Development of Carbon Nanotube based radiation detectors

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Multi Wall Carbon NanoTubes (MWCNT) are one-dimensional structures with diameters ranging between 5 and 30 nm and lengths up to hundreds of micrometers. They show a metallic-like behaviour and enhanced field emission capability; their sensitivity to the radiation is very peculiar, depending on their diameter and chirality. The wide range of their bandgaps (from 0.4 to ≈ 6 eV) makes these devices very suitable for fabrication of sensors of electromagnetic radiation, from UV to IR. This opens the possibility to build wide sensitive range radiation detectors for space researches and environmental controls. Preliminary tests have been performed by exposing a prototype detector made up of a MWCNT carpet grown by the CVD technique between two gold electrodes on a 500 micrometer thick silicon substrate to pulsed UV, visible and IR radiation. First results on the charge generated by pulsed laser and continuous lamp illumination are reported.

Keyword: Carbon Nanotube; radiation detector; sensitivity

INTRODUCTION

Carbon nanotubes (CNT) are empty cylinders made of carbon atoms [1,2]. They are neither semiconductor nor metals, and are classified as semimetals [3]. For Single Wall Carbon NanoTubes (SWCNT) the geometrical parameters, fixed by the (*n, m*) chiral vectors of the structure, set their electronic states [4]. Nanotubes characterized by *n-m=3p* (*p* positive or null integer) behave like metallic conductors. Other combinations of *n* and *m* indexes lead to semiconductors with an energy band gap depending on the tube diameter according to the relation: $E_{gap}=2y_0a_{cc}/d$, where $y_0 = 0.1$ eV, $a_{cc} = 0.142$ nm and d is the tube diameter. Thus, in semiconductor single wall nanotubes, the energy band gap ranges between 0.4 and 0.7 eV [5]. Multi Wall Carbon NanoTubes (MWCNT) appear as several concentric SWCNTs with various tube diameters, ranging from 5 to 30 nm, and/or with randomly distributed values of the chiral vectors. MWCNTs exhibit generally a conductor-like behaviour.

Several experiments have already shown that semiconductor carbon nanotubes exhibit fluorescence properties in the near-infrared wavelength region (between ≈ 1 and ≈ 1.5 μm) [6]. This property is due to the electronic configuration of semiconductor carbon nanotubes and seems promising for applications in devices used in biophysics and in other fields of physics.

Furthermore, CNTs can be grown in nano-patterned areas, allowing one to easily build nano-patterned devices sensitive to the radiation. This growing process, obtained by means of the nanolithography of catalyst on the substrate, is very chip and allows patternization on large areas. In this way large and nano-pixel photocathodes could be obtained.

Results presented in this paper have been obtained in the frame of GINT's *(Gruppo INFN per le Nano Tecnologie)* programme [7], funded by INFN. The GINT's programme is addressed to study the possibility to build a nano-patterned photocathode working in a wide radiation range using carbon nanotubes. Towards this aim MWCNTs have been produced and their response to the incident pulsed laser and continuous lamp radiation, ranging from UV to IR, has been studied.

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The CNT device

The device used for this investigation is shown in Fig. 1. MWCNTs are grown, by the CVD technique, on a flat surface between two gold electrodes deposited on a 500 μm tick silicon substrate insulated by means of 30 nm thick Si_3N_4 layer. A drain voltage applied between the electrodes collects drift charges through the nanotube layer. Under illumination, extra charges are generated in the CNT layer and collected by the applied electrical field. The outgoing signals can be registered by a digital oscilloscope and studied to evaluate the charge amount generated as a function of the laser wavelength and intensity. The comparison with the device response in the absence of light allows the study of CNTs photoresponse
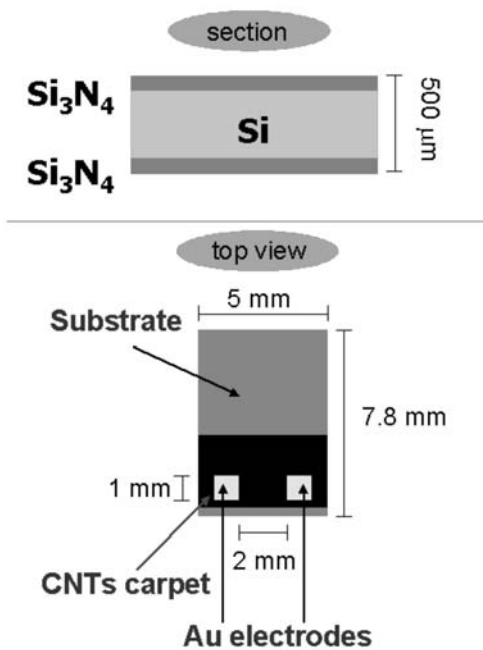
Figure 1: The CNT device used.

The I-V characteristic measured in absence of light (dark current) is reported in Fig. 2. The Ohmic behaviour of the device comes out to be very clear; a value of 9.4 kΩ can be extracted for the layer resistance.

