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Unconventional $-\log T$ dependent resistivity in $Sm_xLa_{1-x}Ta_2Al_{20}$

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Abstract
We have succeeded in growing $Sm_xLa_{1-x}Ta_2Al_{20}$ single-crystals, and performed electronic transport measurements to investigate the characteristic features of conduction electron scattering relevant to the field-insensitive $-\log T$ dependence of the resistivity. We have found that the $-\log T$ dependence of the resistivity remains in a Sm-dilute compound with $x = 0.01$. This finding provides clear evidence for that the $-\log T$ dependence is caused by a local single-ion Kondo effect. We have measured magnetoresistance (MR) of $SmTa_2Al_{20}$ up to 14 T and confirmed that the positive MR continues to develop with decreasing temperature even at 0.5 K, evidencing a large deviation from Kohler's rule. This fact indicates that strong wave-vector dependence of conduction-electron scattering by $4f$ electrons still remains at 0.5 K in the magnetically ordered state.

Keywords: Kondo effect, Sm, La substitution, field-insensitive, x-ray diffraction, magnetoresistance

1 Introduction

Strongly correlated electron systems with cage structures (e.g. filled skutterudites) have attracted much attention due to a variety of interesting phenomena such as heavy fermion (HF) behavior, unconventional superconductivity and rattling motion of guest ions. In recent years, another class of cage compounds with the general chemical formula RTr_2X_{20} (R = rare earth, Tr = transition metal, X = Al, Zn, and Cd), which crystallizes in a cubic structure with the space group $Fd\bar{3}m$, have attracted much attention, where R site has a cubic point symmetry T_d [1, 2, 3, 4, 5]. Remarkable phenomena found in this series include the HF state with the extremely large Sommerfeld coefficient of $\gamma = 8$ J/mol K^2 in $YbCo_2Zn_{20}$ [6, 7] and a nonmagnetic Kondo effect and quadrupole-fluctuation-mediated superconductivity in $PrTr_2Al_{20}$ (Tr = Ti and V) [8, 9].

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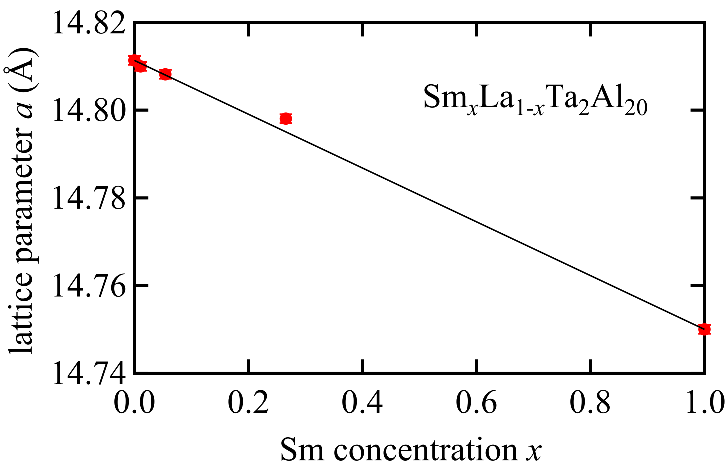

Figure 1: (Color online) Lattice parameter a as a function of Sm concentration x. The solid line represents the Vegard's law.

