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Report on my staying in the Kinso Laboratory, Division of Chemistry, Graduate School of Science

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Abstract. This article is a report for my staying as a Distinguished Visiting Professor in the Division of Chemistry, financed by the International Research Unit of Advanced Future Studies. A summary of scientific activity and some reserach results done during my staying is provided. The future collabration between Kinso Laboratory and my group is discussed in details.

Keywords: Fe-based superconductors, Topological Materials, Itinerant Magnetism, Heavy-Fermion Compound.

1. Research Activity:

I worked as a distinguish visiting professor at the Kinso Laboratory, Division of Chemistry, Graduate School of Science, Kyoto University from Feb. 1 to March 31, 2016. This research stay was financed by the International Research Unit of Advanced Future Studies. I would like to express my appreciation to the Division of Chemistry for hosting me, to the International Research Unit of Advanced Future Studies for providing financial support, and to Professor Yoshimura for inviting me.

During these two months, I have a chance to discuss with Prof. Yoshimura about unconventional superconductors, itinerant magnetism and topological materials research. I learnt a lot of about magnetism and neutron magnetic resonance (NMR) theory, and technique for preparing various materials and measurements from Prof. Yoshimura's group. Prof. Yoshimura and I discussed about the future collaboration between his group and my group in Zhejiang University on these fields.

In the past two months, I took part in three workshops and give two talks in these meeting. One is on the International Symposium on Advanced Future Studies, held on Feb., 12, 2016, Kyoto. My talk topic is the Exploration of (Fe,Ni)-Chalcogenide Superconductors: Fe-vacancy order, new AFM states and SC, As we know, cuprates and Fe-based compounds are two families with highest superconducting (SC) transition temperatures. A common feature in both families is that the superconductivity emerges as antiferromagnetic (AFM) long range order is suppressed. While the parent compound of cuprates is a Mott insulator where the electron repulsion is strong, the parent compound of Fe-based materials is metallic implying weak or moderate electron correlation. A key strategy to develop a unified picture for the Fe- and Cu-based high temperature superconductivity

(HTSC) is to explore the possibility to tune the Fe-based compound into an insulator. The relationship between the antiferromagntic ground state in the Fe-chalcogenides, which is different from that in the Fe-pnictides, and superconductivity is another issue. In this presentation, firstly, I talked about our discovery of superconductivity with T_C = 14 K, determining of the lattice, magnetic structures in the parent of Fe(Te,Se,S) system and the correlation between bi-collinear AFM order and superconductivity in this system. Secondly, I discussed about our efforts on searching for new Fe-chalcogenides with AFM insulating behaviour, such as $La_2O_3Fe_2Se(S)_2$ compounds. Third, I reported our discovery of superconductivity above 30K in (Tl,K,Rb)Fe_xSe_2 system, which the onset SC transition temperature is as high as 40K. While the compound with more Fe vacancies shows an AFM insulator behaviour, which may be associated with the Fe-vacancy ordering in the crystals. Our discovery represents the first Fe-based HTSC at the verge of an AFM insulator. A review on the results of Fe-vacancy super-lattice, magnetism and superconductivity in (Tl,K,Rb)Fe_xSe_2 system was presented in this talk. Finally, I reported the our recently discovery of superconductivity in the $TlNi_2(Se,S)_2$ system.

The second is on the International Workshop on Advanced Future Studies, held on March 14-16, 2016, Kyoto. My talk topic is Topological Materials. It is well known that the discovery and classification of distinctive phases of matter is the main purpose of the condensed-matter physics. Over the past 30 years, the study of the quantum Hall effect has led to a different classification paradigm based on the notion of topological order. The quantum Hall effect defines a topological phase in the sense that certain fundamental properties are insensitive to smooth changes in material parameters and cannot change unless the system passes through a quantum phase transition. In the past 10 years, a new field has emerged in condensed-matter physics based on the realization that the spin-orbit interaction can lead to topological insulating electronic phases, and on the prediction and observation of these phases in real materials. And many new topological materials have been discovered, such as 3D topological insulators, Bi_2Se_3; 3D topological semimetals, Cd_2As_3; 3D Weyl semimetals TaAs, NbP, TaP, NaAs. In this presentation, I first gave a review about this new matter state, such as what is topological material, the relationship between the topology in mathematics and topological materials in the condensed matter physics et al. Secondly, I showed an example to explain how to construct the topological materials. Finally, I reported our recent work on the Weyl semimetal TaP.

