Photocurrent properties of freely suspended carbon nanotubes under uniaxial strain

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The photocurrent properties of freely suspended single-walled carbon nanotubes (CNTs) are investigated as a function of uniaxial strain. We observe that at low strain, the photocurrent signal of the CNTs increases for increasing strain, while for large strain, the signal decreases, respectively. We interpret the nonmonotonous behavior by a superposition of the influence of the uniaxial strain on the resistivity of the CNTs and the effects caused by Schottky contacts between the CNTs and the metal contacts. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3159472]

Carbon nanotubes (CNTs) have attracted considerable attention because of their compelling optoelectronic[1–7] and electromechanical properties.[8–25] For instance, laser-induced excitonic transitions,[3,4] bolometric effects,[6,7] and photodesorption of adsorbed molecules[2] can give rise to a photoconductance in CNTs. Furthermore, electric fields at the Schottky contacts between CNTs and metal contacts can separate optically excited electron-hole pairs, causing a photocurrent across electrically contacted CNTs.[5,6]

At the same time, the electromechanical properties of CNTs have been studied both by locally manipulating CNTs with the tip of an atomic force microscope[11–13] and by applying uniaxial[14–16] and torsional[17,18] strain to the CNTs. Theorists have modeled the electronic behavior of the mechanically deformed CNTs by an enhanced electronic scattering at defects,[12,19,20] a structural induced alteration of the CNTs' band gap,[14–17,19,21,22,25] and by a mechanical induced transition from sp^2 to sp^3 hybridization of the carbon bonds.[10,24]

Here, we report on the photocurrent properties of freely suspended CNTs as a function of statically applied uniaxial strain. We observe a rise of the photocurrent signal, which is generated at Schottky contacts between the CNTs and their bracing source and drain electrodes, of up to ~150% for uniaxial strain values in the range of 0.3 to 1.2%. We detect a decrease of the photocurrent signal for strain values exceeding this magnitude. We explain the nonmonotonous behavior by a superposition of the effect caused by Schottky contacts and the decrease of the conductance for increasing mechanical deformation.[9–12,22,24]

Figure 1(a) shows a schematic side view of the device for applying strain to the suspended CNTs. Two L-shaped fittings are bonded to both sides of a piezoelectric stack, which extends uniaxially if a voltage V_{PIEZO} is applied to it.[16] The fittings support a silicon substrate which features a center gap of about 70 to 80 μm width. The gap is prepared by optical lithography and KOH etching, such that one side of the gap is open, and that the gap can be expanded by the piezo [Fig. 1(b)]. The CNTs are grown via electric-field assisted chemical vapor deposition,[26] such that they are mounted on two 100-nm-thick Au pads (source and drain electrode) on top of an insulating SiO_2 layer. Scanning electron microscope (SEM) images demonstrate that the suspended CNTs are stretched, when V_{PIEZO} is applied to the piezoelectric stack [Figs. 1(c)–1(e)]. In particular, ripples at $V_{PIEZO}=0$ V [e.g., triangles in Fig. 1(c)] are straightened at $V_{PIEZO}>0$ V and eventually the CNTs are uniaxially strained [see dashed line and black triangle in [Figs. 1(c)–1(e)].[16] From the SEM images, we can estimate that the particular sample is stretched by about 1.4 % at $V_{PIEZO}=35$ V.

Then, photocurrent images of the suspended CNTs are acquired by recording the change of the source-drain current ΔI^{DC} across the CNTs at a finite source-drain bias V_{SD}, when the CNTs are illuminated.[5] To this end, the light of a mode-locked titanium:sapphire laser is focused through the objective of a microscope onto the surface of the sample. The typical laser spot diameter is 2 μm, and the typical power density is ~0.5 kW/cm^2. Scanning the laser spot across the

