Project Title:

Computational Studies of Muon Locations, Electronic Structures and Electron Transport in High Tc Superconductor, Organic, Organometallic and Biological Systems

Name:

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1. Background and purpose of the project, relationship of the project with other projects

Muon Spin Rotation/Resonance/Relaxation (μSR) experimental technique is a powerful method to study the properties of materials at microscopic level. The μSR method can be applied to study different aspects of semiconductors, organic magnets, high Tc superconductor and other molecular compounds. Various physical phenomena can be investigated such as hyperfine interactions, magnetism and electron transport. In many instances, the interpretation of μSR results requires further analysis through computational studies so that the underlying physics behind the observed properties could be understood. Therefore, the collaboration between experimental works and computational studies is crucial to achieve the ultimate goals of our group's research endeavour.

One of the main objectives for the computational works is to investigate the most probable stopping sites for muon and muonium. The calculated electronic structures of the muonated system from the computational works could be used to study the hyperfine interactions. Further, the comparison between the electronic structure of the pure and muonated systems would reveal much information about interactions between muon and its local environment.

The μSR experiments have been conducted at the RIKEN-RAL Muon Facility in the United Kingdom and the Paul Scherrer Institut (PSI) in Switzerland, while the computational studies require the excellent supercomputing facilities provided by the Advanced Center for Computing and Communication at RIKEN.

Our group has been working on several research projects under the G16031 ACCC awarded project involving muon in different host materials. These include:

i) Organic magnets which are ß′- $Et_nMe_{4-n}P[Pd(dmit)_2]_2$ and κ-$(BETDT\text{-}TTF)_2Cu[N(CN)_2]Cl$ systems

ii) Antiferromagnetic La_2CuO_4 and $YBa_2Cu_3O_6$

iii) Vanadium Thiobromide ($V_4S_9Br_4$) strongly correlated system

iv) $CeRu_2Al_{10}$ which is a Kondo semiconductor and a strongly correlated material.

v) Metal-organic hybrid materials which are

$(C_2H_5NH_3)_2CuCl_4$ and $(C_6H_5(CH_2)_2NH_3)_2CuCl_4$.

vi) Pyrochlore Iridates $R_2Ir_2O_7$(R= Nd, Sm)

vii) Synthesized simple and short strand DNA

viii) λ-$(BETS)_2GaCl_4$

ix) Kitaev Honeycomb material α-$RuCl_3$

2. Specific usage status of the system and calculation method

We used three software to conduct our computational studies which are:

i) Gaussian 09

ii) NBO for Gaussian 09

iii) Vienna Ab initio Simulation Package (VASP) ver. 5. 4. 1 software for band structure, spin structure and supercell calculation. This software is owned by Advanced Meson Science Laboratory and is installed at HOKUSAI and RICC.

3. Result

i) Organic magnets which are β′- $Et_nMe_{4-n}P[Pd(dmit)_2]_2$ and κ-$(BETDT\text{-}TTF)_2Cu[N(CN)_2]Cl$ systems

We have performed first principle computational studies on the β′-$X[Pd(dmit)_2]_2$ and κ-$(BETDT\text{-}TTF)_2Cu[N(CN)_2]Cl$ in the antiferromagnetic state. As of now, we have completed all the calculations for three pure systems. All the results obtained are as expected and 75% of the objectives have been achieved. At present, we are focusing on the hyperfine interactions for the muonated systems.

ii) Antiferromagnetic La_2CuO_4 and $YBa_2Cu_3O_6$

We have reproduced the band-structure of La_2CuO_4 and $YBa_2Cu_3O_6$ by using Density Functional Theory (DFT) method in order to understand the electronic structure of those systems. We also performed the electrostatic potential calculations to study the muon sites. Currently, our focus is on improving the dipole field calculations.

iii) Vanadium Thiobromide ($V_4S_9Br_4$) strongly correlated system

We performed the calculations on $V_4S_9Br_4$ system in the AFM state. We were unable to complete the Periodic Boundary Condition (PBC) approach using Gaussian 09 due to the limitation in the available resource memory.

iv) $CeRu_2Al_{10}$ which is a Kondo semiconductor and a strongly correlated material.

We have determined the electronic structure of the system by calculating the band structures of $CeRu_2Al_{10}$ in order to decide the suitable characteristic in the input file of VASP.

v) Metal-organic hybrid materials which are $(C_2H_5NH_3)_2CuCl_4$ and $(C_6H_5(CH_2)_2NH_3)_2CuCl_4$.

