

Evidence for a hybridization gap in noncentrosymmetric CeRuSi₃M. Smidman,¹ D. T. Adroja,^{2,3,*} E. A. Goremychkin,² M. R. Lees,¹ D. M. Paul,¹ and G. Balakrishnan^{1,†}¹*Department of Physics, University of Warwick, Coventry CV4 7AL, United Kingdom*²*ISIS Facility, STFC, Rutherford Appleton Laboratory, Chilton, Didcot, Oxfordshire OX11 0QX, United Kingdom*³*Highly Correlated Matter Research Group, Physics Department, University of Johannesburg, Auckland Park 2006, South Africa*

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Inelastic neutron scattering (INS) and specific heat measurements have been performed on the intermediate valence compound CeRuSi₃, which is isostructural to the noncentrosymmetric pressure-induced superconductors CeRhSi₃, CeIrSi₃, and CeCoGe₃. INS measurements at 7 K reveal a broad peak at (58.5 ± 1.4) meV, while at 300 K, broad quasielastic scattering is observed. This indicates a large Kondo temperature of $T_K \sim 680$ K. The magnetic contribution to the specific heat (C_{mag}) has a value of $\gamma = 62.5(1)$ mJ/mol K² at low temperatures and above about 100 K can be well accounted for by the Coqblin-Schrieffer model with a characteristic temperature of $T_0 = 680$ K, which is further evidence that CeRuSi₃ is in the intermediate valence regime.

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I. INTRODUCTION

There has been considerable recent interest in the coexistence of superconductivity and magnetism in heavy fermion compounds. In particular, there are several heavy fermion systems where the electronic ground state can be tuned via a nonthermal parameter such as doping, pressure, or magnetic field [1]. These systems are often explained in the framework of the Doniach phase diagram [2], due to competition between the intersite Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction, which leads to magnetic ordering of the cerium $4f$ electrons and the onsite Kondo interaction, which favors a nonmagnetic ground state. In the region where magnetic order is suppressed, a superconducting dome is often observed [3]. Magnetic fluctuations are believed to play an important role in mediating the superconductivity in both heavy fermion and high-temperature superconductors.

Several members of the CeTX₃ (T = transition metal, X = Si or Ge) series display antiferromagnetic order but become superconducting at sufficiently large pressures. These compounds crystallize in the noncentrosymmetric, tetragonal BaNiSn₃ type structure (space group $I4mm$). Due to a lack of inversion symmetry and a finite antisymmetric spin-orbit coupling, the spin degeneracy of the conduction bands is lifted and the superconducting state is a mixture of spin singlet and triplet states [4,5]. For example, at ambient pressure CeRhSi₃ orders antiferromagnetically at 1.6 K but becomes superconducting for $p > 1.2$ GPa [6,7]. The superconducting state displays large and anisotropic upper critical fields, reaching up to 30 T for magnetic fields along the c axis [8].

It is therefore desirable to characterize CeTX₃ compounds at ambient pressure, to examine the role played by the competition between the RKKY and Kondo interactions in determining the ground state properties and to identify the proximity of the system to quantum criticality. However, several compounds such as CeFeGe₃ [10], CeRuSi₃ [11], and CeCoSi₃ are reported to have nonmagnetic ground states and a broad maximum is observed in the magnetic susceptibility

at around 50, 150, and 200 K respectively. In these systems, the hybridization between the $4f$ and conduction electrons is believed to be strong enough that the Kondo interaction is dominant and magnetic order is entirely suppressed. Within the Kondo lattice model, strong hybridization leads to the formation of renormalized bands and a hybridization gap, shown schematically in Fig. 1 [9,12]. If the Fermi level lies within the gap, insulating behavior in the resistivity is observed, while if it lies on top of the lower hybridized band, the system is metallic despite the presence of a gap. For sufficiently strong hybridization, the system would be expected to enter the intermediate valence regime and this has been observed at low temperatures in resonant inverse photoemission spectroscopy measurements of the CeCoGe_{1-x}Si_x system [13].

