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$Sm_2Ru_3Sn_5$: A Non-centrosymmetric Cubic Member of the $Ln_2M_3X_5$ Family

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Abstract

We present an optimized synthetic method for $Sm_2Ru_3Sn_5$ and investigate its physical properties and electronic structure. $Sm_2Ru_3Sn_5$ was prepared by arc-melting stoichiometric ratios of the elements and confirmed by single crystal and powder X-ray diffraction. An antiferromagnetic transition was observed at T_N = 3.8 K. A modified Curie-Weiss fit to the data in the range 50 - 150 K yielded a Curie-Weiss temperature: θ_{CW} = -36.6 K and an effective magnetic moment: μ_{eff} = 0.83 μ_B, in agreement with a Sm^{3+} oxidation state. Field-dependent magnetization up to H = 7 T at 2 K showed a maximum response of 0.06 μ_B, significantly lower than the expected Sm^{3+} saturation moment (0.71 μ_B). Resistivity measurements indicate metallic behavior, and analysis of the magnetic entropy from the heat capacity revealed a doublet ground state due to crystal electric field splitting. The electronic structure and density of states were calculated with density function theory (DFT) and further supported by the local density approximation with dynamical mean-field theory (LDA+DMFT). Our experimental and computational results highlight localized Sm^{3+} moments and suggest a possible interplay between Ruddelman-Kitel-Kasuya-Yosida (RKKY) and Kondo interactions, positioning $Sm_2Ru_3Sn_5$ as a promising material for studying topology and complex physical phenomena.

Keywords: Intermetallic compounds, Transition metals, Materials science, Samarium, Lanthanides

1. Introduction

Intermetallic compounds with samarium (Sm) can exhibit a +2 or +3 oxidation state. Notable properties of Sm^{3+} compounds include the Kondo effect in $SmSn_3$,[1] heavy fermion behavior in Sm_3Te_4,[2] large magnetoresistance of over 100% at H = 9 T for $SmPd_2Ga_2$,[3] and multiple magnetic transitions are observed in Sm_2Sn_3.[4] A subset of Sm compounds are also recognized for their topology, where spin-orbit coupling (SOC) and *d-f* orbital hybridization can lead to nontrivial topological surface states. In SmB_6, the combination of topology and SOC creates a strong spin-orbit torque without exclusive surface conduction, a property that is intrinsic to topological insulators.[5-6] Another example is SmS which exhibits a pressure-induced valence instability, fluctuating from a Sm^{2+} to a Sm^{3+} state at 6.5 kbar.[7] The change in valence is accompanied by a first-order phase transformation, where the high pressure golden phase (*g*-SmS) has a topological band structure containing Dirac cones.[8] $SmCoIn_5$ is another example of an intermediate-valent compound, where antiferromagnetic ordering dominates and Kondo coherence is not observed; however, charge fluctuations from the crystal field highlight a competition between Kondo and Ruderman-Kittel-Kasuya-Yosida (RKKY) interactions.[9] Finally, a large topological Hall effect was observed in the Weyl semimetal SmAlSi which can be varied with magnetic field, supporting a Weyl-mediated, spiral magnetic ordering.[10] Our interest in finding a compound with high symmetry, topology, and SOC was influenced by the aforementioned examples.

In our 2023 survey of the $Ln_2M_3X_5$ compounds (Ln = lanthanide, or actinide; M = transition metal; X = Si, Ga, Ge, In, Sn), eight different structure types were identified.[11] Recently, $Ce_2Au_3In_5$ was identified as a Weyl-Kondo semimetal.[12] The authors predicted a number of strongly correlated topological semimetals and experimentally validated $Ce_2Au_3In_5$ (space group # 31, $Pmn2_1$). Of the $Ln_2M_3X_5$ family of compounds, consisting of over 230 analogues, at least 15 examples are reported to have polymorphism or structural phase transformations, particularly the analogues with Groups 8 and 9 transition metals. Our group has studied $Pr_2Co_3Ge_5$ which exhibits valence instability from nearly Pr^{4+} to Pr^{3+} upon a monoclinic to orthorhombic structural phase transition.[13] Another notable $Ln_2M_3X_5$ analogue is polymorphic $Sm_2Ru_3Ge_5$ that adopts both the tetragonal $Sc_2Fe_3Si_5$

