Recent theory [1-4] has predicted the existence of optically-inactive, or “dark” excitons in single-walled carbon nanotubes (SWNTs) below the first optically-active, or “bright” exciton state. This could trap much of the exciton population and contribute to the low quantum yield. Dark states arise due to the doubly-degenerate conduction and valence bands of SWNTs as well as the characteristic strong Coulomb interactions among charge carriers in low-dimensional systems.

We have measured near-infrared polarized photoluminescence (PL) on micelle-suspended SWNT samples in magnetic fields up to 45 T at room temperature (Fig. 1a) and PL excitation on an aligned gelatin SWNT film at low temperature up to 35 T (Fig. 1b). PL measurements were performed using the 45 T Hybrid Magnet, while PL excitation (PLE) was done in a 35 T resistive magnet.

We demonstrate that a magnetic field can significantly increase the PL of semiconducting SWNTs by “brightening” the dark exciton state. At low temperature, the PL intensity increased, or “brightened”, with magnetic field. At high temperature, the PL peaks split into two peaks, the amount of which was proportional to the amount of flux threading the tube.

A magnetic field applied to the tube axis removes the valley degeneracy by lifting the time-reversal symmetry producing two equally-bright excitonic states at high magnetic fields [5]. This degeneracy lifting mixes different parity excitonic wave functions providing excitons trapped in the dark state with a radiative recombination pathway, thus brightening the transition. At low temperatures, this is manifest as an increase in PL intensity due to the exciton population being trapped in the lower, dark state, as well as a red shift proportional to the applied field. At high temperature, there is a finite population in the upper, bright state before the field is applied resulting in a two split peaks once the symmetry is broken by a magnetic field. This work clearly demonstrates the existence of dark excitons in SWNTs.