Although inelastic neutron scattering (INS) investigations have been performed on several CeTX₃ compounds for (T = Co, Rh, or Ir; X = Si or Ge) [14–16], as far as we are aware there are no reports of such measurements on isostructural CeRuSi₃. The aim of the present work is to investigate the magnetic INS response of CeRuSi₃ to probe the nature of the $4f$ electrons; that is, whether they display localized or itinerant behavior. The existence of a hybridization gap at low temperatures is deduced and this is studied using INS and specific heat measurements. Furthermore, this study will provide an ideal comparison with existing INS data of CeTX₃ compounds, and characterizing the strength of the hybridization between the $4f$ and conduction electrons may allow a greater understanding of the role of this hybridization in determining the nature of the ground state.

II. EXPERIMENTAL DETAILS

Polycrystalline samples of CeRuSi₃ and LaRuSi₃ were prepared by arc-melting stoichiometric quantities of the constituent elements on a water cooled copper hearth, in an argon atmosphere. The samples were flipped and melted several times to improve homogeneity and then wrapped in Ta foil, sealed in evacuated quartz tubes and annealed at 900 °C for two weeks. Powder x-ray diffraction measurements were performed using a Bruker D5005 diffractometer. Lattice parameters of $a = 4.2106(2)$ and $4.2597(3)$ Å, and $c = 9.9204(7)$ and $9.9382(9)$ Å, were obtained for CeRuSi₃

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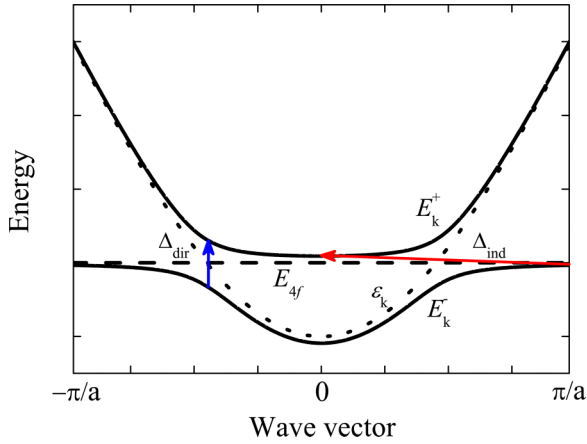


FIG. 1. (Color online) A schematic diagram of the hybridized bands E_k^+ and E_k^- of the Kondo lattice which result from the hybridization of the conduction and 4f electrons, following Ref. [9]. The unhybridized conduction (ϵ_k) and 4f (E_{4f}) bands are shown by the dashed and dotted lines respectively. A finite hybridization opens a gap between the bands and the direct (Δ_{dir}) and indirect (Δ_{ind}) gaps are indicated by arrows.

and LaRuSi₃ respectively. A small number of impurity peaks are present in the patterns of both compounds, the largest of which has an intensity of $\sim 4\%$ of the largest sample peak. Inelastic neutron scattering measurements were performed on the MERLIN time-of-flight spectrometer in the ISIS facility at the Rutherford Appleton Laboratory, U.K. The samples were wrapped in Al foil and cooled to 7 K in a closed cycle refrigerator in He-exchange gas, to thermalize the sample temperature. Measurements were made at 7 and 300 K with $E_i = 30$ and 200 meV selected via a Fermi chopper. Specific heat measurements were performed between 1.8 and 350 K using the two-tau relaxation method with a Quantum Design Physical Property Measurement System (PPMS).

III. RESULTS AND DISCUSSION

A. Inelastic neutron scattering

Color-coded plots of the INS intensity [$S(Q, \omega)$] in absolute units of $\text{mbsr}^{-1}\text{meV}^{-1}\text{f.u.}^{-1}$ (f.u. = formula units) for $E_i = 200$ meV are shown in Fig. 2. The scattering of CeRuSi₃ and LaRuSi₃ at 7 K are shown in Figs. 2(a) and 2(b) respectively. Extra intensity at low $|Q|$ can be identified in the CeRuSi₃ plot, which is evidence for a magnetic contribution to the scattering. Although this is most intense at around 50 meV, it extends up close to 100 meV, whereas the scattering is negligible for LaRuSi₃ at these energy transfers, apart from at high $|Q|$. The scattering of LaRuSi₃ at 300 K [Fig. 2(d)] is similar to that observed at low temperatures. However, although magnetic scattering can still be identified in the measurements of CeRuSi₃ at 300 K [Fig. 2(c)], it has shifted to lower energies. The low $|Q|$ scattering is significantly reduced above 50 meV, suggesting a change in behavior with increasing temperature.