and the orthorhombic $U_2Co_3Si_5$ structure types,[14-15] where each polymorph has been linked to charge density wave behavior.[15-16] While both polymorphs exhibit Sm^{3+} character, the magnetism of each manifests differently. The tetragonal polymorph exhibits a ferromagnetic transition at 7 K;[15] however, the orthorhombic polymorph of $Sm_2Ru_3Ge_5$ has an antiferromagnetic transition at 7 K.[17] Sm-containing compounds, particularly those adopting the $Ln_2M_3X_5$ structures, offer a rich system to study complex structural and magnetic phenomena. For these reasons, $Sm_2Ru_3Sn_5$ was chosen to be the focus of this study as a potential candidate for strongly correlated phenomena. While quite different from the other $Ln_2M_3X_5$ structure types, $Sm_2Ru_3Sn_5$ (*a* = 9.4606(8) Å; $I\bar{4}3m$)[18] adopts a structure that is a non-centrosymmetric variation from the binary cubic, centrosymmetric Ru_3Sn_7 structure (*a* = 9.332 Å; $Im\bar{3}m$).[19] Since a topological band structure has been established for Ru_3Sn_7 analogues,[20] an investigation into the noncentrosymmetric analogue could reveal new states that were unrealized in the centrosymmetric environment Furthermore, there is no understanding on the impact of *f*-elements to physical properties of the Ru_3Sn_7 structure type. Herein, we report an investigation of the synthesis, physical properties, and electronic structure of $Sm_2Ru_3Sn_5$ to understand the introduction of *f*-orbitals to the Ru_3Sn_7 structure type.

2. Results and Discussion

2.1. Synthesis

A previous synthesis of $Sm_2Ru_3Sn_5$ arc-melted and annealed samples at 600 °C for 30 days, where Ru and $SmRuSn_3$ impurities were observed in 15 wt% and 5 wt%, respectively.[18] Single crystals suitable for X-ray diffraction were isolated from the surface of the annealed samples. Similarly, we also synthesized $Sm_2Ru_3Sn_5$ by arc-melting stoichiometric amounts of Sm, Ru, and Sn. Annealing seemed to have no impact on the formation of $Sm_2Ru_3Sn_5$; however, an important experimental condition to the formation of $Sm_2Ru_3Sn_5$ was the particle size of Ru used in the experiment. Larger pieces of Ru introduced inhomogeneous regions to the boule, resulting in areas of different stoichiometry that contributed to the formation of byproducts. Ru powder was vital for consistent formation of $Sm_2Ru_3Sn_5$ without any byproducts. A small homogeneity range of formation was observed, and any deviation in stoichiometry from the 2:3:5 (Sm:Ru:Sn)

ratio results in the formation of Ru_2Sn_3 and $SmRuSn_3$ as impurities. Attempts to grow single crystals with the flux method from excess Sn led to the formation of Ru_3Sn_7 and $SmRuSn_3$, excess Ga or In flux led to the formation of both RuX_3 and SmX_2 (X = Ga, In), and excess Bi flux led to the formation of Ru_3Sn_7 and Ru_2Sn_3.

2.2. Crystal Structure

$Sm_2Ru_3Sn_5$ crystallizes in the cubic space group $I\bar{4}3m$ (No. 217) with room temperature unit cell parameter: ***a*** = 9.4575(5) Å. Details of the single crystal data collection and refinement parameters are outlined in Table 1. $Sm_2Ru_3Sn_5$ is an ordered noncentrosymmetric variant of the centrosymmetric Ru_3Sn_7 structure, consistent with the previously reported lattice parameter ***a*** = 9.4606(8) Å.[18] The Ru_3Sn_7 structure can be visualized as a collection of doubled, condensed square antiprisms that form interpenetrating frameworks (Figure 1a), consisting of three atomic positions: Ru1 (12***e***, 4 *m . m*), Sn1 (12***d***, $\bar{4}$ *m* . 2), and Sn2 (16***f***, . 3 *m*).[21-22] The doubled square antiprisms of Ru_3Sn_7 are connected by a Sn_8 cube with the Sn2 site at the vertices. To compare the Sn2 sites to the structure of $Sm_2Ru_3Sn_5$, the cube can also be described as two congruent, interpenetrating tetrahedra (Figure 1b). The crystal structure of $Sm_2Ru_3Sn_5$ consists of four atomic positions: Sm1 (8***c***, . 3 *m*), Ru1 (12***e***, 2 . *m m*), Sn1 (12***d***, $\bar{4}$ . .), and Sn2 (8***c***, . 3 *m*), where the fractional atomic coordinates are outlined in Table 2. In $Sm_2Ru_3Sn_5$, the tetrahedra are incongruent—one made from four Sn2 atoms and one made from four Sm1 atoms. This motif of two, interpenetrating Ln_4 and Sn_4 tetrahedra (Figure 1c) is also present in the $Ho_5Co_6Sn_{18}$, where the Ho tetrahedral sublattice is associated with geometric frustration, and the spins order noncollinearly.[23] While the distance between the Sm atoms in $Sm_2Ru_3Sn_5$ are too far to be considered within bonding distance ($d_{Sm\text{-}Sm}$ = 5.373(1) Å), it is comparable to the interatomic distance in the $Ho(2)_4$ tetrahedron ($d_{Ho\text{-}Ho}$ = 5.06 - 5.15 Å) for $Ho_5Co_6Sn_{18}$.

