Exciton luminescence of single-walled carbon nanotubes investigated by single nanotube spectroscopy

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Recent discovery of efficient photoluminescence (PL) from isolated semiconducting single-walled carbon nanotubes (SWNTs) has stimulated considerable efforts in understanding optical properties of SWNTs. The semiconducting SWNTs are one-dimensional (1D) systems with direct-gap band structures and their unique optical properties are determined by the dynamics of 1D excitons because of the extremely strong electron-hole interactions (excitonic effects) in 1D systems. The PL energy of SWNTs depends strongly on their diameter (chiral index). However, the diameter and temperature dependences of the PL spectral shape of SWNTs are not fully investigated.

In this work, we have studied PL spectra of about 200 different isolated single SWNTs (diameter: 0.75-1.25 nm), using a home-built scanning confocal optical microscope [1]. The SWNTs samples were synthesized on patterned Si substrates by an alcohol catalytic chemical vapor deposition (ACCVD) method and were excited with a continuous-wave He-Ne laser (1.959 eV). Single nanotube spectroscopy provides intrinsic spectra with no inhomogeneous broadening, and gives us substantial understanding of exciton properties in SWNTs.

The PL spectral shapes of the single SWNTs can be approximately fitted by single Lorentzian functions. In addition, the Stokes shift is very small for each nanotube species. These indicate that the observed PL peaks of single SWNTs correspond to the zero-phonon lines of 1D free excitons, and the spectral linewidth is determined by the homogeneous broadening. From the results of single nanotube measurements, it is found that the PL linewidth clearly becomes broader as the tube diameter decreases. Our observation suggests that the exciton-phonon interaction becomes stronger with a decrease of the diameter, i.e., with an increase of the surface curvature. This is consistent with recent numerical results that certain phonon modes in SWNTs are enhanced compared with graphite because of their surface curvature [2]. From the temperature and chirality dependence of the PL spectral linewidth, the mechanism of the exciton-phonon interactions will be discussed.

[1] K. Matsuda *et al.*, Appl. Phys. Lett. **86**, 123116 (2005).
[2] J. Jiang *et al.*, Phys. Rev. B, **71**, 045417 (2005).