Especially, in these workshops, many talks given by the professors from various field, such as economic, psychology, information, ecological, language, biology, physics, chemistry, let me know that the researches in various fields have some comment characteristics. It is very important to exchange with each other at the same platform. Some new good ideas may be burst out after discussions. I should give my gratitude to the International Research Unit of Advanced Future Studies offering these opportunities.

At the same time, I also took part in the Workshop of the Low Temperature Centre, Graduate School of Science, and the each seminar every week in the Prof. Yoshimura's group. I learnt many things from the speakers. For example, Haraguchi's work on the spin-liquid behaviour in the spin-frustrated Mo_3 cluster magnet $Li_2ScMo_3O_8$ in contrast to magnetic ordering in isomorphic $Li_2InMo_3O_8$ told us a new way to realize magnetic ground state. Of course, in these seminars, I gave the students some suggestions according to my previous experience, which may be helpful for their research in the future.

2. Research Results

During the two months when I stay in Kinso Laboratory, after discussing with Prof. Yoshimura, I finished the two papers. One is about the research on "the unsaturated positive and negative magnetoresistance in Weyl semimetal TaP", which has be published in Sci. China-Phys. Mech. Astro. 59, 657406 (2016). In Weyl semimetal (WSM) phase, the bulk electronic bands disperse linearly along the momentum direction through a node, called the Weyl point, in a three-dimensional (3D) analog of graphere. It can be viewed as an intermediate phase between a trivial insulator and topological insulator. A number of candidates for a WSM have previously been proposed, such as $Y_2Ir_2O_7$ and $HgCr_2Se_4$ compounds in which the magnetic order breaks the time-reversal symmetry, and the $LaBi_{1-x}Sb_xTe_3$ compound in which fine-tuning the chemical composition is necessary to break the inversion symmetry. However, none of these compounds have been used to realize a WSM experimentally. This is because the magnetic domain is not large or it is very difficult to tune the chemical composition within 5%. Very recently, the theoretical proposal for a WSM in a class of stoichiometric materials including TaAs, TaP, NbAs, and NbP that break crystalline inversion symmetry was confirmed in the experiments, except for TaP. This was due to the difficulty of growing large crystals of TaP. The exotic transport properties exhibited by these materials has ignited extensive interest in both the condensed matter physics and material science communities, especially because of their extremely large magnetoresistance (MR) and ultrahigh mobility of charge carriers.

TaP crystallizes in a body-centered tetragonal lattice with the non-symmorphic space group *I4*1*md,* which lacks inversion symmetry. We grew successfully single-crystal TaP by using a chemical vapor transport method. Then, we measured its longitudinal resistivity (ρ_{xx}) and Hall resistivity (ρ_{yx}) at magnetic fields up to 9 T in the temperature range of 2–300K. At 8 T, the magnetoresistance (MR) reached 3.28×10^5 % at 2 K, 176 % at 300 K. Neither value appeared saturated. We confirmed that TaP is a hole-electron compensated semimetal with a low carrier concentration and high hole mobility of $\mu_h = 3.71 \times 10^5$ cm^2/V s, and found that a magnetic-field-induced metal-insulator transition occurs at room temperature. Remarkably, because a magnetic field (H) was applied in parallel to the electric field (E), a negative MR due to a chiral anomaly was observed and reached -3000% at 9 T without any sign of saturation, either, which is in contrast to other Weyl semimetals (WSMs). The analysis of the Shubnikov-de Haas (SdH) oscillations superimposed on the MR revealed that a nontrivial Berry's phase with a strong offset of 0.3958, which is the characteristic feature of charge carriers enclosing a Weyl node. These results indicate that TaP is a promising candidate not only for revealing fundamental physics of the WSM state but also for some novel applications.

