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Magnetic Brightening of Dark Excitons in Carbon Nanotubes

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To gain insight into the internal energy structure and radiative properties of excitons in single-walled carbon nanotubes (SWNTs), we have studied photoluminescence (PL) from individualized HiPco and CoMoCAT samples as a function of magnetic field (B) and temperature (T). The PL intensity increased, or "brightened," with B applied along the tube axis and the amount of brightening increased with decreasing T. These results are consistent with the existence of a dark state below the first bright state [1]. In the presence of time reversal symmetry, exchange-interaction-induced mixing between excitons in two equivalent valleys (the K and K' valleys) is expected to result in a set of exciton states, only one of which is optically active. This predicted bright state, however, is not the lowest in energy. Excitons would be trapped in the dark, lowest-energy state without a radiative recombination path. When a tube-threading B is applied, addition of an Aharonov-Bohm phase modifies the circumferential boundary conditions on the wave functions and lifts time reversal symmetry [2,3]. This symmetry breaking splits the K and K' valley transitions, lessening the intervalley mixing and causing the recovery of the unmixed direct K and K' excitons, which are both optically active. We have calculated PL spectra through B-dependent effective masses, populations of finite-k states, and acoustic phonon scattering, which quantitatively agree with the observations. These results demonstrate the existence of dark excitons, their influence on the PL quantum yield, and their elimination through symmetry manipulation by a B. This work was performed in collaboration with J. Shaver, S. Zaric, O. Portugall, V. Krstic, G. L. J. A. Rikken, X. Wei, S. A. Crooker, Y. Miyauchi, S. Maruyama, and V. Perebeinos and supported by the Robert A. Welch Foundation, the NSF, and EuroMagNET.

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