Table 1. Single Crystal Data Collection and Refinement Parameters for $Sm_2Ru_3Sn_5$

Formula	$Sm_2Ru_3Sn_5$	
Space Group	$I\bar{4}3m$	
Temperature (K)	298	160
a (Å)	9.4575(5)	9.4438(6)
V (Å^3)	845.92(13)	842.25(16)
Z	4	
θ (°)	3.1 - 30.5	
μ (mm^{-1})	33.23	33.37
Measured Reflections	19123	21240
Independent Reflections	272	272
R_{int}	0.073	0.082
$\Delta\rho_{max}$, $\Delta\rho_{min}$ (e$^-$/Å^3)	0.57, -0.62	0.89, -1.11
R [$F^2 > 2\sigma(F^2)$]	0.011	0.015
wR_2 (F^2)	0.024	0.030

$R = \sum\big||F_o| - |F_c|\big| / \sum |F_o|$

$wR_2 = \left(\sum[w(F_o^2 - F_c^2)^2] / \sum[w(F_o{}^2)^2]\right)^{1/2}$

Table 2. Fractional Atomic Coordinates and Displacement Parameters (T = 298 K)

Site	Wyckoff	*x*	*y*	*z*	U_{eq} (Å^2)
Sm1	8*c*	0.20088(4)	0.20088(4)	0.20088(4)	0.01085(15)
Ru1	12*e*	½	0.14990(8)	½	0.00727(16)
Sn1	12*d*	¼	0	½	0.00986(15)
Sn2	8*c*	0.38416(6)	0.38416(6)	0.38416(6)	0.00776(17)

Electron counting has been discussed in relation to the Ru_3Sn_7 structure type as a way to rationalize the chemical composition of substituted analogues.[21, 24-25] For $Sm_2Ru_3Sn_5$ and Ru_3Sn_7, the valence electron concentrations are 50 (5 e$^-$/atom) and 52 (5.2 e$^-$/atom), respectively. While VEC is a useful tool to identify trends within a given structure type or class of intermetallics, normalization of VEC to the transition metal sites offers insight into the bonding.[25] When using the 18-*n*+*m* rule,[26] Ru_3Sn_7 has 17.33 electrons/Ru. Assuming Sm^{3+}, the VEC drops to 16.67 electrons/Ru in $Sm_2Ru_3Sn_5$. The reduced VEC implies either an increase in the Ru—Ru bonding within the structure (the *n* term), or a reduced Sn—Sn interaction (the *m* term). A change in the degree of Ru—Ru bonding can be ruled out, since there is a minimal contraction of 0.064(2) Å for the Ru—Ru contacts in $Sm_2Ru_3Sn_5$ (compared to Ru_3Sn_7). Coupled with this change (shown in Figure 1) is a

similar amount of expansion in the Sn2—Sn2 connectivity due to the replacement of half the Sn2 atoms (. 3 *m* site symmetry) in the Ru_3Sn_7 structure with Sm ($Sm_2Ru_3Sn_5$: d_{tet} = 3.100(6) Å; Ru_3Sn_7: d_{cube} = 3.020(1) Å). It is possible that the change in the *m* term is more heavily influenced by the introduction of Sm—Sn interactions ($d_{Sm1\text{-}Sn2}$ = 3.004(5) Å) between adjacent tetrahedra; however, deviations from the ideal VEC could also be explained by a high density of states at the Fermi level. To further understand the impact of this structural distortion on the chemistry of $Sm_2Ru_3Sn_5$, we investigated the physical properties and electronic structure.

