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Carbon nanotube-based cold cathodes for a new generation of X-ray systems

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Summary. — We present here a new device capable of generating X-rays based on a novel concept of electron source: carbon nanotube-based cold cathodes. In particular, we developed lithographic-based synthesis protocols for obtaining aligned and vertically oriented carbon nanotubes (CNTs) grown by CVD. We report some results obtained in our lab on the characterizations of field emission properties of CNT-based cold cathodes in order to demonstrate that these emitters have the capability to produce self-focused electron beams. Finally we report the results carried out by developing a reliable proof-of-concept of X-ray tube with CNT cold cathodes in the framework of the FP7-SME project NANORAY.

PACS 73.63.Fg – Nanotubes.
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PACS 87.59.-e – X-ray imaging.

1. – Introduction

Nowadays breakthrough in X-ray devices is required in terms of reliability, cost effectiveness and performances, but the technology used to produce X-rays has changed very little since the discovery by Wilhelm Roengten in 1895. As known, current devices still utilize electron beams produced by thermoionic emission from a heated metallic filament. The conventional X-ray tubes are of some limitations related to the features of

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the electron emission into vacuum. The thermoionic emission is characterized by slow response times, high power consumption, short tube lifetimes, and a broad spatial radiation distribution. Moreover, X-ray systems using a thermoionic cathode normally require a cooling system, making it difficult to construct a compact tube.

In recent years, a new scenario has been opened by the availability of nanostructures characterized by efficient field-electron emission and feasibility to fabricate the so-called cold cathodes [1]. Cold electron sources based on field emission (FE) of carbon nanotubes (CNTs) are presently considered as a best choice for generation of free electrons in vacuum, and FE CNT-based vacuum devices are promising for integration in high-frequency, high-power and size-reduced electronics [2-4]. CNT-based cathodes suggest novel technology to fabricate X-ray sources characterized by a fast response time, drastic reduction in size/weight and better definition of generated images [5].

Another interesting feature to be considered is a quick response of the cathodes based on FE processes and the possibility to fabricate an array of miniaturized cathodes to generate a scanning X-ray beam [6-8]. This would result in a 3D X-ray imaging, obtained activating sequentially different electron sources at different angles with respect to the object, without moving the source, and therefore without using high-cost precision mechanics [9].

This last application needs the manufacturing of small-area cathodes with microsized emission spots. To engineer emitting sources with the characteristics required for the above-reported applications, innovative solutions for the preparation of CNT deposits have been proposed and settled in our labs [10]. The synthesis of CNT deposits with controlled density, height and size distribution of the emitters is carried out using a series of different CVD reactors. Small-area cathodes able to produce reduced emission spots are fabricated varying the geometries, from wires and tips to planar substrates patterned by lithography [11].

Taking advantage of the expertise in fabricating CNT-based emitting systems with tailored features, we thought it worthwhile to test such cathodes using an innovative X-ray tube assembly able to overcome the limitations of the conventional sources. In the framework of the FP7-SME we started to develop a NANORAY, a proof-of-concept of a non-conventional device capable to generate X-rays by means of a novel concept of cold cathode, based on CNTs selectively grown upon *ad hoc* synthesized nanostructures. The project NANORAY is carried out in collaboration with industrial partners (Selex-SI of Finmeccanica Group, Italy, NT-MDT, Russia, Nanocyl, Belgium, X-Tek of Nikon Metrology Group, United Kingdom, Sineurop Nanotec - Germany) involved in pursuing an effective nanotechnology transfer. The project objectives are to produce X-ray portable systems with high image resolution and cost effectiveness.

2. – The cathodes: preparation and testing

The catalysed synthesis of carbon nanotubes is carried out using various chemical vapour deposition (CVD) techniques, using CH_4/H_2 mixtures as reactants. By selecting the synthesis procedures both SWCNT or MWCNT deposits can be produced. The substrates are conductive Si sheets with both flat un-patterned or flat patterned shape but also metallic wires and tips (W, Ta, steel). The morphology of the deposits is studied using a Hitachi S-4000 Field-Emission Scanning Electron Microscopy (FE-SEM) and the structural characteristics by micro-Raman spectroscopy ($\lambda = 514.5\,\text{nm}$).