Measurements reported in the following have been obtained using: 1) a Q-Switched Nd-YAG laser (532 nm and 1064 nm, 20 ns pulse duration, 10 Hz repetition rate); 2) a nitrogen laser (337.1 nm wavelength, 300 ps pulse duration, 10 Hz repetition rate); 3) a continuous 633 nm laser; 4) a continuous white lamp as exciting sources.
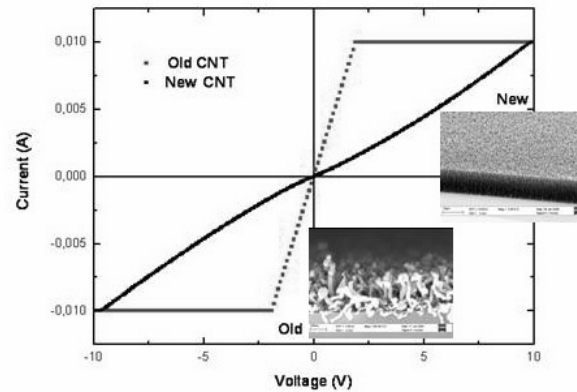

Figure 2: I-V plot for the used sample and for an older one. In the insets SEM images of the surfaces.

CNT characteristics

Fig. 3 shows typical pulses obtained firing with the Nd-YAG laser (a) and with the nitrogen laser (b), at various drain voltage values. Oscilloscope traces on the left parts of the figure show the dark current level, from which the I-V plot of Fig. 2 has been obtained. A long tail is evident at all applied voltages, probably due to the photo-generated charge that relaxes over the capacitance of the device structure.
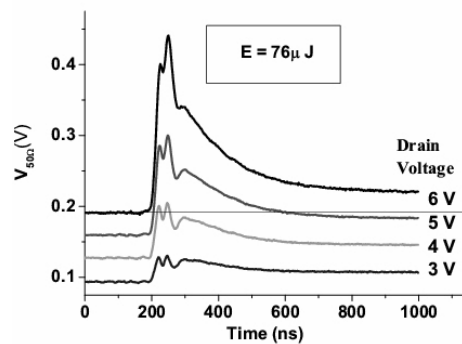
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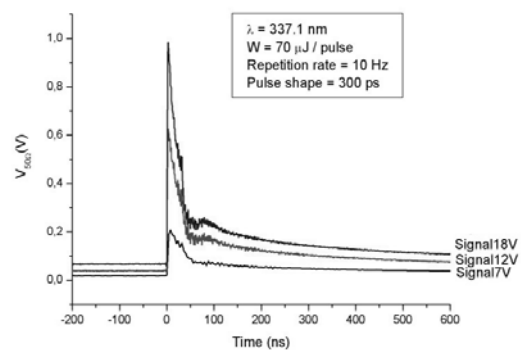
b

Figure 3: Pulse characteristics at various drain voltages

This effect is still evident in Fig. 4, where a comparison among pulses obtained at various wavelengths at a drain voltage of 10 V is shown. The sensitivity to the radiation clearly depends on the radiation wavelength, showing its maximum intensity in the UV region.
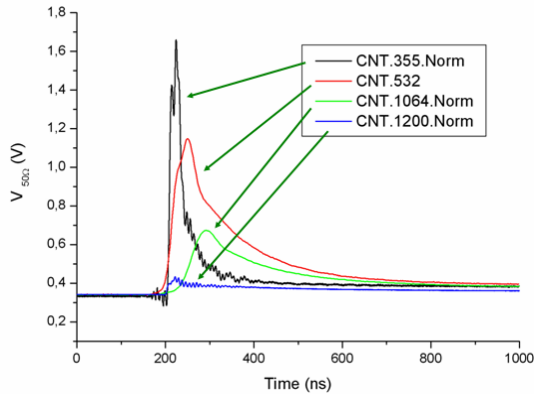

Figure 4: Comparison among pulses obtained at various wavelengths at V_{drain} = 10 V.

The integrated area of signals is proportional to the drained charge. For the 337.1 nm wavelength, the collected pulses have been also amplified by means of a EG&G 579 Fast Filter Amplifier, stretched and fed into a MCA. The recorded spectra are reported in Fig. 5, showing an increasing FWHM vs. V_{drain} and a pulse "saturation" at 20 V. In Fig. 6 the 20 V spectrum is compared with the spectrum recorded by using a Centronic OSD 15-5T photodiode, showing that part of the peak widths is due to the laser jitter.
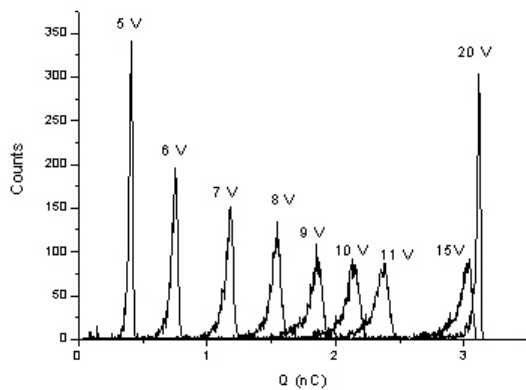

