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Study on the electronic state of rare earth pnictides which show the charge-ordering

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Yb_4As_3 is known as a mixed valence compound with Yb^{2+} and Yb^{3+} ions in a ratio of about 3:1. It shows a charge-ordering(Fig.1) below around 290K. Magnetic properties of Yb_4As_3 have been well understood, however, transport properties are not comprehended, for example, the origin of its hole carrier. To understand the transport properties, an investigation of the electronic state is important. If the ratio of Yb^{2+} and Yb^{3+} ions is exact 3:1, Yb_4As_3 should become a semiconductor or semi-metal. On the other hand, if the ratio deviates from 3:1, the carrier may appear. In this case, a relationship between magnetic and transport properties may be expected. Therefore, purpose of our study is to clarify the electronic state of the whole charge-ordering rare earth pnictides which Yb_4As_3 represents.

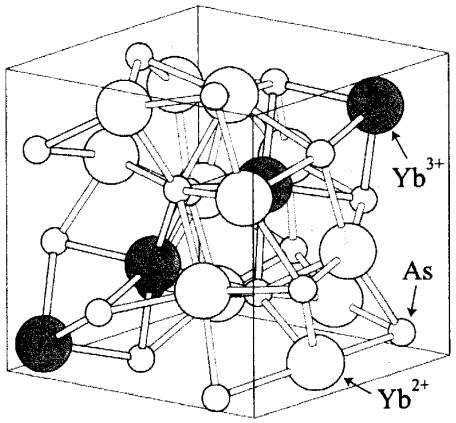

Fig.1: The charge ordered state of Yb_4As_3.

In Yb_4As_3, only hole Fermi surface formed by p-band of Arsenic which was observed by Shubnikov de Haas (SdH) effect until now. In our study, we tried to clarify the whole electronic state near the Fermi level using two different mixed crystals.

The first mixed crystal is $Yb_4(As_{1-x}Se_x)_3$ in which electron-doping effect is expected. With increasing Selenium concentration, the magnetic susceptibility decreases and the lattice constant increases. These results indicate the valence change from Yb^{3+} to Yb^{2+} with increasing Selenium concentration. On the other hand, the results of the SdH and Hall effect measurements indicate the increase of hole carrier number with increasing Selenium concentration. However, the increase of hole carrier number is one order smaller than the increase of Yb^{2+} ions. Therefore, the main effect of Selenium-substitution is considered to be electron doping to the 4f-level of Ytterbium.

The second mixed crystal is $Yb_4(As_{1-x}Sb_x)_3$. The SdH oscillation was observed in the Antimony 5%(Fig.2) and 8% substituted samples. The observed Fermi surface expands with increasing Antimony concentration. As this tendency agrees with the result of the Hall effect measurement, the observed Fermi surface is considered to correspond to the hole surface. Furthermore, the increase of the hole carrier number corresponds to the increase of the Yb^{2+} ions estimated from the magnetic susceptibility.

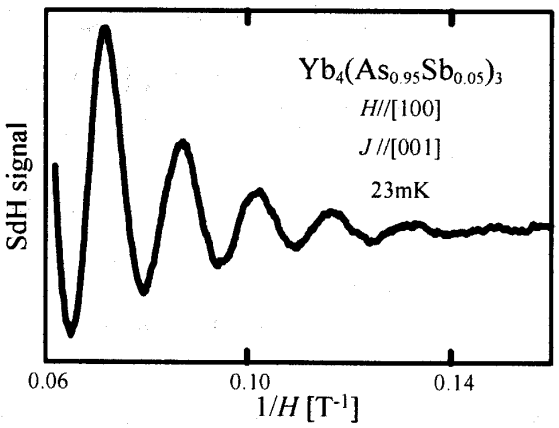

Fig.2 : The SdH oscillation of $Yb(As_{0.95}Sb_{0.05})_3$.

From the above mentioned, in Yb_4As_3, it is concluded that the deviation of the ratio of the Yb^{2+} and Yb^{3+} ions from 3:1 creates the hole carrier on the p-band of Arsenic.

The SdH oscillations were observed in Sm_4Bi_3, which shows the charge-ordering transition. As the carrier number estimated from observed four Fermi surfaces is good agreement with the result of the Hall effect measurement, they are considered to be the hole surfaces. The electron surface was not detected. As the change ratio of the magnetic susceptibility corresponds to the change of hole carrier number at the charge-ordering temperature, the similar electronic state as that of Yb_4As_3 is considered to be realized in Sm_4Bi_3.

The electronic state of Yb_4Sb_3, which is related with the high temperature valence fluctuating phase of Yb_4As_3, is clarified by the de Haas-van Alphen effect measurement. In Yb_4Sb_3, as the relative position of the 4f-band to the top of the p-band shifts lower compared to that of Yb_4As_3, the system falls into the valence fluctuating state due to a strong hybridization between the 4f-band and p-band.

We searched for a new charge-ordering rare earth pnictides. Its discovery may give us new information concerning the origin of the charge-ordering formation. Eu_4As_3 has been pointed out to be one of charge-ordering rare earth pnictides, however, its physical properties has not been clear because its crystallizing method had not been established yet. We clarified its melting behavior and established the crystallization method. Using obtained crystal we found that Eu_4As_3 shows the same charge-ordering transition at around 340K as those of Yb_4As_3 and Sm_4Bi_3. The charge-ordering of Eu_4As_3 indicates that the orbital degrees of freedom is not concerned with the formation of the charge-ordering. The positive and large Hall coefficient suggest that Eu_4As_3 may have the same electronic state as those of Yb_4As_3 and Sm_4Bi_3. At around 18K, it indicates a ferromagnetic transition. From the analysis of the magnetic susceptibility, it is cleared that a ferromagnetic interaction between Eu^{2+} and Eu^{3+} plays an important role in its magnetism.