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The magnitude of the magnetic exchange interaction in the heavy fermion antiferromagnet CeRhIn₅

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We have used high-resolution neutron spectroscopy experiments to determine the complete spin wave spectrum of the heavy fermion antiferromagnet CeRhIn₅. The spin wave dispersion can be quantitatively reproduced with a simple frustrated J_1 - J_2 model that also naturally explains the magnetic spin-spiral ground state of CeRhIn₅ and yields a dominant in-plane nearest-neighbor magnetic exchange constant $J_0 = 0.74(3)$ meV. Our results pave the way to a quantitative understanding of the rich low-temperature phase diagram of the prominent CeTIn₅ ($T = \text{Co, Rh, Ir}$) class of heavy fermion materials.

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The strength of magnetic interactions in heavy fermion (HF) materials represents a key parameter that controls the delicate interplay of localized and itinerant electronic degrees of freedom, which drives a multitude of fascinating strongly correlated electron phenomena in these materials [1, 2]. This competition between localized f electrons of Ce, Pr, Yb or various actinide elements on one hand and the conduction electrons on the other, is qualitatively described by the Doniach phase diagram [3]: On one end the magnetic Ruderman-Kittel-Kasay-Yosida (RKKY) interaction couples neighboring local magnetic moments associated with the f -electrons via the conduction electrons and leads to the development of long-range antiferromagnetic (AFM) order. In the other limit, the Kondo interaction [4] drives the local demagnetization of the f -electron state that is quenched by the spins of the surrounding conduction electrons. This results in delocalization of the f -electrons into the conduction band and thus the formation of a paramagnetic heavy Fermi liquid state [5].

Frequently the most interesting situation occurs near the boundary between the AFM and the demagnetized heavy Fermi liquid state at which Kondo and RKKY interactions mutually cancel [3]. At this point the ordering temperature is suppressed to zero resulting in a quantum critical point (QCP) at which the associated long-wavelength quantum critical magnetic fluctuations are believed to lead to the emergence of novel states of matter [1, 2], with the most prominent one being unconventional superconductivity [6, 7]. This mechanism is also relevant for other classes of unconventional superconductors such as the cuprates or iron-pnictides [8, 9].

Apart from this prototypical QCP scenario in which the AFM order parameter becomes critical, more recently so-called local quantum criticality has been invoked theoretically. In the latter, the local Kondo effect becomes critical at a QCP resulting in local instead of long-wavelength magnetic fluctuations [10]. It is further proposed that the position of the QCP may be controlled via the amount of magnetic frustration in a material [11, 12]. Because HF materials are usually not geometrically frustrated the magnetic frustration would likely originate from a frustration between nearest and further-

neighbor magnetic interactions, highlighting that quantitative knowledge of the magnetic interactions is critical for HF compounds.

Despite the key role of magnetic interactions for the understanding of HF physics, so far little attention has been devoted to determine them quantitatively in HF materials, and few neutron spectroscopy studies reporting spin wave dispersions have been carried out so far [13–17]. Here we report an extensive neutron spectroscopy study carried out on the HF antiferromagnet CeRhIn₅ that reveals the strength of the RKKY interaction in this system. CeRhIn₅ belongs to the intensively studied family of HF materials CeTIn₅ ($T = \text{Co, Rh, Ir}$) for which external pressure P , magnetic field H , and chemical substitution have been demonstrated to tune the ratio between RKKY and Kondo interactions, in turn allowing to access multiple AFM and superconducting phases, as well as various QCPs [18].

While our study has important implications for the understanding of HF materials in general, its aim is to shed more light on the complex interplay between antiferromagnetism, unconventional superconductivity and quantum criticality in CeRhIn₅ by probing the strength of the magnetic exchange interactions. Below $T_N = 3.8$ K, CeRhIn₅ exhibits incommensurate AFM order with a propagation vector $\mathbf{k} = (0.5 \ 0.5 \ 0.297)$ [19]. The AFM order can be suppressed via the application of pressure leading to a QCP at $P_c = 23$ kbar around which a broad superconducting dome emerges with a maximal $T_c = 2.3$ K [20, 21]. While the recent report of a spin resonance in the superconducting state of CeCoIn₅ illustrates that magnetic fluctuations are relevant for the superconductivity observed in the CeTIn₅ family [22], transport [21] as well as de Haas-van Alphen measurements [23] suggest that other critical degrees of freedom such as charge fluctuations may also be present at the QCP.

