

Screening Of Excitons In Single, Suspended Carbon Nanotubes

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We study suspended carbon nanotubes using resonant Raman spectroscopy before and after immersion in water and observe red shifts up to 30 meV in the optical transition energies. We thus quantify the effect of screening.

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By virtue of their geometry, the electronic and vibrational properties of carbon nanotubes are inherently very sensitive to changes in the environment which can affect the nanotube in a number of ways. Quantifying these effects is of great import to a wide range of applications, especially transport. Here we report for the first time shifting of the electronic energy levels due almost entirely to screening of the Coulomb interaction in single CNTs suspended across trenches. Theorists predict the presence of excitons with large binding energies in carbon nanotubes and recent two-photon experiments have confirmed just that. Given binding energies in the hundreds of milli-electron volts and knowing that these coulomb energies are highly sensitive to screening, one might expect that the measured optical transition energies would change significantly with dielectric environment. But in fact the literature reports variations on a scale of just a few tens of milli-electron volts across dielectric environments as different as CNT bundles in solution, CNTs in SDS encapsulation, and individual nanotubes suspended in air. We explain this apparent contradiction below.

We use RRS to probe the underlying electronic structure of single, CVD grown CNTs suspended across trenches. The Stokes peak heights for each Raman active phonon mode are plotted against laser excitation energy. The resulting curves, one per phonon mode, are referred to as resonance excitation profiles, (REPs). The curve will be, in general, double peaked where the lower energy peak corresponds to resonance of the incoming photon with an electronic level, “incoming resonance”, and the higher energy peak corresponds to resonance of the outgoing, or scattered, photon, “outgoing resonance.” By fitting a one phonon, exciton mediated REP line shape, we determine the underlying electronic transition resonant energy. The nanotube is initially measured in dry N₂ in an enclosed chamber. Water vapor is added to the nitrogen and the humidity is measured with a hygrometer. Finally, the sample is directly immersed in water. For each phonon mode, we then have a REP predicting E_{ij} for each dielectric environment and thus have a measure of the shift in the electronic level with increasing ϵ . In figure 1, we see results for outgoing resonances of the G+ tangential and 1734 wavenumber phonon modes. Our data show monotonically increasing red shifts with increasing dielectric environment.

All data was taken at room temperature and mechanical strain is negligible. Charge transfer has been shown to be associated with a change in the tangential phonon energy which we do not observe. Therefore we ascribe the observed shifts solely to dielectric screening. Coulomb interactions are expected to be large in 1D systems. By modeling the carbon nanotube as a 1D quantum wire in an external dielectric we show how dielectric screening indeed leads to large energy shifts of the electron-electron and electron hole interaction energies yet these energy shifts conspire to counteract each other leading to small changes in the observed optical transition energies. Our data agrees with published two photon data that measure exciton binding energies that are several hundred meV in a screened environment. We further assert that, using our model, our data demonstrates binding energies that are 1 eV in an unscreened environment and that the band gap renormalization (electron electron interactions) is larger still.

Figure 1: (a) G+ outgoing resonance (b) mode 1734 outgoing resonance

