Megagauss Absorption Spectroscopy of Single-walled Carbon Nanotubes

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Single-walled carbon nanotubes are 1D nanostructures consisting of rolled sheets of graphene. The electronic band structure of these molecules is completely determined by the precise way that their constituent sheet of graphene is rolled, that is the n and m indices from reciprocal lattice vectors along the zig-zag and armchair edges of the sheet. Metallic SWNTs (satisfying \( (n-m) \% 3 = 0 \)) have first absorption bands in the visible, while semiconducting (satisfying \( (n-m) \% 3 = 1 \) or \( 2 \)) have first absorption bands in the NIR and second in the visible. Individualization of SWNTs in aqueous surfactant solutions or polymer films allows for clear observation of these spectroscopic features [1].

When these tubes are threaded by an amount of magnetic flux the periodic boundary conditions on their circumferential electronic wavefunctions are modified [2]. This addition of an Aharonov-Bohm phase periodically modulates the band structure, opening a gap in metallic tubes and shrinking the gap of semiconducting tubes resulting in spectroscopically detectable changes of peak shifts and splitting. In addition to the spectroscopic changes, a magnetic field will align these rod-like molecules. Metallic tubes are paramagnetic along their axis while semiconducting tubes are diamagnetic with a larger perpendicular magnitude of the susceptibility. These factors result in the alignment of all SWNTs to an applied magnetic field. In solution, many factors affect the alignment of SWNTs; temperature of the solution, viscosity of the solution, length distribution of the SWNTs and magnetic susceptibility anisotropy.

The Single Turn Coil Project (STP) 300 T magnet was used for these experiments. The STP magnet utilizes a fast 250 kJ capacitor bank with 24 cells. Hold off charges of ~40-50 kV are used to build up charge, which is dissipated in a single turn, 10 mm diameter copper coil. The field pulse duration is on the order of ~2-5 \( \mu \)s, with a maximum of up to 300 T depending on hold off voltage, coil diameter, and number of capacitor cells used. A typical experiment for this series was 166 T in a 10 mm coil at 35 kV.

Two experiments were attempted with this magnet system. Experiment 1 was an attempt at high field micro-second absorption spectroscopy in a fiber coupled, transmission sample holder. A Cordin streak camera, coupled to the output of a 300 mm Acton monochromator with a 150 or 300 g/mm grating blazed in the visible was used to measure the magnetic field dependent absorption spectra of SWNTs. Use of the streak camera with a 500 ns / mm streak rate on its 18 mm intensified ccd allowed for continuous transmission data collection for the duration of the magnetic field pulse. Both Xe and QTH lamps were used as excitation sources. A high field, sample in, transmission streak was to be taken and normalized zero field, sample out, transmission streak to calculate absorption spectra. Image analysis software (Igor Pro) was used to bin the data from the streak camera experiment into ~100-200 ns slices for a magnetic field resolution of ~ 10 T. Unfortunately, technical issues with triggering the camera during a high field shot prevented completion of the experiment. The STP magnet generates large amounts of high frequency interference to which the electron beam-based streak camera is highly sensitive to. Zero field absorption spectra with ~100 ns resolution was successfully gathered, proving the feasibility of the experiment once the interference issues are addressed.

Experiment 2 was field dependent single frequency detection of nanotube molecular motion in solution. Using a similar sample holder, a laser (\( \lambda_{\text{ex}} = 532 \text{ nm or } 635 \text{ nm} \)), polarizer, and a fast rise time single channel Si detector time dependent transmission spectra were taken continuously during the magnetic field pulse in the Voigt geometry (light propagation \( \perp \mathbf{B} \)). As the magnetic field is applied, the transmission either increased (polarizer // \( \mathbf{B} \)), or decreased (polarizer \( \perp \mathbf{B} \)). Being 1D nanostructures, SWNTs absorb light polarized parallel to their axis strongly, thus when the polarizer is parallel (perpendicular) to the field, and they align, the light transmission is decreased (increased). Experiments were also preformed in the Faraday geometry (light propagation // \( \mathbf{B} \)), which allowed for omission of the polarizer with results similar to the Voigt (\( \perp \)) experiment. This experiment successfully reproduced results of Shaver et al. [3] performed in with the Berlin Megagauss generator magnet and confirms SWNT motion in solution is on the \( \mu \)s time scale.