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Ultrafast and Nonlinear Optical Spectroscopy of Carbon Nanotubes[1]

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Single-walled carbon nanotubes (SWNTs) provide a variety of unique opportunities for studying the dynamics and interactions of one-dimensional (1-D) electrons and phonons. We have carried out a series of ultrafast and nonlinear optical experiments on SWNTs, revealing novel properties of high- density 1-D excitons as well as coherent lattice vibrations.[2] We have shown that there exists an upper limit on the density of 1-D excitons in SWNTs, which results in photoluminescence saturation. Using a model based on diffusion-limited exciton- exciton annihilation, we provided realistic estimates for the exciton densities in the saturation regime. We also predicted and demonstrated that there is an optimum temperature at which the exciton density can be maximized, due to the existence of a dark exciton state. Using ultrashort pulses, we have also investigated the dynamics of coherent phonons (CPs) in SWNTs, including both the low frequency radial breathing mode and high frequency G-mode phonons. Pulse shaping techniques allowed us to generate and detect CPs in SWNTs in a chirality-selective manner, which provided insight into the chirality dependence of light absorption, phonon generation, and phonon-induced band-structure modulations. Finally, we observed novel large- amplitude CPs through near-band-edge excitations as well as strongly polarization-dependent CP signals in highly-aligned SWNTs.

[1]This work was performed in collaboration with Y. Murakami, A. Srivastava, T. A. Searles, L. G. Booshehri, E. H. Hároz, D. T. Morris, J.-H. Kim, K.-J. Yee, Y.-S. Lim, G. D. Sanders, C. J. Stanton, and R. Saito.

[2]Y. Murakami and J. Kono, Phys. Rev. Lett. **102**, 037401 (2009); Phys. Rev. B **80**, 035432 (2009); A. Srivastava and J. Kono, Phys. Rev. B **79**, 205407 (2009); J.-H. Kim *et al.*, Phys. Rev. Lett. **102**, 037402 (2009); G. D. Sanders *et al.*, Phys. Rev. B **79**, 205434 (2009); Y.-S. Lim *et al.*, ACS Nano **4**, 3222 (2010); L. G. Booshehri *et al.*, arXiv:1007.3144v1.