

화학과 대학원 세미나

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장소: Zoom

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연사: 권오훈 (울산과학기술원 화학과) 교수



Exploring photophysical and photochemical processes using ultrafast spectroscopy and microscopy

Oh-Hoon Kwon

*Department of Chemistry, Ulsan National Institute of Science and Technology
Ulsan 44910, Korea*

Visualizing structural rearrangements at a molecular level is essential for understanding the function of matter. Once isolated, the intermediate and transitional structures of a molecule along a chemical reaction can be determined by ultrafast spectroscopy with a femtosecond temporal precision. For condensed matter made of countless atoms and organized molecular units, local structural imperfections at nanoscales become prevalent such that the physical processes of each singularity do not proceed as those in bulk and even diverge. Because the ensemble nature of spectroscopic measurements cannot reveal the characteristic transition of each nanoscopic structure, the simultaneous spatiotemporal imaging is requisite for disentangling the complex processes, hierarchically spanning small-amplitude, ultrafast atomic displacements to collective structural rearrangements at larger spatial and longer temporal scales.

Recently, our group has achieved a sub-nanometer spatial resolution in ultrafast electron microscopy (UEM) [1], successfully imaging vibrating individual gold nanorods by integrating a direct electron detection camera for the first time, but with limited time resolution of several picoseconds [2]. Introducing an energy filter to UEM can advance the time resolution to the domain of atomic motion. Imaging transient structures with femtosecond temporal precision was made possible by gating imaging electrons of narrow energy distribution from dense chirped photoelectron packets. Presented are the concept and proof-of-principle demonstration of the energy-filtered UEM achieving the temporal resolution limited by the briefness of an optical

excitation pulse, *i.e.*, 500 fs in this study, filming ultrafast insulator-to-metal phase transition of vanadium dioxide [3]. Our approach leads the access of electron microscopy to the timescale of elementary nuclear motions visualizing the onset of structural dynamics of matter at nanoscales.

Also, we have uniquely combined cathodoluminescence (CL) with UEM [4]. A synergistic use of the two methods is essential because CL and UEM realize required spectral and spatiotemporal sensitivity, respectively. For nitrogen-related color centers in nanodiamonds, we demonstrate the measurement of CL lifetime with the local sensitivity of 50 nm and the time resolution of 100 ps. The emitting state of the H3 color center (N2V) was found to populate by hole transfer from the NV center excited by free carrier transfer diffusing across diamond lattices upon electron beam excitation. The technical advance achieved in this study will deliver new concepts for specific control over energy conversion relevant to quantum dots and single photon sources.

References

- [1] Y.-J. Kim and O.-H. Kwon, *ACS Nano* **2021**, *15*, 19480-19489.
- [2] Y.-J. Kim, S. Ji, J. Weissenrieder, and O.-H. Kwon, *submitted*.
- [3] Y.-J. Kim, Y. Lee, K. Kim, and OH Kwon, *ACS Nano* **2020**, *14*, 11383-11393.
- [4] Y.-J. Kim, H. Jung, S. W. Han, and O.-H. Kwon, *Matter* **2019**, *1*, 481-495.