In the $\mathrm{Sm}\,Tr_2\mathrm{Al}_{20}$ family (Tr = Ti, V, Cr, and Ta), unusually field-insensitive phase transitions [T_x = 6.5 K (Tr = Ti), 2.9 K (V), 1.8 K (Cr), and 2.0 K (Ta)] and HF behavior insensitive to the magnetic field have been discovered [10, 11, 12]. The HF behavior in these compounds includes the $-\log T$ dependence of resistivity and large Sommerfeld coefficients of $\gamma = 0.15$ J/mol K^2 (Tr = Ti), 0.72 J/mol K^2 (V), 1.0 J/mol K^2 (Cr), and 3 J/mol K^2 (Ta). The quasiparticle mass enhancement has been confirmed by a recent de Haas-van Alphen experiment; the cyclotron effective mass of $SmTi_2Al_{20}$ is enhanced several times compared with that in $LaTi_2Al_{20}$ [13]. The origin of the field-insensitive HF behavior has not been unveiled yet. The crystalline-electric-field (CEF) ground state of these Sm compounds is expected to be a Γ_8 quartet of the $J = 5/2$ multiplet [10, 11, 12]. Since the size of the ordered Sm moment is smaller than that expected for a Γ_8 state and a Γ_8 quartet includes higher-rank multipolar degrees of freedom, the contribution of higher-rank multipoles to the ordering has been suggested [10, 12]. The magnetic susceptibilities of the compounds show weak temperature dependences, which are evidently different from those of both free Sm^{2+} and Sm^{3+} ions. X-ray absorption spectroscopy (XAS) studies revealed that Sm ions in the $\mathrm{Sm}\,Tr_2\mathrm{Al}_{20}$ family are in mixed valence states with an average Sm ion valence of 2.87 with no significant T dependence between 7 and 300 K [14]. These results suggest that valence fluctuations play a role in the field-insensitive nature as discussed for $SmOs_4Sb_{12}$, another field-insensitive HF compound [15, 16]. In order to investigate the characteristic features of conduction electron scattering relevant to the field-insensitive $-\log T$ dependence of resistivity observed in $SmTa_2Al_{20}$, we are studying the La-substitution effect on the transport properties. In this proceeding, we report preliminary results of our electrical resistivity measurements using single crystals of $Sm_xLa_{1-x}Ta_2Al_{20}$. For x=1, we have measured magnetoresistance (MR) up to 14 T down to 0.5 K.

2 Experimental

Single crystals of $Sm_xLa_{1-x}Ta_2Al_{20}$ (x = 0.01, 0.05, 0.25, and 1) were grown by the Al self-flux method. The starting atomic ratio of the elements [Sm (3N), La (3N), Ta (3N5), and Al (4N)] were x:1-x:2:150. X-ray powder-diffraction experiments were carried out using a Rigaku SmartLab powder diffractometer equipped with a Cu Kα_1 monochromator. A silicon standard was used for an accurate determination of the lattice parameter. It was confirmed that all of $Sm_xLa_{1-x}Ta_2Al_{20}$ samples crystallize in the $CeCr_2Al_{20}$-type structure. The lattice parameter a increases almost linearly with decreasing Sm concentration x, as displayed in Fig. 1. Electrical resistivities were measured using a physical property measurement system [PPMS; Quantum Design (QD)] combined with a QD ^{3}He optional probe down to 0.5 K by a standard four-probe method.

3 Results and discussion

Figure 2(a) shows the temperature dependence of electrical resistivity ρ of $Sm_xLa_{1-x}Ta_2Al_{20}$ in zero field. Expanded views of $\rho(T)$ for x = 0.25 and 0.01 at low temperatures are shown in Figs. 2(b) and (c), respectively. $\rho(T)$ for $LaTa_2Al_{20}$ is also plotted in Fig. 2(c) [17]. These figures show that the $-\log T$ dependent term in $\rho(T)$ becomes weaker with decreasing Sm concentration. It is remarkable that the $-\log T$ dependence is observed in the most diluted sample of $x = 0.01$. Since $\rho(T)$ for $LaTa_2Al_{20}$ is almost flat below 10 K, the $-\log T$ dependence for $x = 0.01$ should be attributed to conduction-electron scattering by Sm $4f$ electrons. This finding provides clear evidence for that the $-\log T$ dependent term is caused by a local single-ion Kondo effect. If the $\rho(T)$ data is compared with a theoretical calculation of $\rho(T)$ for a single-impurity Kondo model [18], the Kondo temperature T_K is estimated to be of the order of 10-50 K; accurate determination of T_K may be difficult since an estimation of the phonon-scattering contribution in $\rho(T)$ is not easy.

