Antiferromagnetic Kondo-lattice systems $Ce_2Rh_3Ge_5$ and $Ce_2Ir_3Ge_5$ with moderate heavy-fermion behavior

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The results of electrical resistivity, magnetic susceptibility, specific heat, isothermal magnetization, and magnetoresistance measurements on $Ce_2Rh_3Ge_5$ and $Ce_2Ir_3Ge_5$ are reported. For both systems, the magnetic contribution to the resistivity shows the logarithmic T dependence in two separate T ranges being typical of a Kondo-lattice system in the presence of crystalline field. A marked peak in the susceptibility and a sharp drop in the resistivity are observed at $T_N = 5.5$ and 9.5 K for $Ce_2Rh_3Ge_5$ and $Ce_2Ir_3Ge_5$, respectively. The total drop in resistivity is as large as 300 $\mu\Omega$ cm. Isothermal magnetization curves at 1.3 K reveal a field induced transition at 23 T in $Ce_2Rh_3Ge_5$ and two transitions at 10 and 15 T in $Ce_2Ir_3Ge_5$. The saturation moment of $0.8\mu_B$/Ce is suggestive of the Γ_7 doublet ground state in both compounds. The specific heat exhibits a λ-type anomaly around T_N, while the values of magnetic entropy at T_N are reduced to 50 and 65% of $R \ln 2$ for $Ce_2Rh_3Ge_5$ and $Ce_2Ir_3Ge_5$, respectively. The linear coefficients of specific heat are 148 and 152 mJ/K^2 mol Ce above T_N. Thus both compounds are classified into moderate heavy-fermion antiferromagnets.
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I. INTRODUCTION

Cerium-based intermetallic compounds often exhibit unusual physical properties which can be described using the Kondo-lattice model.[1] In this model, the ground state is determined by the competition between the intersite Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction and the onsite Kondo coupling. The characteristic energies for these two interactions are $T_{\mathrm{RKKY}} \propto (J\rho_c)^2$ and $T_K \propto \exp(-1/J\rho_c)$, where J denotes the strength of coupling between the $4f$ electron and conduction electrons and ρ_c is the density of states of the conduction electrons at the Fermi level. When RKKY interaction dominates ($T_{\mathrm{RKKY}} > T_K$), the system orders magnetically. For comparable strength of these two interactions ($T_{\mathrm{RKKY}} \approx T_K$), the system shows Kondo-type behavior but still orders magnetically. When Kondo-type interaction dominates ($T_K > T_{\mathrm{RKKY}}$), the system is demagnetized due to the Kondo effect and the ground state is nonmagnetic. $CeAl_2$ and CeB_6 are typical examples of Kondo-lattice compounds which order antiferromagnetically whereas $CePd_2Ga_3$ orders ferromagnetically.[2–4] Kondo compounds with a magnetic ground state show strongly reduced ordered moments. On the other hand, $CeCu_6$ is a typical example with a nonmagnetic ground state.[5] The Kondo compounds often show very large linear coefficient of heat capacity and are called heavy-fermion systems.

In this paper we report the magnetic and transport properties of $Ce_2Rh_3Ge_5$ and $Ce_2Ir_3Ge_5$ and their nonmagnetic analogs $La_2Rh_3Ge_5$ and $La_2Ir_3Ge_5$. The materials were reported to form in the orthorhombic $U_2Co_3Si_5$-type structure[6] which is an intergrowth of the $CaBe_2Ge_2$- and the $BaNiSn_3$-type structures. From the magnetic susceptibility measurement down to 4.2 K, earlier it was reported that the ground state of $Ce_2Rh_3Si_5$ and $Ce_2Ir_3Si_5$ are nonmagnetic but $Ce_2Ir_3Ge_5$ undergoes an antiferromagnetic order below $T_N = 10$ K and $Ce_2Rh_3Ge_5$ did not show any signature of magnetic ordering.[7] These results hinted at the possibility of $Ce_2Rh_3Ge_5$ to lie in the borderline between the magnetic and nonmagnetic ground states and hence we expected this to be a potential candidate for heavy-fermion system. Further the system CeM_2X_2 (X = Si, Ge) exhibits many interesting properties such as heavy-fermion superconductivity in $CeCu_2Si_2$ and metamagnetism in $CeRu_2Si_2$.[8,9] Therefore $R_2M_3X_5$ (R = rare earth, M = transition metal, X = Si, Ge) are expected to show interesting physical properties. Indeed, $Ce_2Ni_3Si_5$ is a valence fluctuating compound,[10] $Ce_2Pd_3Ge_5$ is an antiferromagnet with $T_N = 3.8$ K,[11] and $Tm_2Fe_3Si_5$ is the only antiferromagnetic superconductor showing reentrance to the normal state below T_N.[12] Furthermore, $U_2Rh_3Si_5$ undergoes a first-order phase transition into a simultaneous spin-quadrupolar ordering.[13]

