Observation of electronic raman scattering in metallic carbon nanotubes

H. Farhat¹, **S. Berciaud^{2,3}**, M. Kalbac¹, R. Saito⁴, T. F. Heinz², M. S. Dresselhaus¹, and J. Kong¹

¹Massachusetts Institute of Technology, Cambridge, MA 02139, USA

²Columbia University, New York, NY 10027, USA

³IPCMS (UMR 7504), Université de Strasbourg and CNRS, F-67034 Strasbourg, FRANCE

⁴Tohoku University, Sendai, 980-8578, JAPAN

berciaud@unistra.fr

Raman scattering spectroscopy, a powerful tool for studying elementary excitations in materials, has been instrumental in the progress of carbon nanotube research. The Raman features that have been studied to date in carbon nanotubes are associated with the phonon modes that scatter light due to electron-phonon coupling [1]. In metallic nanotubes, low energy electron-hole pairs are another important type of excitation [2], but their inelastic scattering of light [3], via the Coulomb interaction, has not been yet observed. Here, we report on a new feature in the Raman spectrum of individual metallic carbon nanotubes, that we attribute to resonant electronic Raman scattering (ERS) from low-energy electron-hole pairs created across the "graphene-like" linear electronic subbands of metallic nanotubes. The ERS spectra exhibitbroad peaks that appear at a constant photon energy due to a resonance of the scattered photons with the optical transition energies of the metallic nanotube. Our results are consistent with the picture that the mild asymmetry in the lineshape of the Raman G-mode feature [4] is the result of an interference between the overlapping ERS and vibrational Raman scattering by the longitudinal optical (LO) phonon mode.

References

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Figure 1: Electronic Raman Scattering in a structure assigned (21,15) metallic carbon

nanotube. The Rayleigh (a) and Raman (b) scattering spectra of an individual, freestanding metallic nanotube are used to determine its (*n*,*m*) indices [5, 6]. The labels in a indicate the optical transitions, while the laser energy for the Raman measurement is indicated with arrow. Likewise, the arrow in the Raman spectrum indicate the energies of the optical transitions as obtained from fitting the Rayleigh scattering spectra using an excitonic model [7]. The ERS feature is the broad peak at ~500 cm⁻¹ in (b). The corresponding scattered photon energy (2.08 eV, see top axis in (b)) match the energy of the M_{22}^{-1} transition. No ERS features are found in the Raman spectrum of semiconducting nanotubes.