To determine the size of the magnetic exchange interaction we have measured the complete spin wave spectrum of CeRhIn₅ using the Cold Neutron Chopper Spectrometer (CNCS) at the Spallation Neutron Source [24]. To overcome the issues related to the high neutron absorption of both Rh

FIG. 1. A map of the magnetic elastic scattering in CeRhIn₅ at a temperature $T = 2.5$ K. Reference data obtained at $T = 20$ K was subtracted from the data (see text). A cut along $(-0.5 -0.5 l)$ shows clear satellite peaks that can be indexed with the propagation vector $\mathbf{k} = (0.5 \ 0.5 \ 0.297)$ (inset).

and In, large single crystals of CeRhIn₅ of typical sizes of $10 \times 10 \times 5$ mm³ were grown via the In self-flux method, and cut and polished to an average thickness of 0.6 mm along the crystallographic c axis to optimize the neutron transmission for cold neutrons. The intensity was further maximized by co-aligning 14 crystals that were fixed on a thin aluminum plate to cover the entire incident neutron beam cross-section of 50×15 mm² at the sample position, resulting in a total sample mass of 2.2 g. The mosaic spread of the sample was verified by x-ray diffraction and was found to be 1.5° illustrating the excellent quality of the mosaic assembly. The sample was aligned with the (110) and (001) reciprocal lattice directions in the scattering plane, which in combination with the detector tubes at CNCS that cover a vertical range of $\pm 15^\circ$ perpendicular to the scattering plane, enabled us to probe the spin wave dispersion along the three main high-symmetry directions ($h00$), ($hh0$) and ($00l$). All measurements reported were carried out at a temperature of 2.5 K and with an incident energy $E_i = 3.315$ meV providing an energy resolution $\Delta E \approx 80 \mu\text{eV}$ at the elastic line. From all plots shown here a reference data set recorded at $T = 20$ K was subtracted to highlight the scattering from magnetic correlations. $T = 20$ K was chosen because the quasielastic magnetic fluctuations in CeRhIn₅ extend to about $3T_N$ [25]. The details of the subtraction are provided in the Supplemental Material [26].

Fig. 1 shows a map of the magnetic elastic scattering as observed in the scattering plane with clear satellite peaks at $\tau \pm \mathbf{k}$ (where τ is a reciprocal lattice vector) that have been previously shown to correspond to an AFM spin-spiral propagating along the crystallographic c -axis [19]. The inset shows a cut along the $(-0.5 -0.5 l)$ direction revealing sharp Bragg peaks with an average FWHM of $0.025 \text{ \AA}^{-1}$ confirming the excellent quality of the assembled sample mosaic. We also note that the background is flat and zero demonstrating that subtracting the $T = 20$ K reference data works reliably. In order to maximize the neutron intensity, all inelastic measurements were carried out around the magnetic zone center $(-0.5 -0.5 \ 0.297)$, where the magnetic form factor for Ce³⁺

is large. This additionally allowed us to keep the angle θ between the crystallographic c -axis and the incident beam as low as possible to limit neutron absorption due to the increased path length of neutrons traversing the samples for higher θ .

Panels (a)-(c) of Fig. 2 illustrate the full spin wave spectrum observed in CeRhIn₅ as measured along the three principal directions ($h00$), ($hh0$) and ($00l$), respectively. The salient features that can be derived from the data without theoretical assumptions are: (i) The bandwidth of the spin wave spectrum is approximately 1.8 meV. While the data shown here only cover energy transfers up to 2.5 meV, we have performed additional measurements with $E_i = 12$ meV that demonstrate the absence of higher spin wave branches to at least 10 meV. (ii) The spin wave spectrum exhibits at least two distinct branches for the ($h00$) and ($hh0$) directions. (iii) There is a small spin wave gap Δ of about 0.25 meV.

