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Extreme High-Field Electron Dynamics in Nanomaterials

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Light-matter interactions and the dynamics of charged particles are of fundamental scientific interest. In the widely applicable semiclassical theory of light-matter interactions, a Hamiltonian for the light-matter interaction would, in general, take the form, $H = H_0 + H_I(t)$, where the Hamiltonian for the matter H_0 is independent of time, whereas the interaction Hamiltonian $H_I(t)$ is time-dependent. Under normal circumstances the interaction energy is much smaller than the internal Coulomb energy (1-10 eV for a typical semiconductor), and the time-dependent perturbation theory is sufficient to describe the dynamical processes of light-matter interactions. The perturbation treatment, however, becomes invalid when the light field strength is comparable or even greater than that of the internal Coulomb field of the material. The ultrafast electron dynamics in condensed matter driven by strong electromagnetic waves are largely unknown, even though high-field charge transport and many-body interactions are of great importance for modern science and technology. Understanding and controlling the high-field electron dynamics is indispensable for next-generation high-speed electronic and photonic applications, for which the operating frequency of devices goes beyond 100 GHz and the electric field inside the devices exceeds 100 kV/cm. Recent advances in high-field terahertz (THz) spectroscopy provide unprecedented opportunities to explore the electron dynamics under the extreme conditions. THz radiation lies in the gap between the infrared band and the microwave band in the electromagnetic spectrum. The THz interactions undergo significant qualitative changes in the high-field regime, in which the interaction energy ($H_I = d \cdot E_{THz}$ for electric dipole interactions) is comparable to or even greater than the internal Coulomb energy (e.g., H_0 1 eV for a band gap and 1 meV for intersubband energy level spacing). Furthermore, the relatively long acceleration time in a THz field allows another possibility of extreme light-matter interactions unique in the THz band: the kinetic energy gained by charge acceleration becomes comparable to the unperturbed Coulomb energy of the material. In this strong interaction regime, intense THz pulses excite electrons far from equilibrium and give rise to qualitative changes in optical and electronic properties. The THz excitation and subsequent relaxation takes place on a picosecond time scale, which can be mapped out in the time domain with sub-picosecond resolution by time-resolved THz spectroscopy. High-field electron dynamics in condensed matter is largely influenced by the properties of the material system. We present a few examples of distinctive phenomena of extreme light-matter interactions in different nanomaterial systems: (i) THz control of light-matter coupling in quantum-well microcavities, (ii) High-field THz responses in graphene, (iii) Anisotropic high-field THz response of free-standing carbon nanotubes, and (iv) THz triggered insulator-to-metal transition in a nanoantenna patterned vanadium dioxide film.