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K. Fu, R. Zannoni, C. Chan, S. H. Adams ...+3 more authors

Published on: 23 Jan 2008 - Applied Physics Letters (American Institute of Physics)

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Eric Polizzi, *University of Massachusetts - Amherst* K. S Yngvesson J. Nicholson S. H Adams C. Chan, et al.



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Citation: Appl. Phys. Lett. **92**, 033105 (2008); doi: 10.1063/1.2837188 View online: http://dx.doi.org/10.1063/1.2837188 View Table of Contents: http://apl.aip.org/resource/1/APPLAB/v92/i3 Published by the American Institute of Physics.

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#### ADVERTISEMENT



#### Terahertz detection in single wall carbon nanotubes

K. Fu, R. Zannoni, C. Chan, S. H. Adams, J. Nicholson, E. Polizzi, and K. S. Yngvesson<sup>a)</sup> Department of Electrical and Computer Engineering, University of Massachusetts, Amherst, Massachusetts 01003, USA

(Received 19 November 2007; accepted 24 December 2007; published online 23 January 2008)

It is reported that terahertz radiation from 0.69 to 2.54 THz has been sensitively detected in a device consisting of bundles of carbon nanotubes containing single wall metallic carbon nanotubes, quasioptically coupled through a lithographically fabricated antenna, and a silicon lens. The measured data are consistent with a bolometric detection process in the metallic tubes and the devices show promise for operation well above 4.2 K. © 2008 American Institute of Physics. [DOI: 10.1063/1.2837188]

Single wall carbon nanotubes<sup>1</sup> (SWNTs) have many potential applications in electronics and photonics.<sup>2</sup> Photoconductive detection is apparently very weak,<sup>3</sup> but Itkis *et al.*<sup>4</sup> have reported a sensitive *bolometric* Near Infrared detector based on a carbon nanotube (CNT) film. Microwave detection using SWCNTs has been extended to 110 GHz.<sup>5</sup> Thus, the question arises whether SWCNTs could be useful as terahertz detectors. We have proposed a very fast terahertz detector based on the hot electron bolometric effect in metallic SWNTs (m-SWNTs).<sup>6</sup> In this letter, we present experimental results on detection of terahertz radiation in *m*-SWNTs. We interpret our results based on a general bolometric model.

We have previously demonstrated microwave detection in single m-SWNTs (Ref. 7) that was ascribed to the nonlinearity associated with the "zero-bias anomaly (ZBA)"<sup>8</sup> in the contact resistance (R) at low bias voltage. The microwave response can be predicted from standard microwave detector theory<sup>9</sup>

$$S_I = (1/4) * (d^2 I/dV^2) * V_{\rm MW}^2 / P_{\rm MW}, \tag{1}$$

$$S_V = S_I * R. \tag{2}$$

Here,  $V_{\rm MW}$  is the peak microwave voltage. The factor  $d^2I/dV^2$  was calculated from the measured *IV* curve. The bias voltage dependence and the magnitude of  $\Delta I$  agreed well with that of Eq. (1).

For the present work, we have fabricated m-SWNT devices by the dielectrophoresis method.<sup>10</sup> Typically, we apply a 5 MHz voltage of about 5 V peak to Au contacts made by UV photolithography such as those shown in Fig. 1. We used nonconductive sapphire or silicon on sapphire substrates. A drop of a suspension of CNTs (Ref. 11) in isopropyl alcohol is applied in the contact area. The CNTs will then drift to the narrow gap in the contacts and attach to these. The process is halted when the dc resistance is sufficiently low. The result is that a small number of bundles of CNTs will be contacted in parallel. The lower resistance of these devices compared with the typical single SWCNTs, from 5 to 50 k $\Omega$ , facilitates matching of microwaves or terahertz radiation to the CNTs. While semiconducting tubes are expected to be present in the bundles, we assume these to have a negligible effect (at dc) due their known higher resistance.

Both structures can be measured in a microwave probe system, a useful diagnostic tool. Figure 1 (left) shows a coplanar waveguide (CPW) and Fig. 1 (right) a log-periodic toothed antenna (LPA). The SWCNT bundles are applied across the smallest gaps in these structures, about 3  $\mu$ m for the CPW and 8  $\mu$ m for the LPA. The CPW is designed for microwave work, while the LPA is designed primarily for terahertz experiments.

