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Self-oscillations of carbon nanotube twist-yarn during field emission

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Electromechanical self-oscillations were induced during field emission from the lateral surface of a twisted nanotube yarn obtained by solid-state spinning from a forest of vertically aligned multi-wall carbon nanotubes (CNT). Oscillations at several kilohertz frequencies were driven by applying a constant voltage between fixed opposite ends of the nanotube yarn and a flat anode. An electromechanical model is developed which explains the observed phenomenon. The model is equally applicable to any mechanically flexible field emitter including single nanoemitters like CNTs, which have shown a self-oscillating behaviour in several previous *in situ* experiments. This phenomenon opens a possibility of developing a new type of devices for conversion of DC voltage into the high frequency electromagnetic oscillations utilizing flexible nanoemitters.

1 Introduction Field emission (FE) of carbon nanotubes (CNTs) has been intensively studied since 1995 [1, 2]. The characteristics of FE from CNT cathodes were found to be very promising; however, they were usually lower than those of conventional field emitters consisting of metallic or silicon tips. Typical CNT cathodes have a lower stability of emission current and the shorter lifetimes. These parameters are crucial for their application in a majority of vacuum electronic devices.

Internal properties of CNTs which determine FE characteristics – work function, conductivity, strength of the atomic bonds – are very similar to those of metallic emitters. Moreover, they have extremely high aspect ratios providing a large electric field on the tips under a relatively low applied voltage. At the same time, due to the high aspect ratio, CNTs may be deflected along the electric field by the Coulomb force acting on the charge induced on the tube tip. The experiments show that the outstanding mechanical properties of CNTs allow them to be reversively bent to large angles without mechanical damage [3]. A unique combination of the mechanical flexibility and the low-voltage FE results in an unexpected behaviour of the nanotubes during FE, which was not observed previously for the stiff metallic tips.

In situ TEM experiments have revealed that when DC voltage was applied to the single-wall CNT bundle, it sometimes suddenly began violent mechanical vibrations while emitting electrons [4]. The origin of these electromechanical oscillations was not understood. For multi-wall CNTs (MWCNTs), similar oscillations were also observed [5]. This phenomenon was attributed to the 'ballistic' type of emission in which electrons are emitted as groups due to the small area of the nanotube tip. These discrete oscillations of the charge cause the variation of the electric force and corresponding deflections of the nanotube. Recently, oscillations driven by a DC voltage were observed for semiconducting SiC nanowires [6]. A mathematical model of the nanowire emission process has been built and a qualitative agreement with the experiment has been demonstrated. However, the proposed model has not revealed which properties of the system are crucial for the initiation of the oscillations. Particularly, in case of the nanoscale systems it remains unclear if a low damping and a high resistance of the emitter are essential for the phenomenon to be observed. The mathematical approximations made in Ref. [6] are applicable only for limited types of the systems and obscure the essence of the observed phenomenon.

In this report, we demonstrate electromechanical oscillations driven by a DC voltage in an MWNT twist-spun yarn [7]. We introduce an empirical model that explains the observed behaviour of flexible field emitters.

2 Field emission experiment MWNT yarns are twisted during the spinning process from the oriented MWNT forests grown by CVD technique (See more details in Ref. [7]). The used yarns were about 10 μm in diameter and have lengths up to several cm, although there was no principal limit for the obtainable yarn lengths. They posses a high electrical conductivity (about 600 S/cm) and a high mechanical strength (over 700 MPa).

Basic FE properties of the MWNT yarns have been reported recently [8]. The experiments with MWNT yarn cathode were carried out in 10^{-7} Torr turbo-pumped vacuum chamber. A glass plate of 60 mm diameter covered with an indium-tin-oxide (ITO) film and a layer of phosphor with the thickness of several microns deposited over the ITO was used as an anode. The phosphor layer luminescence allowed characterization of the spatial distribution of emission sites over the cathode surface. The inter-electrode distance was varied in a wide range (up to several centimetres) with a precision screw translator. The FE current was obtained by applying DC voltage up to several kilovolts between the electrodes.

When the voltage was applied to the yarn fixed at one end on a flat steel substrate it became aligned along the electric field, perpendicular to the anode. The observed electron emission pattern on the phosphor anode consisted of lines and arc segments (see Ref. [8] for more details). We believe that these lines may originate from oscillations of the individual tubes protruding from the yarn tip. The oscillations may have the same nature like those observed in *in situ* experiments with individual emitters [4–6] mentioned in the introduction part. Moreover a high current noise and a relatively low current stability, usually observed for the CNT cathodes, may be also caused by these oscillations.

