Coexistence of metallic and nonmetallic properties in the pyrochlore Lu$_2$Rh$_2$O$_7$

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Transition metal oxides of the 4d and 5d block have recently become the targets of materials discovery, largely due to their strong spin–orbit coupling that can generate exotic magnetic and electronic states. Here, we report the high-pressure synthesis of Lu$_2$Rh$_2$O$_7$, a new cubic pyrochlore oxide based on 4d$^5$Rh$^{4+}$, and characterizations via thermodynamic, electrical transport, and muon spin relaxation measurements. Magnetic susceptibility measurements reveal a large temperature-independent Pauli paramagnetic contribution, while heat capacity shows an enhanced Sommerfeld coefficient, $\gamma = 21.8(1)$ mJ/mol-Rh K$^{-2}$. Muon spin relaxation measurements confirm that Lu$_2$Rh$_2$O$_7$ remains paramagnetic down to 2 K. Taken in combination, these three measurements suggest that Lu$_2$Rh$_2$O$_7$ is a correlated paramagnetic metal with a Wilson ratio of $R_W = 2.5$. However, electric transport measurements present a striking contradiction as the resistivity of Lu$_2$Rh$_2$O$_7$ is observed to monotonically increase with decreasing temperature, indicative of a nonmetallic state. Furthermore, although the magnitude of the resistivity is that of a semiconductor, the temperature dependence does not obey any conventional form. Thus, we propose that Lu$_2$Rh$_2$O$_7$ may belong to the same novel class of non-Fermi liquids as the nonmetallic metal FeCrAs.

npj Quantum Materials (2019) 4:9; https://doi.org/10.1038/s41535-019-0148-1

INTRODUCTION

The electronic and magnetic properties of many transition metal oxides can be understood by considering three energy scales: (i) the on-site Coulomb repulsion, Hubbard $U$, (ii) the band width, $W$, and (iii) the spin–orbit coupling, $\lambda$. The strong-coupling limit of this paradigm occurs for 3d oxides where Hubbard repulsion dominates ($U/W \gg 1$) and spin–orbit coupling ($\lambda \propto Z^3$) is negligible. As a consequence, Mott insulating states are observed in many materials that one would expect to be metals from band theory, as exemplified by the parent compounds of the high-temperature cuprate superconductors. The opposing limit occurs for 5d oxides, where the d electron orbitals are far more spatially delocalized such that $U$ decreases while $W$ increases ($U/W \approx 1$). At the same time, the large mass enhancement brings spin–orbit coupling to the forefront, giving rise to remarkable correlated states such as topological insulators, Weyl semi-metals, and spin–orbit assisted Mott insulators. Intermediate to these two limits is the 4d oxides, where $U$, $W$, and $\lambda$ can all be comparable in magnitude.

The chemically versatile pyrochlore oxides, $A_2B_2O_7$, are an excellent family of materials to explore the competition between $U$, $W$, and $\lambda$. In this structure, the $A$ site is usually occupied by a rare earth cation, while the $B$ site can be occupied by transition or post-transition metals from the 3d, 4d, or 5d block. Consequently, a wide assortment of electronic behaviors are observed, ranging from band insulators to correlated metals. Pyrochlores are also of significant interest for their magnetic properties. The corner-sharing tetrahedral arrangement of the $A$ and $B$ cation sublattices is prone to intense geometric frustration, which has been extensively studied in the insulating families. The metallic pyrochlore oxides provide an intriguing platform for studying the interplay of geometric frustration and metallic degrees of freedom, an emerging topic that has begun to garner significant attention. Whereas frustration in local moment systems can straightforwardly be identified via the so-called frustration index, $f = |\langle \lambda_{A\mu}\rangle|$. No equivalent metric has yet been established for itinerant magnetic materials. Hence, how to unambiguously identify the presence of frustration in an itinerant system remains an open problem. A recent study broached this topic by using inelastic neutron scattering to extract exchange couplings which were then compared to a local moment picture.