Each tube is assumed to be modeled by the equivalent circuit introduced and analyzed by Burke<sup>12</sup> (see Fig. 2). The m-SWCNT at the center of Fig. 2 is modeled as a transmission line (the unit cells shown are repeated periodically). The contacts are modeled by a resistance parallel with a capacitance. Our microwave characterization work indicates that the total contact capacitance is of the order of 10 fF, in agreement with other recent measurements.<sup>13</sup> The resistance is dominated by the contact resistance and the ZBA effect is still evidenced in the upward curvature of the IV curves, similar to that found for the single tubes.<sup>7</sup> The propagation velocity on the TL is about 0.01-0.02 times the velocity of light, interpreted as the velocity of a "Tomonaga-Luttinger plasmon" wave. It is predicted that resonances will occur on the TL at terahertz frequencies as its electrical length is a multiple of half wavelengths.<sup>12</sup>

For the terahertz measurements, a device chip with dimensions  $6 \times 6 \text{ mm}^2$  was inserted in a fixture that allowed quasioptical coupling to terahertz radiation, as well as bias input and detector output through a coaxial cable and a bias tee. Gold bond wires were used to connect to the contact pads of the LPA. The fixture was mounted in a liquid helium dewar.

A 4 mm diameter ellipsoidal silicon lens was attached to the substrate for quasioptical coupling to the antenna,<sup>6,14</sup> as shown in Fig. 3. The device was biased through a 100 k $\Omega$ 



FIG. 1. (Color online) Microwave (left) and terahertz (right) structures for coupling to the CNTs.

<sup>&</sup>lt;sup>a)</sup>Electronic mail: yngvesson@ecs.umass.edu.

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FIG. 2. Equivalent circuit model for a m-SWCNT, see Ref. 12.

sensing resistor that configured a Keithley supply as a constant voltage source. A lock-in amplifier was connected across that resistor in a balanced mode in order to record the detected change in current through the device. Terahertz radiation was introduced through the silicon lens from a terahertz gas laser that has a typical output power of 2-5 mW. The laser was modulated at 1 kHz by inserting an acoustooptic modulator after the CO<sub>2</sub> pump laser.

Using this configuration we have demonstrated detection in CNT bundles at five different frequencies from 0.694 to 2.54 THz. We measured the response of two devices with quite different resistances: Device A, 430 k $\Omega$ , and device B, initially 7 k $\Omega$ , later 20 k $\Omega$ , all given at 300 K and low bias voltage. Here, we discuss the results for device B in detail.

This device initially had a room temperature resistance of 7 k $\Omega$  ("device B1") which after about one month changed to 20 k $\Omega$  at 300 K ("device B2"). Many experiments were then performed during which the *IV* curves at a given temperature stayed the same.

A summary of all terahertz detections obtained so far is given in Fig. 4. The terahertz power was measured outside the window of the dewar, and the response was linear in power. There is a roughly 3 to 4 dB optical loss between the dewar window and the antenna terminals. It is clear that there is a general type of detection process that works for a wide range of terahertz frequencies. The higher resistance device A has more than an order-of-magnitude lower responsivity.

Our hypothesis is that the detection process at terahertz frequencies is of the bolometric type, similar to that in Ref. 4. A bolometer is a device that has a temperature-dependent resistance R(T) and a heat capacity C. The bolometer is thermally connected through a thermal conductance  $G_{\text{th}}$  to a heat reservoir at temperature  $T_0$ . As the bolometer is heated by the terahertz power and biased by the dc current  $I_0$ , its temperature is increased from  $T_0$  to  $T_0+\Delta T$ . If we define the factor  $b=(1/R)^* dR/dT$  then the voltage responsivity of the bolometer will be (neglecting electrothermal feedback)<sup>15</sup>





FIG. 4. (Color online) Summary of the responsivities of all terahertz detections obtained, plotted versus frequency.

$$S_V = \Delta V/P_{\text{terahertz}} = \frac{I_0 * R * b}{[G_{\text{th}} + i\omega C]} (V/W).$$
(3)

The thermal time constant of the bolometer is determined by  $\tau_{\text{th}} = C/G_{\text{th}}$ . The near infrared bolometer recently demonstrated by Itkis *et al.*,<sup>4</sup> uses a CNT film containing a network of randomly oriented CNTs. This paper showed convincingly that it is essential for achieving a high bolometer responsivity that the CNT film be suspended and not touching the substrate. Suspending the bolometer minimizes  $G_{\text{th}}$ .

Next, we discuss how a bolometric process can explain our measured data. The equivalent circuit (Fig. 2) contains a capacitance parallel to the contact resistance which our microwave probing shows to be at least 10 fF, large enough that it effectively shunts the contact resistance at terahertz frequencies. We estimate that we have about ten metallic SWCNTs in parallel. Simulation of the circuit in Fig. 2 (for a single m-SCWNT) shows that the mismatch loss right at the resonance frequencies may have large peaks, if the damping is weak.<sup>12</sup> For our sample, we expect *m*-SWCNTs to vary somewhat in length, however, which will tend to wash out the resonances. We find an approximate estimate of the average mismatch loss of 12 dB. Our measured data in Fig. 4 are not inconsistent with this simple model, but clearly many further measurements are required for detailed comparison with the theoretical models. Some CNTs may also be quasimetallic (qm-SWCNTs) and have a bandgap corresponding to terahertz frequencies,<sup>16</sup> which would provide a second efficient mechanism for terahertz absorption in such tubes.