As regards the planar substrates, we present in fig. 1 the FE-SEM image of a large-area deposit formed by SWCNTs laying on a planar substrate. The observations of

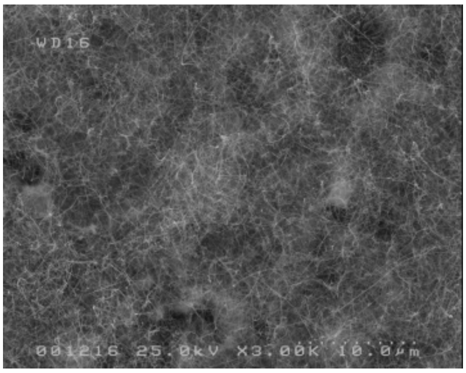

Fig. 1. – Planar deposit of SWCNTs.

many coated samples show that our synthesis conditions are able to produce also homogeneous deposits formed by densely packed bundles of aligned nanotubes perpendicularly anchored to the substrate [10]. The Raman analysis indicates that SWCNTs were the predominant product of the synthesis.

In fig. 2 A) and B) are shown some FE-SEM images of carbon nanotube deposits on $10 \times 10\,\mu$m and $20 \times 20\,\mu$m squared patterns prepared by sputtering thin layers of Ni/Ti catalyst.

Figure 3 evidence the morphology of SWCNT bundles grown on a wire-shaped W substrate.

For each cathode typology the emitted current, the emission stability and the life cycle have been tested. The field emission functional properties were studied by means of a custom-designed apparatus [12]. The measurements were carried out at room temperature and working pressure of 10^{-7} mbar, using as anode a Mo sphere with a diameter of 1.00 ± 0.05 mm. All the samples have been tested at a variable anode-cathode distance, determined by a capacitive approach using an LRC meter (Stanford Research SR715 model). The current was measured by a Keithley 6485 picoammeter with rms noise of

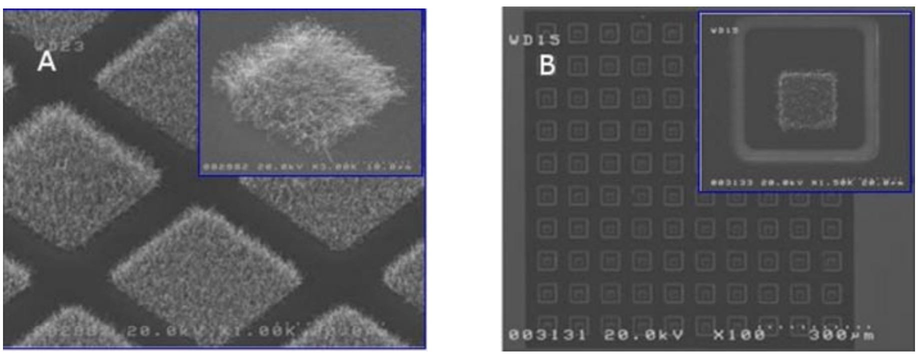

Fig. 2. – FE-SEM images of carbon nanotubes deposits on: A) 10×10 and B) $20 \times 20\,\mu$m squared patterned substrates. The inset of the figure evidences some details of the nanotube deposits.

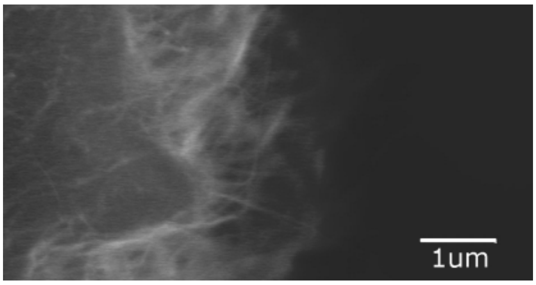

Fig. 3. – FE-SEM images of SWCNT bundles grown on a wire-shaped W substrate.

about 1 pA at lowest current. In fig. 4 A), B), C), D) are reported the experimental I/V curves for the cathodes imaged in figs. 1-3 and in the insets the corresponding Fowler-Nordheim behaviour. Several geometries of CNT arrays are under study in order to establish their effectiveness in producing and focalizing electron beams.