In the rare earth pyrochlores, the 4f electrons from the $A$-site cation are highly localized and do not participate in electrical conduction. Thus, the electronic properties of pyrochlore materials are largely governed by the choice of $B$-site cation. Insulating states are expected and observed in cases where there are no partially filled d-electron orbitals, such as when $B = \text{Tl}^{4+}$, $\text{Ge}^{4+}$, $\text{Sn}^{4+}$, and $\text{Zr}^{4+}$. Due to their dominant Hubbard $U$, insulating or semiconducting states are also found in the majority of cases where $B$ has a partially occupied 3d shell, such as $B = \text{Mn}^{4+}$ and $\text{Cr}^{4+}$. Semiconducting states are found for a number of pyrochlores with partially filled 4d or 5d-shells, such as those...
with $B = \text{Mo}^{4+}, \text{Ru}^{4+},$ and $\text{Pt}^{4+}$. Pyrochlores with metallic states are fewer in number, with notable examples being found amongst the $B = \text{O}^{3+}, \text{Re}^{5+},$ and $\text{Ir}^{4+}$ families. In these systems, large spin–orbit coupling results in fascinating correlated electron phenomena. Meanwhile, $\text{Cd}_2\text{Re}_2\text{O}_7$ is a moderately heavy fermion metal, while 4$d^5$ Re$^{4+}$ has been previously reported,19,20 their characterization has been complicated by the difficulties in stabilizing rhodium in its 4$+$ oxidation state, which we overcome using high-pressure methods. In the pyrochlore lattice, Rh$^{4+}$ sits at the center of a trigonally distorted octahedral oxygen environment with $D_{3d}$ point group symmetry. As schematically shown in Fig. 1a, the crystal electric field and moderately strong spin–orbit coupling ($\lambda \approx 0.19$ eV for Rh in contrast with $\lambda \approx 0.40$ eV for Ir$^{4+}$) split the $d$ electron states into a filled $j_{\text{eff}} = 3/2$ band and a half-occupied $j_{\text{eff}} = 1/2$ band. Thus, akin to the 5$d^5$ iridates,24 the 4$d^4$ rhodates can have a $j_{\text{eff}} = 1/2$ magnetic degree of freedom, leading to a spin–orbit induced Mott insulating state.23 Both theoretical calculations and direct spectroscopic measurements have confirmed the applicability of the $j_{\text{eff}} = 1/2$ picture in other rhodates.

![Fig. 1](image-url)

**Fig. 1** a The 4$d^4$ Rh$^{4+}$ cations in Lu$_2$Rh$_2$O$_7$ sit at the center of a trigonally distorted ($D_{3d}$) octahedral oxygen environment. Strong spin–orbit coupling will further split the $t_{2g}$ states into a filled $j_{\text{eff}} = 3/2$ and a half-filled $j_{\text{eff}} = 1/2$ state; however, this limit may not be fully reached for Rh$^{4+}$ with moderate spin–orbit coupling. b Powder x-ray diffraction pattern and Rietveld refinement for Lu$_2$Rh$_2$O$_7$ measured at $T = 300$ K with a copper $K_{\alpha 1}$ wavelength ($\lambda = 1.5406$ Å). The Bragg peak positions for the $Fd\bar{3}m$ pyrochlore phase are indicated by the upper set of crosses. The lower set of crosses are from a minor RhO$_2$ impurity accounting for 2% of the sample volume. The results of the refinement are shown in Table 1.

### RESULTS

Under strong oxidizing conditions, Lu$_2$Rh$_2$O$_7$ crystallizes in the pyrochlore structure (space group $Fd\bar{3}m$). The powder x-ray diffraction pattern, which is presented in Fig. 1b, was analyzed via Rietveld refinement, giving a cubic lattice parameter of $a = 10.0265$ (1) Å. The ratio of the ionic radii for Lu$^{3+}$ (0.136 nm) and Rh$^{4+}$ (0.121 nm) is within the stability range for the pyrochlore structure. Thus, significant levels of A/B site mixing are neither expected nor observed. The only adjustable coordinate within the pyrochlore structure is the O2$_x$ position, which we refine to $x = 0.3296(5)$. When $x = 0.3125$, the oxygen environment at the $B$-site is an ideal octahedron (point group $O_h$). Deviations from this value induce a compression along the local [111] axis, as shown in Fig. 1a, such that the six equidistant oxygen ions have alternating bond angles of 93.5$^\circ$ and 86.5$^\circ$ with the central Rh$^{4+}$ cation (point group $D_{3d}$). A small RhO$_2$ impurity, comprising 2.1(2)% of the total sample volume, was included as a second phase in the Rietveld refinements (a tetragonal structure with space group $P4_2/mnm$). RhO$_2$ is a high-pressure metallic oxide phase which is insoluble in aqua regia and could not be removed from our samples. The results of the Rietveld refinement, including the goodness-of-fit parameters, are listed in Table 1.