We have performed first principle calculations on the organic-inorganic hybrids of $(C_2H_5NH_3)_2CuCl_4$ (Cu-EA) and $(C_6H_5CH_2CH_2NH_3)_2CuCl_4$ (Cu-PEA) to study the muon sites in those systems from the view point of the total energy. In order to find the ground state, we performed four calculations which are nonmagnetic and three ferromagnetic states with different orientation of magnetic moment (in *a*-, *b*-, and *c*- direction, respectively). At this moment, we have finished the calculation on the hybrid systems.

vi) Pyrochlore Iridates $R_2Ir_2O_7$(R= Nd, Sm)

The results of our calculations indicate that the

all-in all-out magnetic structure can be most convincingly explained by the μSR results. Furthermore, we found that the lower limits of the sizes of magnetic moments were estimated to be 0.12 μB and 0.2 μB for Ir and Nd moments in $Nd_2Ir_2O_7$, and 0.3 μB and 0.1 μB for Ir and Sm moments in $Sm_2Ir_2O_7$, respectively. Further analysis indicates that the spin coupling between Ir and Nd/Sm moments is ferromagnetic for $Nd_2Ir_2O_7$ and antiferromagnetic for $Sm_2Ir_2O_7$.

vii) Synthesized simple and short strand DNA

We have carried out first principle calculations to examine the possible muon stopping sites in individual of DNA bases; Adenine (10 muonium sites), Guanine (11 muonium sites), Thymine (9 muonium sites) and Cytosine (8 muonium sites). From the calculations, we obtained an information on the electronic structures of the bases such as the relative energies, atomic charges, bond order and the hyperfine coupling constant at each muonium site in the DNA bases.

vii) λ-$(BETS)_2GaCl_4$

At this stage, calculation results give us a hint that the mixture of *s*- and *d*- wave pairing symmetry is strongly possible on λ-$(BETS)_2GaCl_4$.

ix) Kitaev Honeycomb material α-$RuCl_3$

In the result based on the first principle calculation, we found that it exhibited the same position of the minimum potential for different spin alignments suggested in the theoretical field, on the center of the honeycomb network. Also, it is revealed that there is no difference between cases of the single layer and of the triple stacked layer of the rhombohedral lattice within the iso-surface of the electrostatic potential for 500 meV.

４． Conclusion

At this stage, we have been able to carry out computational works that form the basis of our attempt for studying the electronic and magnetic characteristics of these novel and intriguing materials. And the results of some of our research have complemented the experimental results obtained from μSR experiments and this can lead to better understanding of these materials. The HOKUSAI GreatWave supercomputer provides important contribution to our research and enable us to conduct a wide range of computationally intensive tasks in various materials.

５． Schedule and prospect for the future

We will need and would like to continue using the HOKUSAI GreatWave supercomputer in the future for our studies on muon in materials. For the new fiscal year, we expect to perform further and more complex calculations on the materials that we are currently studying. Thus, we would require more powerful computing facilities to enable us to perform the calculations on supercell systems.

Usage Report for Fiscal Year 2016

Fiscal Year 2016 List of Publications Resulting from the Use of the supercomputer

[Publication]

1. Asih, R., Adam, N., Mohd-Tajudin, S. S., Sari, D. P., Matsuhira, K., Guo, H., Wakeshima, M., Hinatsu, Y., Nakano, T., Nozue, Y., Sulaiman, S., Mohamed-Ibrahim, M. I., Kumar, P. B., and Watanabe, I. (2017). Magnetic Moments and Ordered States in Pyrochlore Iridates $Nd_2Ir_2O_7$ and $Sm_2Ir_2O_7$ Studied by Muon-Spin Relaxation. *Journal of the Physical Society of Japan*, 86(2), 024705.

[Proceedings, etc.]

1. Watanabe, I., Yoon, S. W., Suprayoga, E., Adam, N., Mohd-Tajudin, S. S., Sari, D. P., Asih, R., Astuti, F., Angel, J., Umar, M. D., Mohamed-Ibrahim, M. I., & Sulaiman, S. (in press). The RIKEN-RAL Muon Facility and the Application of Muons for Studies of Magnetic Properties of Nano-Materials. *AIP Conference Proceedings*.

[Oral presentation at an international symposium]

1. Asih, R., Adam, N., Mohd-Tajudin, S. S., Sari, D. P., Matsuhira, K., Guo, H., Wakeshima, M., Hinatsu, Y., Nakano, T., Nozue, Y., Sulaiman, S., Mohamed-Ibrahim, M. I., Kumar, P. B., & Watanabe, I. (September, 2016). Magnetic Ordered States in Pyrochlore Iridates $Nd_2Ir_2O_7$ and $Sm_2Ir_2O_7$ Studied by μSR. *The 8th International Conference on Highly Frustrated Magnetism (HFM 2016).* Taiwan.
2. Adam, N., Mohd-Tajudin, S. S., Tanida, H., Sera M., Devashibhai, A. D., Takabatake, T., Sulaiman, S., Mohamed-Ismail, M. I., & Watanabe, I. (June, 2016). Magnetic State of the Kondo Semiconductor CeM_2Al_{10} (M=Ru,Os).*Rare Earth International Conference 2016.* Symposium conducted at Hokkaido, Japan.
3. Asih, R., Adam, N., Mohd-Tajudin, S. S., Sari, D. P., Matsuhira, K., Guo, H., Wakeshima, M., Hinatsu, Y., Nakano, T., Nozue, Y., Sulaiman, S., Mohamed-Ibrahim, M. I., Kumar, P. B., & Watanabe, I. (June, 2016). Magnetic Ordered States in Pyrochlore Iridates $R_2Ir_2O_7$ (R = Nd and Sm) Investigated by μSR. *The Rare-Earths International Conference 2016*. Symposium conducted at Hokkaido, Japan.
4. Sari, D. P., Asih, R., Mohd-Tajudin, S. S., Adam, N.,Hiraki, K., Ishii, Y., Takahashi, T., Nakano, T., Nozue, Y ., Sulaiman, S., Mohamed-Ibrahim, M. I., & Watanabe, I. (2016). μSR Study of Organic Superconductor λ-$(BETS)_2GaCl_4$. *3rd International Conference on Functional Materials Science*

2016 (ICFMS2016) "Trends in Functional Materials: From Fundamental to Applications". Bali, Indonesia.

