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Ultrafast and CW Optical Probes of Photoexcitations in Semiconducting Carbon Nanotubes[1]

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Ultrafast dynamics of photoexcitations in semiconducting single walled carbon nanotubes (S-NT) in the form of films and D_2O solutions, have been investigated using low intensity high repetition rate femtosecond laser system based on Ti-sapphire optical parametric oscillator in the spectral range from 0.13 to 1.05 eV. Various transient photoinduced bleaching (PB) and photoinduced absorption (PA) bands were observed, which also show pronounced polarization memory. The PA spectrum does not show Drude free carrier absorption, but instead is composed of two PA bands below the PB of the lowest S-NT absorption band. Also the PB spectrum exactly follows the cw photoluminescence (PL) emission spectrum. We therefore conclude that the primary photoexcitations in S-NT are *excitons* polarized along the NTs. We thus conjecture that the S-NT absorption bands are excitonic in origin; this conclusion is also supported by electroabsorption measurements that show quadratic Stark shift of the absorption bands. In addition, we found that the PL emission is also polarized; from the average PL polarization degree and ps transient polarization memory decay we estimate the PL lifetime in isolated S-NTs in solution to be of the order of 500 ps. Non-radiative decay processes dominate this relatively long PL lifetime. From the PL lifetime and the minute PL quantum efficiency we estimate the radiative PL lifetime in S-NTs to be $\sim$ μsec. This shows that the lowest lying exciton in S-NTs has small oscillator strength, and is in fact dark. The similarity of the PA spectrum in S-NTs and π-conjugated polymers indicates that quasi-1D excitons dominate the photophysics in both organic semiconducting materials. From this and the photon energy of the PA band we estimate the binding energy of the lowest exciton in S-NTs with diameters $\geq$ 1 nm to be $\sim$0.4 eV; in good agreement with two-photon absorption spectroscopy.

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