Optoelectronics and Terahertz Electronic Transport
in Individual Single-Walled Carbon Nanotube Devices

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Abstract:
The excellent electronic and optical properties of single-walled carbon nanotubes (SWNTs) strongly motivate the use of these quasi-one-dimensional (1D) materials in optoelectronic [1] and high frequency electronic [2] applications. By studying individual SWNT devices, we aim to uncover novel physical phenomena and establish a foundation for future applications in carbon nanoelectronics. Here, we review our recent work that investigates SWNTs as the active element in photovoltaic devices and ultra-fast nanoelectronics in the Terahertz frequency regime. We demonstrate that nanotube gated p-n junctions behave as sensitive nanoscale photodetectors and show intrinsic response to ultrafast optical excitation pulses. Due to the gate-controlled n-type (electron) and p-type (hole) regions of the nanotube, these devices provide an ideal system for probing carrier dynamics and interactions of electrons and holes in carbon nanotubes. Also, we describe the first terahertz electrical measurements of SWNT transistors performed in the time domain [3]. We observe a ballistic electron resonance that corresponds to the roundtrip transit of an electron along the nanotube with a picosecond-scale period. The electron velocity is found to be constant and equal to the Fermi velocity, showing that the high-frequency electron response is dominated by single-particle excitations rather than collective plasmon modes. By studying SWNTs in gated p-n junction and FET geometries, we demonstrate the practical capabilities of nanotube devices and reveal new physics in these 1D materials.

Summary of Research:
Devices are fabricated using standard nanolithography techniques. Briefly, molybdenum electrodes, deposited on a highly doped silicon substrate with a layer of thermal oxide, form the split gates above which an additional 100 nm SiO₂ layer is deposited. Iron nano-particle catalyst sites are defined and NTs (with diameters of 1-3 nm) are grown by chemical vapor deposition. Finally, metallic electrodes (5 nm Cr, 50 nm Au) form the contacts for the NT. After fabrication, the devices are wire bonded and placed in an optical cryostat. Electronic characteristics are monitored while the device is illuminated under focused laser illumination at temperatures down to 4°K.

Photocurrent is measured in the p-n junction device under illumination by ultrafast (≈180 fs) laser pulses. The photocurrent vs. optical power density is $\lambda = 810$ nm and $V_{sd} = 0.0V$. The photocurrent is linear below ~ 15 pA, but becomes nonlinear above this value. Using a two-pulse time delay setup, we probe the real-time response of the nanotube p-n junction to ultrafast pulses. At low incident optical power, we observe a very sharp (half width ~ 350 fs) inverse peak centered at zero delay. At higher optical power, we observe a broader peak (half width ~ 860 fs) that persists for increasing optical power. The observed inverse peaks can be fit by exponential functions and decay times can be extracted.
At low powers, the inverse peak shows a single decay constant of 350 fs, while at higher powers, we extract both a fast (350 fs) and slow (860 fs) decay time.

From the photocurrent saturation, we expect that the two time-correlated pulses will add in a nonlinear fashion, leading to the observed inverse peaks. We attribute the fast decay component to the instrument detection limit, while the slow decay is attributed to the nanotube p-n junction response. The time resolution of the measurement is limited by the convolution of two 180 fs pulses, resulting in an inverse peak at all powers with a decay time of ~ 350 fs. At higher powers, however, the slow decay (860 fs) results from the intrinsic nanotube p-n junction response. We speculate that this response time corresponds to the charge carrier time-of-flight in the high electric field generated in the p-n junction, but the detailed nature of this response is unclear, and will be the focus of future investigations.

We fabricate the transistor with both a metal topgate and global silicon backgate on a silicon-on-sapphire (SOS) substrate. The terahertz source composed of a microwave transmission line excited by a short-pulse laser is fabricated directly on the silicon layer. The ground line of the transmission line is connected to the drain electrode of the nanotube transistor.

The terahertz signal is generated by biasing the transmission line with a DC voltage, $V_{\text{Line}}$, and photo-exciting the transmission line gap with a femtosecond Ti:Sapphire laser. Each ultra-short laser pulse produces a unipolar voltage pulse with pulse width of 1-1.5 ps, corresponding to a broadband terahertz signal. The fast voltage pulse is transmitted along the transmission line to the nanotube transistor, applying a voltage pulse across the nanotube device. We then record the DC current response to the terahertz pulses, $I_{\text{THz}}$. To access the time domain, each laser pulse is split into two before illuminating the transmission line gap, resulting in two picosecond voltage pulses with controllable delay time $t_d$. Measuring $I_{\text{THz}}$ as a function of the delay time yields information on the device response at picosecond time scales.

We can use the fast response time of nanotube device to probe ballistic electron transport through the nanotube in the time domain. The nanotubes are gated in a p-n-p configuration, by electrostatic n-doping the section underneath the topgate while maintaining the rest of the nanotube p-doped by the backgate. For all three devices, the $I_{\text{THz}}$ vs. delay time $t_d$ show an inverse peak at $t_d = 0$ and at least one set of satellite peaks symmetrically located around the central peak.

We interpret these peaks as a ballistic electron resonance in the SWNT cavity bounded by p-n junctions. The electrons excited by the first terahertz pulse traverse the first p-n junction, propagate along the length of the nanotube, and are reflected at the second p-n junction. At $t = t_r$, the returning electrons meet the incoming electrons excited by the time-delayed pulse. Larger nonlinear current is generated in the same manner as at $t = 0$, resulting in satellite peaks. The observed period $t_r$ therefore corresponds to the roundtrip transit time of an electron within the nanotube cavity. Our result represents the first direct time domain measurement of electron transit in a carbon nanotube.

To determine the velocity of the excitation, we plot $t_r$ for three devices vs. their topgate lengths. The linear fit of $t_r$ vs. $L_g$ yields an electron velocity $v = 2L_g/t_r \approx 0.8 \pm 0.1 \times 10^6$ m/s. This is in good agreement with the Fermi velocity of electrons in metallic carbon nanotubes, $0.8 \times 10^6$ m/s, but significantly less than that expected for the plasmon mode ($v_{p}\approx 2.7 \times 10^6$ m/s for $g = 0.3$). This demonstrates that single-particle excitations, not the plasmon mode, dominate the high frequency response in our measurement.

References: