Contribution ID: 92

Type: Rapid

Ultrafast Terahertz Spectroscopy of Carbon Nanomaterials

Tuesday, 24 July 2012 12:15 (12 minutes)

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Confined carriers in carbon nanomaterials - fullerenes, nanotubes, and graphene –exhibit a rich variation in electronic and optical properties. Understanding their excitations, correlations and carrier dynamics is of fundamental interest and can yield insight for future high-speed device applications. Here, we present experiments that employ broadband ultrafast THz pulses to investigate single-walled carbon nanotubes and graphene. Photoexcitation of semiconducting nanotubes leads to the emergence of an induced conductivity around 1.7 THz, whose frequency, chirality enhancement, and temperature dependence agree with the observation of intra-excitonic THz transitions. The picosecond decay of this conductivity follows a bi-molecular kinetics, revealing exciton-exciton annihilation dynamics. Moreover, we have carried out first ultrafast mid-IR-pump, THz probe experiments of exfoliated graphene. An ultrabroadband THz probe pulse spanning up to 20 THz was employed to cover both intra- and interband transitions and map the Dirac fermion distribution functions. We have observed transient conductivity changes in single-layer graphene on Si/SiO2 and will discuss its complex spectral response and fast picosecond relaxation. These experiments provide insights into low-energy excitations and dynamics of carbon nanomaterials, motivating studies with intense THz fields for resonant excitation.

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 Session Classification:
 Graphene II

Track Classification: Graphene