II. EXPERIMENTAL

Polycrystalline samples of $R_2M_3Ge_5$ (R = La, Ce; M = Rh, Ir) have been prepared by arc melting the constituent elements (Ce 99.99%, La 99.99%, Rh 99.95%, Ir 99.9%, Ge 99.999% in purity) in stoichiometric ratios on a water cooled copper hearth. The samples were flipped after each melting and were melted several times to ensure homogeneity. The net weight-loss for each sample was less than 0.5%. The melted buttons were wrapped in Ta foil and annealed in an evacuated quartz tube at 900 °C for one week. The phase purity of the samples was checked by metallographic examination and powder x-ray diffraction using Cu K_α radiation. We found that the samples were single phase and form in the

TABLE I. Lattice parameters and unit-cell volume for $R_2M_3Ge_5$ (R=La, Ce; M=Rh, Ir).

Compounds	a (Å)	b (Å)	c (Å)	V (Å^3)
$La_2Rh_3Ge_5$	10.141	12.187	6.039	746.350
$Ce_2Rh_3Ge_5$	10.10	12.111	5.998	733.681
$La_2Ir_3Ge_5$	10.233	12.03	6.10	750.928
$Ce_2Ir_3Ge_5$	10.197	11.938	6.07	738.911

$U_2Co_3Si_5$-type structure. Electrical resistivity measurements were carried out using conventional dc four-probe technique in the temperature range 1.3–300 K. The heat capacity was measured for 1.7–30 K using an adiabatic heat-pulse method. A superconducting quantum interference device magnetometer (Quantum Design) was used for susceptibity measurement. Isothermal magnetization measurements up to 58 T were carried out using a pulsed magnet at Osaka University.

III. RESULTS AND DISCUSSION

The lattice parameters for $Ce_2M_3Ge_5$ and $La_2M_3Ge_5$ (M=Rh, Ir) are given in Table I. The parameters are in agreement with those reported in the literature.[6] Figure 1 shows the magnetic susceptibility χ of the Ce compounds in the temperature range 2–300 K plotted as χ^{-1} vs T. In the high-temperature range above 100 K a Curie-Weiss behavior is visible which leads to an effective moment $2.6\mu_B$ for both compounds, being close to the value expected for Ce^{3+} ions. It is to be noted that the effective magnetic moment value of $1.8\mu_B$ for both was reported in Ref. 7. Strong deviation from the Curie-Weiss behavior is seen below 70 K for $Ce_2Rh_3Ge_5$ and below 90 K for $Ce_2Ir_3Ge_5$. This could be attributed to the crystalline electric field effects. From the high-temperature part of the χ^{-1} vs T curve, the paramagnetic Curie temperatures θ_p is deduced to be -111 K for $Ce_2Rh_3Ge_5$ and -160 K for $Ce_2Ir_3Ge_5$. Such large negative values of θ_p are often found in Kondo compounds, and the absolute value of θ_p is related to the Kondo temperature for

