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Two-photon photoluminescence and exciton binding energies in single-walled carbon nanotubes

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The optical properties of carbon nanotubes are thought to be dominated by excitonic effects. Exciton binding energies between 100 meV and 1 eV have been predicted, depending on diameter, chiral angle, and surrounding medium. Experimental proof for these large binding energies, however, has been missing so far. Here we present direct experimental evidence that the elementary optical excitations of carbon nanotubes are strongly Coulomb-correlated, quasi-one dimensionally confined electron-hole pairs, stable even at room temperature. By comparing one-photon and two-photon luminescence excitation spectra, we probe excitonic states with distinctly different wavefunction symmetry [1]. The energy splitting between one-photon active and two-photon active states is a distinct fingerprint of excitonic interactions in carbon nanotubes and binding energies between 300 and 500 meV are derived. Our results are strongly supported by *ab initio* calculations of two-photon absorption spectra [2]. We discover that for all the tubes studied, excitonic effects are very strong and are crucial for the one-photon as well as the two-photon spectra in both the peak shapes and positions. The implications of these results for the fluorescence yield and lifetime of single-walled carbon nanotubes [3] will be discussed. [1] J. Maultzsch et al., Phys. Rev. B 72, Rxxx (2005); cond-mat/0505150. [2] E. Chang, G. Bussi, A. Ruini, and E. Molinari, Phys. Rev. Lett. 92, 196401 (2004). [3] A. Hagen et al., Phys. Rev. Lett. 95, 197401 (2005).