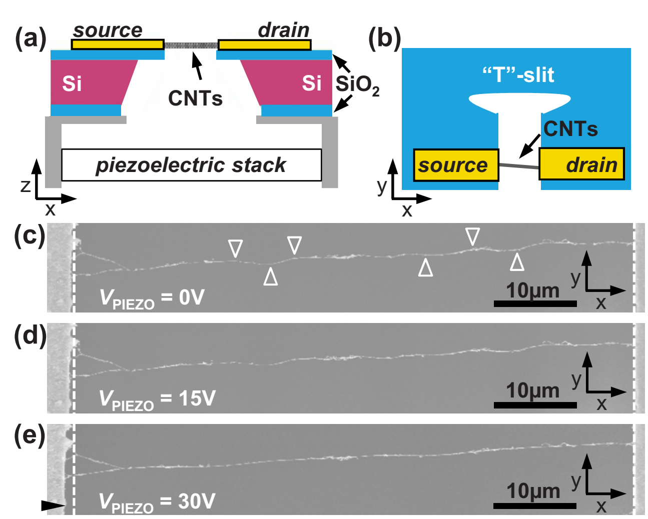

FIG. 1. (Color online) Schematic side (a) and top (b) view of a Si/SiO_2 sample with a "T"-slit and freely suspended single-walled CNTs mounted on a piezoelectric stack. Applying a voltage V_{PIEZO} to the piezoelectric stack allows applying uniaxial strain to the CNTs. [(c)–(e)] SEM images of CNTs at $V_{PIEZO}=0$, 15, and 30 V at RT.

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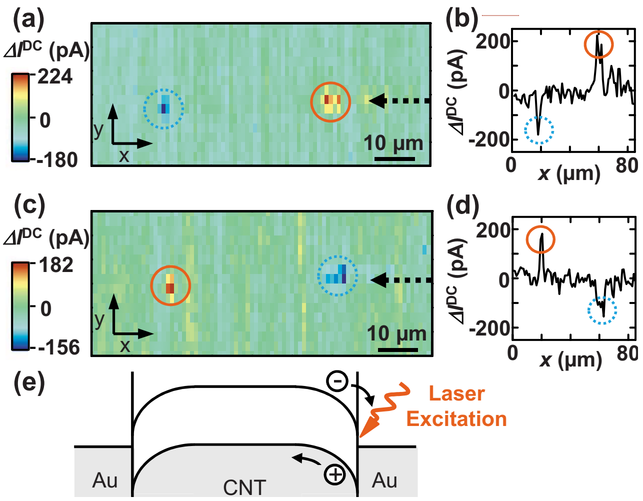

FIG. 2. (Color online) (a) Photocurrent image of the CNTs (λ_{LASER} =830 nm, V_{SD}=−50 mV, RT): a minimum (dashed circle) and maximum (circle) occur close to the left and right electrode, respectively. (b) Single trace along dashed line in (a). (c) Photocurrent image for the reversed bias as in (a) (λ_{LASER}=800 nm, V_{SD}=+50 mV, RT). (d) Single trace along dashed line in (c). (e) Schematic of Schottky barriers between CNTs and the electrodes.

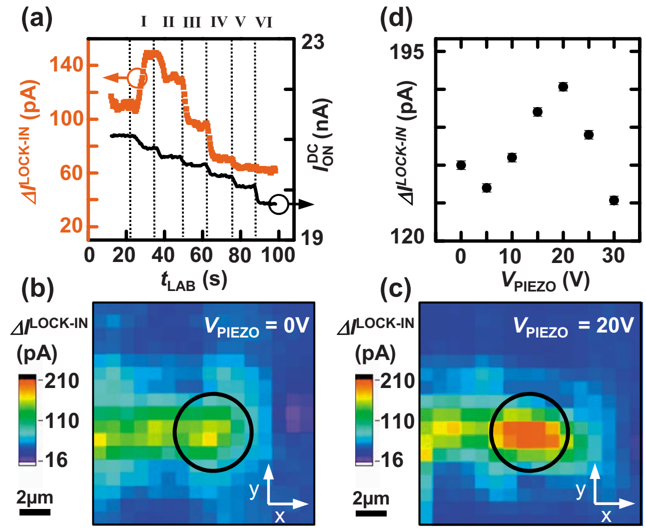

FIG. 3. (Color online) (a) I_{ON}^{DC} and $\Delta I^{LOCK\text{-}IN}$, simultaneously measured, across CNTs as a function of laboratory time, when V_{PIEZO} is increased from 0 to 30 V in six steps of 5 V (λ_{LASER}=714 nm, V_{SD}=+50 mV, f_{CHOP} =912 Hz, RT). [(b) and (c)] Photocurrent image of $\Delta I^{LOCK\text{-}IN}$ of a bundle of CNTs close to a metal contact (λ_{LASER}=714 nm, V_{SD}=+50 mV, f_{CHOP} =912 Hz, RT). (d) Maximum values of $\Delta I^{LOCK\text{-}IN}$ taken from encircled area in (b) and (c) and further photocurrent scans as a function of V_{PIEZO}.

