First-principles simulations of ultrafast phase transition in condensed-matter systems

M.-X. Guan[1,2], X.-B. Liu[2], E. Wang[2], D.-Q. Chen[2], J. T. Sun[1,2], and S. Meng[2]

[1] *Centre for Quantum Physics, Key Laboratory of Advanced Optoelectronic Quantum Architecture and Measurement (Ministry of Education), School of Physics, Beijing Institute of Technology, Beijing 100081, China*

[2] *Beijing National Laboratory for Condensed Matter Physics and Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China*

*email: mxguan@bit.edu.cn

Abstract

The crystal structure of a solid largely dictates its electronic, optical and mechanical properties. Laser driven structural engineering allows the manipulation of functional properties at high speed and beyond what maybe possible in equilibrium. We developed a first-principles approach (*time-dependent ab initio propagation*, TDAP) that aims at providing robust dynamic simulations of light-induced, highly nonlinear phenomena by real time calculation of combined photonic, electronic and ionic quantum mechanical effects within a time-dependent density functional theory (TDDFT) framework. By comparing the theoretical and experimental results, the approaches have been demonstrated effective and efficient in treating ultrafast quantum dynamical processes.

Here, we show strength of this method in revealing the explicit structural phase transition pathways in wide range of quantum materials. Firstly, the temporal characters of laser-driven phase transition from 2H to 1T' has been investigated in the prototype $MoTe_2$ monolayer. This process is found to be induced by fundamental electron-phonon interactions, with an unexpected phonon excitation and coupling pathway closely related to the nonequilibrium relaxation of photoexcited electrons. The order-to-order phase transformation is dissected into three substages, involving energy and momentum scattering processes from optical to acoustic phonon modes in subpicosecond timescale. Secondly, an unexpected orbital-selective photoexcitation in type-II Weyl material WTe_2 is reported under linearly polarized light (LPL), inducing striking transitions among several topologically-distinct phases mediated by effective electron-phonon couplings. The symmetry features of atomic orbitals comprising the Weyl bands result in asymmetric electronic transitions near the Weyl points, and in turn a switchable interlayer shear motion with respect to linear light polarization, when a near-infrared laser pulse is applied. Consequently, not only annihilation of Weyl quasiparticle pairs, but also increasing separation of Weyl points can be achieved, complementing existing experimental observations.

Bibliography

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