Cuts of $S(Q, \omega)$ were made by integrating across low and high values of $|Q|$. Low $|Q|$ cuts are displayed for $E_i = 30$ and 200 meV in Figs 3(a) and 3(b) respectively and the

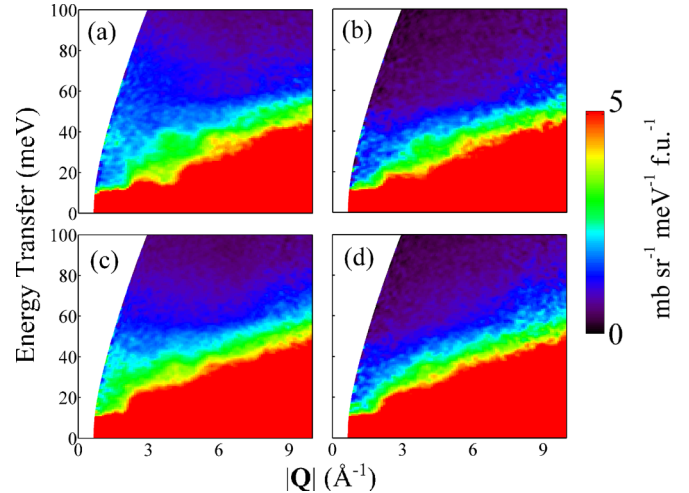


FIG. 2. (Color online) Color-coded plots of the INS intensity with $E_i = 200$ meV for (a) CeRuSi₃ at 7 K, (b) LaRuSi₃ at 7 K, (c) CeRuSi₃ at 300 K, and (d) LaRuSi₃ at 300 K.

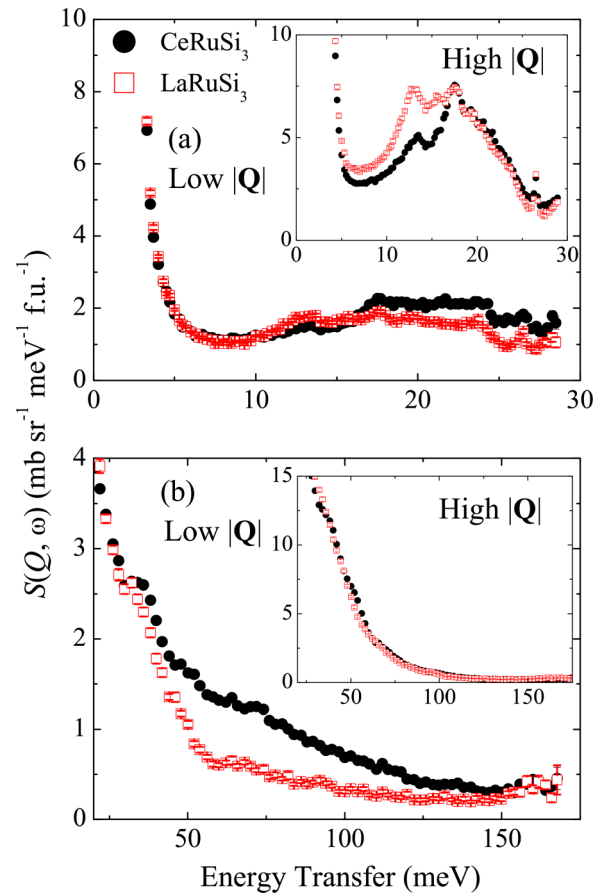


FIG. 3. (Color online) Low $|Q|$ cuts of $S(Q, \omega)$ for CeRuSi₃ and LaRuSi₃ at 7 K with incident energies of (a) 30 meV and (b) 200 meV, with the corresponding high $|Q|$ cuts shown in the insets. The low $|Q|$ cuts were integrated over $0-4 \text{ \AA}^{-1}$ and $0-6 \text{ \AA}^{-1}$, while the high $|Q|$ cuts were integrated over $4-7 \text{ \AA}^{-1}$ and $12-18 \text{ \AA}^{-1}$ for $E_i = 30$ and 200 meV respectively.