In order to investigate the observed spin wave in detail we show constant momentum and energy transfer cuts for the ($h00$) direction in Figs. 3(a) and (b), respectively. For the energy scans in Fig. 3(a) the spin wave peaks were fitted using a Lorentzian line shape. The scan at the zone center (blue squares) clearly reveals a small spin wave gap $\Delta = 0.25(3)$ meV. Further, as indicated by the three arrows, there are actually not two but three distinct spin wave branches, which is in agreement with a model for the spin waves as demonstrated below. For higher momentum transfers (red circles in Fig. 3(a)) the two lower branches overlap and cannot be resolved with the resolution of this experiment. The peaks in the momentum transfer scans in Fig. 3(b) were fit with a Gaussian line shape. Similar cuts were also performed for the ($hh0$) symmetry direction, and the peak positions extracted from all cuts are shown as symbols in panels (d)-(e) of Fig. 2 overlaid on the theoretical calculation of the full spin wave spectrum. We note that for the ($00l$) direction the resolution available at CNCS is not sufficient to perform such fits.

We now turn to the calculation of the spin wave spectrum illustrated in Fig. 2. The interaction between the localized Ce magnetic moments is mediated by the conduction electrons, and therefore is described by an RKKY exchange interaction $J(\mathbf{q})$. As illustrated in Fig. 1 and 2, the magnetic scattering is maximal at the magnetic zone center $\mathbf{q} = \mathbf{k} = (0.5 \ 0.5 \ 0.297)$. The associated spin spiral ground state configuration can be reproduced with the following minimal J_1 - J_2 model, for which we employ a total of three magnetic exchange constants as illustrated in Fig. 2(g): $J_0 > 0$ is the intralayer nearest-neighbor (NN) antiferromagnetic interaction, J_1 is the interlayer NN interaction, and J_2 is the interlayer next NN interaction. We therefore obtain the following model Hamiltonian

$$\mathcal{H} = \sum_{i < j} J_{ij} [(S_{x,i} S_{x,j} + S_{y,i} S_{y,j}) + \delta S_{z,i} S_{z,j}], \quad (1)$$

with $J(\mathbf{q}) = 2J_0[\cos(2\pi q_x) + \cos(2\pi q_y)] + 2J_1 \cos(2\pi q_z) + 2J_2 \cos(4\pi q_z)$. Here $\mathbf{S}_i$ is a spin 1/2 operator representing the effective magnetic moment of the Γ_7^2 doublet. Because the energy gap to the first excited doublet ($\Delta_{\Gamma_7^1} \simeq 7\text{meV}$)

FIG. 2. The measured spin wave spectrum compared to calculations based on a simple J_1 - J_2 model that also describes the antiferromagnetic spin-spiral ground state of CeRhIn₅ (see text). Panels (a)-(c) show the experimental spin wave spectrum obtained at a temperature $T = 2.5$ K for the three principal directions $(h00)$, $(hh0)$ and $(00l)$, respectively. $T = 20$ K reference data was subtracted from each set (see text). The red, green and black solid lines and symbols denote the three branches $\omega_0(\mathbf{q})$, $\omega_0(\mathbf{q} + \mathbf{k})$, and $\omega_0(\mathbf{q} - \mathbf{k})$ of the dispersion corresponding to our spin wave model (see text for details) and have been fitted to our data. Panels (d)-(f) show calculations of the full magnetic susceptibility for the same directions as the data presented in (a)-(c) (see text). The circles correspond to the spin wave peak positions extracted from momentum and energy transfer scans as the ones shown in Fig. 3. In (g) we denote the three magnetic exchange constants used for our calculation.

is much bigger than the exchange constants J_{ij} [29], we can restrict our model to the lowest-energy doublet. Because the observed magnetic moments rotate in the tetragonal basal plane of CeRhIn₅ [19], we assume an easy-plane exchange anisotropy: $\delta < 1$. Further, the exchange interaction is frustrated along the c -axis because $J_2 > 0$, as confirmed by the observed period of the spiral ground state. The ratio of J_1 and J_2 is then fixed through the component of the magnetic propagation vector along the c -axis: $\cos(2\pi k_z) = -J_1/4J_2$ [30], yielding $J_2/J_1 = 0.809$.

