RICE UNIVERSITY

Ultrafast Spectroscopy of (6,5) Carbon Nanotubes

by

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Contents

Illustrations

Tables

Chapter 1

Optical Properties of Single-Wall Carbon Nanotubes

Carbon nanotubes are:

- Allotrope of graphene
- 1-D structures meaning that electrons are confined in a single dimension
- exist in terms of many chiralities, some are semiconducting and others are metallic
- interesting because they let us explore the physics of 1-D structures.
- Due to the higher degree of confinement with respect to convention 3-D structures, different phenomena may occur.

1.1 Optical Selection Rules

- Selection rules dictate which transitions can occur in the presence of certain conditions
- light polarized parallel to carbon nanotube excites one type of transition
- light polarized perpendicular to carbon nanotube excites another set of transitions

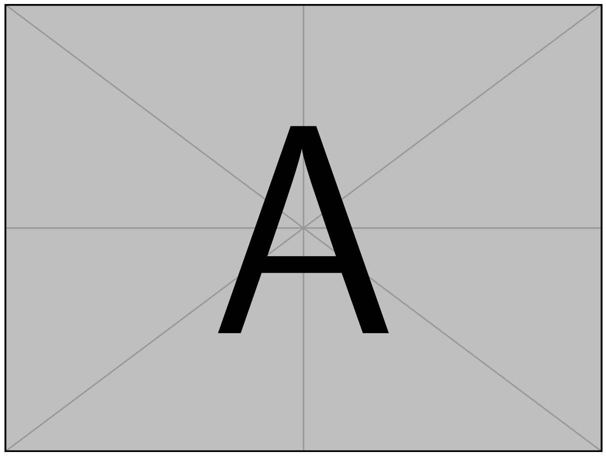

Figure 1.1 : Figure of carbon nanotube bandstructure. Arrows drawn in figure to show allowed transitions.

1.2 Excitons in Carbon Nanotubes

- all optical excitations in carbon nanotubes lead to direct creation of excitons

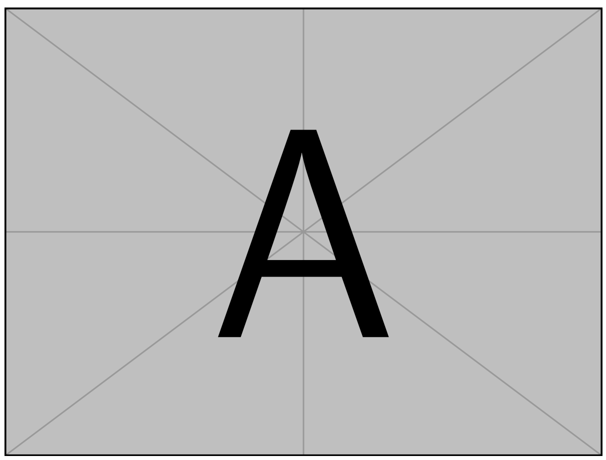

Figure 1.2 : Figure of GaAs absorbance at low-T and high-T to show weak binding energy of excitons. Next to this is a figure of (6,5) absorbance at room temperature

Chapter 2

Experimental Procedures

2.1 (6,5) Carbon Nanotube Sample Properties

Start with ComoCat solution containing many different chiralities. Do gel chromatography to create (6,5) enriched sample. Using DOC as surfactant to prevent nanotubes from clumping together to form bundles.. About 4 percent of solution is DOC. Sample is contained in a quartz cuvette with an optical path length of 1 mm.

Here, absorbance is defined as

$$A = \log_{10}(-T), \tag{2.1}$$

where T represents the transmission through the sample. This is absorbance spectrum of sample. We can see E11, phonon sideband, and E22 within this spectral window. Other peaks in the sample emerge from exciton resonances of (9,1) and (6,4) nanotubes.

2.2 Optical Pump-Probe Spectroscopy

2.2.1 Experimental Apparatus

Laser is Clark MXR-2010, 1 KHz repetition rate, 150 fs pulse duration, and central wavelength of 775 nm.

All of the power is delivered to the OPA. OPA generates signal and idler beams. BBO crystal added to generate SHG of signal which is used as the resonant E22 pump

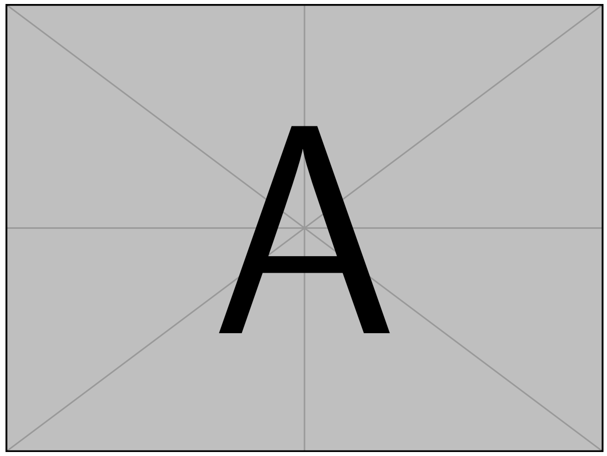

Figure 2.1 : Absorbance spectrum of (6,5) sample.

excitation. Signal of OPA is used to generate white light continuum in a sapphire crystal. Wavelength separator used to seperate SHG and fundamental signal. A motorized stage in the path of optical pump beam is used to alter the optical path length of pump beam.

