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Exciton transition energy in photoluminescence of single-walled carbon nanotubes

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Photoluminescence excitation (PLE) spectroscopy of single-walled carbon nanotubes (SWNTs) has been extensively studied for characterization of their unique electronic properties due to the one-dimensionality. The precise evaluation of exciton transition energy is essential for experimental assignments of optical absorption, photoluminescence, and resonant Raman scatterings. Here, several experimental topics related to exciton transition energy observed photoluminescence spectroscopy are discussed, namely, excitonic phonon side-band, absorption of cross-polarized light, dielectric environmental effect. Detailed DFT-level theoretical studies are desired to understand and predict these phenomena.

We have demonstrated the excitonic phonon side-band peaks in the PLE spectra of micelle-suspended SWNTs by employing the SWNTs from ^{13}C [1]. By comparing photoluminescence excitation spectra of SW^{13}CNTs and normal SWNTs, the excitonic phonon sideband due to strong exciton-phonon interaction was clearly identified with the expected isotope shift. In addition to the direct experimental proof of the strong exciton-phonon interaction, we also found low-intensity "pure electronic" features whose origin has never been elucidated.

We ascribed these peaks to the cross polarized absorption by the polarized-PLE experiment on partially aligned SWNTs in a gelatine film. Then, we further studied polarized PLE spectra of various (n, m) nanotubes in surfactant suspension. Using a simple theory for PL anisotropy, we have obtained decomposed PL maps for parallel and perpendicular polarization from two PL maps measured by so-called L-format method. Distinct absorption peaks corresponding to E_{12} and E_{21} transitions for perpendicular polarization were observed [2]. Observed E_{12} and E_{21} energies were considerably blue-shifted compared to the qualitative values predicted within a single-particle theory. The results indicate a smaller exciton binding energy for perpendicular excitations than for parallel excitations.

Optical transition energies of SWNTs are affected up to 80 meV by the change of environment materials around SWNTs [3], which is known as an environmental effect. Environmental dielectric screening effects on exciton transition energies in SWNTs have been studied quantitatively in the range of dielectric constants from 1.0 to 37 by immersing SWNTs bridged over trenches in various organic solvents by means of photoluminescence excitation spectroscopy. With increasing environmental dielectric constant ε_{env}, both E_{11} and E_{22} exhibited a red shift by several tens meV and a tendency to saturate at a $\varepsilon_{env} \sim 5$ without an indication of significant (n,m) dependence [4]. The red shifts can be explained by dielectric screening of the repulsive electron-electron interaction. We make a simple model for the relation between dielectric constant of environment and a static dielectric constant describing the effects of electrons in core states, σ bonds and surrounding materials [5]. Although the model is very simple, calculated results well reproduce experimental transition energy dependence on dielectric constant of various surrounding materials.

References:

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