Temperature Dependence of the Optical Transitions in Single-walled Carbon Nanotubes

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Single-walled carbon nanotubes (SWCNTs) are promising candidates for a variety of electronic and optical applications. Optical transitions play a central role in the understanding of carbon nanotubes [1]. 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 [2,3].

There is a strong debate on how much optical transitions are dominated by excitons in metallic SWCNTs as well. In general, binding energies are higher in nanotubes due to their one-dimensional character in comparison to bulk semiconductors. Theoretical calculations show that excitons exist even in metallic SWCNTs with binding energies predicted around 50 meV, depending on the nanotube chiral index (n,m) and the transition energy E_{ii} [4,5].

Here we present results on the temperature dependence of the transition energies E_{ii} of SWCNTs. Tunable Raman spectroscopy has been used to investigate the radial breathing mode (RBM).

We performed measurements of the optical transition energy E_{ii} at different temperatures for both metallic and semiconducting SWCNTs in the range of 300 K and 873 K.

In case of semiconducting SWCNTs, we observe a monotonic decrease of E_{ii} with increasing temperature. Metallic SWCNTs show a different behavior, which can be divided into three parts. First we observe a downshift of E_{ii} as a function of temperature. At higher temperatures, we observe an upshift in the transition energy (blueshift) at approximately 573 K. At even higher temperatures there is again a downshift similar to the first one.

We suggest that this behavior can be interpreted in terms of excitons in metallic nanotubes, which are dissociated into free electron-hole pairs at temperatures related to the exciton binding energies.

References:

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