The magnetic susceptibility of Lu$_2$Rh$_2$O$_7$ measured in an $H = 1$ T field is presented in Fig. 2. We have corrected the data for core diamagnetism using the values tabulated in ref. 31, which gives $\chi_0 = -1.54 \times 10^{-8}$ emu/F.U. There is no appreciable difference between data collected under zero-field cooled or field cooled conditions. Lu$_2$Rh$_2$O$_7$ is paramagnetic down to the lowest measured temperature, with no evidence for magnetic ordering or spin freezing down to 2 K. The susceptibility is dominated by a large nearly temperature independent Pauli paramagnetic contribution. Below 10 K, the intrinsic susceptibility is obscured by a Curie-tail. We observe a shallow, broad hump in the susceptibility centered near 50 K. As can be seen in Fig. 2, the inverse susceptibility between 100 and 300 K is reasonably well-described by a Curie–Weiss equation, $\chi = \frac{C}{T - \theta}$, where we obtain $\theta = -1393(2)$ K and $\chi_0 = \sqrt{4C} = 3.1(1)$ μemu/F.U. Clearly, this magnitude of $\theta$ is not physically meaningful as the susceptibility is dominated by the Pauli paramagnetic contribution. However, attempts to model the susceptibility to a modified Curie–Weiss equation failed.

### Table 1. Refined lattice parameters and temperature factors for Lu$_2$Rh$_2$O$_7$ where the goodness of fit parameters are $R_b = 7.34, R_{wp} = 10.9, \omega_R = 6.27$ and $\chi^2 = 3.02$

| Site | $x$ | $y$ | $z$ | $\theta_{iso}$ | $\beta_{iso}$ |
|------|-----|-----|-----|----------------|---------------|
| Lu   | 0.5 | 0.5 | 0.5 | 0.52(4)        | 16d           |
| Rh   | 0   | 0   | 0   | 0.28(5)        | 16c           |
| O1   | 0.375 | 0.375 | 0.375 | 0.42(2)        | 8b            |
| O2   | 0.3296(5) | 0.125 | 0.125 | 0.42(2)        | 48f           |
law, \( \chi = \chi_0 + \chi_0' \), failed to produce a unique solution. Instead, we proceed by subtracting the Curie-tail contribution, which we fit to \( \chi \propto T \), allowing us to estimate the low-temperature Pauli paramagnetic contribution as \( \chi_0' = 7.6 \cdot 10^{-4} \text{ emu/mol-Rh} \). This value is nearly an order of magnitude larger than in typical metals, signaling strong electron correlation effects. The induced magnetization at 2 K and 7 T is only 0.01 \( \mu_\text{B/Rh} \), consistent with an itinerant paramagnetic state (inset of Fig. 2). The susceptibility and magnetization data for Lu2Rh2O7 bear a striking resemblance to that of SrRhO3, another Rh\(^{4+}\)-based paramagnetic metal.\(^{31}\) The unusual properties of SrRhO3 have been argued to emerge as the result of proximity to a quantum critical point.\(^{33}\)

The heat capacity of Lu2Rh2O7 is presented in Fig. 3, where no phase transitions are observed between 2 and 300 K. For comparison, we plot alongside this data the scaled heat capacity of Lu2Ti2O7, which is an insulating lattice analog with no unpaired \( d \) electrons.\(^{34}\) The heat capacity of Lu2Ti2O7 was scaled by \( \mu = \sqrt{M_{\text{LRO}}/M_{\text{LTO}}} \) where \( M_{\text{LRO}} \) and \( M_{\text{LTO}} \) are the molecular weights of Lu2Rh2O7 and Lu2Ti2O7, respectively. These two data sets are largely consistent, confirming that the high-temperature heat capacity of Lu2Rh2O7 can be accounted for by lattice degrees of freedom. The scaled heat capacity of Lu2Ti2O7 is observed to slightly exceed that of Lu2Rh2O7 below 50 K. This difference can be understood to originate from anharmonic effects, which are known to be significant in rare earth pyrochlores,\(^{35}\) and will vary between compounds dependent on the details of the phonon interactions. Indeed, the heat capacity of Lu2Ti2O7 is also observed to exceed that of Dy2Ti2O7 in the same temperature interval.\(^{36}\) At the lowest temperatures, Fermi liquid behavior is observed for Lu2Rh2O7, where \( C/T \) is linear with respect to \( T^2 \) up to \( T = 10 \) K (inset of Fig. 3). The fitted value for the Sommerfeld coefficient is \( \gamma = 21.8(1) \text{ mJ/mol-Rh K}^2 \), which is large and comparable to values observed in other strongly correlated rhodates and ruthenates.\(^{37-39}\)

