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Ultrafast Spectral Diffusion of the First Subband Exciton in Single-Wall Carbon Nanotubes DANIEL SCHILLING, TOBIAS HERTEL, Julius-Maximilans University of Wurzburg — The width of optical transitions in semiconductors is determined by homogeneous and inhomogeneous contributions. Here, we report on the determination of homogeneous linewidths for the first exciton subband transition and the dynamics of spectral diffusion in single-wall carbon nanotubes (SWNTs) using one- and two-dimensional time resolved spectral hole burning spectroscopy. Our investigation of highly purified semiconducting (6,5)-SWNTs suggests that room temperature homogeneous linewidths are on the order of 4 meV and are rapidly broadened by an ultrafast sub-ps spectral diffusion process. These findings are supported by our off-resonant excitation experiments where we observe sub-ps population transfer reflecting the thermal distribution of energy levels around the first subband exciton transition. The results of temperature-dependent spectral hole burning experiments between 17 K and 293 K suggest that homogeneous linewidths are due to exciton interaction with low energy optical phonons, most likely of the radial breathing mode type. In contrast, we find that inhomogeneous broadening is determined by an electronic degree of freedom such as ultrafast intra-tube exciton diffusion which is characteristic and unique for excitons in these one-dimensional semiconductors.

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