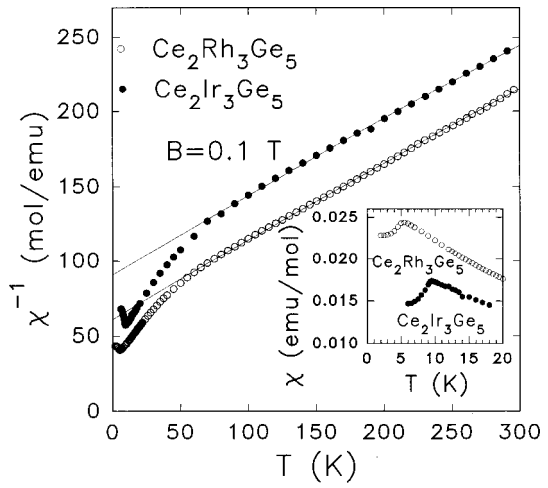

FIG. 1. Inverse susceptibility (χ^{-1}) as a function of temperature for $Ce_2Rh_3Ge_5$ and $Ce_2Ir_3Ge_5$ measured in a field of 0.1 T. The inset shows χ vs T at low temperatures.

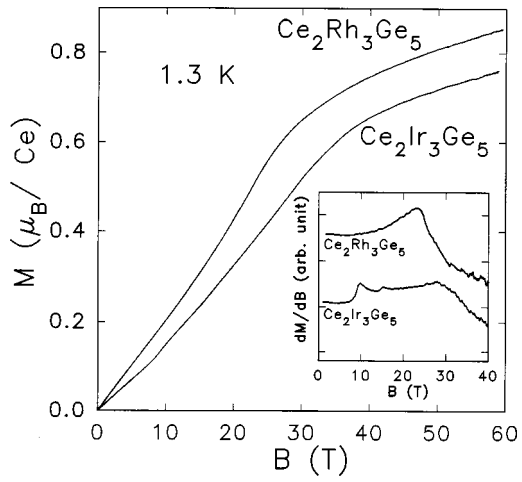

FIG. 2. Isothermal magnetization of $Ce_2Rh_3Ge_5$ and $Ce_2Ir_3Ge_5$ measured at 1.3 K. Inset shows the dM/dB vs B at the same temperature.

the overall crystal-field levels as $T_K \sim |\theta_p/2|$.[14] From this relation we estimate T_K to be 56 and 80 K for $Ce_2Rh_3Ge_5$ and $Ce_2Ir_3Ge_5$, respectively. The data of $\chi(T)$ at low temperatures are shown in the inset of Fig. 1. Occurrence of the peaks at 5.5 and 9.5 K, respectively, indicates the magnetic ordering. The peak in the susceptibility without difference between the zero-field-cooled and field-cooled data suggests antiferromagnetic nature. This is further confirmed by the measurements of isothermal magnetization $M(B)$. In Fig. 2, M vs B at 1.3 K is linear up to 20 T for $Ce_2Rh_3Ge_5$ and up to 8 T for $Ce_2Ir_3Ge_5$, which is consistent with the antiferromagnetic nature of the ground state. For $Ce_2Rh_3Ge_5$ we notice that the magnetization curve has an inflection point at 23 T where the dM/dB vs B curve has a maximum, as is shown in the inset. In the case of $Ce_2Ir_3Ge_5$ two anomalies exist at 10 and 15 T, respectively. The saturation moment is $0.75\mu_B$/Ce for $Ce_2Ir_3Ge_5$ which is very close to $0.71\mu_B$/Ce expected for Γ_7 doublet ground state. In the case of $Ce_2Rh_3Ge_5$ the saturation moment is $0.85\mu_B$/Ce. The slightly larger value than that expected for the Γ_7 doublet is possibly due to field induced mixing up of the first excited state.