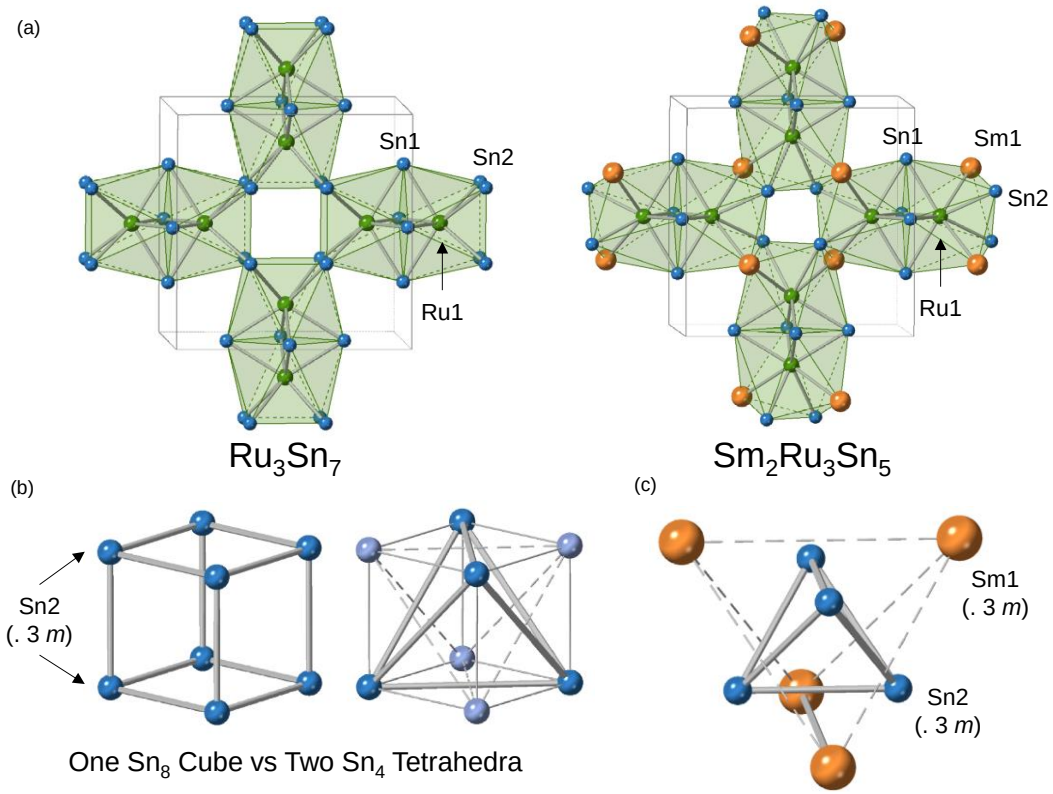

Figure 1. (a) Views of the face-sharing square antiprisms (Ru_3Sn_7) and distorted 8-vertex polyhedra ($Sm_2Ru_3Sn_5$). (b) Sn2 (16*f*) atomic sites in Ru_3Sn_7 can be visualized as either one Sn_8 cube or two Sn_4 Tetrahedra. (c) Sm1 (8*c*) and Sn2 (8*c*) atomic sites in $Sm_2Ru_3Sn_5$ that form two incongruent tetrahedra.

2.3. Physical Properties

Figure 2 shows the magnetic susceptibility and field-dependent magnetization measurements of $Sm_2Ru_3Sn_5$. An antiferromagnetic transition at T_N = 3.8 K is observed (shown in Figure 2a). The inverse susceptibility (Figure 2b) shows a slight curvature that

is consistent with a temperature independent magnetic term. The magnetic susceptibility data was fit from 50 - 150 K with a modified Curie-Weiss law: $\chi = \chi_0 + C/(T - \theta_{CW})$, where the temperature-independent term: $\chi_0 = 0.0026$, C is the Curie constant, and the Curie-Weiss temperature: $\theta_{CW} = -36.6$ K. The effective moment from the fit was 0.83 μ_B, consistent with the expected effective moment for Sm^{3+} ($\mu_{eff} = 0.84\ \mu_B$). In systems with strong spin-orbit coupling (SOC), the preferred alignment directions of spins can conflict with each other, causing the original Curie-Weiss law to break down from temperature-dependent local magnetic moments.[27] The combination of SOC and frustration could lead to fractionalization of fermionic excitations, known as a quantum spin liquid.[28] The modified Curie-Weiss law accounts for a more accurate θ_{CW} by introducing a constant to better fit temperature-independent contributions to the data. $SmPd_2Al_3$ is a compound with a strongly anisotropic magnetic response.[29] Subsequent magnetometry and neutron diffraction experiments illuminated a geometrically frustrated Sm lattice with

