7. Background of Research:

Discovery of high Tc superconductivity resulted an intensive research in strongly correlated electron systems (SCES) in the vicinity of Mott insulators and also it has been successful in bringing novel quantum phenomena such as metal to insulator transitions, normal to magnetic or superconducting transitions etc. Mott insulators are one of the manifestation of strong correlation among the SCES at equilibrium and are expected to exhibit novel phenomena because of their highly sensitive ground states due to many-body interaction. Mott insulators contain a large number of carriers that are "frozen" because of the strong mutual Coulomb repulsion. In reality, most of the natural process occur far from equilibrium. Therefore, the pertinent question is: what happens if one drives the Mott insulator under nonequilibrium (transient or steady) conditions? The frozen electrons of the Mott insulator can be changed into mobile carriers by many ways: by shining an intense laser or applying an electric field or passing a DC current. In recent past, Prof. Maeno's group (Kyoto University) in collaboration with Prof. F. Nakamura's group (Kurume Institute of Technology) discovered that the Mott insulator Ca_2RuO_4 becomes a metal at room temperature if a small electric field of about 50 V/cm is applied. This insulator-metal transition is accompanied by a strong first-order structural transition, in which the RuO_6 octahedra become prolonged along the inter-layer *c*-axis. Furthermore, the induced metallic state can be maintained all the way to low temperatures if one keeps the current flowing. In the JSPS research that I performed at Kyoto, our approach was to control the Mott insulator precisely under non-equilibrium steady state (NESS) by passing a steady DC current. Such electronic method of controlling Mott insulator is very important as it has scientific as well as technological advantages.

8. Research methodology:

We choose layered Ruthenates (Ca_2RuO_4 and $Ca_3Ru_2O_7$) as the Mott insulating systems. Ca_2RuO_4 has a moderate Mott gap. On the contrary $Ca_3Ru_2O_7$ is a metallic system but becomes Mott insulating (with a tiny gap) with 0.5% Ti substitution at the Ru site. For this study, we used high quality single crystals samples grown using a floating-zone technique. Several high quality crystals were grown at Kurume Institute of Technology (F. Nakamura) as well as at Kyoto University in collaboration with NIMS (N. Kikugawa). We have also sent our few of our grown crystals to USA (Stony Brook University) and Korea (Seoul National University) for collaborative works. We made a sample holder for simultaneous measurements of magnetization and resistivity under DC current compatible with a commercial SQUID magnetometer (Quantum Design, MPMS XL). We performed most of the experiments at Kyoto University. We also used IMR, Tohoku high magnetic field facility to measure the high magnetic field behavior.

9. Results/impacts:

<u>Ca_2RuO_4</u>:

Under flowing DC current the insulating nature of Ca_2RuO_4 is strongly suppressed and becomes a Mott semimetal state. The antiferromagnetic ground state of Ca_2RuO_4 is completely suppressed by 3 mA (1 A/cm^2) current. Most importantly, we discovered the largest non-superconducting diamagnetism at low temperature (below 50 K) with the application of 5 mA (1.5 A/cm^2) current and the system concomitantly goes into the semimetallic phase. To explain our results theoretically, we collaborated with T.Oka and S. Kitamura (Max Plank Institute, Dresden), and K. Kuroki (Osaka University). We find that origin of diamagnetism: current reduces the Mott gap and indirect electron and hole pockets emerge from the upper and lower Hubbard bands with very low effective mass. Thermally excited quasiparticles arising from these states determine a large orbital diamagnetism. We published this work in *Science* Journal. Further we found that the diamagnetism is weaker when current is passed along c_o than b_o axis. We found that diamagnetism vanishes at high field (above 7 T, 65 mA) at IMR Tohoku. We also notice that diamagnetism disappears at very low temperature which matches with our theoretical picture of thermally excited light mass quasiparticles governing diamagnetism in Ca_2RuO_4.

<u>$Ca_3Ru_2O_7$:</u>

With a tiny (0.5%) amount of Ti substitution at the Ru site, the metallic $Ca_3Ru_2O_7$ becomes Anderson-Mott insulator with 8 orders higher in resistivity. The metal to insulator transition (T_{MI}) and the antiferromagnetic ordering temperature (T_N) is found to be 50 K and 56 K respectively. Under DC current $Ca_3(Ru_{0.995}Ti_{0.005})_2O_7$ the resistivity and magnetization in the metallic region remained unaffected. Only the insulating region (below 50 K) is affected by the Dc current. The insulating region becomes less resistive with an increase in the magnetization. But below 20 K the magnetization starts to diminish and finally becomes negative below 10 K. The minimum current required for setting diamagnetism in $Ca_3(Ru_{0.995}Ti_{0.005})_2O_7$ is 0.1 A/cm^2 which is one order lower than in the case of Ca_2RuO_4. More importantly, diamagnetism in $Ca_3(Ru_{0.995}Ti_{0.005})_2O_7$ can be in-situ switched ON and OFF by varying the current. We successfully performed 10^5 switching cycles between On and OFF state. Currently this work is under review in Physical Review Letters Journal.

