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# NEUTRON SCATTERING STUDY OF $Ce_3Au_3Sb_4$

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## ABSTRACT

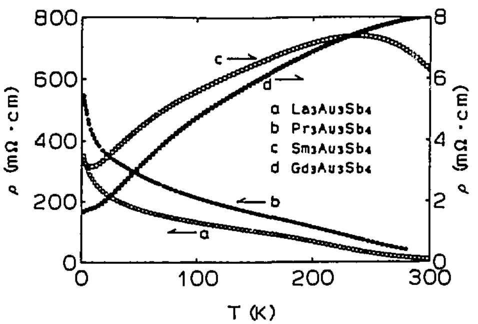
Rare-earth compounds with an $Y_3Au_3Sb_4$-type crystal structure are semiconductors or semi-metals. Among them, $Ce_3Au_3Sb_4$ is a semiconductor with an activation energy of about 640 K and shows no magnetic order down to 1.5 K. The magnetic part of the specific heat for $Ce_3Au_3Sb_4$ obtained by subtracting the value for $La_3Au_3Sb_4$ from the total specific heat of $Ce_3Au_3Sb_4$ shows a broad peak at around 10K, the origin of which is well explained by the crystalline-field splitting determined by neutron scattering.

## INTRODUCTION

From our experimental studies of new rare earth compounds with an $Y_3Au_3Sb_4$-type crystal structure, we have found that $La_3Au_3Sb_4$ and $Ce_3Au_3Sb_4$ are narrow gap semiconductors and the valence of the Ce atom is a well-defined 3+ state[1),2)]. These experimental findings are also consistent with results of the band calculation on $La_3Au_3Sb_4$; the valence band comes mainly from the Sb-5p electrons and is filled by the transfer of three electrons from a La atom and one electron from an Au atom, leading $La_3Au_3Sb_4$ chemically to be $La_3^{3+}Au_3^{1+}Sb_4^{3-}$ and to be a semiconductor with an energy gap of 1900 K[1),3)]. Then, the frame of the electronic structure of $Ce_3Au_3Sb_4$ may be the same as that of $La_3Au_3Sb_4$, but a localized $4f^1$ level is located within a filled valence band in the former. On the other hand, to study the mechanism of the appearance of Kondo resonance in Kondo insulators, theoretical studies have been performed from two extreme cases, i.e., one from nonmagnetic impurities, so-called Kondo holes, in Kondo insulator[4)] and the other from a single magnetic impurity in non-f nonmetallic system[5)6)]. Experimentally, however, systematic studies have not yet been done so far. In this sense, $Ce_3Au_3Sb_4$ is a good candidate for the study of the structures within a band gap in Kondo insulator. Therefore, it is important to investigate the ground-state properties of $Ce_3Au_3Sb_4$ itself more in detail. In this paper, we will present the experimental results of the electrical resistivity and the specific heat of $R_3Au_3Sb_4$ (R=La, Ce, Pr, Sm, Gd). Then, we will mention about an anomaly of the specific heat observed in $Ce_3Au_3Sb_4$. Finally, we will show that this anomaly can be well explained by crystalline-field splitting determined by recent neutron scattering.

## RESULTS AND DISCUSSION

First, we present the transport properties of non-f $La_3Au_3Sb_4$. As shown in Fig.1, $La_3Au_3Sb_4$ shows semiconducting behavior. Activation energy estimated above 200 K is 1540 K, which may correspond to the energy gap of $La_3Au_3Sb_4$, in agreement with the result of band calculation[1]. As for the



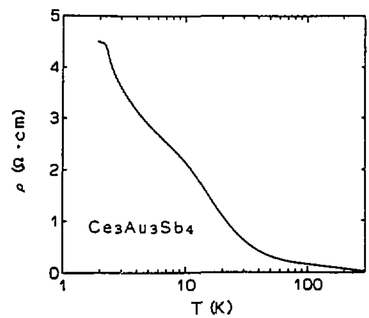




**Fig.1.** $\rho$ **(T) curves of** $R_3Au_3Sb_4$ **(after ref.2). Fig.2.** $\rho$ **(T) curve of** $Ce_3Au_3Sb_4$**.**

heavier rare-earth members, $Pr_3Au_3Sb_4$ also shows semiconducting behavior, whereas $Gd_3Au_3Sb_4$ is metallic and the resistivity of the latter is two orders of magnitude smaller than that of the former. The present results indicate that $R_3Au_3Sb_4$ systems show a gradual evolution from semiconducting towards semi-metallic behavior as R is varied from La to Gd[2]. In Fig.2, we present temperature dependence of electrical resistivity of $Ce_3Au_3Sb_4$. The resistivity is 26 mΩ cm at room temperature and increases up to 4.5 Ω cm at 2 K and 15 Ω cm at 0.5 K[7]. The ratio of resistance R(0.5 K)/R(300 K) is 577. On the other hand, the temperature dependence of magnetic susceptibility $\chi$ (T) above 150 K can be well described by the Curie-Weiss law with $\Theta_p$ of 0.4 K and effective number of Bohr magneton of 2.45. The latter value is comparable with 2.54 for the free ions of $Ce^{3+}$. This result suggests that the valence of the Ce ions in $Ce_3Au_3Sb_4$ is 3+[1]. These results are the reason why we consider the frame of the electronic structure of $Ce_3Au_3Sb_4$ to be the same as $La_3Au_3Sb_4$ with a $4f^1$ level in the filled valence band.

