Strongly correlated electron behavior in single crystalline U$_2$Os$_3$Al$_9$

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Abstract. We report the magnetic properties of a single crystal of a new compound U$_2$Os$_3$Al$_9$ which crystallizes in the well known Y$_2$Co$_3$Ga$_9$ type orthorhombic structure with space group Cmcm. The susceptibility of U$_2$Os$_3$Al$_9$ shows a peak at 7 K typical of antiferromagnetic ordering. The susceptibility in the paramagnetic state is anisotropic, the easy axis of magnetization lying in the ab-plane of the orthorhombic crystal lattice. The magnetization at 2 K, measured up to a maximum field of 160 kOe, shows a metamagnetic transition near 118 kOe when the field is aligned along [010] in addition to a small metamagnetic transition near 25 kOe. The bulk antiferromagnetic ordering of the uranium ions at T$_N$ = 7 K is confirmed by a peak in the heat capacity with $\Delta C$ nearly 7 J/U.mol K. An extrapolation of the heat capacity data from the paramagnetic regime to T = 0 gives an enhanced electronic specific heat coefficient of 120 mJ/U.mol K$^2$. The electrical resistivity of U$_2$Os$_3$Al$_9$ shows a negative temperature coefficient between 300 and T$_N$ which is a signature of spin fluctuations in a narrow band or a Kondo type of interaction. The data thus suggest the presence of strong electron correlations in this compound.

1. Introduction
The study of ternary compounds of the type R$_2$T$_3$X$_9$ crystallizing in the orthorhombic Y$_2$Co$_3$Ga$_9$-type structure has revealed that Yb, Ce and U based analogues show strongly correlated electron behaviour due to the hybridization of 4f/5f electrons with the itinerant conduction electrons [1, 2, 3, 4]. A host of ground states ranging from Pauli paramagnetic heavy fermion, dense Kondo lattices to magnetically ordered state has been reported for T = Co, Ru, Rh and Ir and X = Al and Ga. Recently we reported magnetic properties of R$_2$Ru$_3$Ga$_9$ (R = Ce, U) compounds [5]. U$_2$Ru$_3$Ga$_9$ shows a negative temperature coefficient of resistivity at high temperatures below 300 K and orders antiferromagnetically at 16 K. Ce$_2$Ru$_3$Ga exhibits valence fluctuating behaviour with a paramagnetic ground state. An extension of our work has led to the discovery of a new iso-structural compound U$_2$Os$_3$Al$_9$. In the present work we report the anisotropic magnetic and transport properties of single crystalline U$_2$Os$_3$Al$_9$ in the temperature range 1.8-300 K and in applied magnetic fields up to 160 kOe.

2. Experiment
A single crystal of U$_2$Os$_3$Al$_9$ was grown in a tetra arc furnace using Czochraski pulling process. About 10 grams of total charge, with at least 3N pure metals, was taken and initially melted a...
few times to ensure proper homogenization. A crystal was then pulled at a rate of 10 mm/hour. An oriented single crystal was cut from the pulled rod with spark erosion and oriented using Laue diffraction. A powder x-ray diffraction taken on a Phillips Pan-Analytical x-ray diffractometer confirmed the phase purity of the material. The actual crystal composition was confirmed using a CAMECA SX100 Electron Probe Microanlyser (EPMA). The DC magnetic susceptibility and the in-field magnetization measurements were performed in the temperature range 1.8-300 K using a Quantum Design (QD) superconducting quantum interference device (SQUID) magnetometer. The temperature dependence of electrical resistivity in the range 1.8-300 K was measured using a home made DC electrical resistivity set up. The heat capacity was performed using a QD physical properties measurement system (PPMS).

3. Results and Discussion

3.1. Crystal Structure

Powder x-ray diffraction pattern revealed the sample to be single phase with Y$_2$Co$_3$Ga$_9$ type crystal symmetry. Lattice parameters obtained from Rietveld analysis using FullProf software [6] package are $a = 13.2375$ Å, $b = 7.6450$ Å and $c = 9.3992$ Å.

3.2. Magnetization

Fig. 1(a) shows the magnetic susceptibility of U$_2$Os$_3$Al$_9$ in temperature range 1.8-300 K in a field of 3 kOe along the three principal axes respectively. Susceptibility is nearly isotropic along a and b axes but anisotropy is observed relative to [001] axis. $\chi_{100}/\chi_{001}$ varies from 1.2 at 300 K to 3.2 at 10 K. Susceptibility at high temperatures varies according to Curie Weiss law $\chi = C/(T - \theta_p)$ and a least squares fitting furnishes $\mu_{eff}$ for H parallel to [100], [010] and [001] axes as 2.82 $\mu_B$, 2.96 $\mu_B$ and 3.02 $\mu_B$, respectively. The corresponding values of $\theta_p$ are -13.6, -19.9 and -148 K. Values of effective magnetic moment are lesser than both U$^{4+}$ (3.58 $\mu_B$) and U$^{3+}$ (3.62 $\mu_B$) moment implying some degree of hybridization between the 5f and ligand electronic orbitals. Negative values of $\theta_p$ indicate an antiferromagnetic exchange interaction between the uranium ions. Indeed, the susceptibility along each of the three axis shows a sharp peak at 7 K associated with the antiferromagnetic ordering of the uranium ions.