Good and reproducible FE behaviour has been observed for all the realized cathodes. Overall results demonstrated that CNT-cathodes, with proper anode-cathode configurations, have the capability to produce self-focused electron beams with a small energy spread.

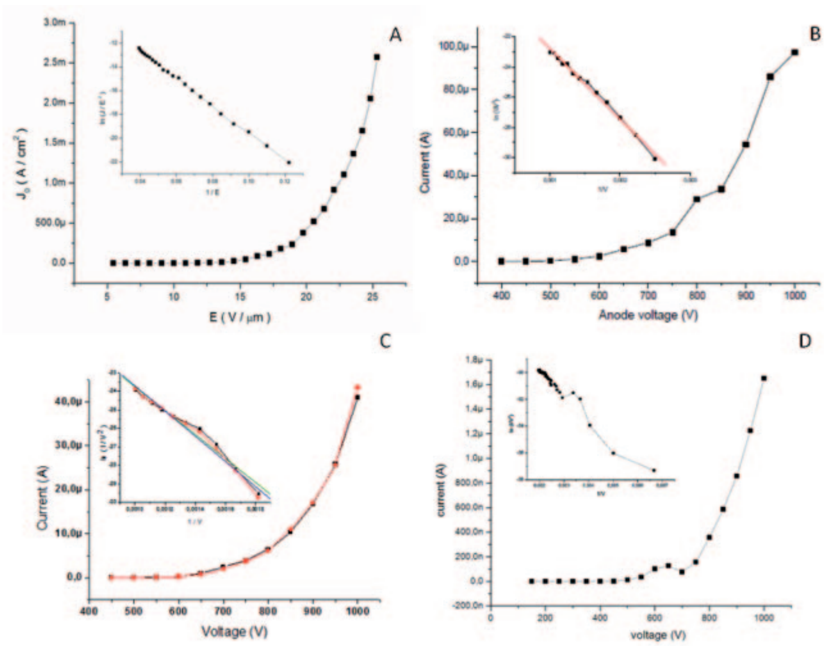

Fig. 4. – A) Plot of the electric field *vs.* area density of the sample depicted in fig. 1. B) Experimental I/V curve of the cathodes in fig. 2A. C) Experimental I/V curve of the cathodes in fig. 2B. D) Experimental I/V curve of the cathodes in fig. 3. In the insets the corresponding Fowler-Nordheim behavior.

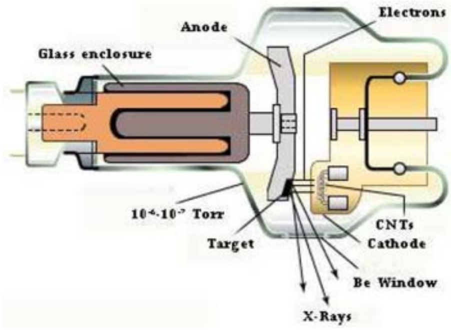

Fig. 5. – Scheme of the X-ray tube.

3. – The X-ray tube

A scheme of the tube prototype is presented in fig. 5.

The main components are the CNT-based cathode, the anode, the grid and the focusing electrodes. The anode is made by Cu. A Be window is used. The critical parameters to be optimized in order to produce a X-ray beam with the features required by the project are: the value of the electric field on the cathode, the voltage applied to the anode, the cathode-grid-anode distances, the grid shape and characteristics (geometry, material, mesh dimension). The design of the devices enables to change independently such parameters, and to modulate therefore the characteristics of the produced X-ray beam.

The electrode configuration of the prototype will allow us to obtain, for anode voltages of 10 kV and 30 kV, minimal focalization spots in the range of 0.1–0.3 mm. An image of an X-ray spot taken outside the tubes at 1 cm from the Be window is shown in fig. 6.

A further advantage of the system is the possibility to tune the output current by changing the acceleration voltage, making it possible to use the same tube for very different applications. The experiments performed up to now clearly indicate that the developed prototype is potentially able to produce X-ray small focal spots for high-resolution imaging.

