and SC states at several different values of energy transfer. The fits are to a single Gaussian function giving a dynamic correlation length (defined as the inverse of the half-width at half maximum) $\xi_{ab} = 9.6 \pm 1.0 \text{ \AA}$ at $\hbar\omega = 0.55 \text{ meV}$. In both the normal and SC phases, the magnetic scattering is peaked at $\mathbf{Q} = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ as for cubic CeIn_3 [11] indicating nearest neighbor AFM correlations within and between a - b planes.

The right-hand panel of Fig. 2 shows the wave vector dependence of the magnetic scattering along the $(\frac{1}{2}, \frac{1}{2}, L)$

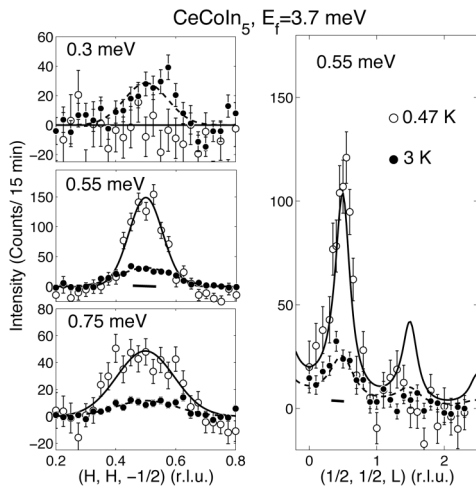


FIG. 2. Constant energy scans along the $(HH\frac{1}{2})$ and $(\frac{1}{2}, \frac{1}{2}, L)$ directions in the normal and superconducting states. A featureless background measured at 15 K has been subtracted from the data. The horizontal bars represent the resolution width. The data has been corrected for absorption effects.

direction. The solid line indicates a fit to $I(\mathbf{Q}) \propto f(Q)^2 \times [1 - (\hat{\mathbf{Q}} \cdot \hat{\mathbf{c}})^2] \sinh(c/\xi_c) / [\cosh(c/\xi_c) + \cos(\mathbf{Q} \cdot \mathbf{c})]$ which represents short-range AFM correlated Ce^{3+} moments polarized along the $[001]$ direction with a dynamic correlation length $\xi_c = 6.5 \pm 0.9 \text{ \AA}$ at $\hbar\omega = 0.55 \text{ meV}$. The squared form factor, $f(Q)^2$, was taken from previous measurements which agree with calculations by Blume *et al.* [12]. The second factor, $(1 - (\hat{\mathbf{Q}} \cdot \hat{\mathbf{c}})^2)$, indicates spin fluctuations polarized along \mathbf{c} which is consistent with the anisotropic susceptibility. In contrast to the Fermi surface which is highly two-dimensional, the ratio of dynamic spin correlation lengths is only $\xi_{ab}/\xi_c = 1.5 \pm 0.4$.

Commensurate AFM correlations distinguish CeCoIn_5 from CeRhIn_5 , which develops AFM long range order with $\mathbf{Q} = (\frac{1}{2}, \frac{1}{2}, 0.297)$ [13]. This is consistent with current understanding of the Fermi surfaces for these materials [5]. While there is considerable modulation and potential for nesting along \mathbf{c} for CeRhIn_5 , the Fermi surface for CeCoIn_5 is comprised of cylinders with a large effective mass and little modulation along \mathbf{c} . It is interesting to note that both commensurate and incommensurate order has been observed in $\text{CeRh}_{1-x}\text{Co}_x\text{In}_5$ ($x = 0.4$) and for similar concentrations in $\text{CeRh}_{1-x}\text{Ir}_x\text{In}_5$. AFM order and SC coexist in both compounds [14,15] indicating a close connection between commensurate AFM spin fluctuations and SC.

