(27a3)

Present status and a plan for single crystal preparation of filled skutterudite compounds

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Filled skutterudite compounds RT_4X_{12} (R=rare-earth; T=Fe, Ru and Os; X=P, As and Sb) have attracted much attention because of their interesting anomalous physical properties, such as metal-insulator transition in $PrRu_4P_{12}$, unusual Pr-based heavy fermion (HF) behavior in $PrFe_4P_{12}$ and HF-superconductivity in $PrOs_4Sb_{12}$. The quality of samples is the most important starting point in the reliable discussion of new exotic phenomena. We have grown high quality single crystals of the filled skutterudites by the Sn-flux method for phosphides and the Sb-self-flux method for antimonides, using high purity elements, 4N(99.99% pure) rare-earth, 4N transition metal and 6N pnictogen. One of the most reliable tests to examine the quality of samples is the de Haas-van Alphen (dHvA) experiment. We have succeeded in observing the dHvA effect in ten filled skutterudites and investigated the anomalous physical properties so far. These single crystals are distributed to other researchers and specialized measurements, such as photoemission spectroscopy, ultrasonic experiment, neutron scattering, nuclear magnetic resonance, thermal conductivity, muon spin resonance etc., have been performed. Their attractive properties have been summarized in Table 1.

Under the ambient pressure, the single crystal growing of filled skutterudites is limited; i.e., we could only grow RFe_4P_{12}, RRu_4P_{12}, RRu_4Sb_{12} and ROs_4Sb_{12} with light rare-earth (R=La-Sm) elements. High-pressure synthesis is a powerful technique to prepare the materials. Shirotani *et al.* have succeeded to synthesize polycrystalline samples of RFe_4P_{12} and RRu_4P_{12} with heavy rare-earth and RRu_4As_{12} with light rare-earth elements by this technique, and investigated the physical properties. In the next stage, we have a plan to grow single crystals using the high-pressure synthesis technique.

Table 1: Physical properties of filled skutterudites with light rare-earth (SC: superconducting, semi-C: semiconducting, semi-M: semimetallic, NMO: non-magnetic order, FM: ferromagnetic, AFM: antiferromagnetic, M-I: metal-insulator transition, HF: heavy fermion, NFL: non-Fermi liquid). There is more information available at "http://skut.phys.metro-u.ac.jp/database/database.html".

R	La	Ce	Pr	Nd	Sm
RFe_4P_{12}	SC*	semi-C	NMO(HF)*	FM*	FM(HF)*
	4.1K	0.107eV	6.5K	2K	1.6K
RRu_4P_{12}	SC*	semi-C	M-I	FM	M-I,AFM
	7.2K	0.074eV	62K	1.6K	16K
RRu_4Sb_{12}	SC*	semi-M*	SC*	FM	
	3.58K	NFL	1.3K	2K	
ROs_4Sb_{12}	SC*	semi-C	SC(HF)*	FM	
	0.74K	0.001eV	1.85K	0.8K	

*dHvA experiment has been done.