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Raman Studies of Exciton and Exciton-Phonon Coupling Behavior in Metallic Single-Walled Carbon Nanotubes

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A scaling law analysis of carbon nanotube transition energies has been found to be valuable in revealing new electronic behaviors for the third and fourth transitions in semiconducting nanotubes. In the work presented here, we discuss resonance Raman data obtained for the E_{11}^M and E_{22}^M transitions of a broad diameter range (0.7 - 4 nm) of metallic carbon nanotubes. We show that 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. Also, 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 Γ 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.