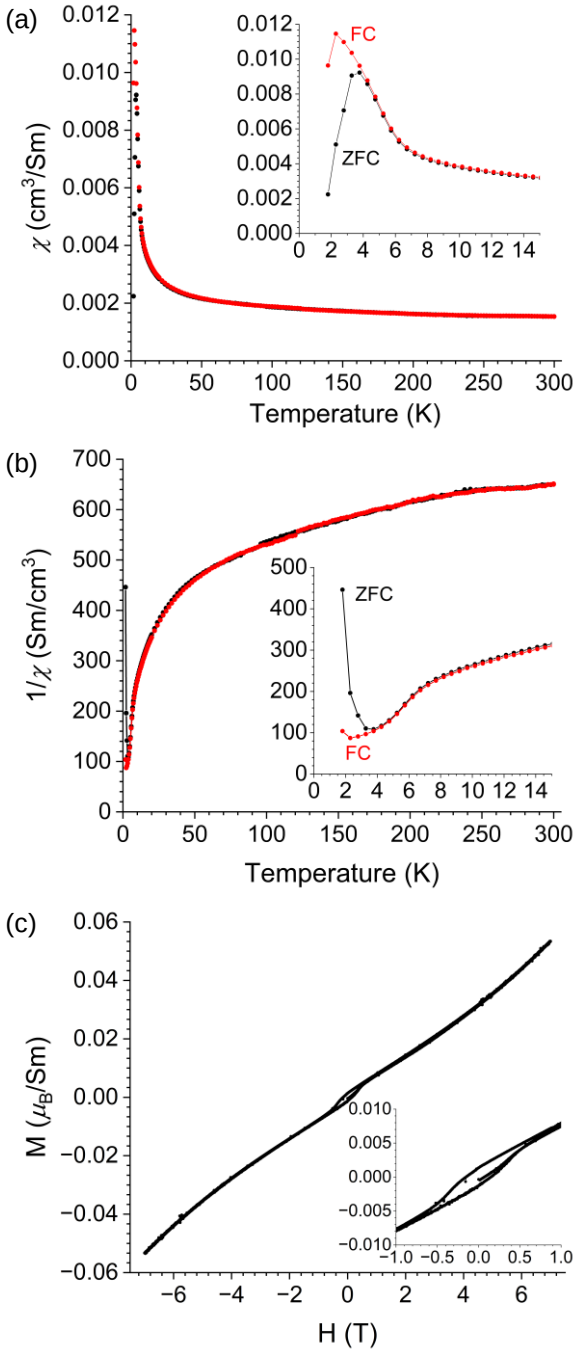

Figure 2. (a) The magnetic susceptibility for polycrystalline $Sm_2Ru_3Sn_5$ at H = 0.1 T. Inset shows the low temperature transition. The red points represent the field cooled (FC) experiment, and the black points represent the zero field cooled (ZFC) experiment. (b) The inverse of the magnetic susceptibility of $Sm_2Ru_3Sn_5$. (c) The field-dependent magnetization for $Sm_2Ru_3Sn_5$ measured at T = 2 K.

complex magnetic behavior that is influenced by the kinetic effect of a sweeping magnetic field.[30] While $Sm_2Ru_3Sn_5$ is cubic, directional dependence of interactions (anisotropy) can also cause frustration. Considering the frustration index ($f = \theta_{CW}/T_N$), $Sm_2Ru_3Sn_5$ has a value: $f = 9.6$. Even though a value of $f > 5$ is accepted for frustrated 3*d* transition metal magnets, similar values for 4*f* magnets should be treated with caution as the θ_{CW} may be potentially inflated due to crystal field effects.[31-32] An alternative explanation to the large difference between θ_{CW} and T_N is that the Sm moments are screened by the conduction electrons. The phenomenon could be mediated by RKKY[33-34] or Kondo interactions.[35-36] These findings indicate $Sm_2Ru_3Sn_5$ may be a good candidate to study low-temperature Kondo screening in a cubic, non-centrosymmetric environment. The MvH measurements (Figure 2c) were collected below the antiferromagnetic transition at 2 K and show a slight hysteresis at low fields as well as a small, field dependent response; however, no saturation was observed up to 7 T with a response that is an order of magnitude lower than the calculated saturation moment (0.71 μ_B).

The resistivity of $Sm_2Ru_3Sn_5$ is shown in Figure 3a. The measured compound is metallic and has a slight curvature, similar to the shape of the resistivity for Ru_3Sn_7.[37] The authors report the non-linearity of the resistivity in Ru_3Sn_7 to be similar to the A15 (Nb_3Sn) compounds and relate the curvature to the presence of *d*-orbitals near the Fermi level. The resistivity $Sm_2Ru_3Sn_5$ at room

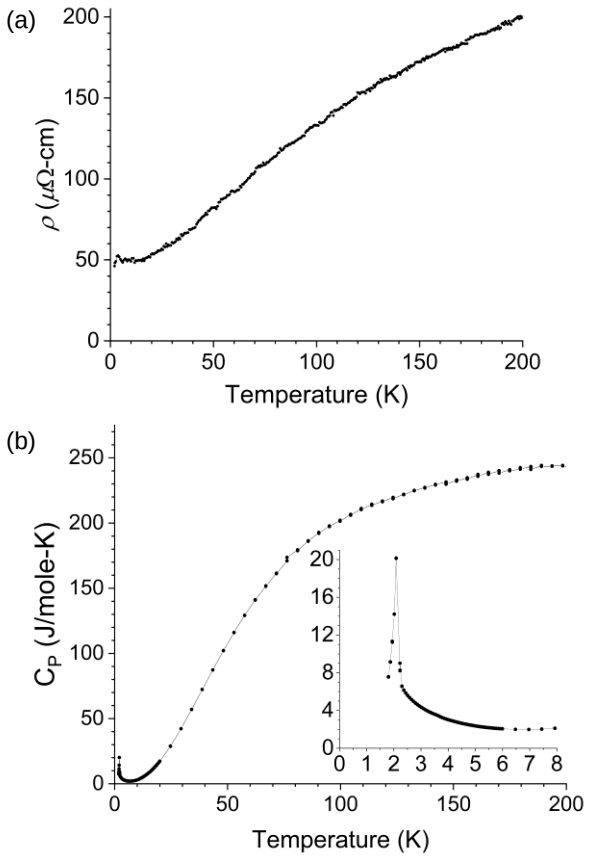

Figure 3. (a) Temperature dependent electrical resistivity of a polished piece of polycrystalline $Sm_2Ru_3Sn_5$ measured using a four-point probe method. (b) Temperature dependent heat capacity for $Sm_2Ru_3Sn_5$ measured on the same sample. Inset shows a sharp feature at 2.1 K, consistent with the antiferromagnetic transition.