<u>Impact:</u> The present work reveals that DC current is a new control tool to control the physical properties of a variety of SCES, with the possibility of inducing novel states which are not accessible by conventional control parameters. We have given a unique direction to enrich this new field namely SCES-NESS.

10. Research Presentations during the period of the fellowship (Name of the conference, title, place, date):

7. International mini-workshop on Nonequlibrium transport and phase transition in novel materials, Current-induced strong diamagnetism in the Mott Insulator Ca_2RuO_4, Nagoya Univ., Nov 26, 2018.
6. 2nd international workshop on quantum materials and topological materials, Peking Univ., China, Sept 24, 2018.
5. International Conference on Magnetism (ICM), Current-induced giant diamagnetism and Mott semimetal behavior in single crystal Ca_2RuO_4, San Francisco, USA, July 15, 2018.
4. JPS Annual meeting, Giant diamagnetism in Ca_2RuO_4 induced by DC current, Tokyo Univ. of Science, Noda, March 22, 2018.
3. TMS-EPiQS 2nd Alliance Workshop: Topological magnets and topological superconductors, Current induced Mott-semimetal behavior and giant diamagnetism in Ca_2RuO_4, Kyoto Univ., Jan 11, 2018.
2. ETH-Amsterdam-Kyoto mini-workshop, Current induced giant diamagnetism in Ca_2RuO_4, Kyoto Univ., Oct 20, 2017.
1. JPS Spring meeting, Switching between Mott-insulating state and diamagnetic state with DC current in $Ca_3(Ru_{1-x}Ti_x)_2O_7$, Iwate University, Sept 21, 2017.

11. A list of paper published during or after the period of the fellowship, and the names of the journals in which they appeared. Attach a copy of each article if available.

[5]. C. Sow, R. Numasaki, G. Mattoni, N. Kikugawa, S. Yonezawa, S. Uji, and Y. Maeno,
In situ control of diamagnetism by electric current in $Ca_3(Ru_{1-x}Ti_x)_2O_7$,
Phys. Rev. Lett. (2nd round review) (2019), Impact Factor: 8.8, arXiv:1902.02515.

[4]. M-C Lee, C H Kim, I Kwak, CW Seo, CH Sohn, F Nakamura, C Sow,
Y Maeno, E-A Kim, TW Noh, KW Kim,
Strong spin-phonon coupling unveiled by coherent phonon oscillations in Ca_2RuO_4,
Phys. Rev. B **(accepted)** (2019), Impact Factor: 3.8, arXiv:1712.03028.

[3]. J. Zhang, A. S. McLeod, Q. Han, X. Chen, H. A. Bechtel, Z. Yao, S. N. G. Corder, T. Ciavatti, H. Tao, M. Aronson, G. L. Carr, M. C. Martin, C. Sow, S. Yonezawa, F. Nakamura, I. Terasaki, D. N. Basov, A. W. Millis, Y. Maeno, M. Liu,
Nano-Resolved Current-Induced Insulator-Metal Transition in the Mott Insulator Ca_2RuO_4,
Phys. Rev. X. **9**, 011032 (2019), Impact Factor: 14.4.

[2]. M. C. Lee, C. H. Kim, I. Kwak, J. Kim, S. Yoon, B. C. Park, B. Lee, F. Nakamura, C. Sow,
Y. Maeno, T. W. Noh, K. W. Kim,
Abnormal phase flip in coherent phonon oscillations of Ca_2RuO_4,
Phys. Rev. B **98**, 161115 (2018), Impact Factor: 3.8, arXiv: 1808.06026.

[1]. C. Sow, S. Yonezawa, S. Kitamura, T. Oka, K. Kuroki, F. Nakamura, and Y. Maeno,
Current-induced strong diamagnetism in the Mott insulator Ca_2RuO_4,
Science **358**, 1084 (2017), Impact Factor: 37.2, arXiv: 1610.02222.

12. Awards during the period of the fellowship (Name of the award, Institution, date etc.)

No such awards received during the JSPS fellowship tenure.