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Saturation of Photoluminescence from Carbon Nanotubes at High Laser Intensities: Exciton-Exciton Annihilation near the Mott Density YOICHI MURAKAMI, Rice University, The University of Tokyo, JUNICHIRO KONO, Rice University — We have carried out a nonlinear photoluminescence excitation (PLE) spectroscopy study of carbon nanotube ensembles using intense, femtosecond, and wavelength-tunable optical pulses. For all PL features we examined, their intensities were seen to saturate at high laser fluence, irrespective of whether the excitation was resonant or non-resonant with the E_{22} transition. As the fluence was increased from the linear regime to the saturation regime, excitation resonances at E_{22} energies gradually broadened and eventually became completely flat at the highest fluence, indicating that the PL intensity became independent of the excitation wavelength. However, the energies and lineshape of PL *emission peaks* did not show any changes throughout the entire range of fluence used. Through absorption spectroscopy at high laser intensities, we also demonstrated that E_{22} *absorption peaks* do not show any shift or broadening even at high laser fluence, indicating that state-filling or scattering is not the cause of the observed “flattening” of the excitation spectra. We developed a model to explain these observations by carefully taking into account the spatial overlap of excitons when the average inter-exciton distance approaches the Bohr radius in the exciton-exciton annihilation process.

Yoichi Murakami
Rice University

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