

Observing Electronic Dynamics within 100 fs in 2H-MoTe₂ by Double-excitation Ultrafast Electron Diffraction

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Transition metal dichalcogenides (TMDs) are two-dimensional layered materials, especially MoTe₂ which exhibits a semiconductor, semimetal and Weyl semimetal phases [1,2]. Understanding the carrier dynamics in semiconducting materials is important because the properties of these compounds are affected by the behavior of hot carriers in the conduction and valence bands. Electronic relaxations have been directly observed by two-photon photoelectron spectroscopy and time-resolved angle-resolved photoemission spectroscopy [3,4]. We proposed a unique method to understand the electronic relaxation dynamics in semiconducting materials. The use of a double-optical-pulse excitation ultrafast time-resolved electron diffraction measurements under saturable absorption conditions [5] enabled to observe electronic relaxation of hot carriers in the conduction band of 2H-MoTe₂ [6]. We found that the momentum relaxation of excited electrons occurred within 100 fs in the conduction band of 2H-MoTe₂. Because the time constant of the momentum relaxation was comparable to the duration of the excitation pulse, it was required to measure the time constant of the electronic scattering more precisely. To measure extremely fast electron scattering (within 100 fs), we need to use an ultrafast time-resolved electron diffraction setup with shorter excitation optical pulses. Our laboratory constructed a setup that generates an ultrashort optical beam (wavelength: 800 nm, duration: 35 fs) and an electron pulse (duration: ~100 fs) with a compression cavity [7]. We introduced the double-pulse excitation system with a Mach-Zehnder interferometer in the pump line for the double-optical-pulse ultrafast time-resolved electron diffraction measurement. We measured the pulse duration of the excitation beam (wavelength: 400 nm) by auto- and cross-correlation to be approximately 62 fs (FWHM). This result shows that we can observe the momentum relaxation of electrons with a time resolution of approximately 30 fs. Then, we measured the fluence range to induce a saturable absorption in the 2H-MoTe₂ sample at approximately 3–8 mJ/cm². After setting up the systems, the electron relaxation in the momentum direction will be investigated by using the ultrafast time-resolved electron diffraction setup, with the electron compression system.

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