For a rough estimation of the magnitude of the $-\log T$ dependent term in $\rho(T)$, we tentatively use the following equation neglecting the phonon scattering part of $\rho(T)$:

$$\rho = \rho_0 - \alpha \log T, \tag{1}$$

where ρ_0 and α are fitting constants [19]. The value of α obtained from the fitting is plotted in Fig. 2(d). The value of α decreases drastically with the La substitution. The x dependence of α can be expressed by $\alpha = cx^n$ with c =11.2 $\mu\Omega$cm and n = 1.43, although the data points are limited. The value of n larger than 1 may be caused by possible correlations among Sm ions in high x region. To clarify further details of the x dependent development of the $-\log T$ term in $\rho(T)$, measurements for samples with other x values are underway. We expect that comparison with the similar $\rho(T, x)$ data reported for typical Ce-based HF compounds $(Ce_xLa_{1-x})Al_2$ and $(Ce_xLa_{1-x})Cu_6$ [20, 21] will help to understand the difference in the Kondo scattering between Ce and Sm ions.

A broad peak appears at $T^* = 2.5$ K for $x = 1$ [see Fig. 2(a)]. This peak can be caused by either (1) short-range ordering associated with the phase transition at $T_x = 2$ K [12] or (2) the development of coherence among Sm ions (in this case, the coherence temperature $T_{coh} \simeq T^*$). For $x = 0.25$, as shown in Fig. 2 (b), a similar broad peak appears at 1.6 K, which is lower than that for $x = 1$. This may indicate that T_x or T_{coh} decreases with decreasing Sm concentration.

For $x = 1$, we have reported in a previous paper that the slope of $-\log T$ dependence of resistivity does not show any noticeable change with varying the magnetic field up to 9 T [12]. In order to further investigate the field insensitivity, the temperature dependence of resistivity

