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Terahertz Photoconductivity of Graphene Nanostructures

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The photoconductive properties of graphene nanoribbons and carbon nanotubes were studied using optical pump-THz probe spectroscopy. A reduction in conductivity of GNRs compared to CNTs was observed.

I. Introduction And Background

Recently a "bottom-up" approach was developed for producing long graphene nanoribbons (GNRs) with well-defined edge structures and widths down to ~1 nm, possessing bandgaps corresponding to visible wavelengths [1]. The structure of these GNRs is shown in **Figure 1**. Carbon nanotubes (CNTs), are structurally very similar to GNRs, only 'rolled up' in the lateral dimension. Here, we studied semiconducting (6,5) type CNTs as shown in **Figure 2**. We present a comparative study of the transient photoconductivity of structurally defined GNRs and semiconducting CNTs, employing optical pump – THz probe spectroscopy to investigate the photogenerated species on sub ps timescales. Here charge carriers are optically excited in the sample material by an ultrashort laser pulse, and probed by a freely propagating, single cycle electromagnetic pulse consisting of frequencies in the 0.3-1.6 THz range. A schematic of the experimental setup is shown in **Figure 3**. The setup is driven by an amplified pulsed laser system which delivers pulses of 800 nm central wavelength and sub - 100 fs duration at a repetition rate of 1 kHz. A portion of these pulses (the *pump* beam in **Figure 3**) is used to photo-excite the sample directly; alternatively, the excitation wavelength can be altered first either by optical doubling crystals or by an optical parametric amplifier (OPA). Another portion of the laser beam (the *generation* beam) generates the THz probe in a ZnTe crystal by a process known as optical rectification [3]. The THz probe is focused on the sample, and then refocused on a second ZnTe crystal where it is combined with a third beam, the *sampling* beam. In this second ZnTe crystal the time-dependent electric field of the THz probe pulse is directly measured using free-space electrooptic sampling (FEOS) – an inverse optical rectification effect [3]. The electric field in the THz pulse is proportional to the ellipticity of the sampling laser beam, which is analyzed optically via a variable wave plate, a Wollaston prism (or polarizing beam splitter), and a pair of photodiodes. The differential voltage on the photodiodes, proportional to the THz field strength, is recorded with lock-in detection as function of the delay between the THz pulse and the sampling laser pulse.

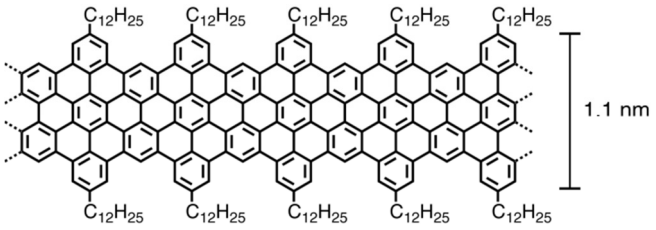

Figure 1 Molecular structure of graphene nanoribbon (GNR) of length 600 nm and effective bandgap 1.88 eV.

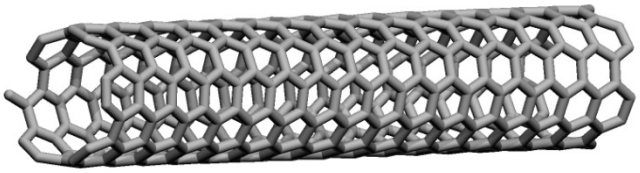

Figure 2 Molecular structure of semiconducting (6,5) carbon nanotube [2].

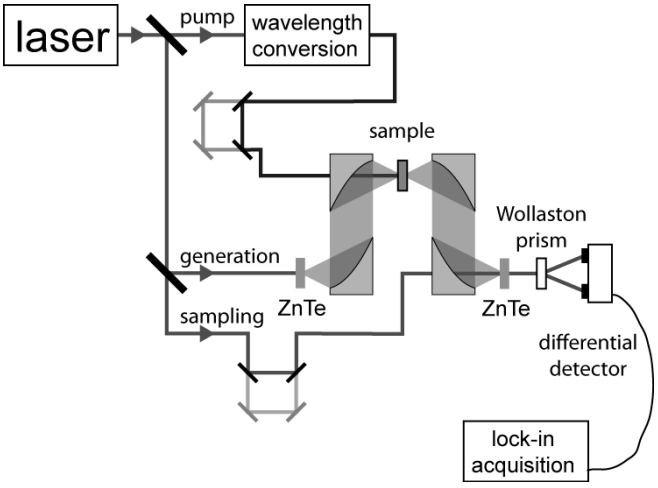

Figure 3 Sketch of optical pump – THz probe experimental setup.

II. Results

Figure 4 shows the frequency-resolved real part of the photoconductivity of the GNRs, measured 300 fs after photoexcitation at 400 nm wavelength. A positive real conductivity is observed, increasing in magnitude with probe

frequency. The real part of the photoconductivity of CNTs, measured 300 fs after photoexcitation at 800 nm wavelength, is also shown in **Figure 4**. It peaks at around 1 THz. The conductivity of CNTs per absorbed photon is more than an order of magnitude higher than for GNRs. This is consistent with theoretical calculations [4] that have predicted significantly larger carrier mobilities for the CNTs than the GNRs for the materials with the bandgaps presented here.

The different possible causes for the different conductivities and the comparison to theory [4] are at the focus of our current work, and will be discussed in more detail in our contribution.

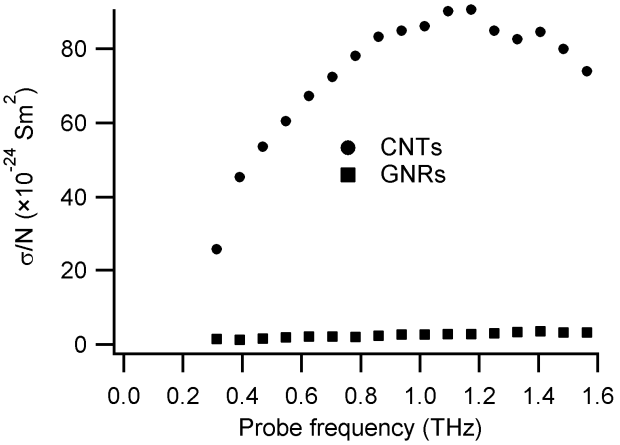

Figure 4 Real conductivity normalized by absorbed photon density for carbon nanotubes (CNTs) and graphene nanoribbons (GNRs).

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