corresponding high $|\mathbf{Q}|$ cuts are shown in the insets. In Fig. 3(a), it can be seen that for energy transfers greater than about 17 meV, the low $|\mathbf{Q}|$ scattering of CeRuSi₃ is greater than that of LaRuSi₃, whereas the high $|\mathbf{Q}|$ scattering is similar for both compounds. Similarly, the low $|\mathbf{Q}|$ scattering of CeRuSi₃ in Fig. 3(b) is greater than that of LaRuSi₃ for energy transfers greater than 35 meV, up to at least 150 meV. This is strong evidence for the presence of a magnetic contribution to the low $|\mathbf{Q}|$ scattering of CeRuSi₃, which is not observed at high $|\mathbf{Q}|$. This is because the magnetic intensity is proportional to the square of the magnetic form factor $[F(\mathbf{Q})]$, which is maximum at $|\mathbf{Q}| = 0$ and has a negligible contribution for $|\mathbf{Q}| > 10 \text{ \AA}^{-1}$. The high $|\mathbf{Q}|$ scattering is similar in both compounds, which is expected since at large momentum transfers, $S(Q, \omega)$ is dominated by phonon scattering, which increases as Q^2 . An exception to this is at energy transfers between 5 and 15 meV, displayed in the inset of Fig. 3(a). Although both plots of the high $|\mathbf{Q}|$ scattering display a peak at around 13 meV, the LaRuSi₃ data is of a greater intensity at these energy transfers. This is likely because the coherent scattering cross section of La is approximately three times that of Ce and this indicates that the phonon scattering in this region is dominated by the rare-earth atoms.

The magnetic contribution to the scattering $[S_{\text{mag}}(Q, \omega)]$ was obtained by subtracting an estimate of the phonon scattering. This is commonly estimated by directly subtracting the spectra of the nonmagnetic reference compound from that of the magnetic compound. However, this method does not adequately remove the contributions from phonon scattering. Instead, the ratio between the high ($S_{\text{ph}}^{\text{hQ}}$) and low ($S_{\text{ph}}^{\text{lQ}}$) $|\mathbf{Q}|$ cuts was calculated for the reference compound LaRuSi₃. If the phonon scattering of the magnetic scattering scales similarly with $|\mathbf{Q}|$, the magnetic scattering can be estimated from the low $|\mathbf{Q}|$ ($S_{\text{CeRuSi}_3}^{\text{lQ}}$) and high $|\mathbf{Q}|$ ($S_{\text{CeRuSi}_3}^{\text{hQ}}$) scattering using

$$S_{\text{mag}} = S_{\text{CeRuSi}_3}^{\text{lQ}} - \frac{S_{\text{ph}}^{\text{lQ}}}{S_{\text{ph}}^{\text{hQ}}} S_{\text{CeRuSi}_3}^{\text{hQ}}. \quad (1)$$

$S_{\text{mag}}(Q, \omega)$ is displayed in Fig. 4 for $E_i = 30 \text{ meV}$. At 300 K a broad quasielastic response is observed, whereas there is no quasielastic scattering at 7 K and the magnetic scattering is

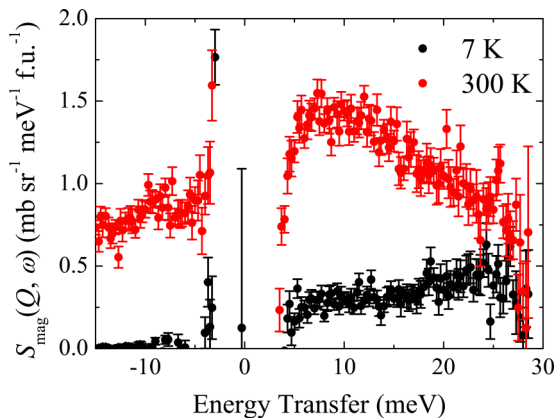


FIG. 4. (Color online) Magnetic scattering of CeRuSi₃ at 7 and 300 K for $E_i = 30 \text{ meV}$, estimated using Eq. (1).

