Magneto-photoluminescence of individual semiconducting single-walled carbon nanotubes

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Since the first experimental evidence [1] of photoluminescence (PL) of semiconducting single-walled carbon nanotubes (SWNTs), studies have been conducted to investigate the optical properties of these nanostructures, motivated by possible applications in the fields of quantum information, biological labeling, opto-electronics or laser technology.

Radiative transition energies in SWNTs are well reproduced by theoretical models which take into account Coulomb interaction within photo-created electron-hole pair, showing the excitonic nature of excited states in these one-dimensional systems [2,3]. The two lowest singlet states play a significant role in the luminescence of SWNTs. The upper one – bright state (B) – is optically active and the lower one – dark state (D) – is a parity forbidden transition. Through the Aharonov Bohm effect, B and D states can be coupled by applying a magnetic field in the direction of the SWNT, so that the lower state acquires oscillator strength and can be studied via PL signal. This opened up the field of magneto-photoluminescence spectroscopy [4,5], as a promising way to investigate the photophysical properties of these states.

We study suspended SWNTs on a lithographed silicon substrate at the individual level using a confocal optical microscopy setup, with a large numerical aperture NA=0.95, operating at cryogenic temperatures down to 2K. A magnetic field up to 7T is produced by superconductive coils. We measure PL spectrum and PL decay of SWNTs under various experimental conditions including changes of the magnetic field, temperature and optical excitation frequency. Figure 1(a) shows the spectacular magnetic brightening of the D state through the emergence of a peak ~10meV below the B state. Figure 1(b) highlights the bi-exponential behavior of the PL decay and the enhancement of the weight of the long-time component due to magnetic coupling. The magnetic brightening of D state is of particular interest because of its tunable oscillator strength.

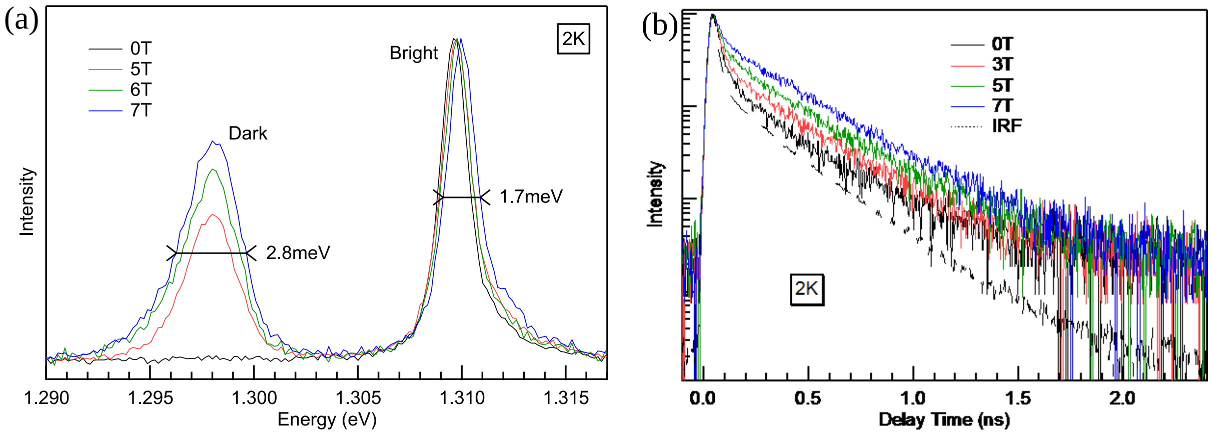

Figure 1: Renormalized luminescence spectrum (a) and decay (b) of two single (6,5) SWNTs at 2K for different magnetic field intensities under optical excitation on S22 transition.

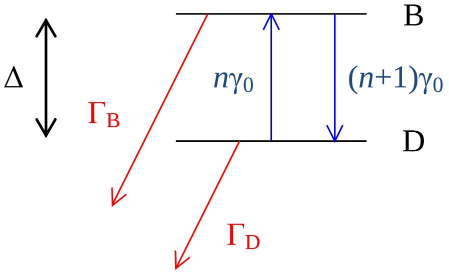

Figure 2: Schematic of the two level model. Γ_D and Γ_B are the decay rates of D and B states respectively. γ_0 is the zero-temperature decay rate from B to D due to phonons. $n=1/(\exp(\Delta/k_BT)-1)$ is the Bose-Einstein phonon occupation number at the energy Δ corresponding to the energy splitting between B and D states.

The simple two level model presented in Fig. 2 leads to a prediction for the magnetic field dependence of PL intensity ratio and energy splitting of B and D states, assuming equal population rates from S22 decay. The bi-exponential behavior observed in the PL decay provides the decay rates of the two relaxation modes. In the low temperature regime ($n \to 0$), the long time τ_L is only related to the D state decay rate ($\tau_L = \Gamma_D^{-1}$) and the short time τ_S derives from the two decay channels of the B state ($\tau_S = (\gamma_0 + \Gamma_B)^{-1}$). Combining the experimental results of the PL spectrum and the PL decay allows a determination of relevant relaxation rates involved in SWNTs PL dynamics.

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