Figures 3(a) and 3(b) respectively display the resistivity ρ of $Ce_2Rh_3Ge_5$ and $Ce_2Ir_3Ge_5$ together with the nonmagnetic La compounds in the temperature range 1.3–300 K. The insets show more detail of the low-temperature part. A broad hump is seen in $\rho(T)$ of $Ce_2Rh_3Ge_5$ around 70 K which could be caused by the interplay between crystal field and the Kondo effect.[15] At low-temperature region, $\rho(T)$ of $Ce_2Rh_3Ge_5$ exhibits a minimum at $\sim$25 K. On further cooling $\rho(T)$ passes through a maximum at 7.5 K and then a sudden drop below T_N=5.5 K. The resistivity of $Ce_2Ir_3Ge_5$ shows a similar behavior but the minimum in $\rho(T)$ is not as pronounced as in $Ce_2Rh_3Ge_5$. A sharp drop in $\rho(T)$ is seen below T_N=9.5 K. For $La_2Rh_3Ge_5$ a drop in $\rho(T)$ and a diamagnetic trend in the magnetic susceptibility have been observed at $\sim$2 K which is suggestive of a superconducting transition. Since the metallographic examination and electron-probe microanalysis of the sample of $La_2Rh_3Ge_5$ revealed the presence of a small amount of $LaRh_2Ge_2$ and $LaRhGe_3$ as impurity phases, it is essential to check whether or not the superconducting signal comes from the impurity phases. We did not find any signature of superconductivity

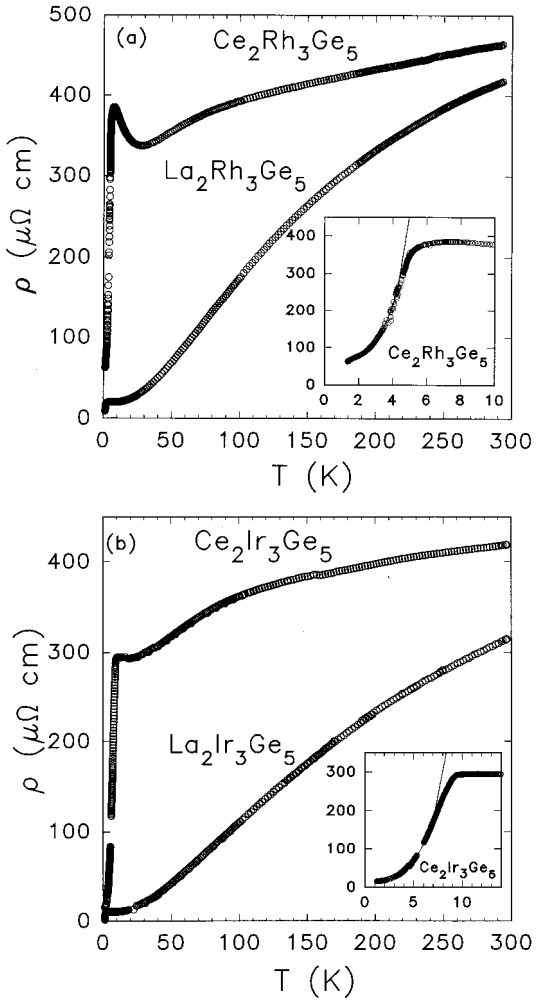

FIG. 3. Electrical resistivity of (a) $Ce_2Rh_3Ge_5$ and (b) $Ce_2Ir_3Ge_5$ together with the lanthanum analogs in the temperature interval 1.3–300 K. Insets show expanded view of the low-temperature part of the resistivity. Solid lines show fit of the data to the equation $\rho = \rho_0 + AT^2 + BT^5$.

down to 1.3 K in $LaRhGe_3$ in agreement with the results of Muro *et al.*[16] However, we found a drop of resistivity in a sample of $LaRh_2Ge_2$ around the same temperature as that found in $La_2Rh_3Ge_5$ and in this case also the resistivity does not go to zero down to 1.3 K. Thus it is not possible from the present investigation to state whether the superconductivity is intrinsic to $La_2Rh_3Ge_5$ or due to the presence of a minor impurity phase.

The magnetic contribution to the resistivity ρ_m is obtained by subtracting the resistivity of the isostructural nonmagnetic $La_2M_3Ge_5$ from that of $Ce_2M_3Ge_5$ (M = Rh, Ir). As shown in Fig. 4, ρ_m varies as $-\ln T$ in two different temperature regions which are separated by a hump at about 60 and 90 K, respectively. According to the model of Cornut and Coqblin,[15] such a behavior can be expected for Kondo-type interaction in the presence of crystal field. The low-temperature $-\ln T$ behavior of ρ_m reflects the Kondo scattering in the crystal-field ground state, whereas that at high temperatures corresponds to the Kondo effect in the whole multiplet of the $J = \frac{5}{2}$ state. The high-T hump or maximum in the ρ_m vs $\ln T$ is a measure of the overall crystal-field splitting. The ratio of the low- and high-T slopes is given by