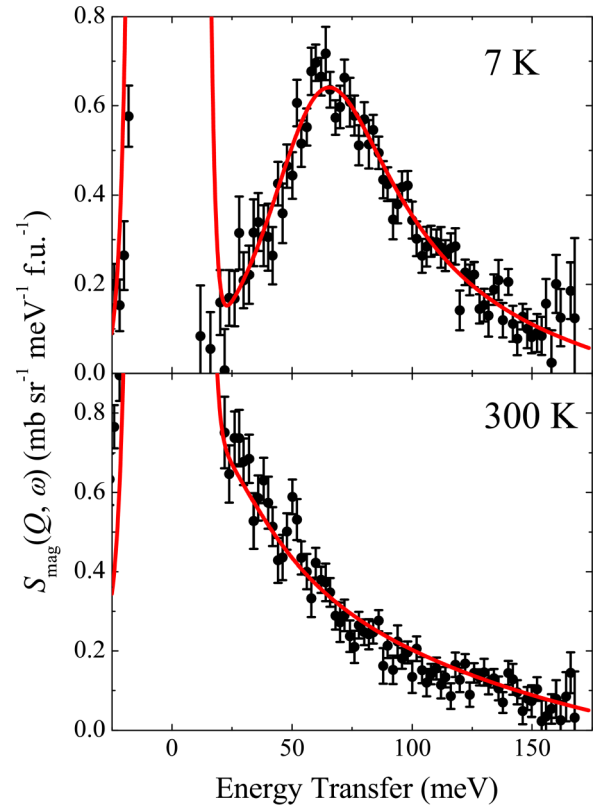


FIG. 5. (Color online) Magnetic scattering of CeRuSi₃ at 7 and 300 K for $E_i = 200 \text{ meV}$, estimated using Eq. (1).

greatly reduced. At 7 K, the magnetic scattering gradually increases with increasing energy transfers up to at least 25 meV and this may be the tail of an excitation centered at higher energies. The magnetic scattering for $E_i = 200 \text{ meV}$ is displayed in Fig. 5, where broad quasielastic scattering is also observed at 300 K. However at 7 K, the magnetic response has shifted to a broad peak centered at energy transfers of around 60 meV. The scattering at 7 K was fitted with a Lorentzian function convoluted with the instrument resolution. A peak center of $(58.5 \pm 1.4) \text{ meV}$ with a linewidth (half-width at half maximum) of $(31.9 \pm 1.3) \text{ meV}$ was obtained. Using the second sum rule $\int S_{\text{mag}}(\omega)/F^2(Q)d\omega = 48.8\mu_{\text{eff}}^2$, an effective moment of $\mu_{\text{eff}} = 1.49\mu_B$ is calculated. Although this is lower than the expected value of $2.54\mu_B$, this result indicates the bulk origin of the magnetic scattering and this suggests there may be an additional magnetic response at energy transfers greater than 170 meV. For the 300 K data, the quasielastic scattering was fitted with a Lorentzian function, centered on the position of the elastic line. A linewidth of $30(3) \text{ meV}$ is obtained, which is similar to the width of the inelastic excitation at 7 K. The observed INS response of an inelastic peak at low temperatures and quasielastic scattering at high temperatures is typical behavior for intermediate valence compounds [12]. The lack of both quasielastic scattering at low temperatures and well defined crystalline electric field (CEF) excitations is also expected for these materials. The position of the inelastic peak indicates the scale of the Kondo temperature [17], giving $T_K \sim 680 \text{ K}$. This is smaller than that of the isostructural, intermediate valence compound CeCoSi₃,