![Fig. 2](image2.png)

**Fig. 2** The magnetic susceptibility of Lu2Rh2O7, which was measured at \( H = 1 \) T, is dominated by a largely temperature independent Pauli paramagnetic contribution. The full symbols and the left hand axis are the susceptibility, \( \chi \), while the open symbols and the right hand axis give the inverse susceptibility, \( \chi^{-1} \). The yellow dashed line is the as-measured susceptibility data, prior to the Curie tail subtraction. The red line is a fit to the Curie–Weiss equation. The inset shows magnetization measurements at 2 and 25 K.

![Fig. 3](image3.png)

**Fig. 3** The heat capacity of Lu2Rh2O7 measured in zero field, with the scaled heat capacity of Lu2Ti2O7 from ref. \(^{34}\) shown for reference. The inset shows a fit of \( C/T = \gamma + \beta T^2 \) over the shaded region, which yields a large Sommerfeld coefficient, \( \gamma = 21.8(1) \text{ mJ/mol-Rh K}^2 \).
Lu$_2$Rh$_2$O$_7$, which was annealed in air at 600 °C for 10 h to minimize conventional four-terminal method on a cold-pressed pellet of evidence for magnetic order or spin freezing down to 2 K. may share an origin with the broad hump in the susceptibility in signal is dominated by slowing spin dynamics. The decrease in $\rho / T$ vs. $T^{-1}$ fitted relaxation rate, $\lambda$, and the stretching parameter, $\beta$, reveal weak spin dynamics and a crossover from more Gaussian-like relaxation to Lorentzian relaxation fluctuating nuclear dipoles whereas at lower temperatures the signal is dominated by slowing spin dynamics. The decrease in $\beta$ may share an origin with the broad hump in the susceptibility in the same temperature interval. Moreover, these $\mu$SR measurements confirm that Lu$_2$Rh$_2$O$_7$ remains paramagnetic, with no evidence for magnetic order or spin freezing down to 2 K.

Electrical resistivity measurements were carried out using a conventional four-terminal method on a cold-pressed pellet of Lu$_2$Rh$_2$O$_7$, which was annealed in air at 600 °C for 10 h to minimize grain boundary effects. X-ray diffraction measurements were performed before and after the annealing protocol to confirm that it did not result in any degradation of the sample. In striking contrast to the magnetic susceptibility and heat capacity, which were measured in numerous polycrystalline samples of rhodates synthesized via the same high-pressure techniques used here. It is also worth mentioning that our minor impurity phase, RhO$_2$, is known to be a metal. Thus, the nonmetallic resistivity is clearly intrinsic to Lu$_2$Rh$_2$O$_7$, a remarkable result in a material that all other probes suggest is metallic.

**Fig. 4**  
(a) Several representative asymmetry spectra from muon spin relaxation measurements on Lu$_2$Rh$_2$O$_7$. Error bars are derived from $\sqrt{N}$ counting statistics. The solid lines are fits to a stretched exponential function. (b) The temperature dependence of the fitted relaxation rate, $\lambda$, and the stretching parameter, $\beta$, reveal weak spin dynamics and a crossover from more Gaussian-like relaxation to Lorentzian relaxation $(-\Delta/2k_B T)$, yielding a very small gap of $\Delta = 37(1)$ meV. There are several factors worth considering in the quantitative interpretation of this data. First is that this sample is a pressed pellet and thus, scattering from grain boundaries may be expected to uniformly increase the resistivity. However, the cold-pressing and annealing protocol followed here has been demonstrated to provide nearly intrinsic transport measurements in other transition metal oxides, including samples that are far softer and more porous than the one measured here. Moreover, even without the cold-pressing technique, conventional metallic resistivity (i.e., $d\rho/dT > 0$) has been measured in numerous polycrystalline samples of rhodates synthesized via the same high-pressure techniques used here. It is also worth mentioning that our minor impurity phase, RhO$_2$, is known to be a metal. Thus, the nonmetallic resistivity is clearly intrinsic to Lu$_2$Rh$_2$O$_7$, a remarkable result in a material that all other probes suggest is metallic.