temperature (~200 $\mu\Omega$-cm) is nearly three times that of the reported value for Ru_3Sn_7 at room temperature (~70 $\mu\Omega$-cm). The more semimetallic character of $Sm_2Ru_3Sn_5$ is consistent with the scattering of the conduction electrons due to a local magnetic moment. It is worth noting that another group recently studied the impact of Sb-doping on the resistivity of Ru_3Sn_7.[38] The authors found that when Sb was substituted for Sn in $Ru_3Sb_{1.75}Sn_{5.25}$, superconductivity was observed at 4.1 K and attribute it to local maxima near the Fermi level in the density of states.

The heat capacity of $Sm_2Ru_3Sn_5$ is shown in Figure 3b. There is a sharp peak at 2.1 K, consistent with the antiferromagnetic transition from field cooled magnetic susceptibility measurement (T_N = 2.3 K). The data was fit with a Lattice-Debye model. After subtraction of the lattice contribution, the magnetic contribution to the heat capacity (C_{mag}) was integrated to find the magnetic entropy (S_{mag}). The magnetic entropy saturates at a value of Rln(2), consistent with a doublet ground state for the measured temperature range. This is lower than a full J = 5/2 expected for a Sm^{3+} compound. A saturation of the magnetic entropy is observed for other Sm^{3+} compounds and is often attributed to crystal electric field effects caused by splitting of the *f* states.[29, 39-40]

2.4. Electronic Structure

Figure 4 shows the band structure and density of states for Ru_3Sn_7 and $Sm_2Ru_3Sn_5$ calculated with density functional theory (DFT). Both materials are metals, in agreement with the measured resistivity. The DOS near the Fermi energy of both materials is mainly composed of Ru *d* and Sn *p* orbitals, while the Sm atoms contribute little to the DOS of $Sm_2Ru_3Sn_5$.

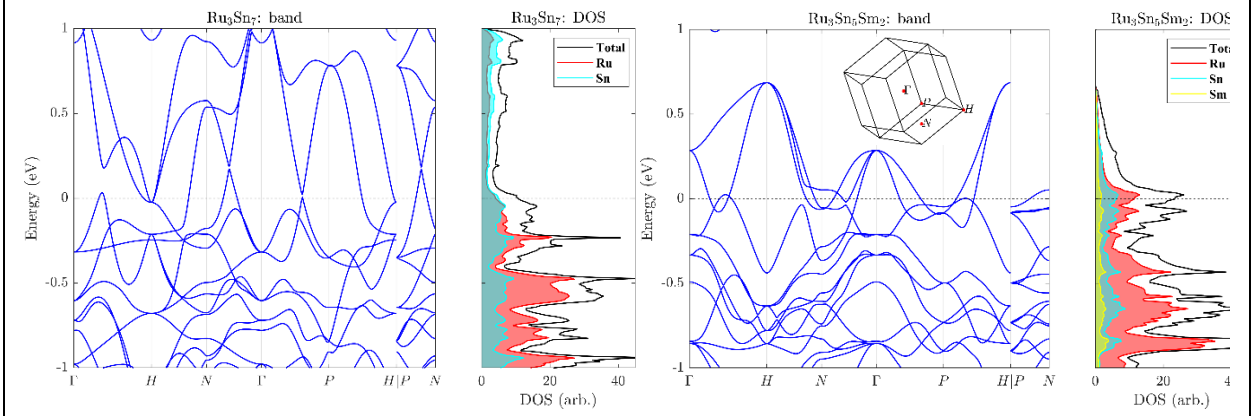

Figure 4. Band structure and density of state (DOS) of Ru_3Sn_7 (left) and $Sm_2Ru_3Sn_5$ (right), respectively. High-symmetry points in the Brillouin zone are shown in the inset of the $Sm_2Ru_3Sn_5$ band structure panel. In the DOS plots, both the total and element-resolved DOSs are shown. The charge neutral point (Fermi energy) is set to zero energy.