CNTs [Fig. 1(b)], the local change $\Delta I^{DC}(\hat{x},\hat{y})=I_{ON}^{DC}(\hat{x},\hat{y}) - I_{BACKGROUND}^{DC}$ is detected for the laser being "ON." $I_{BACKGROUND}^{DC}$ describes the direct current value, when the samples are illuminated far away from the CNTs. In Fig. 2(a), $\Delta I^{DC}(\hat{x},\hat{y})$ is plotted using a linear false color scale as a function of the coordinates $\hat{x}$ and $\hat{y}$. We observe two oppositely signed resonances in ΔI^{DC} close to the contacts; as can be also inferred from the maximum (circle) and minimum signal (dashed circle) in the corresponding line cut along the $\hat{x}$ coordinate [Fig. 2(b)]. The resonances change their sign, when the source and the drain contacts are switched; i.e., the current amplifier for detecting ΔI^{DC} is connected first to drain [Figs. 2(a) and 2(b)] and then to source [Figs. 2(c) and 2(d)]. Hereby, we confirm recent reports that a Schottky contact between CNTs and metal pads can give rise to a photocurrent.[1,5] In this process, the electron-hole pairs locally created by photoexcitation are separated due to the local built-in electric field at the Schottky contact [Fig. 2(e)], and a maximum (minimum) photocurrent signal can be detected when the region at the reverse-biased (forward-biased) Schottky contact is illuminated.[5]

Recent reports show that the resistivity of CNTs increases for increasing mechanical deformation.[9–12,22,24] In turn, one expects that the photocurrent I_{ON}^{DC}, which is generated at the Schottky contacts [as depicted in Figs. 2(a) and 2(c)], decreases due to the increased global resistivity of strained CNTs. Figure 3(a) depicts data of such a strain-induced change of photocurrent as a function of the laboratory time. Here, the CNTs are optically excited at a position of maximum photocurrent [such as at the circle in Fig. 2(a)] and V_{PIEZO} is increased from 0 to 30 V in six steps of 5 V. We detect that I_{ON}^{DC} decreases monotonously for increasing voltage steps. The measurement is reversible in a way, that I_{ON}^{DC} reaches similar values for each step VI to I, when V_{PIEZO} is decreased from 30 to 0 V (data not shown). Hereby, we interpret the decrease of I_{ON}^{DC} for increasing V_{PIEZO} to reflect an increase of the CNTs' global resistivity ρ_{CNT} due to mechanical deformation.[9–12,22,24]

To increase the experimental sensitivity, we chop the exciting laser field at a frequency f_{CHOP} and additionally amplify the resulting current $\Delta I^{LOCK\text{-}IN} = I_{ON}^{LOCK\text{-}IN}(\hat{x},\hat{y},f_{CHOP},V_{PIEZO}) - I_{OFF}^{LOCK\text{-}IN}(f_{CHOP},V_{PIEZO})$ across the sample for the laser being "ON" or "OFF," respectively, by a current-voltage converter in combination with a lock-in amplifier at the reference frequency f_{CHOP}. This technique allows us to distinguish a small optically induced change of $\Delta I^{LOCK\text{-}IN}$ as a function of V_{PIEZO} [approximately a few tens of pA per step in Fig. 3(a)] from the larger change of I_{ON}^{DC}, which is induced by the global change of the CNTs' resistivity as a function of V_{PIEZO} [~260 pA per step in Fig. 3(a)]. We find that $\Delta I^{LOCK\text{-}IN}$ first increases (step I) and then decreases (steps II to VI). Again, the measurement of $\Delta I^{LOCK\text{-}IN}$ is reversible, when V_{PIEZO} is decreased from 30 to 0 V (data not shown). For large strain values, we can assume that the increased resistivity equally dominates the amplitudes $I_{ON}^{LOCK\text{-}IN}$ and $I_{OFF}^{LOCK\text{-}IN}$. In turn, we can explain the decrease and the saturation of $\Delta I^{LOCK\text{-}IN} = I_{ON}^{LOCK\text{-}IN} - I_{OFF}^{LOCK\text{-}IN}$ for large strain values phenomenologically [steps IV to VI of Fig. 3(a)]. From SEM images such as in Figs. 1(c)–1(e), we can estimate the strain value for the maximum photocurrent to be ~0.3% at V_{PIEZO}=5 V [step I in Fig. 3(a)]. Most strikingly, the relative change of $\Delta I^{LOCK\text{-}IN}$ as a function of V_{PIEZO} is significantly larger than the one of I_{ON}^{DC}, i.e., $\Delta I^{LOCK\text{-}IN}(V_{PIEZO}=5\text{ V})/\Delta I^{LOCK\text{-}IN}(V_{PIEZO}=0\text{ V}) \approx 148\%$, $\Delta I^{LOCK\text{-}IN}(V_{PIEZO}=30\text{ V})/\Delta I^{LOCK\text{-}IN}(V_{PIEZO}=0\text{ V}) \approx 55\%$, and $I_{ON}^{DC}(V_{PIEZO}=30\text{ V})/I_{ON}^{DC}(V_{PIEZO}=\text{ V}) \approx 94\%$. This observation makes it plausible that the origin of the photocurrent, i.e., the built-in electric fields close to a Schottky contact, is altered when the samples are stretched.