Moreover, in many advanced applications (from medical imaging to XRF spectroscopy) the concurrent requirement of a reduced spot size and an increased X-radiation density is a key point. In such a context, a challenging solution is represented by the

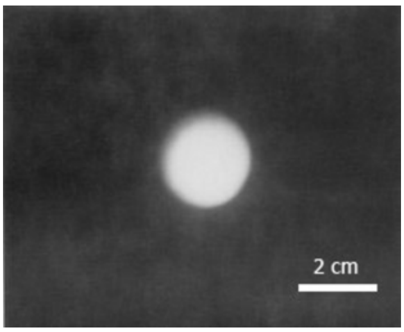

Fig. 6. – Image of X-ray spot generated by using CNT-based cathode as the source of electrons.

use of polycapillary optics [13]. In this system proper configuration of the lenses allows getting quasi-parallel X-ray beams from divergent sources, or convergent beams from quasi-parallel sources, allowing a wider freedom of parameters in the design of the X-ray source for specific applications. In such a context we are also investigating the possibility to replace the glass capillary structures with ropes made by parallel CNT that some theoretical predictions have been shown to be effective in transporting, focusing and deflecting beams of X-ray.

4. – Conclusions

The outstanding electron field-emission properties of carbon nanotubes enable fabricating cold cathodes as a possible alternative for the replacement of thermoionic electron sources for X-ray generation. In this context the fabrication of CNT-based cathodes is a topic of wide interest, due to the unique combination of electrical properties with toughness and chemical inertness, and, thus, to the possibility to use such cathodes also in harsh environments.

From the preliminary experiments, the most relevant features of X-ray tube prototypes assembled with CNT-based cathodes are:

- potential for miniaturization;
- reduced focal spot ($< 0.3\,\mathrm{mm}$) and ultrahigh image resolution;
- very low power consumption;
- instantaneous response time;
- production of pulsed X-ray radiation with programmable width and repetition rate;
- long working life.

It is interesting also to mention that in one of the experimental runs we have registered some phenomenon of diffraction that cannot be explained within classical approximation; the assuming explanation might result in orientational features of X-ray propagation in aligned nanotubes, namely, in X-ray channeling in nanotubes [14, 15].

An interesting application exploiting the quick response of cold cathodes is the 3D X-ray imaging, obtained irradiating the object from different angles, activating sequentially different electron sources. Applications of the proposed device are envisaged in medical and archaeometric diagnostics, fluorescence spectrometry, security, quality control in electronics, aerospace and micro/nano mechanics.

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REFERENCES

[1] Sugie H. *et al.*, *Appl. Phys. Lett.*, **78** (2001) 2578.
[2] Bogue R., *Sensor Rev.*, **30** (2010) 276.
[3] Yue G. Z. *et al.*, *Appl. Phys. Lett.*, **81** (2002) 355.
[4] Sessa V. *et al.*, *Proc. SPIE: Nanotechnology III*, **6591** (2007) 65910M.
[5] Senda S. *et al.*, *Appl. Phys. Lett.*, **85** (2004) 5679.

[6] Schwoebelt P. R., Holland C. E. and Spindt C. A., *IEEE Vacuum Electronics Conf. (2006)*, p. 511.
[7] Gupta S., *Mater. Res. Soc. Symp. Proc.*, **963** (2006) 187.
[8] Wang S. *et al.*, *Mater. Res. Soc. Symp. Proc.*, **1065** (2008) 21.
[9] Zhang J. *et al.*, *Appl. Phys. Lett.*, **86** (2005) 184104-1.
[10] Orlanducci S. *et al.*, *Chem. Phys. Lett.*, **367** (2003) 109.
[11] Terranova M. L. *et al.*, *Chem. Phys. Lett.*, **381** (2003) 86.
[12] Boscolo I. *et al.*, *J. Vac. Sci. Technol. B*, **25** (2007) 1523.
[13] Kumakhov M. A., *Proc. SPIE*, **4155** (2000) 212.
[14] Okotrub A. V. *et al.*, *JETP Lett.*, **81** (2005) 34.
[15] Dabagov S. B., *Nanosci. Nanotechnol. Lett.*, **3** (2011) 24.