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Report on my stay at Kinso Laboratory, Division of Chemistry, Graduate School of Science

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Abstract. This article is a report of my 5-week stay as a Distinguished Visiting Senior Lecturer at the Division of Chemistry, Graduate School of Science supported financially by the International Research Unit of Advanced Future Studies. A summary of scientific activities and some avenues for future research collaboration between Kinso Laboratory and my group is discussed.

Keywords: Magnetoresistance, structural quantum phase transition, superconductivity, high pressure

1. Research Activity

I worked as a distinguish visiting senior lecturer at Kinso Laboratory, Division of Chemistry, Graduate School of Science, Kyoto University from 7 May 2018 to 9 June 2018. This research stay was funded by the International Research Unit of Advanced Future Studies. It is my great honour to have the opportunity to visit Kinso Laboratory for such an extended period. I would like to express my gratitude to the Division of Chemistry for hosting me, to the International Research Unit of Advanced Future Studies for providing financial support, and to Professor Kazuyoshi Yoshimura for inviting me.

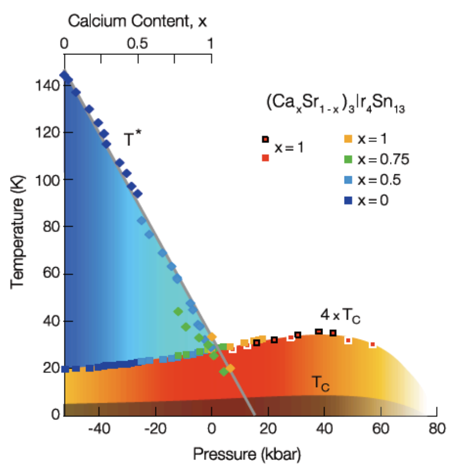
During my 5-week stay, I have the opportunity to interact with Prof. Yoshimura, Assoc. Prof. Ueda, Asst. Prof. Michioka, and students of their group. My group at The Chinese University of Hong Kong (CUHK) is interested in measuring high quality single crystals under multiple extreme conditions. Kinso Laboratory has the capability of growing high quality single crystals. Therefore, we possess complementary research interests and skills. In fact, I have enjoyed several years of productive collaboration with Kinso Laboratory, and have published several papers with Prof. Yoshimura [1-6]. One of the key objectives of this visit is to identify future compounds to continue our collaboration.

During my stay, I gave three talks. On 22 May 2018, I gave a seminar on our recent discovery of a quasilinear quantum magnetoresistance in CrAs [7] at Department of Physics, Kyoto University. On 25 May 2018, I gave a seminar at Department of Chemistry, Kyoto University, to describe my work with Prof. Yoshimura on 3-4-13 systems [1-6]. I thank Prof. Youichi Yanase and Prof. Yoshimura for organising the seminar at Department of Physics and Department of Chemistry, respectively. On 18 May 2018, I have the priviledge of giving an invited talk at “The Second International Workshop on

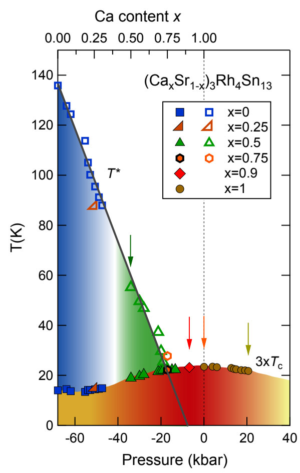
Itinerant-Electron Magnetism". This workshop was organised to celebrate Prof. Yoshimura's 60th birthday. Thus, it was a delightful experience to describe my work with him over the past few years on such a special occasion. After the workshop, we had a party at Kyoto Brighton Hotel. As a foreigner, I find the party a very unique experience, and it offers a nice chance to experience traditional Japanese custom and culture. Importantly, at both the workshop and the party, I met many former students of Prof. Yoshimura, many of them are well established, highly regarded scientists in their own research field. These interactions may open up future collaborative projects between Hong Kong and Japan.

More informally, I interacted with several students of Kinso Laboratory. I have been interested in a class of quasi-skutterudite compounds with $M_3T_4Sn_{13}$ stoichiometry, where M=Ca, Sr, La and T=Ir, Rh, Co. This class of materials exhibits an interesting interplay between superconductivity and structural order. For example, $Sr_3Ir_4Sn_{13}$ shows an anomaly in the electrical resistivity at T^*=147 K, which was identified as a structural transition. When Sr is substituted by Ca, or when pressure is applied, T^* decreases rapidly while the superconducting transition temperature T_c increases. T^* eventually reaches 0 K when a physical pressure of ~18 kbar acts on $Ca_3Ir_4Sn_{13}$ (see Figure 1a). In 2012, together with the authors of Ref. [1], we identified the formation of a structural quantum critical point in material class.