Pump and probe beams are focused onto surface of the sample. This is a non-collinear geometry, meaning that pump and probe do not propagate toward the sample in a parallel trajectory. The probe is collected into an optical fibre which sends this beam to a spectrometer equipped with a charge-coupled device (CCD) camera. Camera allows to measure the intensity of several wavelengths of probe beam simultaneously.

Setup is equipped with an optical shutter placed in the optical path traveled by the pump beam. At each time delay, measure transmission of the probe beam with the pump beam blocked and with the pump beam unblocked by the shutter.

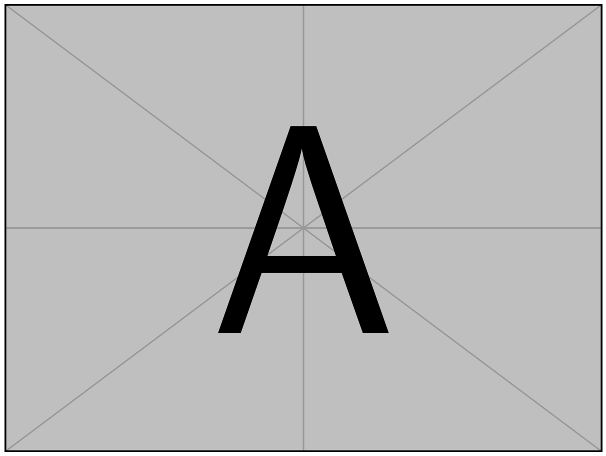

Figure 2.2 : Schematic Diagram of the Experimental Apparatus

2.2.2 Pump and Probe Spot Sizes

Pump and probe spot sizes measured using a knife edge scan. For this, mounted a razor blade on a delay stage placed in the sample position as shown in Figure 2.4. Measure transmitted power as a function of delay stage positioning. Average power drops as razor blade cuts into beam diameter.

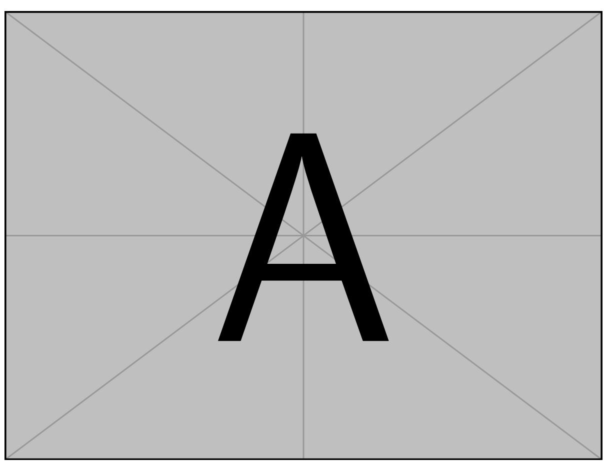

Figure 2.3 : Spot Size Measurement Setup

Assuming that the beam diameter can be approximated as a gaussian distribution, we can compute the spot size by fitting this data with the equation

$$P = P_0 + P_{max}/2 \left(1 - \mathrm{erf}\left(\frac{\sqrt{2}(x - x_0)}{w}\right)\right), \tag{2.2}$$

which this represents the cumulative distribution of a Gaussian measured and used to obtain the beam diameter w. Here P_0 represents the baseline of the power meter observed when the beam is fully blocked, erf is the standard error function, P_{max} is the maximum power of the beam, x is coordinate describing lateral position of the razor blade and x_0 represents position of razor blade when the razor blade blocks 50% of the total power.

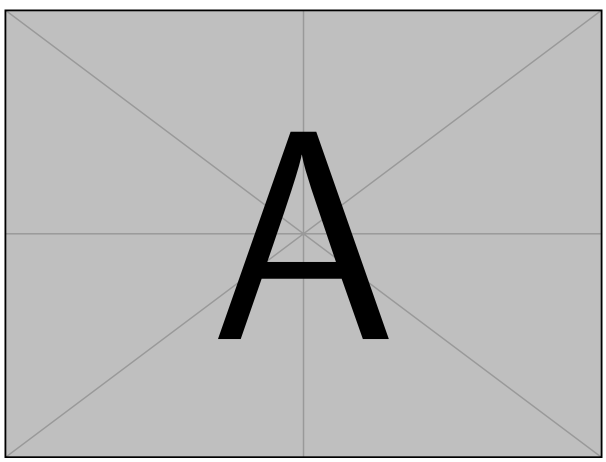

Figure 2.4 : Example measurement of beam diameter for pump and probe. Solid lines are fits to the data using function defined in equation

Chapter 3

Ultrafast Carrier Dynamics of (6,5) After Resonant E_{22} Pumping

3.1 Experimental Results

3.2 Analysis and Discussion

Figure 3.1 : Saturation of the change in the E_{11} region‘