The in-plane and out-of-plane magnetic susceptibility for the spin Hamiltonian (1) are (see Supplemental Material [26]),

$$\chi_{\nu\nu}(\mathbf{q}) = -\frac{S^2}{4\hbar^2} \left[\frac{\delta J(\mathbf{q} - \mathbf{k}) - J(\mathbf{k})}{(\omega + i\gamma)^2 - \omega_0^2(\mathbf{q} - \mathbf{k})} + \frac{\delta J(\mathbf{q} + \mathbf{k}) - J(\mathbf{k})}{(\omega + i\gamma)^2 - \omega_0^2(\mathbf{q} + \mathbf{k})} \right], \quad (2a)$$

$$\chi_{zz}(\mathbf{q}) = \frac{S^2[-A(\mathbf{q}) + J(\mathbf{k})]}{\hbar^2[(\omega + i\gamma)^2 - \omega_0^2(\mathbf{q})]}, \quad (2b)$$

with $\nu = x, y$ and

$$\hbar\omega_0(\mathbf{q}) = S\sqrt{[-J(\mathbf{k}) + \delta J(\mathbf{q})][-J(\mathbf{k}) + A(\mathbf{q})]}, \quad (3)$$

where $A(\mathbf{q}) = [J(\mathbf{q} + \mathbf{k}) + J(\mathbf{q} - \mathbf{k})]/2$. Here we have introduced a phenomenological damping constant γ to account for the damping of spin wave by magnon-electron and magnon-magnon interactions. The spin wave dispersion is obtained from the poles of the magnetic susceptibility.

Finally, due to the small size of the magnetic Brillouin zone along the c -direction, Umklapp scattering occurs at the zone boundary, resulting in two additional spin wave branches $\omega_0(\mathbf{q} + \mathbf{k})$, and $\omega_0(\mathbf{q} - \mathbf{k})$, respectively. In principle, one expects additional branches at $\omega_0(\mathbf{q} \pm m\mathbf{k})$ with $m = 0, 1, 2, 3, \dots$, however, higher order Umklapp scattering is suppressed by the translational symmetry of the incommensurate magnetic spiral [31]: any propagation of the helix by multiples of $\mathbf{k}$ can be compensated by changing the phase of the spiral by 2π . This is consistent with our data, as we indeed observe three distinct spin wave branches (Figs. 2 and 3).

The parameters $J_0 = 0.74(3)$ meV, $J_1 = 0.09(2)$ meV and $\delta = 0.82(2)$ were determined by fitting the dispersion (open symbols in Fig. 2(d) and (e)) to Eq. 3. Introducing a small, but non-resolution limited spin wave damping of $\hbar\gamma = 0.15$ meV, additionally allows us to explain the full spin wave spectrum of CeRhIn₅ (Eq. 2) as demonstrated in Fig. 2. We note, that a small discrepancy exists between this model and our data, which is the observed spin wave gap Δ , that we have accounted for by shifting the calculated dispersion by Δ . A gap is in principle not expected for an incommensurate spin spiral, which should always exhibit a gapless phason mode (Goldstone mode) due to the translational invariance [31]. However, the presence of the gap is indeed consistent with specific heat measurements [32]. The existence of a gap Δ due to defects is discussed in the Supplemental Material [26].

We will now discuss the implications of our results for understanding the phase diagram of CeRhIn₅. A mean-field calculation based on the RKKY exchange interaction deter-

FIG. 3. Constant momentum (a) and energy transfer (b) cuts for the $(h00)$ direction. The intensity of the elastic line in (a) is scaled by a factor of 30. The different scans are shifted vertically for clarity. The arrows denote the various spin wave branches extracted from fits (solid lines, see text for details).

mined in the experiments here yields an antiferromagnetic ordering temperature $T_N^{\text{MF}} = J(\mathbf{k})S(S+1)/3 \simeq 9.1$ K. Considering that thermal fluctuations [25] tend to suppress the mean field ordering temperature by a factor of two for a quasi two-dimensional system like CeRhIn_5 ($J_1/J_0 \sim 0.13$) [33], this is consistent with the experimental value of $T_N = 3.8$ K. We can compare this energy with the single-ion Kondo temperature $T_K = 0.15$ K that has been determined from $\text{Ce}_{1-x}\text{La}_x\text{RhIn}_5$ with dilute amounts of Ce [34]. Thus, at ambient pressure, the RKKY interaction is the dominant energy scale, and CeRhIn_5 exhibits well-localized f -electrons with local magnetic moments. This observation is consistent with de Haas-van Alphen measurements [23] that describe a small Fermi surface that does not include f -electrons. However, T_K estimated via NMR measurements is 5 K [35], and the real value of the Kondo scale is likely in between those extremes. Further high-resolution neutron spectroscopy measurements above T_N will be required to determine T_K unambiguously.

