Time-resolved Optical Studies of Colossal Magneto-Resistance and Charge-Density Wave Materials

YUHANG REN

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Advisor: Gunter Luepke, Associate Professor of Applied Science

Abstract

This thesis presents measurements of collective modes and ultrafast carrier relaxation dynamics in charge-density-wave (CDW) conductors and colossal magneto-resistance (CMR) manganites by means of femtosecond time-resolved optical spectroscopy. In these experiments, a femtosecond laser pump pulse excited a broad frequency spectrum of low-energy collective modes and electron-hole pairs via interband transitions in the material, thereby changing its optical properties. The low-energy collective excitations and quasiparticle relaxation and recombination processes were monitored by measuring the resulting photoinduced absorption as a function of probe pulse wavelength and time delay after photoexcitation. Therefore, the technique enabled direct real-time measurements of non-equilibrium low-energy collective excitations and quasiparticle recombination dynamics.

First, we developed a general model to describe the photogeneration and detection mechanism of collective modes based on light absorption in two-color pump-probe experiments. The excitation of the density wave states can be well described by a spatially and time-dependent order parameter, which includes phase and amplitude excitations (phasons and amplitudons). The excitation mechanism was different from previous pump-probe transmission experiments in weakly absorbing crystals, in which the wave-vector of the excited modes was determined by the phase-matching condition of the pump beam. Since the absorption depth in CMR and CDW conductors is ~100 nm for wavelengths in the visible to infrared range, a broad spectrum of collective modes with frequencies down to a few GHz were excited and propagated normal to the surface into the material. According to the model, the dispersion of the long-wavelength phason and amplitudon were measured by changing the probe wavelength.

Second, we performed the first pump-probe spectroscopy from the ultraviolet to mid-infrared wavelength range to study low-frequency collective excitations, including temperature evolution, dispersion, damping, and anisotropy of amplitude mode and transverse phason in quasi-one dimensional CDW conductors, $K_{0.3}MoO_3$ and $K_{0.33}MoO_3$ on ultrafast time scale. The transverse phason exhibited an acoustic-like dispersion relation in the frequency range from 5 – 40 GHz. The phason velocity was strongly anisotropic with a very weak temperature dependence. In contrast, the amplitude mode exhibited a weak (optic-like) dispersion relation with a frequency of 1.66 THz at 30K.

Third, we presented femtosecond time-resolved infrared spectra from doped perovskite manganite thin films and single crystals. A low-energy collective mode was observed and discussed in terms of the opening of a pseudogap resulting from charge/orbital ordering phases. The softening of the collective mode cannot be explained solely by electronic instability. A cooperative Jahn-Teller type distortion of the MnO_6 octahedra coupled to the collective mode was necessary to explain our results. Moreover, the quasiparticle dynamics in the vicinity of the metal-insulator transition was strongly affected by the presence of a pseudogap, phase separation and percolation, which are strongly dependent on temperature. Furthermore, a very long-lived relaxation process was observed, both in the metallic and insulating phase but which is absent in the paramagnetic phase. Therefore it is ascribed to a slow spin relaxation process. The dynamics of the spin system was further investigated in strained and unstrained thin films, which showed a strong strain effect.