probe these properties, we investigate the third-order, coherent anti-stokes (CAS) response of individual SWCNTs using a dual color, four-wave-mixing technique. Despite SWCNTs being nanoscale objects much smaller than the wavelength of light, the CAS response allows single SWCNTs to be optically imaged with good spatial resolution [1]. The absence of a pronounced Raman signature shows that the CAS response is dominated by electronic, rather than vibrational, dynamics. Indeed, the CAS signal allows for straightforward optical discrimination between metallic and semiconducting SWCNTs. However, variability exists in the CAS intensity that may depend on electrical contact resistances, charge carrier densities, and local physiochemical effects. Moreover, identification of the third-order CAS response in SWCNTs requires proper accounting of other non-linear processes including two-photon fluorescence. [1] H. Kim et al, Nano Lett. 9 2991-2995 (2009)

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Exciton effects on Raman and photoluminescence of single wall carbon nanotubes

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Resonance Raman spectroscopy and photoluminescence have been used for optical characterization and investigation of excitonic properties of single wall carbon nanotubes (SWNTs). Experiments and theories have been demonstrated a diameter and chirality dependence for resonance Raman and photoluminescence intensity of SWNTs. Excitonic effects change the intensity and optical transition energy of the photoluminescence and resonance Raman peaks. To investigate excitonic effects in resonance Raman and photoluminescence, we need to consider and calculate the intensity in the exciton picture. In this study we discuss the excitonic effects of the photoluminescence and resonance Raman intensity, and the dependence of the photoluminescence and resonance Raman intensity on diameter and chirality. Here we use the exciton-phonon and exciton-photon matrix elements in the framework of the tight-binding scheme in order to calculate the resonance Raman and photoluminescence intensity in the exciton picture. The exciton energy and exciton wave function coefficient of SWNTs are calculated by solving the Bethe-Salpeter equation in which the one particle energies are given by the tight-binding method. The screening from environment and nanotubes itself is expressed by a dielectric constant. In the calculation we change a dielectric constant to consider environmental effects. The behavior of the calculated intensity in the exciton picture reproduces the experimental results.

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Evidence for excitons in metallic carbon nanotubes by temperature dependent Raman scattering

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Single-walled carbon nanotubes (SWCNTs) are promising candidates for a variety of applications [1]. Optical transitions play a central role in understanding the fundamental properties of carbon nanotubes and for optoelectronic devices. Both theoretical and experimental studies revealed that light absorption excites strongly correlated electron-hole pairs in semiconducting nanotubes, known as excitons, with binding energies of several hundred meV [2-5]. We present resonant Raman measurements of the optical transition energy Eii in metallic and semiconducting nanotubes at different temperatures. In semiconducting nanotubes, the transition energy decreases at high temperatures, as observed many semiconducting systems. In metallic nanotubes, on the other hand, Eii first decreases and then increases again. We interpret this unusual behavior of metallic nanotubes in terms of excitons in metallic nanotubes. We suggest that the excitons in metallic nanotubes, due to their small binding energy, dissociate into free electron-hole pairs at temperatures related to the exciton binding energy. Our results are in good agreement with theoretical predictions and experimental absorption measurements [6]. [1] S. Reich, C. Thomsen and J. Maultzsch, Carbon Nanotubes: Basic Concepts and Physical Properties (Wiley-VCH, Berlin, 2004) [2] Maultzsch et al. PRB 72, 241402 (2005) [3] Wang et al. Science 208, 838 (2005) [4] Spataru et al. PRL 92, 077402 (2004) [5] Deslippe et al. Nano Letters 8, 1626 (2007) [6] Wang et al. PRL 99, 227401 (2007)

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Raman intensities of the radial-breathing mode in carbon nanotubes: (n,m) abundances versus scattering efficiencies

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Nanotube ensembles show strong variations in Raman intensities of the different tubes (n,m) which can either be interpreted as variations in the (n,m) abundances or variations in the scattering efficiencies. In order to address this issue we collected Raman intensities of the radial breathing mode from a large number of different tubes (n,m) (HiPCO). To collect Raman intensities from resonantly excited tubes we varied the excitation energy between 1.15 and 2.15 eV. We observe systematic variations of the Raman intensities for different (n,m) including dependences on the optical transition, diameter, family and chiral angle. Here we focus on intensity variations depending on family and and chiral angle which we contribute to variations in the exciton-phonon coupling. In particular, we observe an intensity minimum for nantubes with chiral angle = 20o . This finding corresponds to a minimum in the exciton-phonon coupling along the K-K direction in the Brillouin zone of graphene. Furthermore we discuss intensity variations which are due to different natural electronic line widths. The line width shows a dependence on diameter and varies between metallic and semiconducting tubes.

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Screening due to inter-wall interaction and exciton properties in double-walled carbon nanotubes

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Exciton properties in double-walled carbon nanotubes are expected to be different from those in single-walled carbon nanotubes because excitons in the inner (outer) tube are screened by electrons/holes in the outer (inner) tube through the inter-wall Coulomb interaction. In this work, we have studied effects of screening due to the inter-wall Coulomb interaction on excitons and optical spectra in semiconducting double-walled carbon nanotubes within the effective-mass theory and a static screened Hartree-Fock approximation. Our calculations clarify that the intra-wall electron-hole interaction is largely suppressed by inter-wall screening effects. The suppression is sensitive to the effective inter-wall distance between the inner and outer tubes and leads to reduce the exciton binding energy as well as the band gap energy. As a result, the exciton energy levels are redshifted slightly from those in the single-walled carbon nanotube with the same diameter. We find that the energy shift of the ground exciton has little dependence on the tube diameter, in contrast to that of the excited exciton. The exciton properties in the case of a metallic outer or inner tube, where the screening can be changed dramatically, will also be discussed.

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Importance of Exciton Coupling on the Upper Density Limit of Optically Created Excitons in SWCNTs

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An important feature of single walled carbon nanotubes (SWCNTs) are their quasi 1-D, chiral structure which gives rise to exitonic photoluminescence from 2/3 of the species. These excitons can be