Iso-thermal magnetization versus the applied magnetic field is shown in Fig. 1(b) for field applied along three principal directions. An overall nearly linear magnetization up to about
100 kOe is consistent with the AFM ordering of uranium moments. The magnetization does not saturate in field as high as 160 kOe. The moment at 160 kOe for \( H \parallel [100], [010], \) and [001] is 0.77, 0.72, and 0.16 \( \mu_B \) respectively. As seen in the susceptibility the moment values for \( H \parallel [100] \) and [010] axes are higher than for \( H \parallel [001] \). For \( H \parallel [010] \) an irreversible jump in the magnetization is observed near 25 kOe in addition to a metamagnetic transition around 118 kOe. Similar behaviour is observed for \( H \parallel [100] \) but the transition at 118 kOe is broader. However for \( H \parallel [001] \) no metamagnetic transition is observed up to the highest field of 160 kOe, which implies that it is the hard axis of magnetisation. For \( H \parallel [010] \) the magnetization was measured at various temperatures (not shown). Metamagnetic transition shifts to lower fields at higher temperatures. A magnetic phase diagram is constructed for \( H \parallel [010] \) as shown in Fig. 2(a).

### 3.3. Electrical Resistivity

Fig. 2(b) shows the temperature dependence of electrical resistivity with current applied along [100], [010] and [001], respectively, from room temperature down to 1.8 K in zero external magnetic field. Some anomalous features of resistivity are noticeable. The magnitude of resistivity at 300 K is high. Although not a common phenomenon, comparable values of resistivity have been previously seen in some uranium systems [7]. The resistivity exhibits a negative temperature coefficient, which is often seen in strongly electron correlated uranium systems. Solid lines in Fig. 2(b) show how the resistivity varies linearly with -\( \ln(T) \). For \( I \parallel [001] \) two such linear regions are observed. Below 7 K the resistivity sharply rises in magnitude which we attribute to the onset of superzone boundaries on the Fermi surface due to the antiferromagnetic ordering at 7 K. The rise in the resistivity below 7 K continues down to the lowest temperature of 1.8 K in our measurement. The negative temperature coefficient of resistivity at high temperatures can tentatively be attributed to the presence of spin fluctuations in a narrow f-band system or to a Kondo-like interaction between the U-5\( f \) moments and the itinerant charge carriers.

### 3.4. Heat Capacity

Fig. 3(a) shows the heat capacity of \( U_2Os_3Al_9 \) in the range 1.8-30 K and adjusted heat capacity of the reference polycrystalline compound \( Lu_2Os_3Al_9 \) which is used as a background for the lattice and conduction electron contributions. Bulk nature of the magnetic transition is confirmed by the occurrence of a peak in the heat capacity near 7 K with a peak height \( \Delta C \) of 7 J/U.mol K.
The jump is reduced compared to the mean field value of 12.5 J/mol-K for spin $S=\frac{1}{2}$. The $5f$ contribution to the heat capacity, obtained by the standard procedure after adjustment is made for the difference in the atomic weights of U and Lu, is plotted in the Fig. 3(b); the corresponding entropy $S_{5f}$ is also plotted. The entropy at $T_N$ is nearly $0.37 R \ln 2$ and attains the value $R \ln 2$ (for spin $\frac{1}{2}$ level) at an elevated temperature of nearly 30 K. These two observations indicate a low U-moment state and support the reduced value of $\mu_{\text{eff}}$ derived from susceptibility. The variation of entropy with temperature nearly 30 K suggest possible Shottky contribution to the heat capacity from excited crystal field split levels. Data above 30 K are required for verification.

$C_{5f}/T$ vs $T$ is increasing with decreasing temperature seen clearly in Fig. 3(b). Assuming the increase is solely due to an increasing density of states associated with a narrow band, an extrapolation of $C_{5f}/T$ vs. $T^2$ data, between 30 and 15 K, to 0 K furnishes a $\gamma$ ($C/T$ as $T\to 0$) of 120 mJ/U.mol.K$^2$, which suggests a substantial enhancement of the effective mass due to electronic correlations.

4. Conclusion
We have successfully grown a single crystal of orthorhombic $U_2Os_3Al_9$ by Czochraski pulling process. The uranium ions in this compound order antiferromagnetically at $T_N = 7$ K at which a gap opens up at the Fermi surface. The bulk nature of magnetic ordering is confirmed by heat capacity. A negative temperature coefficient of resistivity, magnetic entropy lower than $R \ln 2$ up to $T_N$ and a relatively high $\gamma$ indicate the presence of strong electron correlations in $U_2Os_3Al$.

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Figure 3. (Color online) (a) Heat capacity vs temperature of $R_2Os_3Al_9$ ($R = U, Lu$) from 1.8 K to 30 K, Lu heat capacity is adjusted for molecular weight difference. Inset shows $C/T$ vs $T^2$ of $U_2Os_3Al_9$. (b) $C_{5f}/T$ and $S_{5f}/T$ vs $T$ for $U_2Os_3Al_9$.