【Grant-in-Aid for Scientific Research (S)】
Broad Section D

Title of Project : Development of Phase-Controlled Near Field Spectroscopy with Extremely High Spatiotemporal Resolution

TAKEDA Jun
(Yokohama National University, Faculty of Engineering Science, Professor)

Research Project Number: 20H05662 Researcher Number : 60202165
Keyword : terahertz, scanning tunneling microscopy, near field, luminescence, mid-infrared pulse

【Purpose and Background of the Research】
Manipulating the structure and function in matters by controlling their electronic and charged states at arbitrary time and space is one of the most important issues in the fields of materials science and nanoscience. In terms of the time resolution, attosecond control of electrons has been achieved in elaborate nanostructures by modulating a carrier-envelope phase of near infrared optical pulses. However, it is difficult to apply this technique to a variety of materials. In terms of the spatial resolution, on the other hand, scanning tunneling microscopy (STM) has been widely utilized as an analytical tool with atomic-scale resolution yet with low time resolution, making it difficult to reveal energy conversion and dissipation in quantum systems.
In this study, we develop terahertz-field-driven scanning tunneling luminescence (THz-STL) spectroscopy by combining the THz-STM and STL techniques to capture the energy dynamics triggered by ultrafast electron tunneling. In addition, we also utilize single-cycle mid-infrared (MIR) near fields to control vibrational states in functional and biological materials. Our final goal is to hybridize these techniques that can manipulate a variety of physical properties of materials in THz to MIR frequency regions (Fig. 1).

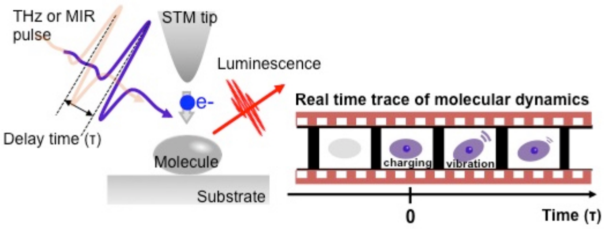

Figure 1 Schematic of phase-controlled terahertz (mid-infrared) field-driven near-field spectroscopy.

【Research Methods】
Conventional STL spectroscopy can measure photons converted from the tunneling electrons of an STM. In our new THz-STL spectroscopy, by injecting a charge using phase-controlled THz pulses that are generated using a $LiNbO_3$ prism with tilted pulse-front configuration, we can trace time-resolved excited-state dynamics in a single molecule. The phase of THz pulses can be tuned by a THz phase shifter originally developed. In addition, we produce phase-stable MIR pulses via optical rectification of 10 fs near-infrared laser pulses compressed by either a hollow fiber or a chirped mirror. After a combination of these field-driven nanoscopic techniques, we try to explore radiative decay processes of a single molecule and trace local structural dynamics of hydrogen-bonded network.

【Expected Research Achievements and Scientific Significance】
Our goal is to establish a new spectroscopic tool with unprecedented spatiotemporal resolutions to capture energy dissipations and conversions among various quanta, which offers prospects for sensing and controlling quantum systems, providing novel insights and advances in nanoscale science and technology.
Our challenge will also provide information on how smaller and how faster we can access the quantum nature of materials, which is the supreme proposition for the development of next-generation nanoelectronics and plasmonic devices with high performances.

【Publications Relevant to the Project】

- K. Yoshioka, I. Katayama, Y. Minami, M. Kitajima, S. Yoshida, H. Shigekawa, and J. Takeda, "Real-space coherent manipulation of electrons in a single tunnel junction by single-cycle terahertz electric fields", **Nature Photon. 10**, pp. 762-765 (2016).
- K. Yoshioka, I. Katayama, Y. Arashida, A. Ban, Y. Kawada, K. Konishi, H. Takahashi, and J. Takeda, "Tailoring single-cycle near-field in a tunnel junction with carrier-envelope phase-controlled terahertz electric fields", **Nano Lett. 18**, pp. 5198-5204 (2018).
- H. Mashiko Y. Chisuga, K. Oguri, H. Masuda, I. Katayama, J. Takeda and H. Gotoh, "Multi-petahertz electron interference in $Cr:Al_2O_3$ solid-state material", **Nature Commun. 9**, 1468 (2018).

【Term of Project】 FY2020-2024

【Budget Allocation】 146,600 Thousand Yen

【Homepage Address and Other Contact Information】
http://www.ultrafast.ynu.ac.jp/
http://www.laser-nanoscience.ynu.ac.jp/en
jun@ynu.ac.jp