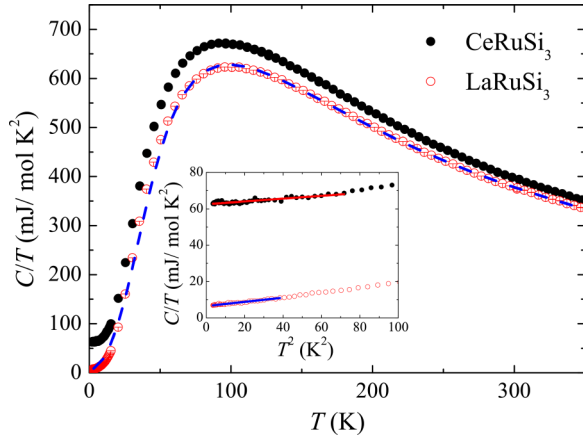


FIG. 6. (Color online) Temperature dependence of the specific heat (C) of CeRuSi₃ and LaRuSi₃. The dashed line shows a fit to the specific heat of LaRuSi₃ described in the text. The inset shows plots of C/T against T^2 with linear fits for both compounds.

where $T_K \sim 990(50)$ K is estimated from a peak in the magnetic INS scattering at 85 meV [16], while T_K of the order of 900 K is estimated from specific heat and magnetic susceptibility measurements [18]. However, T_K is larger than for the magnetically ordered CeRhSi₃ and CeIrSi₃, which has been estimated to be around 100 K for both compounds [6]. It should also be noted that the threshold for the onset of magnetic scattering is predicted to be 0.8 times the peak energy [19], but no clear threshold is observed. The increase of the magnetic scattering at around 20 meV in Fig. 4 suggests that scattering from the low energy tail of the peak near 60 meV is present down to at least these energy transfers.

B. Specific heat

The specific heats of CeRuSi₃ and LaRuSi₃ are displayed in Fig. 6. The specific heat of CeRuSi₃ is consistently larger than that of LaRuSi₃, indicating the presence of a magnetic contribution. As shown in the inset, the residual value of C/T at zero temperature is larger in CeRuSi₃. A linear fit to C/T against T^2 gives $\gamma = 62.5(1)$ mJ/mol K² for CeRuSi₃, while a smaller value of $6.5(1)$ mJ/mol K² is obtained for LaRuSi₃, indicating an enhancement of γ due to hybridization between $4f$ and conduction electrons.

The magnetic contribution to the specific heat from the $4f$ electrons (C_{mag}) was calculated by subtracting the specific heat of LaRuSi₃ by two methods. Firstly, the specific heat of LaRuSi₃ was directly subtracted from that of CeRuSi₃. Secondly, the specific heat of LaRuSi₃ was fitted with the sum of γT and an estimate of the phonon contribution (C_{ph}), where γ was fixed to the low temperature value. C_{ph} was fitted using the sum of a Debye and Einstein term [21]. A Debye temperature of $\theta_D = 314$ K and an Einstein temperature of $\theta_E = 569$ K were obtained, with a 73% contribution from the Debye term. This is displayed by the dashed line in Fig. 6 and shows that the specific heat of LaRuSi₃ can be accounted for with the sum of a phonon contribution and a linear electronic term. The two estimates of C_{mag} are displayed in Fig. 7. In both plots, C_{mag} displays a broad peak at around 200 K. An additional feature is observed at around 50–60 K. In the

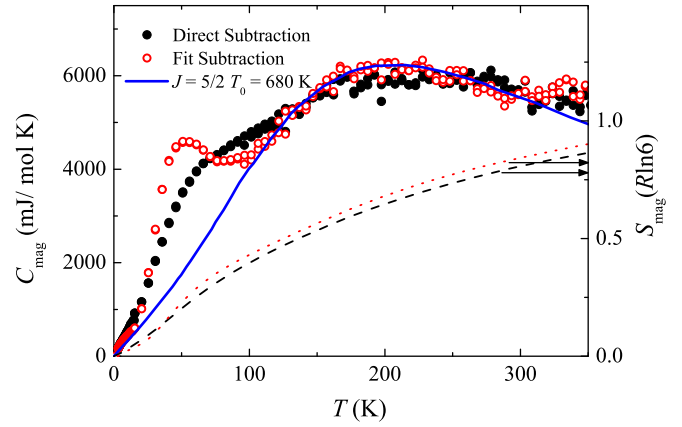


FIG. 7. (Color online) Magnetic contribution to the specific heat (C_{mag}) of CeRuSi₃ estimated by a direct subtraction of the specific heat of LaRuSi₃ and of a fit to the LaRuSi₃ data described in the text. The solid line shows the specific heat of Coqblin-Schrieffer model for $J = \frac{5}{2}$ from Ref. [20], with a characteristic temperature of $T_0 = 680$ K. The temperature dependence of the entropy in units of $R \ln 6$ are shown by the dashed and dotted lines for the direct and fit subtractions respectively.

