Geometry Dependence of Magnetic Brightening in Stretch -Aligned SWNT Films

Jonah Shaver (Rice University), Scott A. Crooker (NHMFL-LANL), Jeffery A. Fagan, Erik Hobbie (NIST), Junichiro Kono (Rice University)

Single-walled carbon nanotubes' (SWNTs) high aspect ratio, valley degeneracy, and hollow molecular structure make them an interesting system for the study of 1D excitons. A theoretically predicted rich exciton manifold includes dark excitonic states, recently observed through measurements of photoluminescence *magnetic brightening* at low temperatures [1], resulting from the interaction of electrons and holes in equivalent energy K-space valleys. In the presence of a magnetic field, the valley degeneracy is lifted, shifting the observed peak positions and lowering the strength of exchange interaction induced mixing between states, resulting in a redistributed oscillator strength in the exciton manifold and a brightening of the lowest lying singlet exciton [2].

We performed low-temperature magneto-photoluminescence (PL) of highly-aligned SWNT films in magnetic fields (***B***) up to 55 T. The magnetic field was generated using the 60 T long pulse magnet powered by a 1.4 GVA motor-generator at the National High Magnetic Field Laboratory in Los Alamos, NM. Polyacrylic acid films containing DNA suspended SWNTs with a narrow chirality distribution (CoMoCAT) were stretch-aligned to an extension ratio of ~4:1, resulting in an order parameter S ~ 0.9. This allowed for the utilization of two well-defined measurement geometries, SWNTs || ***B*** and SWNTs ⊥ ***B***. Photoluminescence, from (6,5) nanotubes, was resonantly excited with a dye laser tuned to 570 nm (or 2.175 eV). Near infrared spectra were dispersed on a monochromator and collected with an InGaAs diode array using a typical exposure plus readout time of 1-2 ms.

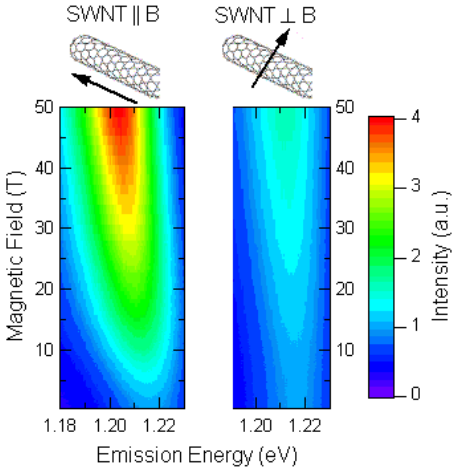

Figure 1: Magneto-PL of stretch-aligned SWNT film parallel and perpendicular to the applied magnetic field at low temperature (1.5 K).

Figure 1 shows a contour map of PL intensity as a function of emission energy and magnetic field at 1.5 K. In the SWNT || ***B*** geometry, the magnetic field increases the PL intensity by nearly 4x and the peak position red shifts significantly. In contrast, the SWNT ⊥ ***B*** geometry only shows a modest increase in PL intensity of less than 1.5x and the peak position is only slightly red shifted. This results from the redistribution of oscillator strength between the low energy dark state and higher energy bright state of the singlet exciton manifold. Excitons confined to dark state at zero field thus have their PL magnetically brightened at high field.

Plotting the peak position as a function of magnetic field in Fig. 2 shows a shift of ~14 meV for the parallel and ~4 meV for the perpendicular geometry at 55 T. An average angle of ~15° can be determined from the order parameter (S), this is used to find the component of the magnetic field projected on the long SWNT axis. When the peak shift is plotted vs. the parallel ***B*** component, the curves collapse on to a single line. This verifies that the PL from excitons in SWNTs is only sensitive to a parallel magnetic field.

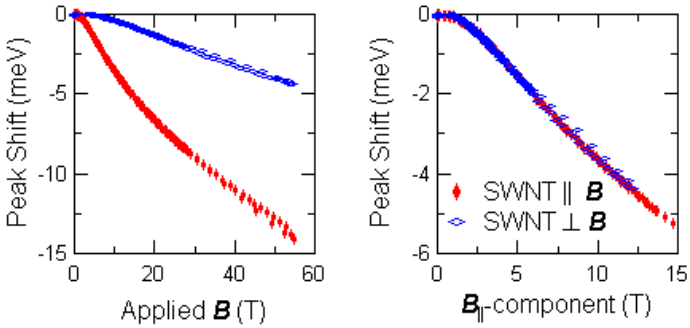

Figure 2: Peak shift as a function of magnetic field plotted vs. applied magnetic field and the parallel magnetic field component. In the latter case, the traces collapse on to the same line.

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References

[1] I. B. Mortimer and R. J. Nicholas, Phys. Rev. Lett. **98**, 027404 (2007), J. Shaver *et al.*, Nano Lett **7**, 1851 (2007)
[2] J. Shaver and J. Kono, Laser and Photonics Reviews **1**, 260 (2007)