Figure 3 shows three constant- \mathbf{Q} scans at $\mathbf{Q} = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ for $T < T_c$. The solid lines are fits to a damped harmonic oscillator response function related to scattering through the fluctuation dissipation theorem. The fitting parameters

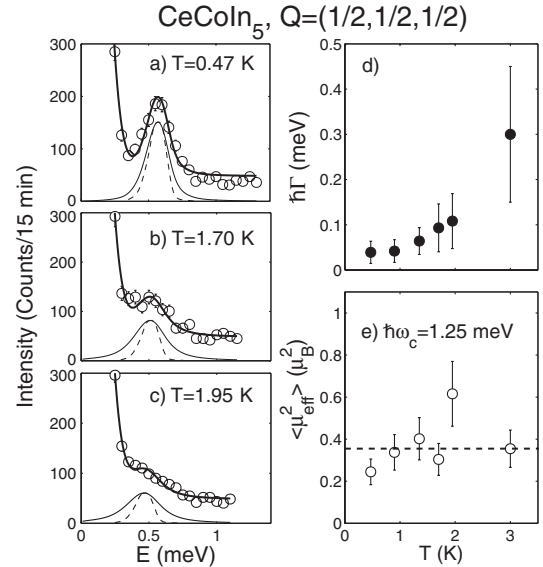


FIG. 3. (a)–(c) Constant- $\mathbf{Q} = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ scans for $T < T_c = 2.3 \text{ K}$ in CeCoIn_5 . The dashed line is the resolution function measured at $\hbar\omega = 0$ and scaled to have the calculated width for finite energy transfers. (d) Half-width at half maximum for inelastic peak from fits described in text. (e) \mathbf{Q} and $\hbar\omega$ integrated intensity below an energy cutoff $\hbar\omega_c = 1.3 \text{ meV}$.

include the staggered susceptibility $\chi'(\mathbf{q})$, the resonance frequency ω_0 , and the relaxation rate, Γ . For the fits in Fig. 3, we fixed the background to be that measured at 15 K where the magnetic scattering is no longer peaked in momentum transfer. The fit provides a good description of the data at all temperatures with a low T peak that broadens and softens on heating. Figure 3(d) shows $\hbar\Gamma$ versus temperature. As T approaches T_c , the peak broadens substantially in energy, less in momentum, indicating a decrease in lifetime of collective spin fluctuations as the SC gap closes.

The total moment sum-rule provides a measure of the fluctuating moment for $E < \hbar\omega_c$:

$$\langle \mu_{\text{eff}}^2 \rangle_{\omega_c} = \frac{\hbar}{\mu_B^2 \pi} \int_0^{\omega_c} d\omega \int_{\Omega_q} \frac{d^3 \mathbf{q}}{\Omega_q} \times \coth\left(\frac{1}{2} \beta \hbar \omega\right) \text{Tr}\{\chi''(\mathbf{q}, \omega)\}$$

Figure 3(e) shows that this quantity is approximately T -independent in the temperature range probed. This indicates that the spectral weight under the resonance peak predominantly comes from low-energies as confirmed through the constant energy scan at $\hbar\omega = 0.3$ meV illustrated in Fig. 2. We note that $\langle \mu_{\text{eff}}^2 \rangle_{\omega_c}$ is comparable to that obtained in the elastic channel of AFM ordered CeIn₃ and CeRhIn₅ [13,16].

Two temperature dependent energy scales can be derived from Fig. 3. Figure 4(a) shows the resonance energy, $\hbar\omega_0$ compared to the temperature dependence of the amplitude of the d -wave SC gap [17] scaled to $\hbar\omega_0$ for $T \rightarrow 0$. $\hbar\omega_0$ varies significantly less with temperature than the d -wave superconducting gap amplitude $2\Delta(T)$ calculated by conventional BCS theory indicating that these may be distinct energy scales.