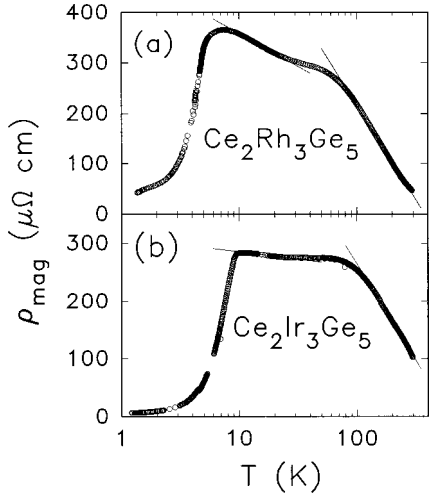

FIG. 4. Magnetic contribution to the electrical resistivity ρ_{mag} vs $\ln T$ for (a) $Ce_2Rh_3Ge_5$ and (b) $Ce_2Ir_3Ge_5$. Solid lines are fits to the equation $\rho_m(T) = \rho_0 - c_k \ln T$.

$(\alpha_1^2 - 1)/(\alpha_2^2 - 1)$ where α_1 and α_2 are the degeneracy of the $4f$ state at low temperatures and high temperatures, respectively. In the case of Ce compounds with noncubic symmetry, the sixfold degenerate multiplet splits into three doublets. Therefore for well-separated crystal-field levels three regions of $\ln T$ dependence of resistivity is expected as was seen in Ce_2Cu_2In.[16] In practice, however, due to crystal-field levels not being well separated the intermediate region might not be seen in the resistivity curve. In such a case the ratio of the low-T and high-T slopes is $\frac{3}{35}$ if the ground state is a doublet and the ratio is $\frac{3}{7}$ if the ground state is a quartet.[15] Thus from the analysis of $\rho_m(T)$ it is possible to guess the degeneracy of the ground state.

We have analyzed the data of $\rho_m(T)$ by the expression $\rho_m(T) = \rho_0 - c_K \ln T$, where ρ_0 is the spin disorder resistivity and c_K is the Kondo coefficient. The fits to the above expression are shown as solid lines in the two regions in Fig. 4. For $Ce_2Rh_3Ge_5$ we find $c_K^{(1)} = 56.7\ \mu\Omega$ cm for 10–21 K and $c_K^{(2)} = 162.8\ \mu\Omega$ cm for 100–300 K. Their ratio is $\sim\frac{3}{9}$ which is close to $\frac{3}{7}$ (quartet) and far off from $\frac{3}{35}$ (doublet). However, the high-field magnetization results suggested the ground state to be a doublet. The ratio being closer to that expected for a quartet state is possibly due to the fact that the first excited doublet lies close to the ground-state doublet in case of $Ce_2Rh_3Ge_5$. This prediction needs to be verified by specific-heat and inelastic neutron-scattering measurements. In Fig. 4(b), two regions for $Ce_2Ir_3Ge_5$ are separated by a hump around 90. We found that the ratio of the two slopes is $\sim\frac{3}{40}$, being more close to $\frac{3}{35}$ than $\frac{3}{7}$. This suggests that the crystal-field ground state in $Ce_2Ir_3Ge_5$ is a doublet which is consistent with the high-field magnetization result. As mentioned above, the temperature at the hump, 60 and 90 K for $Ce_2Rh_3Ge_5$ and $Ce_2Ir_3Ge_5$, respectively, can be considered as the energy of the overall crystal-field splitting. We point out here that these temperatures are close to the values of the temperature below which the susceptibility deviates from the Curie-Weiss behavior.

