Saturation of Photoluminescence from Carbon Nanotubes at High Laser Intensities: Exciton-Exciton Annihilation near the Mott Density

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Nonlinear photoluminescence excitation (PLE) spectroscopy of individualized carbon nanotube ensembles has been carried out using wavelength-tunable femtosecond optical pulses. All the PL features we examined were seen to saturate at high laser fluence irrespective of the excitation wavelength. As the fluence was increased from the linear to the saturation regime, we found that excitation resonances at E22 energies gradually broadened and eventually became completely flat, where the PL intensity became independent of the excitation wavelength (Fig. 1).

Through absorption spectroscopy at high laser intensities, we demonstrated that *state-filling or strong 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 in the exciton-exciton annihilation process. The developed model shows an excellent agreement with the observed saturation behavior as shown by Fig. 2 up to near the Mott density, where the average inter-exciton distance approaches the Bohr radius. *Based on the saturation curve analysis, we estimated for the first time the density of excitons in carbon nanotubes* for the nonlinear regime extended up to near the Mott density.

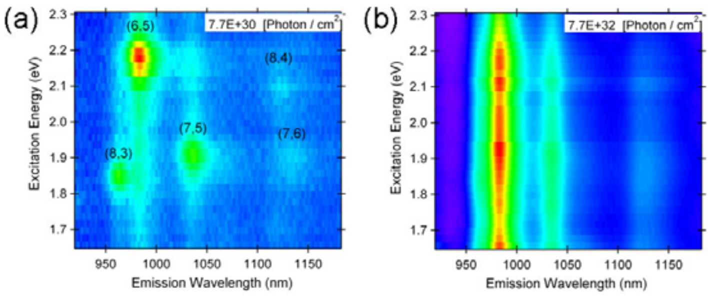

Fig. 1. PLE maps for the (a) linear (7.7E30 photon/cm^2) and (b) saturated (7.7E32 photon/cm^2) regimes. (Sample: micelle-suspended CoMoCAT in D_2O)

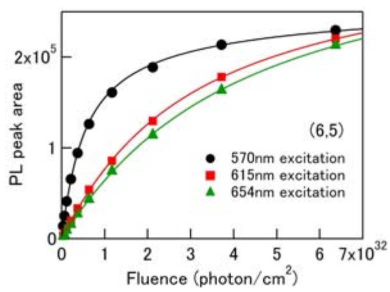

Fig. 2. Dependence of the PL peak area of (6,5) tubes on the laser fluence at various excitation wavelengths. Solid lines are the behaviors fitted by the developed model.

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