**Fig. 5** The electrical resistivity of Lu$_2$Rh$_2$O$_7$ is typical of a semiconductor in magnitude but with an abnormal temperature dependence. The inset shows $\ln(\rho)$ vs. $T^{-1}$, where the red line indicates a fit to thermally activated Arrhenius behavior yielding a gap of $\Delta = 37(1)$ meV.

Our characterizations have revealed that Lu$_2$Rh$_2$O$_7$ possesses a highly unusual non-Fermi liquid state. While both heat capacity and magnetic susceptibility indicate a finite density of states at the Fermi energy, the temperature dependence of the resistivity appears more consistent with a semiconducting gap. Moreover, even for a semiconductor, the temperature dependence of the resistivity is unconventional. It is instructive to consider a comparison with the "nonmetallic metal" FeCrAs. First, an important structural commonality is that in both materials, the magnetic constituent(s) are connected in triangular motifs that can be prone to geometric magnetic frustration. In the case of Lu$_2$Rh$_2$O$_7$ this is the corner-sharing tetrahedral pyrochlore sublattice and in the case of FeCrAs the non-magnetic Fe atoms occupy a triangular network of trimers while the magnetic Cr atoms form a distorted Kagome network. In regards to their electronic properties, the magnetic susceptibility of both materials is dominated by a largely temperature independent Pauli paramagnetic contribution. Secondly, both materials have a Sommerfeld coefficient, as determined by heat capacity measurements, significantly larger than typically expected for a $d$ electron system, $\gamma = 21.8(1)$ mJ/mol-Rh K$^2$ and 31.6 mJ/mol K$^2$ for Lu$_2$Rh$_2$O$_7$ and FeCrAs, respectively. However, the Wilson ratio $R_w = 4\pi^2 k_B^2 T / (3 g\mu_B)^2$ for FeCrAs, $R_w = 4 - 5$ is larger than our computed value of $R_w = 2.5$ for Lu$_2$Rh$_2$O$_7$. A third commonality is the temperature dependence of the resistivity, where $d\rho/dT$
is negatively, nominally indicative of a gap in the density of states. However, the room temperature resistivity for FeCrAs is approximately 0.4 mΩ-cm, approximately three orders of magnitude smaller than our value of 0.4 Ω-cm. Another significant difference between these two materials is that the Cr moments in FeCrAs undergo a long-range antiferromagnetic ordering transition at $T_N = \text{125 K}$,27 while no magnetic order is observed in Lu$_2$Rh$_2$O$_7$ down to at least 2 K.

Several compelling explanations have been put forward to explain the nonmetallic metallic state in FeCrAs, which are worth examining in the current context of our results on Lu$_2$Rh$_2$O$_7$. The first proposal, which comes from Rau and Kee, suggests a hidden spin liquid state for the non-magnetic Fe trimers.51 They propose that the electrons residing on these trimers are close to a metal-insulator quantum critical point, such that charge fluctuations strongly renormalize the transport behavior while only minimally affecting the thermodynamic properties. A second proposal that does not rely on proximity to a Mott insulating state, but instead attributes the unusual qualities of FeCrAs to it being a Hund’s metal.52 In a Hund’s metal, non-Fermi liquid properties emerge due to strong intra-atomic exchange in a multiorbital material, such as ruthenates and iron pnictides.53 The result of strong Hund’s coupling in these materials is that a subset of the $d$ orbitals are pushed towards the local limit while maintaining strong coupling to other itinerant electrons. Several arguments make this second scenario less favorable in the case of Lu$_2$Rh$_2$O$_7$. First, Hund’s coupling generally plays a weaker role in 4$d$ $\text{Rh}^{4+}$.54 Secondly, this model is expected to give a large Wilson ratio,28 as indeed observed in FeCrAs where $R_W = 4–5$,27 whereas in Lu$_2$Rh$_2$O$_7$, $R_W = 2.5$. Recent inelastic neutron scattering experiments have revealed high energy itinerant-like spin excitations, which Plumb et al. propose may account for the nonmetallic resistivity.55 Further study of the spin dynamics in Lu$_2$Rh$_2$O$_7$ are called for, a task which would be greatly benefited by the availability of large single crystal samples. While no single crystals of rhodate pyrochlores have been reported to date, a route similar to that used for Sr$_3$RhO$_4$ may prove fruitful.38