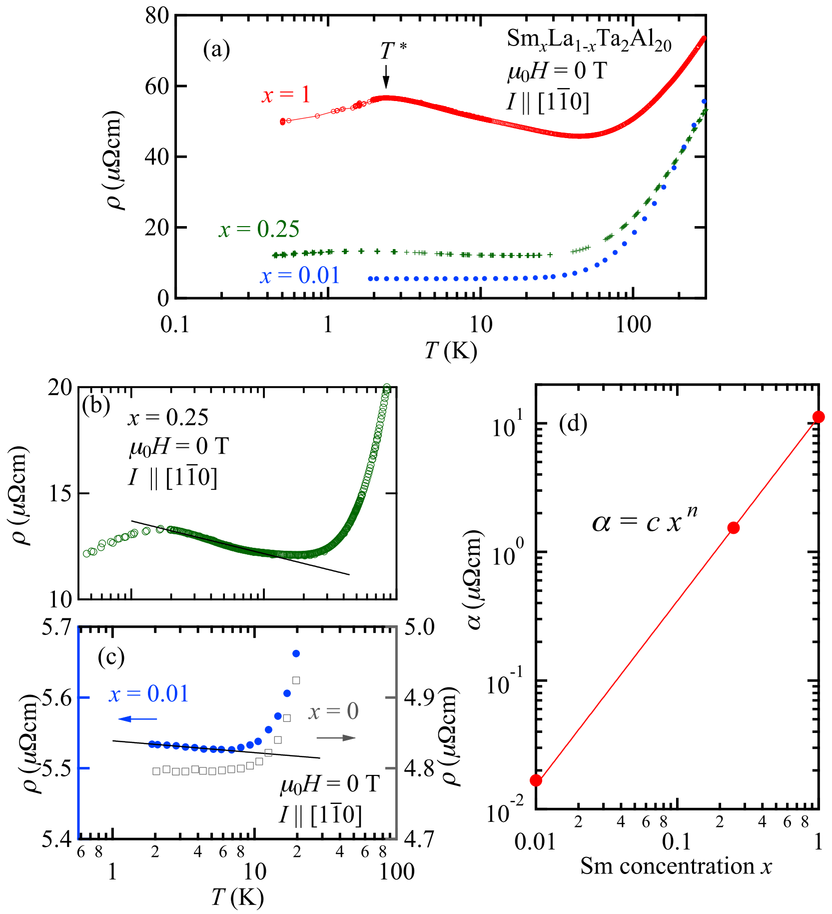

Figure 2: (Color online) (a) Temperature dependence of resistivity $\rho(T)$ of $Sm_xLa_{1-x}Ta_2Al_{20}$ (x = 0.01, 0.25, and 1). Expanded views of $\rho(T)$ for (b) $x = 0.25$ and (c) $x = 0.01$ at low temperatures in zero field. In order to show the contrast between $\rho(T)$ for $x = 0.01$ and that for $x = 0$, $\rho(T)$ for $LaTa_2Al_{20}$ is also plotted in (c) [17]. The solid lines show the fitting results. The fitting range is 2 - 6 K for $x = 0.01$, 3 - 10 K for $x = 0.25$, and 5 - 20 K for $x = 1$. (d) The slope of $-\log T$ dependence, α, is plotted as a function of Sm concentration x. The solid line represents $\alpha = cx^n$ with $c = 11.2$ $\mu\Omega$cm and $n = 1.43$.

in 14 T was measured down to 0.5 K, which is shown in Fig. 3(a) along with those in 0 and 9 T. We have confirmed that the slope of $-\log T$ dependence of resistivity is not suppressed by magnetic field at least up to 14 T. Figure 3(b) shows the temperature dependence of MR, $\Delta\rho/\rho(0)$ [$\equiv (\rho(H) - \rho(0))/\rho(0)$] in 9 and 14 T. MR is always positive over the investigated temperature range, being in marked contrast to the negative MR observed in Ce-based HF compounds [21]. The positive MR has been confirmed to exhibit a H^2 dependence in the low-field condition in the temperature range between 2 and 300 K [22]. This observation clearly shows that the ordinary MR due to the cyclotron motion of conduction electrons far exceeds the expected field suppression of the Kondo effect.

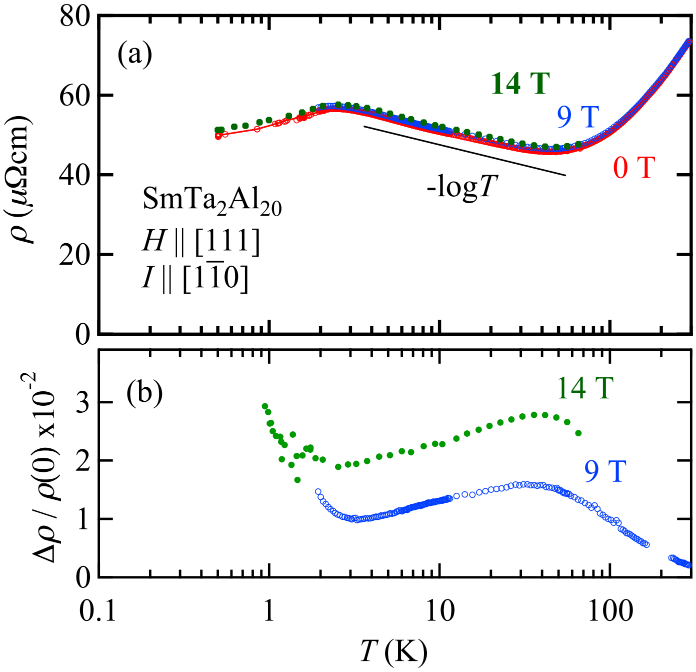

Figure 3: (Color online) Temperature dependence of (a) resistivity of $SmTa_2Al_{20}$ in 0, 9, and 14 T and (b) MR $\Delta\rho/\rho(0)$ [$\equiv (\rho(H)-\rho(0))/\rho(0)$] in 9 and 14 T. The data in 9 T is reproduced from Ref. 22. The solid line is a guide to the eye. Below 0.9 K, accurate determination of MR in 14 T was difficult using excitation currents low enough to avoid Joule heating.

