Spectral features due to dark exciton in photoluminescence map of single-walled carbon nanotubes

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Features in Photoluminescence Map of SWNTs Over the past seven years photoluminescence (PL) spectroscopy has emerged as an important characterization tool for determining the (n, m) chirality and population of semiconducting SWNTs. This technique is essential for the chirality selective growth and/or the separation depending on chirality. Under excitation with the appropriate wavelength of light, SWNTs exhibit characteristic peaks with well-defined emission from their corresponding excitonic states. Besides the quantum yield of the photoluminescence, sound understanding of extra features mainly due to dark exciton are necessary, in order to use this PL map for the determination of chirality population of SWNTs. Such features are shown in Fig. 1, where the sample contains only (7,5) and a trace of (6,5); at least within PL spectroscopic measure. Optical absorption by excitonic phonon sideband due to mixing of dark exciton and K phonon (marked as E_{11}+K and E_{22}+K) is well discussed [1]. And, the cross-polarized absorption marked as E_{12}^{+} is confirmed [2]. Here, the phonon sideband in emission (E_{11}–K) and quasi-dark exciton in cross-polarized excitation (E_{12}^{-}) are discussed.

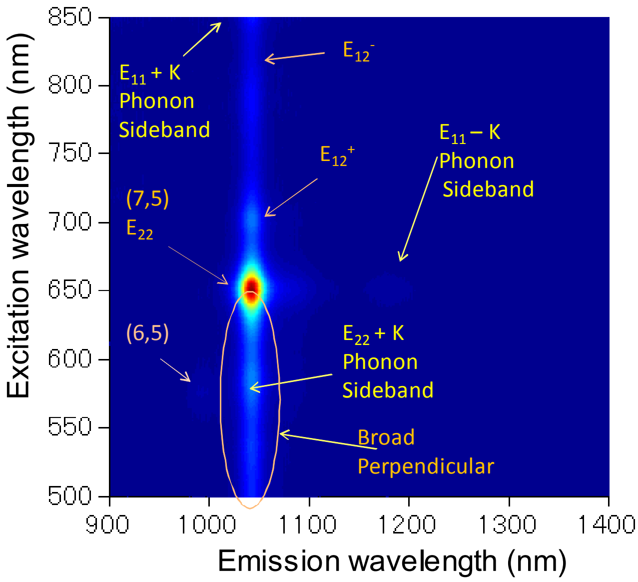

Fig. 1 Various features in photoluminescence map

Phonon Sideband in Absorption and Emission We performed detailed PL spectroscopy studies of three different types of single-walled carbon nanotubes (SWNTs) by using samples that contain essentially only one chiral type of SWNT, (6,5), (7,5), or (10,5). The observed PL spectra unambiguously show the existence of an emission sideband at ~ 145 meV below the lowest singlet excitonic (E_{11}) level. We find that the energy separation between the E_{11} level and the sideband is almost independent of the SWNT diameter. Based on this, we ascribe the origin of the observed sideband to coupling between K-point phonons and dipole-forbidden dark excitons [3].

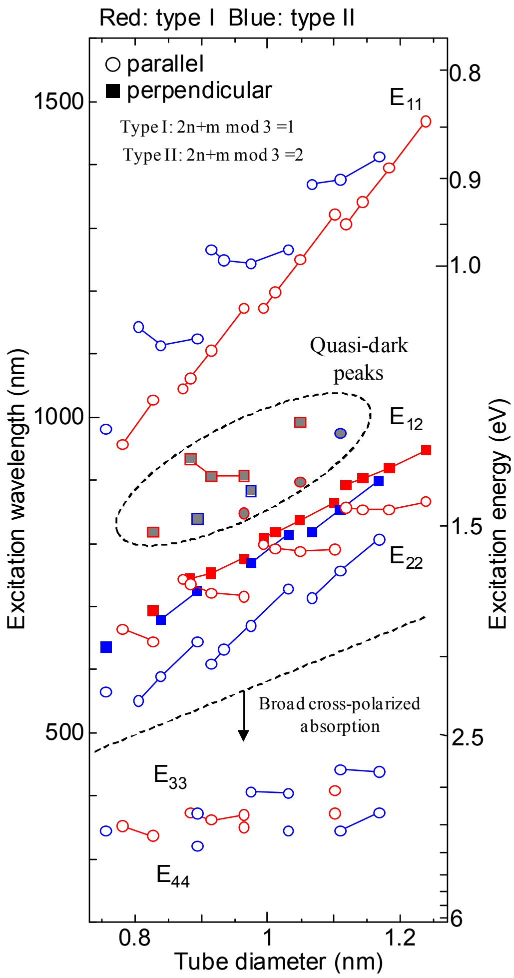

Fig. 2 Kataura plot including cross-polarized absorption.

Quasi-Dark Exciton in Cross-Polarized Absorption We performed the direct observation of the spin-singlet transverse dark excitons in SWNTs through the polarized photoluminescence excitation spectroscopy as shown in Fig. 2. We studied the exciton optical absorption polarized transverse to the nanotube axis. The intrinsic asymmetry between valence and conduction bands lifts the degeneracy of the spin-singlet transverse excitons at two equivalent K and K' valleys in momentum space, leading to "brightening" of transverse dark exciton states. The energy splitting between transverse bright and dark states were about 200-300 meV, much larger than the bright-dark splitting of longitudinal excitons. The chirality-dependent spectral weight transfers to the transverse dark states from the bright states were clearly observed.

References

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