Another interesting feature of the resistivity is the large drop below T_N in both compounds. Not only the ratio of $\rho_m(T_N)/\rho_m(1.3\,\mathrm{K})$ is as large as 7 and 25, respectively for

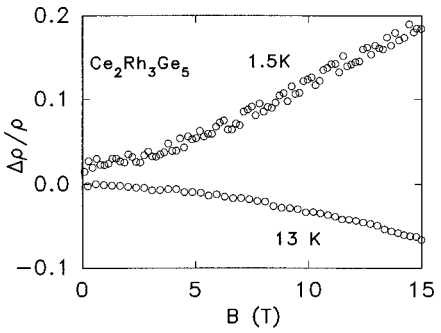

FIG. 5. Longitudinal magnetoresistance of $Ce_2Rh_3Ge_5$ at 1.5 and 13 K.

$Ce_2Rh_3Ge_5$ and $Ce_2Ir_3Ge_5$, but also the value of total drop across the magnetic transition is enormously large as ~ 300 $\mu\Omega$ cm for the both. Such a large drop in $\rho_m(T)$ below T_N was previously observed in Ce- and U-based compounds such as $CeAl_2$, $CePb_3$, $CeAg_2Sb_2$, CeB_6, UPd_2Al_3, and UPd_2Ga_3.[17–23] The large drop was attributed to reduction of the spin-disorder resistivity and possibly also to coherence effects. The importance of coherence in the magnetically ordered state has been demonstrated in UPd_2Al_3 and UPd_2Ga_3.[22,23] In these systems, the resistivity below T_N follows the expression $\rho(T)=\rho_0+AT^2+BT^5$ with a large value of $A\sim 1\ \mu\Omega\,\mathrm{cm\,K^{-2}}$. Our data for $Ce_2Rh_3Ge_5$ and $Ce_2Ir_3Ge_5$ also could be fitted well with this expression below 4.2 and 7.0 K, respectively, as is shown by solid lines in the inset. The fits provide the value of A as 4.9 and 1.8 $\mu\Omega\,\mathrm{cm\,K^{-2}}$ for $Ce_2Rh_3Ge_5$ and $Ce_2Ir_3Ge_5$, respectively. These values are comparable to those found in heavy-fermion compounds.

To examine the presence of coherence in the magnetically ordered state, we measured the magnetoresistance. Figure 5 shows the longitudinal magnetoresistance of $Ce_2Rh_3Ge_5$. At 1.5 K well below $T_N=5.5$ K, the magnetoresistance is positive, whereas it is negative at 13 K in the paramagnetic state. In dilute Kondo systems the magnetoresistance is negative at all temperatures which is due to freezing out of spin-flip scattering by the magnetic field. However, in Kondo-lattice systems like $CeAl_3$ and $CeCu_2Si_2$,[24,25] the magnetoresistance becomes positive at low temperature due to destruction of the coherent state by the application of field. For $Ce_2Rh_3Ge_5$, the magnitude of the positive magnetoresistance is as large as 18% at 15 T. We believe that this is due to addition of two components; one is the destruction of the coherent state by the application of magnetic field and the other is reduction of spin disorder scattering associated with the increase of the magnetization for one of the antiferromagnetic sublattice.

Figure 6 shows the specific heat of $Ce_2M_3Ge_5$ (M=Rh, Ir) and their nonmagnetic analogs below 20 K. The specific heat of the Ce compounds exhibits a λ-type anomaly around T_N which indicates magnetic phase transition. The jump ΔC due to magnetic ordering is much reduced compared to the expected value of 12.48 J/K mol for a purely magnetic system with $S=\frac{1}{2}$.[26] It has been shown theoretically that the magnitude of ΔC follows a systematic reduction as a function of T_K/T_N.[27] Using this model, we estimated the Kondo temperature to be 6 K for $Ce_2Rh_3Ge_5$ and 5 K for $Ce_2Ir_3Ge_5$. The magnetic entropy at T_N reaches to values of 50% and 65% of $R\ln 2$, respectively. Thus there is a considerable reduction of magnetic entropy due to the Kondo effect. In the inset, the specific-heat data is plotted as C/T vs T^2. From the data at temperatures 15–20 K, the linear coefficient is estimated to be $\gamma=148$ and 152 mJ/K^2 mol Ce for $Ce_2Rh_3Ge_5$ and $Ce_2Ir_3Ge_5$, respectively. These values are of the order of that found in moderate heavy-fermion systems exhibiting magnetic order. More remarkable is the fact that even in the magnetically ordered state the large γ values are maintained. In the range 1.8–4.0 K, the specific heat of $Ce_2Rh_3Ge_5$ is well represented by $C(T)=\gamma T+\beta T^3$ with $\gamma=103$ mJ/K^2 mol Ce. For $Ce_2Ir_3Ge_5$, we find $\gamma=67$ mJ/K^2 mol Ce for 1.8–8 K. This suggests that strongly correlated electronic states coexist with magnetic order in these systems, which is consistent with our interpretation of the resistivity and magnetoresistance above mentioned.