In order to estimate the increase in temperature due to absorption of terahertz radiation we note that Pop<sup>17</sup> found a quite large thermal conductance from a single *m*-SWCNT to oxide covered silicon substrates, g=0.17 W K<sup>-1</sup> m<sup>-1</sup> at 300 K. Similarly, Maune *et al.*<sup>18</sup> obtained a value of 0.26 W K<sup>-1</sup> m<sup>-1</sup> for SWCNTs on sapphire substrates, as used in this research. We will use the latter value for our estimates below. Note that the above  $G_{th}$  represents heat conducted *directly from the CNTs to the substrate*. For ten parallel tubes, 8  $\mu$ m long, we estimate total thermal conductances of 2.1 × 10<sup>-5</sup> W/K at 300 K and 5.4 × 10<sup>-6</sup> W/K at 77 K. These values are expected to be modified for a bundle of tubes. To further test the model based on Eq. (3) we have measured the resistance of device B2 for a range of tempera-



FIG. 5. (Color online) Plots of dR/dT vs bias voltage for several different temperatures. These plots were derived from *IV* curves such as that for 4.2 K shown in the inset.

tures from 300 to 4.2 K, and calculated dR/dT from these data (see Fig. 5).

We can then predict the bias voltage dependence of the detected voltage response from Eq. (3), while using  $G_{\text{th}}$  as an adjustable parameter. We find good fits for four laser frequencies, examples of which are shown in Fig. 6, further supporting the hypothesis that the device detects by a bolometric process. On the other hand, as is also shown in Fig. 6, the measured response does not fit the prediction based on  $R^* d^2 I/dV^2$  [Eqs. (1) and (2)], so the detection processes at terahertz and microwaves are different. This is consistent with our circuit model since in the microwave case the non-linearity is provided directly by the contact resistance, which is effectively shunted at terahertz frequencies. In terahertz detection, the actual CNTs are heated leading to a change of the dc resistance in the same manner as in our resistance measurements versus temperature.

The measured responsivity shown in Fig. 6 is based on the power outside the dewar window. If we estimate the total of the mismatch loss and the optical loss to be 16 dB, we then find revised values of  $G_{\rm th}=6\times10^{-6}$  W/K (4.2 K) and  $5\times10^{-5}$  W/K (77 K), within an order of magnitude of the estimates above.<sup>19</sup> The temperature dependence of  $G_{\rm th}$  is consistent with theory (see Mingo and Broido).<sup>20</sup> The re-

#### Measured Measured **Bolometer Mode** Bolometer Mode 10 2 0 Diode mode 0 Diode Mode (M/N)NS N/2/2 0 -2 Vbias(mW) Vbias(mV) -3 1000 .500 ò 1000 1000 -500 ò 500 1000 500 (a) (b)

FIG. 6. (Color online) Fit of the measured responsivity vs bias voltage to that predicted from Eq. (3) (bolometer mode) and Eqs. (1) and (2) (diode mode). The data are for (a) a laser frequency of 1.395 THz at 77 K and (b) a laser frequency of 0.694 THz at 4.2 K.

sponsivity decreases by a factor of two at 25 K compared with 4.2 K, indicating a potential for operation well above liquid helium temperature.

Further, we estimate the thermal time constant to be  $\tau_{\rm th}$  = 1.5 ps at 77 K with a similar value at 4.2 K. The measurements do not yet allow us to verify the value for  $\tau_{\rm th}$ . We modulate the laser at 1 kHz and find that the detected signal decreases when the modulation frequency is increased in the range of 5–15 kHz. This is consistent with the maximum rate at which our terahertz gas laser can be modulated, as verified by using a Schottky diode detector.

Based on the work of Itkis *et al.*<sup>4</sup> and Pop *et al.*,<sup>17</sup> we expect the bolometer responsivity to be much larger (by an estimated two to three orders of magnitude) for *m*-SWCNTs suspended in vacuum across a trench. In that case the thermal conduction will be entirely confined to the SWCNT itself. The responsivity and the thermal time constant may be traded against each other by adjusting  $G_{\text{th}}$  as was done, for example, in Ref. 21.

In conclusion, we report detection of terahertz radiation over a wide frequency range in bundles of CNTs containing m-SWCNTs. The experimental data are consistent with a general bolometric model. While much detailed work remains to clarify and optimize the properties of m-SWCNT terahertz detectors, we have demonstrated the advantage for such work of employing lens/antenna coupling to the CNTs, as proposed in Ref. 6.

This work was supported by NSF under grant Nos. ECS-0508436 and ECS-0725613.

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