In parallel to the flat anode, the 1 cm long and 14 μm diameter yarn was fixed at two ends on a flat steel substrate. When the voltage was applied, the electron emission pattern on the anode was quite uniform (Fig. 1a) with some inhomogeneities due to variation of the field enhancement factor of the different emission sites along the yarn. The cathode possesses the remarkable FE characteristics: a threshold field less then 1 V/μm for the current density 10^{-9} A/cm^2 and a high value of the field enhancement factor (above 7000).

In case when the cathode was more flexible, that is we used a yarn with a smaller diameter and a bigger length, a non-uniform electron pattern was observed on the anode (Fig. 1b). The pattern was much brighter in the yarn centre than at its fixed ends. This means that the central part of the yarn was deflected towards the anode. The deflection is caused by the Coulomb force acting on the charge induced on the yarn surface. Despite applying a DC voltage, the FE current was far from constant. The current had a strictly periodic form in time, with a repetition frequency of several kilohertz depending on the length and diameter of the yarn (Fig. 2). The oscillations were very stable (at least during 1 h) with small amplitude changes.

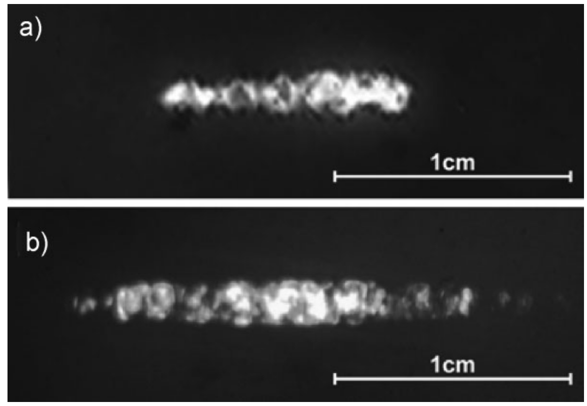

Figure 1 The electron patterns on the phosphor anode of MWNT yarn cathode fixed at two ends with different dimensions: (a) 1 cm length and 14 μm in diameter, (b) 2.5 cm length and 10 μm in diameter. In case of the more flexible yarn (b) the pattern is brighter in centre, which results from the bending of the central part of the yarn in the direction to the anode.

3 Discussion The observed undamped electromechanical oscillations of the MWNT yarn are driven by a constant voltage and are excited independently of initial conditions. This type of oscillations belongs to self-oscillations – a common natural phenomenon observed in various fields of physics. The examples are a heart beat, a human voice, the violin string vibrations, the electrical vacuum tube generators, etc. As stated above, the electromechanical self-oscillations of flexible field emitters have been observed for a wide range of micro- and macroscale objects with significantly different properties. The important

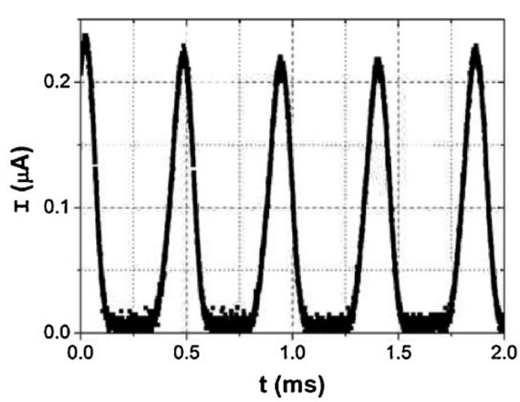

Figure 2 The time dependence of the FE current from lateral side of the MWNT yarn (2.5 cm length and 10 μm in diameter) at DC voltage of 800 V. The repetition frequency is 2.2 kHz.

characteristics being essential for self-oscillations are only a stable field electron emission and a mechanical flexibility. Therefore, only these two properties should be taken into account in development of the basic model of the self-oscillating field emitter.

