Optical Absorption and Photoluminescence Excitation Spectroscopy of SWNTs

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Optical absorption and photoluminescence excitation (PLE) spectroscopy of singlewalled carbon nanotubes (SWNTs) have been extensively studied for characterization of their unique electronic properties due to the one-dimensionality. In our previous studies, we have demonstrated the excitonic phonon side-band peaks in the PLE spectra of isolated SWNTs by employing the SWNTs from ¹³C [1]. Then, the PL peaks for cross-polarized excitation to the nanotube axis are further identified by polarized PLE spectroscopy [2]. The measured resonance energies for perpendicular excitations were considerably larger than the values predicted within a single-particle theory. These results indicate a smaller exciton binding energy for perpendicular excitations than for parallel excitations [2]. In addition to the physical interest in unique excitonic features of 1-D material [3-5], clear identification of absorption and PL features is very important for the estimation of chirality distribution of produced nanotubes [6].

In this study, we focus on optical absorption and polarized PLE spectra for UV-Vis range [2]. In PL for perpendicular excitation, we found small but nonzero intensity tails above the distinct peaks of parallel excitation. PL intensities corresponding to the perpendicular excitation were even comparable to those for the parallel excitation in a certain energy region. This result indicates that one can not neglect the contribution of perpendicular excitations in optical measurements of SWNTs for UV-Vis range beyond E_{22} absorption. In addition, we studied details of optical absorption of SWNTs in UV range by comparing conventional optical absorption and PLE spectroscopy. In an optical absorption spectrum of SWNTs, there are two major peaks corresponding to parallel (~ 4.5 eV) and perpendicular (~ 5.25 eV) excitation to the SWNT axis [2]. As we compared absorption and PLE spectra. Since only semiconducting SWNTs contribute to a PLE spectrum, this result indicates that ~ 4.5 eV peak is only from metallic nanotubes, even though we cannot completely neglect the possibility of very slow relaxation time to the E_{11} excitonic state for the ~ 4.5 eV peak.

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