Ultrafast terahertz probes of interacting dark excitons in chirality-specific semiconducting single-wall carbon nanotubes

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Abstract

Ultrafast terahertz (THz) spectroscopy accesses the dark excitonic ground state in resonantly excited (6,5) single-wall carbon nanotubes (SWNTs). This is feasible via internal, direct dipole-allowed transitions between the lowest lying dark-bright pair states around 6 meV. An analytical model reproduces the response, thus enabling a quantitative analysis of the transient densities of dark excitons and the unbound *e-h* plasma, the oscillator strength, the transition energy renormalization and the dynamics. Stable quasi-1D multi-exciton states emerge rapidly even with increasing off-resonance photoexcitation. They evolve uniquely from a predominant dark exciton population to complex phase-space filling of both dark and bright pair states. These are distinctly different from dense 2D and 3D excitons, which are characterized by the ionization of free carriers and slow formation.