Next, we present experimental results of the specific heat C of $R_3Au_3Sb_4$. As is seen from Fig.3, C of $Sm_3Au_3Sb_4$ and $Gd_3Au_3Sb_4$ show $\lambda$-type anomaly peaking at 2 and 10.3 K, respectively, which correspond to magnetic ordering temperatures. It is interesting to note that there is a minimum at about 3 K in the C(T) curve of $Ce_3Au_3Sb_4$; a sharp upturn in C/T is observed at low temperatures with C/T exceeding 2.5 J/mole $K^2$. The origin of this remains a future task. Another prominent feature is that the magnetic part of the specific heat of $Ce_3Au_3Sb_4$ obtained by subtracting the value for $La_3Au_3Sb_4$ from the total specific heat of $Ce_3Au_3Sb_4$ shows a broad peak at around 10 K. A plausible explanation on the origin of it is that the peak comes from Schottky anomaly due to the crystalline-field splitting into three doublets, because local symmetry of rare-earth site in $Y_3Au_3Sb_4$-type structure is tetragonal $\bar{4}$ site.

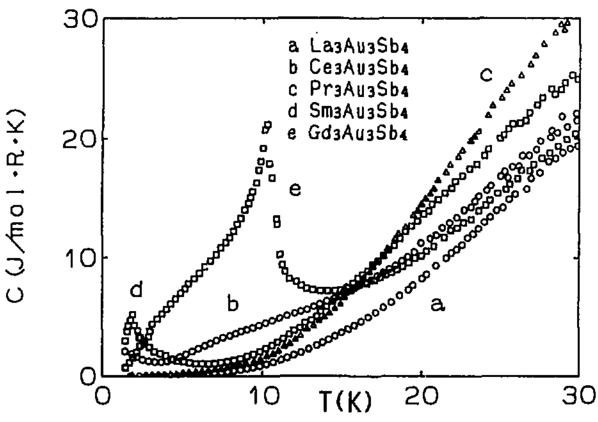



**Fig.3. Specific heat C vs T for $R_3Au_3Sb_4$ (R=La, Ce, Pr, Sm, Gd) (after ref.2)**

In fact, the inverse magnetic susceptibility $1/\chi$ vs T curve deviates slightly from the Curie-Weiss law below 150 K[1] and we predicted that overall splitting of the J=5/2 state into three doublets by the crystallne field may be less than 150 K[1].

To determine the crystalline-field splitting of $Ce_3Au_3Sb_4$, we performed neutron scattering experiment by using the spectrometer LAM-D at the spallation neutron source KENS in the National Laboratory for High Energy Physics. Well-defined crystal field excitations were observed at 22 and 100 K. Crystal-field excitation levels are at 2.26, 10.37 and 12.47

meV. Based on the temperature dependence of the intensity, these excitations are attributed to be of magnetic origin and they correspond to crystalline-field transitions between the three doublets of $Ce^{3+}$ (J=5/2). Analysis of the specific heat by this crystalline field is shown in Fig.4. Overall features of the magnetic specific heat are quite well explained by the crystalline field.

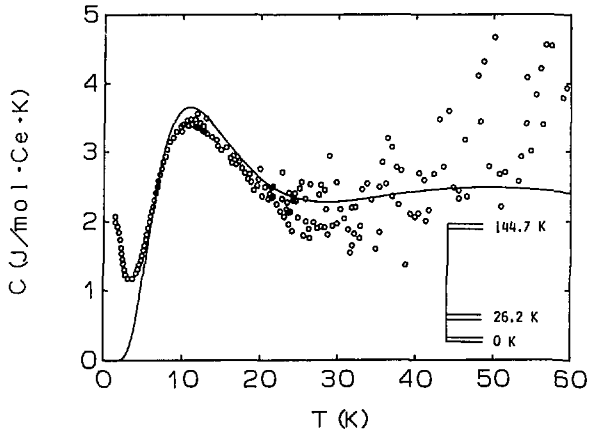



**Fig.4.Observed magnetic specific heat of $Ce_3Au_3Sb_4$ (open circles) and calculated one(solid line) based on the experimental results of neutron excitation(inset).**

As is shown in Fig.4, it is interesting to note that the magnetic specific heat increses below 3 K. To determine whether the upturn of the specific heat below 3 K is due to a Kondo resonance in a Kondo insulator or due to onset of magnetic ordering, elastic neutron scattering experiments are now in progress.

## R E F E R E N C E S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