The T dependence of the energy of the $4f$ electron system can be extracted from the corresponding spin susceptibility via the first moment sum-rule [18] $\hbar^2 \int_{-\infty}^{\infty} \omega d\omega \mathcal{S}^{\alpha\alpha}(\mathbf{q}, \omega) = \frac{1}{2} \langle [S_{\mathbf{q}}^{\alpha}, \mathcal{H}_{4f}, S_{\mathbf{q}}^{\alpha}] \rangle$. While single ion terms produce \mathbf{q} -independent contributions, Fig. 2 shows strongly \mathbf{q} -dependent changes in $\mathcal{S}^{\alpha\alpha}(\mathbf{q}, \omega)$ through T_c . This indicates that intersite terms dominate. Neglecting all but isotropic RKKY exchange yields

$$\Delta \langle \mathcal{H}_{\text{RKKY}} \rangle \approx \frac{-\hbar^2}{4\pi(g\mu_B)^2} \int_0^{\infty} \omega \text{Tr}\{\Delta \chi''(\mathbf{q}_0, \omega)\} d\omega. \quad (1)$$

Here $g = 0.86$ is the calculated Landé factor and we assume $\mathbf{q}_0 \cdot \mathbf{d} = \pi$ for all displacement vectors, \mathbf{d} , separating spins with bilinear exchange interactions. Introducing the cutoff $\hbar\omega_c = 1.25$ meV, $\Delta \langle \mathcal{H}_{\text{RKKY}} \rangle$ was calculated from the fit parameters and Eq. (1) and plotted versus T in Fig. 4(b). Also shown is the much smaller overall reduction in electronic energy derived by integrating phonon subtracted specific heat data (solid line).

The present neutron data associate CeCoIn₅ with various magnetic SC with a spin resonance. Most notably, the cuprate superconductors YBa₂Cu₃O_{6+x} and

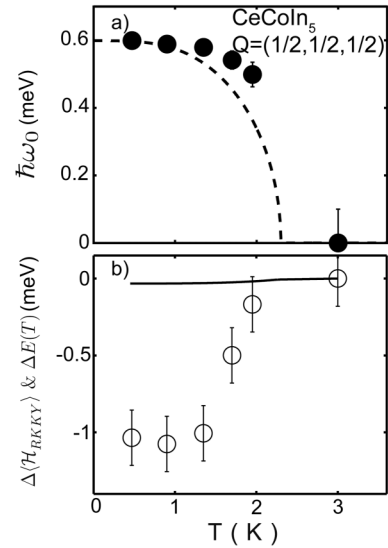


FIG. 4. (a) Spin resonance energy versus T compared to the scaled d -wave BCS superconducting gap amplitude described text. (b) thermal variation for $T < 3$ K of the exchange energy, $\Delta \langle \mathcal{H}_{\text{RKKY}} \rangle$, derived from inelastic neutron scattering compared to the overall electronic energy derived from specific heat data: $\Delta E(T) = \int_0^T C(T') dT' - \int_0^{3\text{K}} C(T') dT'$. Note there is an overall 50% systematic error bar on the normalization of the data for $\Delta \langle \mathcal{H}_{\text{RKKY}} \rangle$ in (b).

Bi₂Sr₂CaCu₂O₈ display a peak in the neutron scattering cross-section near $\hbar\omega = 41$ meV which has been associated with formation of a d -wave superconducting ground state [19–21]. A sharp spin resonance has also been observed in UPd₂Al₃ for $\hbar\omega = 0.35$ meV $\approx 2.3k_B T_c$ [22]. Independent experimental work has established that the SC gap function of all these materials undergoes a sign change $\Delta(\mathbf{q} + \mathbf{Q}_0) = -\Delta(\mathbf{q})$, where \mathbf{Q}_0 is the wave vector where the spin resonance cross section is greatest. Empirically, a sign change in $\Delta(\mathbf{q})$ thus appears to be a necessary condition for a spin resonance. Assuming line nodes extending along the c axis, our observation of a spin resonance at $\mathbf{Q}_0 = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ for CeCoIn₅ thus indicates $d(x^2 - y^2)$ symmetry for the SC order parameter. This result is consistent with recent angular dependent point contact tunneling data [23–25] and theoretical analysis of specific heat data in a magnetic field [26].