In the low-field condition, the value of $\Delta\rho/\rho(0)$ at a constant $\mu_0H/\rho(0)$ develops by an order of magnitude with decreasing temperature from 300 to 2 K [22]; this behavior is called "a deviation from Kohler's rule (DKR)". This behavior reflects the development of a wave-vector dependent relaxation time (or anisotropic scattering) of conduction electrons by Sm $4f$ electrons with decreasing temperature. The most anomalous feature of DKR in $SmTa_2Al_{20}$ is that DKR continues to develop with decreasing temperature; in conventional metals, DKR appears in a middle temperature range (e.g., Al shows a maximum at $\sim$ 20 K) [23]. To investigate this anomaly further, we have performed a MR measurement at 0.5 K. In a Kohler plot of Fig. 4, the data of $\Delta\rho/\rho(0)$ measured at 0.5 and 2 K lie almost on the same curve within the experimental uncertainty. This fact indicates that the feature of the anisotropic scattering of conduction electrons by Sm $4f$ electrons still remains at 0.5 K in the ordered state.

Instead of Kohler's rule, the MR data of $SmTa_2Al_{20}$ have been found to satisfy the so-called "modified Kohler's rule (MKR)", i.e., $\Delta\rho/\rho(0) \propto \tan^2\theta_H$, where θ_H represents the Hall angle, except for a middle temperature range where $\theta_H \simeq 0$ [22]. Since MKR is satisfied by some of the high-T_c cuprates and Ce$TrIn_5$ HF compounds [24, 25], this rule is usually considered to reflect the nature of quasiparticle scattering in two-dimensional electron systems with hot spots on the Fermi surface [26]. However, the above-mentioned observation in $SmTa_2Al_{20}$, providing the first example of MKR satisfied in three-dimensional electron systems, suggests that MKR is not limited in two-dimensional electron systems and $SmTa_2Al_{20}$ has hot spots on the Fermi

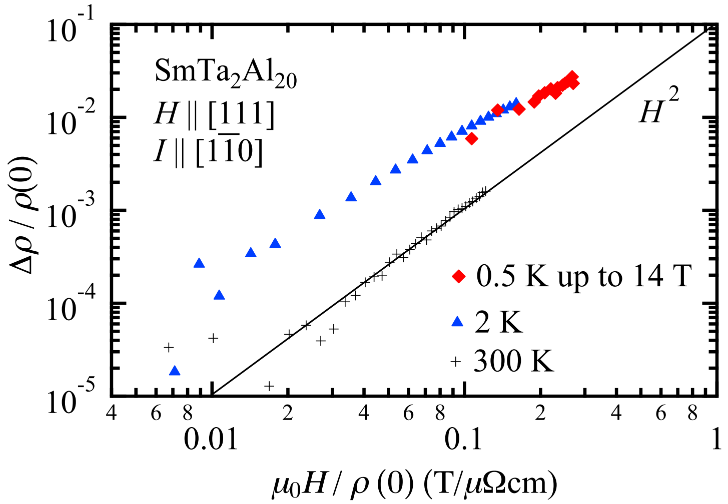

Figure 4: (Color online) Kohler plot of the $\Delta\rho/\rho(0)$-vs-$\mu_0 H/\rho(0)$ data measured at 0.5, 2, and 300 K. The data at 2 and 300 K are reproduced from Ref. 22.

surface.

In summary, we have succeeded in growing single crystals of $Sm_xLa_{1-x}Ta_2Al_{20}$ ($x = 0.01$, 0.05, 0.25, and 1) and performed electronic transport measurements. The $-\log T$ dependence of resistivity remains even in the most dilute sample with $x = 0.01$, clearly evidencing that the $-\log T$ dependence is caused by a local single-ion Kondo effect. In $SmTa_2Al_{20}$, the field-insensitive nature of the $-\log T$ dependent resistivity has been confirmed to remain up to 14 T. The magnetoresistance (MR) $\Delta\rho/\rho(0)$ is always positive; the expected field suppression of the Kondo scattering is overwhelmed by the positive MR caused by the cyclotron motion of conduction electrons. The temperature dependence of MR reveals that the deviation from Kohler's rule (DKR), characterizing the anisotropic conduction-electron scattering by Sm $4f$ electrons, still remains large at 0.5 K in the magnetically ordered state. MR studies on other Sm-based strongly correlated electron systems are highly desired to clarify the relation between the field-insensitive behaviors and the anomalies in DKR.

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