Ultrafast spectroscopy of a Dirac semimetal Cd_3As_2 driven by intense multi-terahertz pulses

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Optical properties of topological Dirac semimetals are attracting growing interest because 3D massless electrons show unique electromagnetic responses with a large interaction volume with light, providing a fascinating platform for studying ultrafast control of matter and promising application in optoelectronics and nonlinear optics such as efficient harmonic generation [1]. Their unique infrared responses and large nonlinearity originate from the gapless band structure, where the interband and intraband transitions occur in the close energy scale and exert influences on each other. To thoroughly derive their novel functionalities, in-depth understanding of nonequilibrium broadband complex response functions is indispensable.

We have developed a phase-stable time-domain spectroscopy system in the multi-terahertz (10-50 THz) range [2] and studied ultrafast dynamics of the broadband response functions in a photoexcited Cd_3As_2 thin film with 30-fs time resolution. We found that photoexcited carriers largely suppress the multi-terahertz refractive index due to the elevated plasma frequency [3]. We also investigated the broadband response function in Cd_3As_2 under the formation of Floquet-Bloch state. Under 30-THz narrowband and linearly polarized pump, we found that the conductivity spectrum shows a highly dispersive line shape with net optical gain. The result is explained by the stimulated Rayleigh scattering, which corresponds to the transitions between the Floquet subbands and is remarkably enhanced by the elevated plasma frequency [4]. In addition, we performed 33-THz circularly polarized pump and THz polarization resolved probe experiments to study the light-induced anomalous Hall effect in Cd_3As_2. Our comprehensive analysis clarified that the microscopic origins of the light-induced anomalous Hall current during and after the pump irradiation are accounted for by field-induced injection current [5] and inverse "isospin" Hall effect [6], respectively.

To manipulate the light-field driven states such as Floquet topological states, the control of the polarization states or the vector field waveforms of the pump pulse are important. We are developing the techniques for generation and detection of vector field shaping in multi-terahertz region. By using polarization modulated electro-optic sampling, the measurement of arbitrary trajectory of electric field vector was realized [7]. We also demonstrated the generation of the tailored counter rotating bicircular multi-THz pulses [8], which would be applied to ultrafast control of topological materials.

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