[Others (Press release, Science lecture for the public)]

1. Adam, N., Mohd-Tajudin, S. S, Tanida, H., Sera, M., Sulaiman, S., Mohamed-Ibrahim M. I., & Watanabe, I. (2016, December). Magnetic State of CeM_2Al_{10} (M=Ru, Os). Theoretical and Experimental Aspects of Advanced Material Sciences. *Emallia Conference*. Symposium conducted at Hokkaido University, Japan.

2. Asih, R., Adam, N., Mohd-Tajudin, S. S., Maeda, S., Matsuhira, K., Wakeshima, M., Hinatsu, Y., Miyake, A., Tokunaga, M., Watanabe, I., Nakano, T., & Nozue, Y. (December, 2016). Investigation of Magnetic Ordered States in the Pyrochlore Iridates $(Nd_{1-x}Ca_x)_2Ir_2O_7$. *Emallia Conference*. Symposium conducted at Hokkaido University, Japan.

3. Mohd-Tajudin, S. S., Nishizaki, T., Kikkawa, A., Adam, N., Suprayoga, E., Mohamed-Ibrahim, M. I., Sulaiman, S., & Watanabe, I. (December, 2016). Muon in high-Tc Superconducting $YBa_2Cu_3O_6$. *Emallia Conference*. Symposium conducted at Hokkaido University, Japan.

4. Asih, R., Mohd-Tajudin, S.S., Astuti, F., Maeda, S., Matsuhira, K., Wakeshima, M., Hinatsu, Y., Miyake, A., Tokunaga, M., Watanabe, I., Nakano, T., Nozue, Y. (September 2016). μSR Study on the Magnetic Ordered State of $(Nd_{1-x}Ca_x)_2Ir_2O_7$. *JPS Meeting Sept 2016*. Japan.

5. Adam, N. (May, 2016). Magnetic State of $CeRu_2Al_{10}$. Experimental and Theoretical Applications of Spin-Resonance Techniques to Advanced Material Sciences. *Hokkaido U. - RIKEN Joint Symposium*. Japan.

6. Asih, R., Adam, N., Mohd-Tajudin, S. S., Maeda, S., Matsuhira, K., Wakeshima, M., Hinatsu, Y., Miyake, A., Tokunaga, M., Watanabe, I., Nakano, T., & Nozue, Y. (May, 2016). Investigation of Magnetic Ordered States in the $(Nd_{1-x}Ca_x)_2Ir_2O_7$ Probed by μSR. *Hokkaido University-RIKEN Symposium*. Japan.

7. Asih, R., Adam, N., Mohd-Tajudin, S. S., Maeda, S., Matsuhira, K., Wakeshima, M., Hinatsu, Y., Miyake, A., Tokunaga, M., Watanabe, I., Nakano, T., & Nozue, Y. (19 – 22 March, 2016). Effect of Ca-substitution on the Magnetic Ordered States of $(Nd_{1-x}Ca_x)_2Ir_2O_7$ Studied by μSR. *JPS 71th Annual Meeting 2016*. Japan.

8. Mohd-Tajudin, S. S., Nishizaki, T., Kikkawa, A., Adam, N., Suprayoga, E., Sulaiman, S., Mohamed-Ibrahim, M. I., & Watanabe, I. (19 – 22 March, 2016). Computational and Experimental Studies of Muon Sites in $YBa_2Cu_3O_6$. *JPS 71th Annual Meeting 2016*. Japan.

9. Sari, D. P., Asih, R., Hiraki, K., Ishii, Y., Takahashi, T., Koretsune, T., Seo, H., Nakano, T., Watanabe, I., & Nozue, Y. (January, 2016). μSR and DFT study of Non-magnetic Anion-based Organic Superconductor l-$(BETS)_2GaCl_4$. *Condensed Molecular Materials Laboratory SEMINAR* . Seminar conducted at Condensed Molecular Material Laboratory - RIKEN (Reizo Kato laboratory), Seminar (Invited Oral talk). Japan.

10. Asih, R., Adam, N., Mohd-Tajudin, S. S., Suprayoga, E., Sari, D. P., Matsuhira, K., Wakeshima, M., Hinatsu, Y., Miyake, A., Tokunaga, M., Watanabe, I., Nakano, T., Nozue, Y. (2016). μSR Study on the Pyrochlore Iridates $R_2Ir_2O_7$ (R= Nd, Sm). *RIKEN-RAL Anniversary at RIKEN*. Japan.