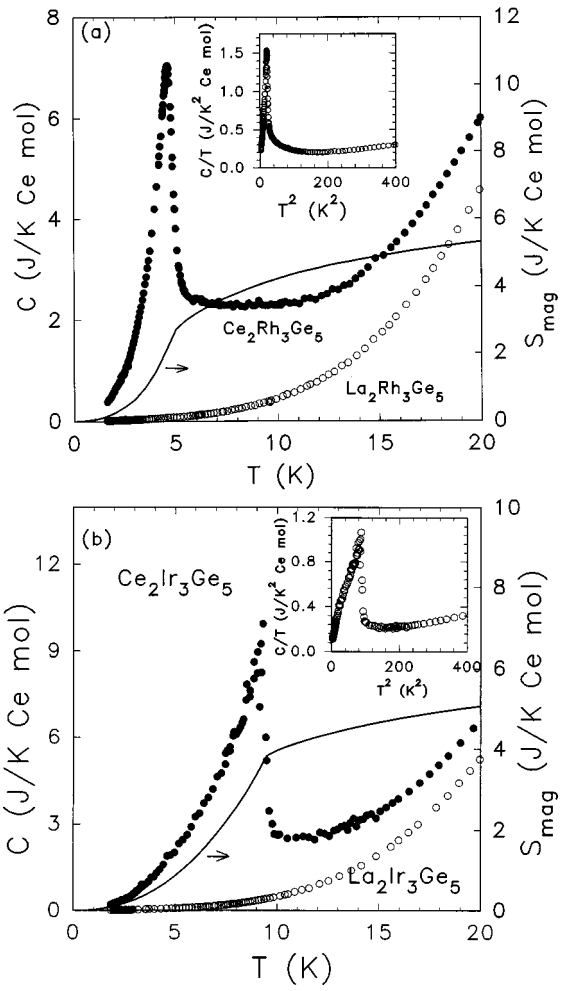

FIG. 6. Specific heat and magnetic entropy of (a) $Ce_2Rh_3Ge_5$ and (b) $Ce_2Ir_3Ge_5$ as a function of temperature. Inset shows C/T vs T^2.

IV. CONCLUSION

We have shown that $Ce_2Rh_3Ge_5$ and $Ce_2Ir_3Ge_5$ are antiferromagnetically ordered Kondo compounds with $T_N=5.5$ and 9.5 K, respectively. The drop of resistivity below T_N is as large as 300 $\mu\Omega$ cm in both compounds, which is attributable to the combined effect of magnetic order and development of coherence in the Kondo lattice. The presence of

coherence effect is supported by the observation of large positive magnetoresistance in the magnetically ordered state. Resistivity data of both compounds show typical temperature dependence expected for a Kondo-lattice compound in the presence of crystal-field splitting. From the analysis of resistivity, specific heat, and high-field magnetization, the ground state is suggested to be the Γ_7 doublet in both compounds. The specific-heat data also reveal the enhancement of $C/T \sim 150\,\mathrm{mJ/K^2\,mol\ Ce}$ in both compounds above T_N. By using the unversal relation between the value of the jump in specific heat as a function of T_K/T_N, the Kondo temperatures for the ground-state doublet is estimated to be 6 K for $Ce_2Rh_3Ge_5$ and 5 K for $Ce_2Ir_3Ge_5$. In the former, the Kondo interaction ($T_K = 6$ K) indeed competes with the magnetic interaction ($T_N = 5.5$ K). Further investigations are essential to determine the magnetic structures and the crystal-field level schemes in these compounds.

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