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Low Temperature, Photoluminescence and Photoluminescence Excitation Studies of Individual Carbon Nanotubes

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Semiconducting, single-walled carbon nanotubes are nanoscale, near-infrared light emitters that have great potential for a wide variety of optoelectronic applications. A detail understanding of fundamental photophysics of nanotubes is essential to fully exploit this potential. To address the nature of fundamental photoexcitations in nanotubes, we perform for the first time low-temperature, photoluminescence (PL) and PL excitation (PLE) studies at the single-nanotube level. In our PL spectra, we observed two types of features. Some of the nanotubes show sharp (sub-meV to a few meV linewidths), symmetric spectral lines that can be attributed to on- dimensional (1D) excitons. On the other hand, we also detect broad (¿ 6meV linewidths), asymmetric peaks that show strong thermal broadening on their high-energy sides. The spectral shape as well as the unusual temperature dependence of these peaks can be explained in terms of the Fermi-edge-singularity effect that arises from many-body interactions of photoexcited carriers with pre-existing population of carriers introduced into nanotubes at the preparation stage (unintentional doping). Our PLE spectra, in addition to features due to a direct excitation to the second electronic transition, exhibit a number of strong phonon-assisted transitions involving the excitation of one or more phonon modes together with the first electronic state. Surprisingly, the phonon replicas are as intense as the zero-phonon transition associated with the second electronic state. In contrast to a small width of emission lines, most of the PLE features are characterized by tens of meV linewidths indicating significant lifetime broadening induced by inelastic electron-phonon scattering. All of these observations suggest that strong electron-phonon coupling gives rise to a significantly more complex structure of nanotube absorption spectra than it is assumed in a simple picture of optical transitions dominated by singularities in the 1D energy spectrum.