

Environment-dependent exciton dynamics of single-walled carbon nanotubes studied by femtosecond excitation correlation spectroscopy

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Since the discovery of efficient photoluminescence (PL) from isolated single-walled carbon nanotubes (SWNT)s [1], their optical properties have been extensively investigated both from fundamental and technological viewpoints. Recently, both theoretical predictions and experimental studies have shown that the optical properties of SWNTs are determined by the dynamics of excitons formed by the strong electron-hole interaction in the quasi-1-dimensional (1D) systems. However, the recombination dynamics of excitons in the quasi-1D SWNTs is not clear and still controversial.

In this work, we have studied PL lifetimes (τ_{PL}) for four different SWNT samples by means of the femtosecond excitation correlation method with a time resolution of about 150 fs [2]. The SWNTs were synthesized by the alcohol catalytic CVD (ACCVD) method and the high-pressure CO (HiPco) method and were suspended in gelatin films or D₂O solutions. The PL lifetime of SWNTs in D₂O solutions ($\tau_{\text{PL}} \sim 22$ ps) is faster than that in gelatin films ($\tau_{\text{PL}} \sim 31$ ps), whereas the PL lifetimes depend scarcely on the synthesis method and the tube diameter of SWNTs. Moreover, the weak temperature dependence of $1/\tau_{\text{PL}}$ for the SWNTs in gelatin films reveals that the relaxation processes of excitons are determined by the interplay of free excitons and weakly localized excitons in potential fluctuations due to changes in the local environment, rather than the intrinsic effects such as the exchange splitting of excitons. Furthermore, the difference of the exciton dynamics between double-walled carbon nanotubes (DWNTs) and SWNTs will be discussed.

[1] M. J. O'Connell *et al.*, Science 297, 593 (2002).

[2] H. Hirori *et al.*, Phys. Rev. Lett. 97, 257401 (2006).