It is also useful to compare our spin wave data on CeRhIn_5 with the related compounds in the family $\text{Ce}_m\text{T}_n\text{In}_{3m+2n}$. Measurements on the cubic material CeIn_3 ($m = 1, n = 0$) reveal a NN exchange interaction $J_0 = 0.35$ meV along the cubic diagonals [15]. The increased superconducting critical temperature T_c in CeRhIn_5 ($m = 1, n = 1$) compared with its cubic building block CeIn_3 ($T_c = 0.2$ K under pressure) is believed to be due to the reduced dimensionality that occurs because the CeIn_3 conducting layers are separated by an TIn_2 layer in the CeTIn_5 family. Here we find that this reduced dimensionality is also reflected in the magnetic interactions, as the NN interaction J_1 along the c -axis is almost 10-fold reduced compared to the NN interaction J_0 within the tetragonal basal plane. In contrast, this trend does not continue for CePt_2In_7 ($m = 1, n = 2$), which exhibits a $T_c = 2.1$ K (under pressure) [36]. Thus, it would be useful to determine the exchange interactions for this compound, and verify if they exhibit less of a two-dimensional character. We note, that for HF systems the magnitude of the effective exchange constants can not easily be compared between two compounds, because they are sensitive towards the degree of f -electron delocalization controlled by the Kondo interaction.

While our results were obtained at ambient pressure they also allow us some insight into the unconventional superconducting state of CeRhIn_5 under pressure. In the case that bosonic degrees of freedom such as the spin waves observed here would be involved in mediating the superconductivity in CeRhIn_5 , it is expected that the coupling between the electronic quasiparticles and the spin waves is retarded [37]. The Fermi velocity of CeRhIn_5 in the superconducting state under pressure was estimated as $v_F \approx 39$ meVÅ [38]. We can use the measured spin wave dispersion to estimate the spin velocity in the tetragonal basal plane as $v_{\text{SW}} = 8(1)$ meVÅ, demonstrating that the magnetic excitations are indeed retarded with respect to the electronic quasiparticles. Notably, the retardation is of similar magnitude as the values identified for CeCu_2Si_2 , for which the superconductivity is believed to be mediated by magnetic fluctuations [17], suggesting that the spin excitations in CeRhIn_5 are a possible candidate for a superconducting glue.

Finally, we note that the frustrated exchange interactions identified in CeRhIn_5 will allow us to study new interesting physics such as micro-phases and competing magnetic structures and their interplay with strongly correlated electron phenomena such as unconventional superconductivity [39]. They further pave the way to quantitatively investigate the recently proposed model for HF materials that attempts to capture the physics of local and conventional quantum criticality in a global zero-temperature phase diagram [11, 12]. Notably, the theory suggest that the global phase diagram may be controlled via two distinct tuning parameters, where the first one is determined via the competition between RKKY and Kondo interactions, and the latter is the amount of magnetic frustration. Careful inelastic neutron scattering measurements in CeRhIn_5 as function of various tuning parameters such as pressure, magnetic field or chemical substitution will allow the characterisation of the amount of frustration throughout the phase diagram. This proposal of a global HF phase diagram currently attracts a lot of interest because it goes beyond the conventional Landau framework of order-parameter fluctuations by considering electron delocalization effects that are at the heart of HF physics and strongly correlated electron materials in general.

In conclusion, we have quantitatively determined the magnitude of the magnetic exchange interactions in CeRhIn_5 , which is a member of the prototypical family CeTIn_5 of HF compounds. This first microscopic measurement of the magnetic exchange interactions in CeRhIn_5 is an important step towards a quantitative understanding of the complex phase diagrams observed in the CeTIn_5 family and related compounds, and will be a key parameter for testing the validity of current and future theories.

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