Time-resolved picosecond photocurrents in contacted carbon nanotubes

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We introduce coplanar stripline circuits to resolve the ultrafast photocurrent dynamics of freely suspended carbon nanotubes (CNTs) in the time-domain. By applying an on-chip pump-probe laser spectroscopy we demonstrate that CNTs, contacted by metal electrodes, exhibit a picosecond photocurrent response. We find a combination of an optically induced ultrafast displacement current, transport of photo-generated charge-carriers at the Fermi velocity to the electrodes, and interband charge-carrier recombination processes to dominate the ultrafast photocurrent of the CNTs [1].

CNTs are promising building blocks for future optoelectronic devices because of their compelling electronic and optical properties. So far, only optical methods have been used to characterize the ultrafast dynamics of photo-generated charge-carriers in CNTs in the time domain. Typical fluorescence decay times of individual CNTs have been measured to be in the range of ten to hundreds of picoseconds. Conventional electronic measurements cannot resolve such ultrafast dynamics because available electronic equipment cannot produce trigger signals and detect transients faster than tens of picoseconds. We introduce an experimental on-chip scheme to measure the photocurrent dynamics of electrically contacted nanosystems in the time domain [1]. The technique applies an ultrafast optical pump-probe scheme to coplanar stripline circuits, and Auston switches sample the photocurrent response of CNTs. The studied CNTs are grown via an electric-field-assisted CVD method, such that a network of CNTs spans two electrodes of a stripline (Fig. 1a). The freely suspended CNTs and the two electrodes form a two-terminal stripline circuit driven by a bias voltage $V_{SD}$ (Fig. 1b). The CNTs are optically excited by a pump-pulse at the second interband transition $E_{22}$ of the semiconducting CNTs in the network with ~160 fs pulse length generated by a Ti:Sa laser. After the excitation, an electromagnetic pulse starts to travel along the stripline. A sampling circuit senses the transient electric field of the travelling pulse at the position of a field probe (Fig. 1b). Here, we utilize an Auston switch based on a low-temperature grown GaAs substrate. The time delay $\tau_{\text{Delay}}$ between the pump and the probe pulse is controlled by a delay stage. Measuring the current $I_{\text{Sampling}}$ in the sampling circuit as a function of $\tau_{\text{Delay}}$ yields information on the optoelectronic response of the CNTs in the stripline circuit with a picosecond time-resolution. Directly after optical excitation, the photo-generated charge-carriers redistribute in order to decrease the local potential differences in the CNT network. This displacement of the

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charge carriers decreases the electric field $E$ in the irradiated region. The current density in the CNTs can be described by a transient displacement current density (1) $j_D = \varepsilon \varepsilon_0 \frac{dE}{dt}$, with $\varepsilon$ and $\varepsilon_0$ the relative and vacuum permittivity. Fig. 2a shows the time-resolved $I_{\text{Sampling}}$ as a function of $t_{\text{Delay}}$ for $|V_{\text{SD}}| \leq 3 \text{ V}$. For $t_{\text{Delay}} \leq 7 \text{ ps}$, $I_{\text{Sampling}}$ equals nearly zero, because during this time-delay, the electro-magnetic pulse generated at the CNTs has not reached the field probe. At $t_{\text{Delay}} \approx 10.6 \text{ ps}$, $I_{\text{Sampling}}$ exhibits a first peak with a FWHM of $\Delta t_{\text{1stPeak}} \approx (2.1 \pm 0.1) \text{ ps}$. We interpret the first peak of $I_{\text{Sampling}}$ to result from an optically induced displacement current, in accordance with equation (1). As depicted in Fig. 2a, we find that $I_{\text{Sampling}}$ also exhibits a second peak (triangle). It is delayed by $t_{\text{2ndPeak}} = (4.8 \pm 0.2) \text{ ps}$ with respect to the first peak at $\approx 10.6 \text{ ps}$. Such a second peak results from the transport of photo-generated charge-carriers to the electrodes, as we recently demonstrated for GaAs in a similar stripline configuration [2]. Here, the laser spot is located in the center between the two electrodes. Therefore, the photo-generated charge-carriers in the CNTs need to propagate $d/2=(4.5 \pm 0.5) \mu\text{m}$, before they reach the metal contacts. Assuming an average group velocity of the photo-generated charge-carriers in the CNT network, we can estimate its value to be $v_{\text{Group}} = d/(2 \cdot t_{\text{2ndPeak}}) \approx (0.9 \pm 0.1) \times 10^6 \text{ m/s}$. This value is consistent with the Fermi velocity in CNTs of $\approx 0.8 \times 10^6 \text{ m/s}$, and it is significantly less than the propagation velocity $v_{\text{Plasmon}}=2.7 \times 10^6 \text{ m/s}$ of plasmon modes in CNTs. Hereby, the measurements suggest that single electron excitations and not plasmon modes dominate the ultrafast optoelectronic response of freely suspended CNTs. As can be seen in Fig. 2b, $I_{\text{Sampling}}$ also exhibits a slow decay component. We utilize the sum of two Lorentzian functions and an exponentially convoluted Gaussian to account for all features in $I_{\text{Sampling}}$ (Fig. 2c). A fitting parameter $\tau_{\text{Slow}}$ describes the exponential decay of the convoluted Gaussian, and it can be associated with the slow decay. We find a value of $\tau_{\text{Slow}} = (248 \pm 2) \text{ ps}$, independent of $V_{\text{SD}}$. We interpret $\tau_{\text{Slow}}$ to be the lifetime of photo-generated charge carriers in the CNT network. We stress that it is outstanding to measure carrier lifetimes in freely suspended, CVD grown CNTs, because such CNTs have typical interband transition energies $E_1$, below $1 \text{ eV}$. This energy range lacks a sufficiently fast photo-detector. The versatility of the presented optoelectronic technique recommends further time-domain experiments on electrically contacted nanoscale systems, such as graphene, quantum wires, and nanowires.


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