東京大学グローバルCOEプログラム　機械システム・イノベーション国際拠点

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Raman Studies of Exciton Behavior in Single-Walled Carbon Nanotubes

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要旨

The realization of photonic, optoelectronic, sensing, and other applications of carbon nanotubes requires a complete understanding of their electronic structure, nature of the optical excited states, and how electronic and phonon structure couple. Resonance Raman spectroscopy will be demonstrated as a sensitive probe of several related behaviors. A scaling law analysis of transition energies will be presented as a route to revealing new electronic behavior, including results for E_{33} and E_{44} energies for small diameter (0.7 to 1.2 nm) nanotubes probed with resonance Raman and photoluminescence excitation spectroscopy in the deep blue to UV (488 nm to 280 nm) excitation regions. We demonstrate that the E_{33} and E_{44} scaling behavior for nanotubes with diameter < 0.9 nm deviates significantly from that previously observed for larger diameter nanotubes. The scaling analysis allows a first demonstration that the E_{33} and E_{44} energy trend lines for a given 2n+m branch do in fact cross over each other. Additionally, our observation of a pronounced negative trend in scaling behavior at small diameters represents a breakdown in the scaling-law physics that may be interpreted as a many-body exciton effect that becomes significant only at high curvatures. Application of the scaling law analysis to transition energies for metallic nanotubes suggests that the transitions are excitonic in nature and that relative scaling of electron self-energies and exciton binding energies in metallic nanotubes closely matches that found in semiconductors. This similarity in behavior can be understood in terms of similar regions of the Brillouin zone being sampled by E_{11}^M and E_{11}^S and E_{22}^S (and by E_{22}^M and E_{33}^S and E_{44}^S). Additionally, for large diameter nanotubes (> 1.3 nm) we now observe the previously elusive upper branch signatures for several chiralities for both E_{11}^M and E_{22}^M excitation. These results are discussed as a consequence of the nodal behavior of exciton-phonon coupling. While theoretical calculations for the (*n,m*) dependent matrix elements predict the RBM intensity should decrease with increasing diameter, the opposite behavior is observed experimentally. We show this to be a consequence of an increase in the resonance Raman broadening factor G as diameter decreases. Finally, we present Raman excitation data from surfactant suspensions highly enriched in metallic nanotubes via density gradient ultracentrifugation. Specifically, we will focus on the evolution of G-band behavior over a wide range of chiralities enabled by these new sample types. The variable behavior of the Breit-Wigner-Fano line in these enriched ensemble samples will be discussed.

主催：　東京大学グローバルCOEプログラム「機械システム・イノベーション国際拠点」事務室
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