The local density approximation with dynamical mean-field theory (LDA+DMFT) calculations support the localized picture (Sm^{3+}) of the *f* electrons in the measured range of temperatures. Figure 5a displays the local density of states at T = 600 K, and the total density of states. The *f* electrons are in a Mott state with fluctuating local moments and no weight at the Fermi level: The local density of states of the *f* electrons displays Hubbard bands in the one electron spectra. Figure 5b displays the probability distribution of the electronic configurations with different numbers of *f* electrons. The dominant occupation is Sm^{3+} with N = 5 electrons, an open shell with a local fluctuating moment. In this material the conduction electrons are scattered by the local moments, and this additional scattering channel provides a possible explanation for the much larger value of the high

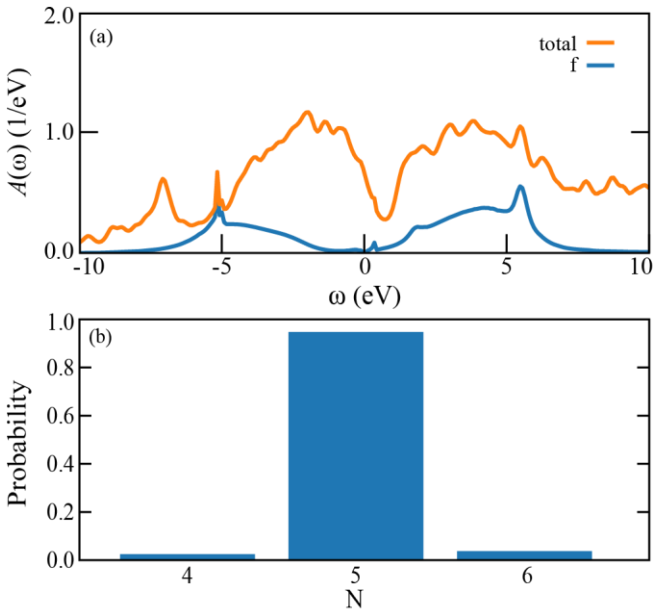

Figure 5. Spectral functions and valence histogram in $Sm_2Ru_3Sn_5$ as predicted within one-shot LDA+DMFT at T = 600 K. There is no Sm-*f* character on the Fermi surface and the valence is almost always in the *N* = 5

temperature resistivity of $Sm_2Ru_3Sn_5$ relative to Ru_3Sn_7. These local moments can be partially Kondo screened at lower temperatures, as it has been observed, for example, in UGe_2.[41] This could account for the reduced magnetic entropy observed in the heat capacity (Figure 3). In this picture, one can interpret the scale of θ_{CW} = -36.6 K extracted from the susceptibility as a Kondo scale.

3. Conclusions

We have successfully prepared $Sm_2Ru_3Sn_5$ and measured its physical properties. Our joint experimental and computational methods support localized Sm *f* electrons. An antiferromagnetic transition is observed at T_N = 3.8 K, and our DFT and LDA+DMFT calculations indicate a dominant Sm^{3+} occupation of the electronic configuration probability distribution. Based on the local density of states (T = 600 K), the *f*-electrons are in a Mott state indicating the presence of Hubbard bands. Notably, the θ_{CW} is an order of magnitude larger than the ordering temperature implying either geometric frustration of the magnetic moments or partial Kondo screening of the Sm magnetic moments at low temperatures. Our analysis of the heat capacity for $Sm_2Ru_3Sn_5$ is consistent with a doublet ground state, implying the presence of crystal field effects which could artificially inflate θ_{CW} and impact the frustration parameter. The geometric frustration could be explained by the tetrahedral Sm sublattice; however, other measurements indicate possible screening of the local Sm moments by the conduction electrons. The reduced moment of approximately 0.06 μ_B/Sm in $Sm_2Ru_3Sn_5$ can be attributed to partial screening of localized magnetic moments by conduction electrons. For comparison, $SmPd_2Al_3$ (where Sm also carries a Sm^{3+} moment) shows a lower than expected magnetic moment of 0.16 μ_B/Sm due to strong crystal field effects.[30] Transport measurements of $Sm_2Ru_3Sn_5$ indicated a higher resistivity than Ru_3Sn_7. This could be explained by local Sm magnetic moments contributing to the scattering of the conduction electrons.

Given the interest in SmB_6 (a topological Kondo insulator) and $SmCoIn_5$ (an antiferromagnet with Kondo screening), low-temperature resistivity measurements to probe $Sm_2Ru_3Sn_5$ for a Kondo resonance would be very useful in confirming Kondo screening of the Sm moments in $Sm_2Ru_3Sn_5$. Our joint experimental and computational methods suggest the presence of Hubbard bands, making it worthwhile to explore the

possibility of suppressing magnetism to observe a Kondo lattice. Given the nature of the magnetism, inelastic X-ray scattering measurements would also be useful in experimentally confirming the electronic structure or the Fermi surface of $Sm_2Ru_3Sn_5$. The competition between RKKY-type interactions and Kondo behavior opens exciting possibilities for studying topology and discovering new quantum materials to explore complex physical phenomena.