Another research is on the ferromagnetic quantum critical behavior in heavy-fermion compounds $CeTi_{1-x}Ni_xGe_3$. In the past 30 years, quantum criticality raises continuously scientist research interest because it is believed to be the origin of the many exotic emergent phenomena in the modern condensed-matter physics, such as quantum phase transition (QPT), new hidden order, unconventional superconductivity and the breakdown of the Fermi liquid model. The QPT, which occurs at zero kelvin, is driven by quantum fluctuations, in contrast to the phase transition emerging at finite temperature, driven by thermal fluctuations. The existence of ferromagnets with a lower T_C makes the ferromagnetic quantum criticality to be visible, because their T_Cs are easily suppressed to zero kelvin by tuning the external parameters, such as chemical composition, or pressure. Most of the experimental studies on the QPT were devoted to the stoichiometric compounds based on ytterbium, uranium or cerium, which show antiferromagnetic (AFM) correlation. On the other hand, ferromagnetic QPT is quite unusual both from the experimental and theoretical point of view. Therefore it is especially attractive to investigate ferromagnetic alloys in which Curie-temperature (T_C) can be driven to zero kelvin by tuning external parameters. For example, in $URhGe_5$ $UCoGe_6$, and UGe_2, the pressure tuned ferromagnetic QPT has been realized. And in $CePd_{1-x}Rh_x$, $CePd_{1-x}Ni_x$, $CeTi_{1-x}V_xGe_3$, $YbNi_4P_2$, $URu_{2-x}Re_xSi_2$ and $UCo_{1-x}Fe_xGe_{13}$, the T_C can be driven to zero kelvin by a chemical substitution.

$CeTiGe_3$ is an anisotropic ferromagnetic Kondo-lattice system with T_C = 14 K at ambient pressure. It crystallizes in the $BaNiO_3$ type structure (hexagonal perovskite, $P6_3/mmc$), which is consist of one-dimensional chains of face shearing Ti-centered octahedra stacked along the *c*- axis. The short distance between Ti-Ti atoms has been taken as a sign of a weak metal-metal bonding. The spins of Ce^{3+} ions with a moment of $(1.5\pm0.1)\mu_B$/Ce order ferromagnetically at low temperatures. It was found that ferromagnetic order in $CeTiGe_3$ is difficult to be suppressed by applying hydrostatic pressure. While $CeNiGe_3$ is an antiferromagnet with a Néel temperature T_N = 5.5 K due to the localized 4*f* magnetic moment. $CeNiGe_3$ crystallizes in the $SmNiGe_3$ type orthorhombic crystal structure. Alloys of $CeTi_{1-x}Ni_xGe_3$ ($0.0 \leq x \leq 0.45$) were synthesized by an arc-melting method under an argon (Ar) atmosphere. X-ray powder diffraction (XRD) pattern for all the samples was recorded at room temperature with a X-ray diffractometer. Analysis of the XRD data was made by using the GSAS suite of Rietveld programs. The magnetization measurements were carried out by a Quantum Design MPMS (SQUID). The magnetic susceptibility of all the samples was measured at a 1000 Oe magnetic field with a process of field-cooling (FC). The resistivity and heat capacity measurements were carried out in a Quantum Design Physical Properties Measurement System (PPMS). It was found that the Curie temperature, T_C, decreases with increasing Ni content, and reaches to zero kelvin near a critical content $x_{cr} = 0.44$. A new phase diagram was constructed based on the results of magnetization (M), resistivity (ρ) and specific heat (C) measurements for this system. The non-Fermi liquid behavior in $\rho(T)$, and $1nT$ relationship in $C(T)$ in the samples near x_{cr}, demonstrate that strong spin fluctuation emerges in these samples, indicating them to be near a quantum critical point (QCP). This work will be published in Phys. Rev. B.

I discussed with Prof. Yoshimura about the future collaboration on these two systems. NMR is the unique tool to determine microscopically the spin state of electrons. Prof. Yoshimura's group has obtained many important results by using NMR measurement, and their works on magnetism by NMR measurements has a board impact in the world. We plan to measure the spin texture structure in the Weyl semimetal TaP by using NMR technique in his group, which may discover some exotic quantum electronic states in this new condensed matter state. Another, in order to explore the mechanism of the non-Fermi liquid behavior near quantum critical point in the $CeTi_{1-x}Ni_xGe_3$ alloys, we are going to observe NMR spectrum at different temperatures in his laboratory. We are looking forwards to obtaining many interesting results through our collaboration on these issues.

3. Summary

During the past two months staying in Kinso Laboratory, I learnt many things and obtained some interesting research results. I would like to thank again the International Research Unit of Advanced Future Studies for providing financial support, Professor Yoshimura for inviting me.