To substantiate the last interpretation we detect photocurrent images of $\Delta I^{LOCK\text{-}IN} = \Delta I^{LOCK\text{-}IN}(\hat{x},\hat{y})$ of a bundle of CNTs close to the location of maximum photocurrent at different V_{PIEZO}. For the particular bundle of CNTs characterized in Figs. 3(b) and 3(c), the region of maximum photocurrent at V_{PIEZO}=0 V is extended toward the "left" [Fig. 3(b)]. The observation of an extended "lobe structure" is consistent with the interpretation, that in a CNT bundle the built-in electric fields due to a Schottky contact are extended to a large region because of intertube transitions with different morphology and varying space charge configurations.[5] Most significantly, at $V_{PIEZO}=20$ V corresponding to an estimated strain value $\sim 1.2\%$, $\Delta I^{LOCK\text{-}IN}$ exhibits locally a new maximum [circle in Fig. 3(c)], which is larger than the photocurrent signal of the general lobe structure. We interpret the new maximum at $V_{PIEZO}>0$ V to reflect large electric fields at the location of maximum strain within the CNT bundle. In Fig. 3(d), the maximum value of $\Delta I^{LOCK\text{-}IN}$ within the circle in Figs. 3(b) and 3(c) is plotted as a function of V_{PIEZO}. We detect that the local maximum value of $\Delta I^{LOCK\text{-}IN}$ first rises and then decreases for increasing V_{PIEZO}, which is consistent with the observations in Fig. 3(a). Hereby, we interpret the nonmonotonous behavior resulting from a superposition of the local effect caused by a mechanically deformed contact region forming a Schottky contact and the globally increasing effect of uniaxial strain on the resistivity of the CNTs.

The increase of ρ_{CNT} as a function of the uniaxial strain is usually explained by an alteration of the CNTs' band gap.[14–17,21,22,25] Reports verified this band gap change to be ~ 12 meV for a maximum strain value and a device geometry similar to the one reported here.[16] We repeated the photocurrent measurements as a function of the uniaxial strain at different laser wavelengths λ_{LASER} (data not shown). First, we detect a spectrally broad maximum of the photocurrent at $\lambda = 830 \pm 20$ nm at $V_{PIEZO}=0$ V, which is consistent with a CNT diameter of about $d_{CNT}=1.10\pm 0.05$ nm.[7] Second, we do not detect any strain induced shift of the resonant excitation energy because the broad maximum at $V_{PIEZO}=0$ V seems to mask a possible shift of the photocurrent resonance at $V_{PIEZO}>0$ V.

In summary, we present spatially resolved photocurrent measurements on freely suspended CNTs. The CNTs are strained by elongating a piezoelectric stack attached to the sample. We observe a rise of the photocurrent signal of up to $\sim 150\%$ for uniaxial strain of about 0.3–1.2% and a decrease of the photocurrent signal for strain values exceeding this value. We explain the nonmonotonous behavior by a superposition of a local effect caused by Schottky contacts and the global influence of uniaxial strain on the CNTs.

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