In 2015, we observed the evidence of structural quantum critical point in another series: $(Ca_xSr_{1-x})_3$**Rh**$_4Sn_{13}$ [2]. Similar to the case of the Ir-series, both T^* and T_c of the Rh-series are also tunable by either applied pressure or the Ca content (*c.f.* Figure 1b). However, one important difference must be noted. As can be seen from Figure 1, T^* extrapolates to 0 K when the Ca fraction is tuned to ~0.9. Therefore, the structural quantum critical point can be reached without the need of applying pressure, in contrast to the Ir-series where physical pressure must be applied to reach the structural quantum critical point. This has a far-reaching impact: the accessibility to the structural quantum critical point without the need of applying pressure enables detailed investigations by a wide range of experimental probes. For instance, we have used heat capacity to examine the Rh-series straddling the structural quantum critical point [3]. Similar studies under pressure (*e.g.* on the Ir-series) would be a lot more challenging. This shows that by clever chemistry, one can explore the same physics with a slightly different set of compounds. Since I have the chance to be in an environment surrounded by professional chemists, we naturally explored various ways to perturb the material system **chemically** in order to widen the material base for further detailed studies of structural quantum criticality. In particular, we would like to develop materials to explore the part of the phase diagram on the right hand side of the structural quantum critical point (the 'quantum disordered' side) at ambient pressure.



(a)



(b)

Figure 1. *Temperature-pressure phase diagram of (a) $(Ca,Sr)_3Ir_4Sn_{13}$ and (b) $(Ca,Sr)_3Rh_4Sn_{13}$. The phase diagrams are adapted from Refs. [1] and [2], respectively.*

I enjoy many 'coffee breaks' with Asst. Prof. Chishiro Michioka. During these breaks, I learnt the physics and chemistry of transition metal dichalcogenides from him. Asst. Prof. Michioka explains the way to synthesize single crystals of transition metal dichalcogenides with various intercalants. We also explored the idea of intercalation using organic molecules, and to prepare transition metal dichalcogenices based misfit-layer compounds for studies under extreme conditions.

Finally, I organised a mini-symposium with Kinso Laboratory on 6 Jun 2018. Three members from CUHK visited Kinso Laboratory and presented their research results, and two members from Kinso Laboratory gave their talks. The topics include some recent inelastic X-ray results on $(Ca_xSr_{1-x})_3Rh_4Sn_{13}$, new high-pressure resistivity data on a strong-coupling superconductor, and some transport and magnetic data on a heavy fermion systems. After the symposium, CUHK members had a tour of Kinso Laboratory. We thank Prof. Michioka for showing us the NMR laboratory and explaining to us the principles behind NMR experiments. We thank Prof. Ueda for showing us the crystal growth facilities, and for giving us some tips on chemical vapour transport technique for preparing large single crystals. From these laboratory visits, we gained some inspirations on several experimental designs that can be useful for our laboratory in Hong Kong.

2. Research Results

During my 5-week stay, I finished and submitted one paper that reports the extremely large magnetoresistance in a particular semimetal RSb (R=rare earth), and the complete determination of its Fermi surface topology. I also discussed with Prof. Yoshimura the content of this paper, and the design of the experiment. I will briefly describe the results of this paper below.

As mentioned in the preceding section, my group recently becomes attracted to the physics of magnetoresistance. Our laboratory in Hong Kong is well equipped for this sort of investigation, which requires high magnetic field and low temperatures. The availability of pressure can tune the bandstructure further, allowing us to extract key component(s) responsible for the observed magnetoresistance behaviour. Using crystals provided by Prof. C. S. Lue's group at National Cheng Kung University, we measured the magnetoresistance of single crystalline RSb. In contrast to CrAs described earlier, RSb exhibits a more conventional quadratic magnetoresistance. However, in RSb, the magnitude of the magnetoresistance is rather large: at 2 K, the magnetoresistance reaches ~28000% at 14 T, and it is non-saturating at 14 T.

On top of the large magnetoresistance, Shubnikov-de Haas oscillations can be resolved. The oscillation spectrum is rich, allowing the determination of the Fermi surface. By comparing with the density functional theory calculations performed by my colleague Prof. J. Y. Zhu and his group at CUHK, we successfully identified all Fermi surface sheets, from which we can calculate the carrier density. We found that the hole carrier density is almost the same as the electron carrier density. Therefore, the RSb we studied is a compensated semimetal, and a two-band model can be applied to explain its quadratic, large, non-saturating magnetoresistance.

In addition to RSb, I also worked on CrP. CrP is the high pressure analog of CrAs. On the CrAs end, we observed a very interesting quasilinear magnetoresistance, which we interpret as coming from a small pocket reaching the extreme quantum limit. In CrP, however, the magnetoresistance is quadratic, and in fact it shares certain similarities with the case of RSb. Although I have not been able to finish this paper during my stay, I have had many enlightening discussions with several colleagues, which help me to finalise the structure of this paper.

Finally, I stayed at SPring-8 with several members of Kinso Laboratory and CUHK on 6-8 June 2018. We used inelastic X-ray scattering to study the lattice dynamics of $(Ca_xSr_{1-x})_3Rh_4Sn_{13}$, and obtained very useful data for several crystals with different Ca content. These data will be analyzed and published in the near future.

3. Summary

My stay at Kinso Laboratory provides a chance to discuss with many scientists at Kyoto University. These discussions are helpful for understanding some of our new data, and for shaping the structure of several papers. The interactions also generate many ideas for future collaboration. Once again, I would like to thank the International Research Unit of Advanced Future Studies for providing financial support, and Professor Yoshimura and his group for hosting me.

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