estimate from the fit subtraction, there is a peak in this region while in the direct subtraction, there is a clear shoulder. The dotted and dashed lines show the temperature dependence of the entropy (S_{mag}) in units of $R \ln(6)$, obtained from integrating C_{mag}/T from the fit and direct subtractions respectively. The entropy reaches $0.9R \ln(6)$ and $0.87R \ln(6)$ at 350 K for the respective plots.

The solid line shows the Coqblin-Schrieffer model for $J = \frac{5}{2}$ as calculated by Rajan in Ref. [20]. The one adjustable parameter in the model is the characteristic temperature T_0 , which was fixed to 680 K, which corresponds to the energy of the INS peak position. The model accounts well for the data at high temperatures. However, below temperatures of about 100 K, there is poor agreement with the data and there is an additional contribution at low temperatures, including the feature at around 50–60 K. This suggests there may be an additional low energy scale in the specific heat. It might be expected that this could lead to two gaps being observed in INS measurements, as is the case for CeFe₂Al₁₀ [22]. If two gaps are present in CeRuSi₃, the weak rise in magnetic scattering above 5 meV at 7 K for $E_1 = 30$ meV shown in Fig. 4 may be due to transitions across the lower energy gap. It would also be interesting to investigate the gap structure of CeRuSi₃ with optical conductivity measurements, which would provide information on the size of the direct or charge gap. The ratio of the charge to the spin gap can provide important information on the dimensionality of the magnetic interactions [12].

IV. CONCLUSIONS

INS and specific heat measurements have been performed on CeRuSi₃ and LaRuSi₃ and the results are consistent with CeRuSi₃ being an intermediate valence compound. The INS measurements of CeRuSi₃ at 7 K indicate the lack of both quasielastic scattering and narrow excitations associated with transitions between CEF levels. Instead a broad inelastic peak

is observed, centered at (58.5 ± 1.4) meV with a linewidth of (31.9 ± 1.3) meV. The opening of such a hybridization gap in the inelastic response which transforms to quasielastic scattering at higher temperatures, indicates that CeRuSi₃ is an intermediate valence compound with $T_K \sim 680$ K.

The magnetic contribution to the specific heat shows a broad peak at around 200 K, and above temperatures of around 100 K, there is good agreement with the Coqblin-Schrieffer model, with $T_0 = 680$ K. However, at lower temperatures there is an additional contribution to the specific heat, which may indicate the presence of a lower energy scale in the compound. The value of γ is $62.5(1)$ mJ/mol K² for CeRuSi₃ compared to $6.5(1)$ mJ/mol K² for LaRuSi₃. This is a moderate enhancement due to electronic correlations and is comparable to the values observed for the isostructural CeRhGe₃ [15] and CeCoGe₃ [23]. Both CeRhGe₃ and CeCoGe₃ order antiferromagnetically and INS measurements reveal well defined CEF levels. The different ground state behavior of CeRuSi₃ results from the larger value of T_K , which is an order of magnitude smaller, at around 10 K in the aforementioned ordered compounds [14,15]. The value of γ is also larger than that observed in CeCoSi₃ of 37 mJ/mol K² and is similar to CeCoGe_{0.75}Si_{2.25} [18], which is in closer proximity to quantum criticality.

There is particular interest in determining whether the properties of intermediate valence compounds can be accounted

for by the single-ion, Anderson impurity model or whether coherence effects arising from the regular arrangement of rare-earth ions needs to be taken into account. In the case of the latter, the system is described by the Anderson lattice model and at low temperatures, renormalized bands with a hybridization gap and large effective masses are expected to emerge. A \mathbf{Q} dependence of the magnetic scattering would be strong evidence for coherent behavior, as observed in CePd₃ [24]. It would therefore be interesting to study the INS response of single crystals of CeRuSi₃ and to compare the results to both the Anderson impurity and Anderson lattice models.

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