Using high-pressure methods, we have synthesized a new pyrochlore Lu$_2$Rh$_2$O$_7$, in which $\text{Lu}^{3+}$ occupies a frustrated corner-sharing tetrahedral lattice. No magnetic order is observed down to 2 K in μSR measurements. Heat capacity and magnetic susceptibility measurements both indicate strong electron correlations, with a large temperature independent Pauli paramagnetic susceptibility of $\mu\text{B} / \text{mol Rh}^4\text{K}^2$. Resistivity measurements, in contrast, suggest nonmetallic behavior ($\rho / \partial T < 0$) in the full measured temperature range, 2–300 K. Thus, Lu$_2$Rh$_2$O$_7$ has a non-Fermi liquid state, sharing many commonalities with the nonmetallic metal FeCrAs.

**METHODS**

High-pressure synthetic techniques are often required to stabilize Rh$^{4+}$ compounds, as Rh$^{3+}$ is the more common oxidation state. The cubic pyrochlore Lu$_2$Rh$_2$O$_7$ was prepared using a belt-type high-pressure apparatus at the National Institute for Materials Science in Tsukuba, Japan. Lu$_2$O$_3$ was thoroughly dried prior to the reaction. Rhodium metal was first oxidized to Rh$_2$O$_3$ by reacting under flowing oxygen at 1000 °C for 72 h. The reaction into the pyrochlore phase was completed using K$_2$O as an oxidizing agent to promote the Rh$^{3+}$ oxidation state. The precursors (Lu$_2$O$_3$, Rh$_2$O$_3$, K$_2$O in a 1:1:3 ratio) were thoroughly ground together and sealed in a platinum capsule. The reaction was completed at 1700 °C and 6 GPa of applied pressure for 2 h, after which the sample was quenched to room temperature before the pressure was released. The final product was repeatedly washed with water to remove KCl and then dried. The resulting 0.5 g sample was made up of sub-micron sized black crystallites. Once prepared, this material is stable in air on a timescale of years.

X-ray diffraction measurements were performed using a PANalytical diffractometer with copper $K_{\alpha1}$ radiation, $\lambda = 1.540560$ Å. Rietveld refinement was performed using FullProf.55 Magnetic susceptibility, resistivity, and heat capacity measurements between 2 and 300 K were completed using a Quantum Design Physical Property Measurement System. Zero field μSR measurements were performed at the M20 surface muon channel at TRIUMF in the LAMPF Spectrometer. The μSR data were fitted over the full measured time interval, up to 9.5 μs, using the musrft software package.

**DATA AVAILABILITY**

The crystal structure of Lu$_2$Rh$_2$O$_7$, as determined by Rietveld refinement of the powder x-ray diffraction data, is available at the Cambridge Crystallographic Data Centre with deposition number CSD 1895045. All other data is available from the authors on reasonable request.

**ACKNOWLEDGEMENTS**

The authors gratefully acknowledge useful conversations with Yipeng Cai, Jonathan Gaudet, John Greedan, Chien-Lung Huang, and Murray Wilson. The authors thank Bassam Hitti and Gerald Morris from TRIUMF for their support with the μSR experiment. A.M.H. thanks the National Institute for Materials Science (NIMS) for their hospitality and support through the NIMS Internship Program. This work was supported by the Natural Sciences and Engineering Research Council of Canada (NSERC) and Canada Foundation for Innovation. C.R.W. is additionally supported by the Canada Research Chair (Tier II) program and the Canadian Institute for Advanced Research. Q.C. and H.D.Z. are supported by the National Science Foundation (NSF-DMR-1350002).

**AUTHOR CONTRIBUTIONS**

The high-pressure synthesis and x-ray diffraction were performed by A.M.H. and M.T. Magnetic susceptibility and electric resistivity measurements, and their analysis, were performed by Q.C., H.D.Z., C.M., P.M.S., J.P.A., A.M.H., and C.R.W. Heat capacity measurements were performed by A.Z.S. and C.R.W. μSR measurements were performed and analyzed by A.M.H. and G.M.L. with assistance from C.D. and Z.G. The manuscript was written by A.M.H. and C.R.W. with input from all authors.

**ADDITIONAL INFORMATION**

Competing interests: The authors declare no competing interests.

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