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Extensive study of cross-polarized exciton absorption of single-walled carbon nanotubes by polarized photoluminescence excitation spectroscopy

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In our previous study [1], we found that some PLE peaks for cross-polarized excitation to the nanotube axis (E_{12} exciton absorption) can be clearly observed in the polarized PLE spectra of micelle-suspended SWNTs with relatively small diameters ($<$ 1 nm). In this report, we have studied polarized PLE spectra of various (n, m) SWNTs in a surfactant suspension with diameters from 0.8 nm to 1.2 nm for a wide excitation range from UV to near IR using Xe lamp and Ti :Sapphire CW laser as light sources. In addition to bright E_{12} (or E_{21}) peaks for many (n, m) SWNTs, we observed small unassigned cross-polarized excitation peaks about 230 meV below the E_{12} peaks. We attribute these peaks to E_{12} dark excitons. For cross-polarized exciton absorption, it is predicted that the bonding combination of E_{12} and E_{21} at K and K′ points becomes an optically active bright exciton and the anti-bonding combination becomes a dark exciton when considering symmetric structure between valence and conduction band [2]. However, the real band structure is known to be slightly asymmetric, and the degeneracy of E_{12} and E_{21} gap is imperfect due to this asymmetry. Hence, we expect that the dark exciton for E_{12} becomes slightly bright and could be observed. We also investigated the family pattern of E_{12}. Since the S1 band and S2 band are on opposite sides of the K point, the family effect is usually opposite for S1 and S2 bands. Hence, for the E_{12} excitation, family effects from the S1 and S2 bands are expected to be always cancelled out. We predict the family pattern of E_{12} using E_{11} and E_{22} just by taking the average of E_{11} and E_{22} with a simple diameter-dependent up-shift from the average value, and compare the observed E_{12} plot with the prediction. The satisfactory agreement of this prediction further supports our assignment of these cross-polarized excitation peaks as E_{12} (or E_{21}).

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