Much theoretical work has been devoted towards understanding the spin resonance in high temperature SC. Associating it with transitions to a bound state between collective spin and itinerant electron degrees of freedom, a coherence factor indicates that enhanced resonance intensity at wave vector \mathbf{Q}_0 requires a sign change in $\Delta(\mathbf{q})$ under the corresponding translation in reciprocal space: $\Delta(\mathbf{q} + \mathbf{Q}_0) = -\Delta(\mathbf{q})$. While the bound state should lie below $2\Delta_0$, $\hbar\omega_0$ may depend on the detailed band structure and the coupling constant between AFM and SC. For Bi₂Sr₂CaCu₂O_{8+ δ} ARPES and STM measurements are consistent with $\Delta_0 \approx 39$ meV [27] while $\hbar\omega_0^{(o)} = 42$ meV and $\hbar\omega_0^{(e)} = 55$ meV for odd and even parity excitations of

the SC bilayer [28] so that on average $(\hbar\omega_0/2\Delta_0) = 0.62$. Recent tunnelling experiments for CeCoIn₅ indicate $\Delta_0 = 0.46$ meV so that $(\hbar\omega_0/2\Delta_0) = 0.65$, though we note that considerable debate on the tunnelling spectrum in CeCoIn₅ remains unresolved [23–25]. The corresponding numbers for UPd₂Al₃ are $\Delta_0 = 0.235$ meV [29] and $\hbar\omega_0 = 0.35$ meV [22] so that $(\hbar\omega_0/2\Delta_0) = 0.74$. The apparent similarity of $(\hbar\omega_0/2\Delta_0)$ over this series of different *d*-wave SC contrasts with $(2\Delta_0/k_B T_c)$, which is 2.8, 4.6, and 6.5, respectively, for UPd₂Al₃, CeCoIn₅, and Bi₂Sr₂CaCu₂O_{8+ δ} .

The wave vector dependence of the spin resonance may be affected by band structure directly as well as RKKY exchange. Indeed both commensurate and incommensurate spin correlations are observed in compounds closely related to CeCoIn₅. The dynamic spin correlation length inferred from the width of constant energy scans is largely unaffected by the transition to SC (see Fig. 2) and short ranged. It is remarkable that the resonance peak remains well-defined in the absence of long-ranged spatial correlations. This fact distinguishes CeCoIn₅ from UPd₂Al₃ which has long range spin order but is similar to the cuprates where a resonance is observed in the absence of long-ranged spin correlations.

It is interesting to contrast CeCoIn₅ with other magnetic SC where a spin resonance has been sought but not found. In Sr₂RuO₄ (0.7 K $< T_c < 1.4$ K) funneling data indicate $2\Delta_0 = 2.2$ meV and the present interpretation of a range of data indicate a triplet *p*-wave SC. For a triplet SC a resonance is expected without a sign change in the SC gap function however for $\hbar\omega > 0.4$ meV magnetic neutron scattering at $\mathbf{Q} = (0.7, 0.3, 0)$ is unaffected by cooling into the SC state [30]. A multiband model where the nesting bands that contribute to the spin susceptibility are only tangentially involved in SC may provide a resolution [31,32]. Early experiments in UPt₃ ($T_c = 0.5$ K) also detected no changes in magnetic neutron scattering for $\hbar\omega > 0.1$ meV [33]. Experiments and theory indicate that the SC gap in UPt₃ has line nodes in the basal plane and point nodes along the sixfold axis; however, there is no final resolution regarding the symmetry of the order parameter or the role of magnetic fluctuations in stabilizing it. The apparent absence of a resonance in SC UPt₃ should be reexamined.

We have investigated low-energy magnetic fluctuations in CeCoIn₅ using neutron inelastic scattering. A strong spin resonance with an energy of $\hbar\omega_0 = 0.60 \pm 0.03$ meV is observed to develop at low temperatures gathering spectral weight from low-energies. The results indicate strong coupling between spin fluctuations and a *d*-wave superconductivity. In addition our analysis of the *T*-dependent spin fluctuation spectrum indicates a large reduction in the RKKY spin exchange energy for $T < T_c$

the relation of which to the development of *d*-wave superconductivity remains to be understood.

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