Nonlinear electronic responses and phase transitions induced by a strong mid-infrared pulse in correlated electron materials

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Developments of femtosecond laser technology enable us to generate a strong frequency-tunable mid-infrared (IR) pulse with a fixed carrier envelope phase (CEP) of electromagnetic wave [1, 2]. We have newly constructed a mid-IR-pump visible-probe spectroscopy system, in which the pump light is a CEP-stable mid-IR pulse with a temporal width of about 100 fs and the probe light is an ultrashort visible pulse with a temporal width of about 9 fs [3]. By introducing a double feedback correction scheme, we have succeeded in eliminating both the CEP drift in mid-IR pulses due to fluctuations in the difference of optical-path lengths of two idler pulses before the differential frequency generation (DFG) process and the drift in pump-probe delay times due to fluctuations in the difference of the overall optical-path lengths of the pump and probe pulses. As a result, the absolute carrier phase of mid-IR pulses can be fixed within 200 mrad and errors in the measurement of phase-sensitive responses can be reduced to within 1 fs over a few tens of hours. Using this system, we have been investigating nonlinear electronic responses and phase transitions induced by a strong mid-IR pulse in correlated electron materials. In this presentation, we will report our recent results focusing on the following three topics.

(1) Observation of nonlinear electronic responses by a resonant excitation of intramolecular vibrations coupled with intermolecular charge transfers in a one-dimensional (1D) spin-Peierls system of an organic molecular material, K-tetracyanoquinodimethane (TCNQ)
(2) Ionic to neutral phase transition induced by a resonant excitation of intramolecular vibrations coupled with intermolecular charge transfers in a quasi-1D organic molecular compound TTF-CA (TTF: tetrathiafluvalene, CA: *p*-chloranil)
(3) Observation of light-dressed states (or Floquet states) formed by a strong mid-IR pulse excitation in a 2D cuprate Mott insulator $Sr_2CuO_2Cl_2$.

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