Relaxation Pathways of Excitations in Semiconducting Carbon Nanotubes

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Experimental studies of the excitation relaxation dynamics in single-walled semiconducting carbon nanotubes indicate changes of the observable signals on a very fast timescale [1, 2]. We present a thorough analysis of one- and two-color transient absorption measurements performed on single- and double-walled semiconducting carbon nanotubes. By combining the currently existing models describing exciton–exciton annihilation—the coherent and the diffusion-limited ones—we are able to simultaneously reproduce excitation kinetics following both E11 and E22 pump conditions. Our simulations revealed the fundamental photophysical behavior of one-dimensional coherent excitons and non-trivial excitation relaxation pathways. In particular, we found that after non-linear annihilation a doubly-excited exciton relaxes directly to its E11 state bypassing the intermediate E22 manifold, so that after excitation resonant with the E11 transition, the E22 state remains unpopulated. A quantitative explanation for the observed much faster excitation kinetics probed at E22 manifold, comparing to those probed at the E11 band, is also provided.

[1] M. W. Graham, J. Chmeliov, Y.-Z. Ma, H. Shinohara, A. A. Green, M. C. Hersam, L. Valkunas and G. R. Fleming, *J. Phys. Chem. B*, **115**, 5201–5211 (2011).
[2] J. Chmeliov, J. Narkeliunas, M. W. Graham, G. R. Fleming and L. Valkunas, *Nanoscale*, **8**, 1618-1626 (2016).