4. Experimental Methods

$Sm_2Ru_3Sn_5$ was prepared via arc-melting. Stoichiometric ratios of the elements were measured and pressed together in a hydraulic press to create a homogeneous environment and prevent Ru powder from coating the interior of the setup. The elements were melted into a single boule under a positive flow of Ar gas. A Zr "getter" was included in the reaction setup to minimize oxidation of the sample. The boule was flipped and re-melted three times for homogeneity. Single-crystals were isolated from a fragment of the arc-melted boule (approximately 0.01 x 0.01 x 0.02 mm) and used to collect X-ray diffraction data at T = 160 K and 298 K (above and below the previously reported temperature of T = 240 K).[18] Data was collected using a Bruker D8 Quest Kappa single-crystal X-ray diffractometer equipped with an Incoatec IμS microfocus source (Mo K_α radiation, λ = 0.71073 Å) and a PHOTON III CPAD area detector. The raw frames were integrated with Bruker SAINT, and the intensities were corrected for absorption with a multi-scan method in SADABS.[42] The intrinsic phasing method in SHELXT was used to generate preliminary crystallographic models,[43] which were finalized with least-squares refinements in SHELXL.[44] The details of the data collection, refinement parameters, and atomic coordinates are described in Tables 1 and 2.

Pieces of the arc-melted boule were also ground in an agate mortar and pestle. The powder was then placed on a low-background sample holder, and the powder X-ray diffraction data were collected on a Bruker D2 Phaser equipped with a Cu K_α source (λ = 1.54184 Å) and a LYNXEYE XE-T detector. Data were collected in the 2θ range 5 – 80 ° at room temperature and were analyzed by a Pawley fit in TOPAS (shown in Figure S1).[45]

Energy-dispersive X-ray spectroscopy (EDS) was performed on a piece of the arc-melted boule with a VERSA 3D focused ion beam scanning electron microscope. The formula obtained from EDS was $Sm_{1.9(5)}Ru_{3.1(4)}Sn_{5.0(2)}$, in good agreement with the nominal composition.

Temperature-dependent magnetization data were collected on polycrystalline $Sm_2Ru_3Sn_5$ using a Quantum Design MPMS system. The sample was zero-field-cooled (ZFC), then measured on warming from 1.8 – 300 K in an external magnetic field of 0.1 T. The field-dependent magnetization data were collected at 1.8 K in a range -7 – 7 T. The electrical resistance and heat capacity were measured on the same sample in a Quantum Design PPMS system. For the electrical resistance, a standard 4-probe method was employed on the same sample, where each probe consisted of 0.002 in. diameter Pt wire attached with silver paste. A current of 500 μA was applied, and resistance was measured on cooling in a temperature range of 300 – 2 K.

The electronic structure was calculated using density functional theory (DFT) as implemented in the Vienna *Ab-initio* Simulation Package (VASP).[46] The Perdew, Burke, and Ernzerhof (PBE) generalized gradient approximation[47] was employed to model the electron exchange–correlation interactions. The Projected Augmented Wave (PAW) method[48] was utilized, treating the Sm *f* electrons as core states. All crystal structures were fully relaxed until the forces on all atoms were below 1 meV/Å. A plane-wave basis set with an energy cutoff of 300 eV was used. The reciprocal space was sampled using a 10×10×10 k-point mesh. Spin–orbit coupling was not included. The density of states was calculated using the tetrahedron method with Blöchl corrections.

To calculate the electronic structure including correlation effects, we used codes in Comsuite.[49] In particular, we conducted one-shot density functional theory + dynamical mean field theory (DFT+DMFT) calculations using *Portobello*,[50] the all-electron code FlapwMBPT[49, 51-52] to solve the fully relativistic DFT problem within the local density approximation, and the continuous-time quantum Monte Carlo code ComCTQMC[53] to solve the quantum impurity problem. The Sm-4*f* shell is treated as correlated, and a spherically symmetric Slater-Condon interaction is applied with Hubbard interaction U = 6.5 eV and Hund interaction J = 0.7 eV. We use the nominal double counting[54] with

nominal occupancy $N_0 = 5$ and conduct the calculations at 600 K. We then computed the spectral functions of the *f* electrons, $A(\omega)$, and the valence histogram, *i.e.*, the projection of the many-body density matrix onto the number of electrons, *N.*

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Accession Codes

Crystallographic data (excluding structure factors) for the structures reported in this paper have been deposited with the Cambridge Crystallographic Data Centre as supplementary publication no. CSD-2421218 - 2421219. Copies of the data can be obtained free of charge on application to CCDC, 12 Union Road, Cambridge CB2 1EZ, UK [fax.: (internat.) + 44 1223/336-033; e-mail: deposit@ccdc.cam.ac.uk].

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