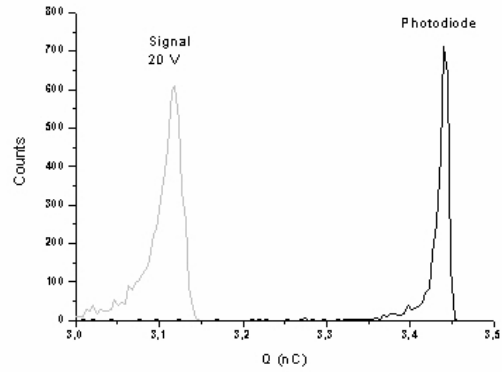**Figure 5.** Spectra of the collected charge at various V_{drain} for the 337.1 nm pulsed laser.

Figure 6: Comparison between the spectra recorded by our device at V_{drain} = 20 V and by a photodiode (see text).

A plot of the charge collected at different drain voltages is shown in Fig. 7. The bias current due to the ohmic conductivity of the CNT layer has been subtracted
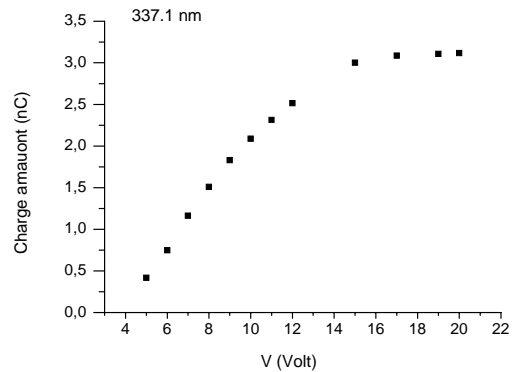

Figure 7: Charge drained vs. the drain voltage at 337.1 nm.

About 10 billion of electrons are collected for each laser pulse. The quantum efficiency strongly depends on the radiation wavelength, assuming its maximum value in the UV region. Unfortunately the device layout does not allow a precise measurement of CNT illuminated area, thus we can only report a relative measurement, in particular in the range between 400 and 850 nm, where an accurate intensity measurement by a monochromator has been performed (Fig. 8 a and b). A lamp light, filtered by a monochromator (a) is chopped and illuminates the CNT layer. The CNT responsivity (b) is calculated by normalizing the drained charge at fixed light intensity. The results show a growing quantum efficiency

going from 400 to 700 nm and a quick decrease between 700 and 850 nm.
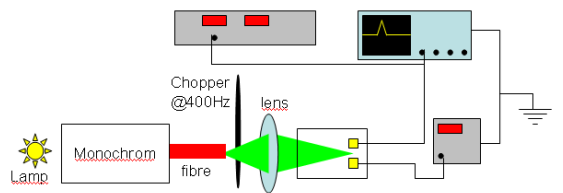
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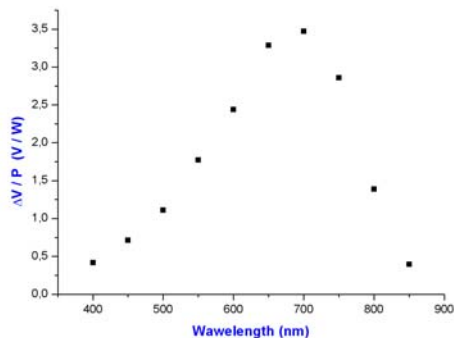
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Figure 8: The apparatus used to measure the device responsivity in the range 400-850 nm (a) and the device responsivity (b).

From Fig.7 and from analogous results obtained with a 633 nm continuous laser, shown in Fig. 9, one can infer that a minimum electrical field is required to efficiently collect the charge produced by illumination. Threshold voltages of about 2.6 V for the 337.1 nm pulsed laser illumination and of about 6 V for the 633 nm continuous laser can be drawn.
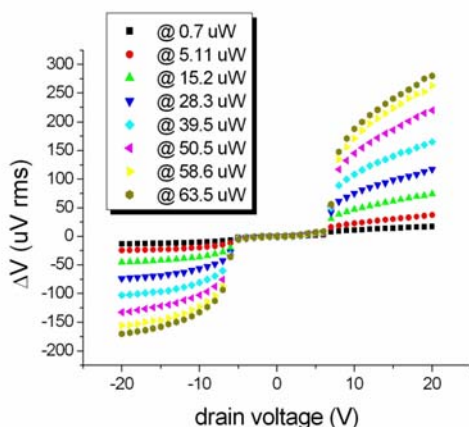

Figure 9: The drained charge induced in the CNT layer with a 633 continuous laser.

5. CONCLUSIONS

We have approached the problem to build a large area, large wavelength range radiation detector with the use of CNTs. The results are very encouraging showing:

1. Good quality aligned MWCNTs have been produced by the CVD technique;
2. The films have shown important photo-conversion effects as a function of the wavelength in a large spectral range; first results about the drained charge have been obtained.

An accurate analysis on CNT photoconductivity properties aiming at understanding the solid state mechanisms accounting for the observed effect is in progress.

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