Quantum light signatures from cavity-embedded carbon nanotubes

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Single-walled carbon nanotubes (SWCNTs) are semiconductors with a direct band gap and are strongly considered for next generation optoelectronic and quantum photonic devices. Their optical recombination is dominated by excitons with inherent strong Coulomb interactions giving rise to exciton binding energies up to 400 meV, about an order of magnitude larger than values found in quantum dots made from compound semiconductors. Electroluminescence of excitons from individual SWCNTs was already demonstrated at room temperature and first photon antibunching signatures of bare SWCNTs recorded liquid Helium temperatures [1] indicate that single photon sources might be feasible which operate at room temperature in the telecom bands by proper choice of the SWCNT chirality.

Besides these outstanding optical properties and device prospects there are fundamental problems to overcome such as the high sensitivity to influences in the surrounding environment of the SWCNT which limits the emission efficiency due to non-radiative recombination, intermittent quantum blinking, and pronounced spectral diffusion from nearby charge fluctuations.

In this talk we will discuss methods how to **e**nhance the light extraction and spectral purity, while simultaneously preventing multi-photon emission, as well as blinking and spectral diffusion in the dielectric environment of a planar cavity [2,3]. Furthermore we will discuss exciton dephasing time measurements recorded for individual SWCNT which are either embedded in a polymer or are free hanging over pillar posts. Compared to what is known from dephasing time measurements of CNT ensembles, our data recorded for individual SWCNTs are significantly longer.

In particular, we found that utilization of nonpolar polystyrene as a cavity dielectric completely removes spectral diffusion and blinking on the ms to multi-second time scale, despite the presence of surfactants, leading to 50-fold enhanced exciton emission and pronounced photon antibunching ($g^2(0)=0.15$) of individual SWCNTs. Furthermore, with these cavity-embedded SWCNTs we have been able to carry out photophysical studies for the first time with nanosecond timing resolution and uncover that fast spectral diffusion processes (1-3 ns) remain that make significant contributions to the spectral linewidth of typically several meV.

We compared our polymer-cavity embedded studies of the exciton photophysics with data from CVD-grown CNTs which bridge an air-gap over pillar posts and which are free from surfactants. In this case it was found that the spectral diffusion signature is absent and the spectral linewidth narrows significantly. Thus this type of residual ns spectral diffusion for the case of polymer embedded SWCNTs seems to be an extrinsic phenomenon inherent to these surfactant wrapped nanotubes.

The reduced linewidth is also in agreement with the measured lengthening of the exciton dephasing time. Values for the free standing CNTs can reach up to several ps and are thus significantly longer than what is typically found from ensemble studies.

The demonstrated enhanced single photon emission from cavity-embedded SWCNTs [3] for temperatures up to 80 K is promising for applications in quantum key distribution. The discovered passivation effect of nonpolar polystyrene with respect to the stability of the optical emission opens a novel pathway towards optoelectronic devices with enhanced performance [2]. Since the exciton dephasing time is largely influenced by the environment, future work aims to explore the intrinsic limit, which according to theoretical calculations should reach into the ns time domain. If these values can be indeed realized than advanced quantum information science protocols relying on indistinguishable photons are within technical feasibility.

References

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