Exploring Novel Physical Properties by Multi-Dimensional Spectroscopy

Division of Advanced Molecular Science (Department of Materials Molecular Science, Electronic Structure)

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Education

- 1988 B.S. Tohoku University 1990 M.S. Tohoku University
- 1991 Ph.D. Tohoku University

Professional Employment

- 1991 JSPS Postdoctoral Fellow, Tohoku University
- 1993 Research Associate, Kobe University
- 1993 Research Associate, Institute for Molecular Science
- 1998 Associate Professor, Kobe University
- 2002 Associate Professor, Institute for Molecular Science
- 2013 Professor, Osaka University
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Awards

- 2001 Young Incentive Award, Japanese Society for Synchrotron Radiation Research
- 2008 The Commendation for Science and Technology by MEXT, Japan Science and Technology Prize (Research Field)
 2008 Morita Memorial Prize

Keywords

Condensed Matter, Biological Materials, Optical Property

Physical and chemical properties of solids, such as conductivity, magnetism, superconductivity and chemical reactions, and also life phenomena, such as redox and photosynthesis, originate from microscopic electronic and molecular vibrations and/or molecular movements based on quantum mechanics in materials and their interactions. To clarify the microscopic states provides us not only the knowledge of the origins of the physical properties and life phenomena but also the clarification of hidden functionalities. The information is useful for the creation of novel functional properties. To visualize the microscopic state, we also develop novel spectroscopic techniques using synchrotron radiation, high brilliant electron beams, and other so-called quantum beams. Recently, we have successfully developed a novel electron spectroscopy, namely resonant electron-energy-loss spectroscopy, as shown in Figure 1, to detect element-specific plasmons. Based on the obtained information of electronic structures, we are aiming to develop novel physical properties of new materials.

Selected Publications

- S. Kimura, T. Ito, M. Sakai, E. Nakamura, N. Kondo, K. Hayashi, T. Horigome, M. Hosaka, M. Katoh, T. Goto, T. Ejima and K. Soda, "SAMRAI: A Variably Polarized Angle-Resolved Photoemission Beamline in the VUV Region at UVSOR-II," *Rev. Sci. Instrum.* 81, 053104 (7 pages) (2010).
- S. Kimura, T. Iizuka, H. Miyazaki, A. Irizawa, Y. Muro and T. Takabatake, "Electronic-Structure-Driven Magnetic Ordering in a Kondo Semiconductor CeOs₂Al₁₀," *Phys. Rev. Lett.* **106**, 056404 (4 pages) (2011).



Member Secretary

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Figure 1. Resonant electron-energy-loss spectroscopy (rEELS) apparatus developed by our group. The apparatus consists of a high-brilliant spin-polarized electron gun and a photoelectron spectrometer.

- S. Kimura and H. Okamura, "Infrared and Terahertz Spectroscopy of Strongly Correlated Electron Systems under Extreme Conditions," *J. Phys. Soc. Jpn.* 82, 021004 (28 pages) (2013). [review]
- K. Hagiwara, Y. Ohtsubo, M. Matsunami, S. Ideta, K. Tanaka, H. Miyazaki, J. E. Rault, P. Le Fèvre, F. Bertran, A. Taleb-Ibrahimi, R. Yukawa, M. Kobayashi, K. Horiba, H. Kumigashira, K. Sumida, T. Okuda, F. Iga and S. Kimura, "Surface Kondo Effect and Non-Trivial Metallic State of the Kondo Insulator YbB₁₂," *Nat. Commun.* 7, 12690 (7 pages) (2016).

1. Non-Trivial Surface States of Samarium Hexaboride at the (111) Surface¹⁾

The peculiar metallic electronic states observed in the Kondo insulator, samarium hexaboride (SmB_6), has stimulated considerable attention among those studying non-trivial electronic phenomena. However, experimental studies of these states have led to controversial conclusions mainly to the difficulty and inhomogeneity of the SmB_6 crystal surface.

In this work, the detailed electronic structure of SmB_6 is revealed by angle-resolved photoelectron spectroscopy measurements of the three-fold (111) surface, where only two inequivalent time-reversal-invariant momenta (TRIM) exists. We observe the metallic two-dimensional state was dispersed across the bulk Kondo gap. Its helical in-plane spin polarization around the surface TRIM indicates that SmB_6 is topologically non-trivial, according to the topological classification theory for weakly correlated systems. Based on these results, we propose a simple picture of the controversial topological classification of SmB_6 .

We expect that this discovery could provide a new, fertile playground for the following researches about the concert effect between electron correlation and topology. It would also be applicable for future spintronic technologies.



Figure 2. (a) Crystal structure of SmB_6 . A shaded area is the (111) surface studied in the current work. (b) Surface electronic structure of SmB_6 obtained by angle-resolved photoelectron spectroscopy. Bright area represents the electrons with corresponding wavenumber (Fermi surface). A solid hexagon is the zone boundary determined from surface periodicity.

2. Relaxation Dynamics of [Re(CO)₂(bpy) {P(OEt)₃}₂](PF₆) in TEOA Solvent Measured by Time-Resolved Attenuated Total Reflection Terahertz Spectroscopy²)

To reveal highly efficient photocatalytic properties of an

artificial photosynthesis material $[Re(CO)_2(bpy){P(OEt)_3}_2]$ (PF₆), we have directly observed the photo-induced relaxation dynamics and reductive quenching process of the photoexcited state on a photosynthesis material in Triethanolamine (TEOA) solvent as an electron donor by time-resolved attenuated total reflection spectroscopy in the terahertz (THz) region. The spectrum of the complex in TEOA has an intermolecular vibrational mode between the complex and TEOA molecules, which reflects the precursor of the reductive quenching process. The intermolecular vibrational mode has three-step relaxation process in a picosecond timescale after photoexcitation, where firstly the triplet metal-to-ligand charge transfer excited state is vibrationally cooled down, secondly the distance between Re and TEOA is reduced by the rotation of TEOA molecules due to dipole-dipole interaction accelerated by heat transfer, and finally electrons transfer from TEOA to Re. These observations provide us the detailed information of the electron transfer process of photocatalytic properties of [Re(CO)₂(bpy){P(OEt)₃}₂](PF₆) in TEOA solvent.



Figure 3. The schematic figures of the temporal evolution from I to III of the interaction between photocatalyst $[Re(CO)_2(bpy){P(OEt)_3}_2]^+$ as a photocatalyst and TEOA molecules as reductants.

References

- Y. Ohtsubo, Y. Yamashita, K. Hagiwara, S. Ideta, K. Tanaka, R. Yukawa, K. Horiba, H. Kumigashira, K. Miyamoto, T. Okuda, W. Hirano, F. Iga and S. Kimura, *Nat. Commun.* 10, 2298 (7 pages) (2019).
- 2) P. N. Nguyen, H. Watanabe, Y. Tamaki, O. Ishitani and S. Kimura, *Sci. Rep.* 9, 11772 (7 pages) (2019).