A mechanical motion of the flexible emitter is governed by an opposite elastic force, a damping intrinsic friction force and an actuating electrostatic Coulomb force acting on the charge induced on the emitter surface. The forces may be determined via the solution of the Laplace and strain-displacement equations for the certain geometry of the emitter. However, to describe the general phenomenon of the self-oscillations we may consider the phenomenological resultant forces in projection on the direction of oscillations (Fig. 3a,b). Assuming that an effective mass m and a charge q are concentrated in the point of the forces application (with coordinate x) the motion is determined by Newton's second law:

$$m\ddot{x} = F_{\text{elastic}} + F_{\text{electric}} + F_{\text{friction}}. \tag{1}$$

In the simplest approximation, flexing of the emitter may by described by Hooke's law $F_{\text{elastic}} = m\omega^2 x$ with the friction force $F_{\text{friction}} = m(\omega/Q)\text{d}x/\text{d}t$, where ω is the fundamental frequency and Q is the phenomenological quality factor of the oscillator. The electrostatic Coulomb force is defined as $F_{\text{electric}} = qE$ where E is the field strength in the vicinity of the emitter surface. The values of the field strength E and the emitter charge q are proportional to the applied voltage V and determined by the geometry of the emitter at given coordinate x. In the first approximation, E and q may be considered to be a linear function of the coordinate $E(x) = (k_{E1} + k_{E2}x)V$ and $q(x) = c(x)V$ with $c(x) = (k_{c1} + k_{c2}x)$ – a mutual capacitance of an emitter and a flat anode.

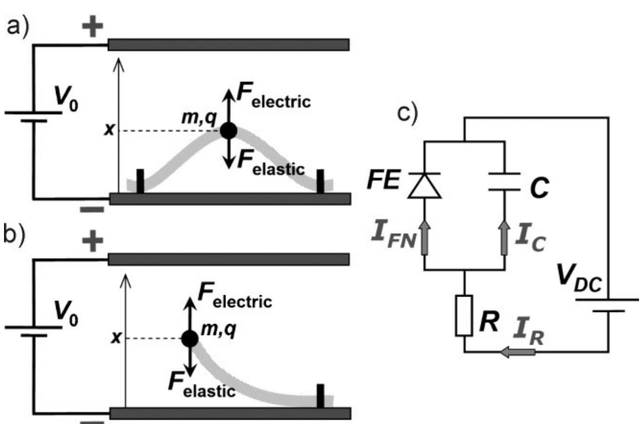

Figure 3 The electromechanical model of the self-oscillating emitter. A mechanical part of the system (a, b) consists of the effective mass m with the charge q moving along the axis x by electrostatic and elastic forces. In case of emitter with two ends (a) and one end (b) fixed, the forces and dynamic equations are the same. An electrical diagram (c) consisting of the emitter, represented by its resistance R, capacity C and FE diode connected to the DC voltage supply.

The constants k_i may be determined from electrostatic calculations or, indirectly, from the experimental data.

When the DC bias voltage V_0 applied to the emitter is high enough to produce an FE current I_{FN}, the voltage on the emitter surface V will be decreased due to the voltage drop on the emitter resistance R. Thus, the electrical part of the system may be represented as the equivalent circuit (see Fig. 3) consisting of the voltage supply, the emitter resistance and the capacity and the diode with a current–voltage characteristic given by the Fowler–Nordheim law $I_{\text{FN}} = C_1(\beta E)^2\exp[-C_2(\beta E)^{-1}]$. The factor β characterizes a difference of the field strength in the point of the FE current outflow and the average field on the emitter surface. The electrical processes in the circuit are, then, described by Kirchhoff's laws:

$$\begin{aligned} V_0 &= I_{\text{R}}R + V, \\ I_{\text{R}} &= I_{\text{FN}} + I_{\text{c}}. \end{aligned} \tag{2}$$

where $I_{\text{c}} = \text{d}q/\text{d}t = \text{d}(cV)/\text{d}t$ is the current due to a charge variation. A combination of Eqs. (1) and (2) gives the final system of equations:

$$\begin{cases} \ddot{x} = -\omega^2 x + c(x)E(V,x)\dfrac{V}{m} - \dfrac{\omega}{Q}\dot{x}, \\ \dfrac{V_0 - V}{R} = C_1[\beta E(V,x)]^2\exp\{-C_2[\beta E(V,x)]^{-1}\} \\ \qquad + c(x)\dfrac{\text{d}V}{\text{d}t} + V\dfrac{\text{d}c(x)}{\text{d}t}. \end{cases} \tag{3}$$

Analysis of the system has shown that at a certain choice of the oscillator parameters and the applied voltage the self-oscillating regime can be achieved. In Fig. 4, an example of the system solution for the parameters corresponding to the MWNT yarn experiment is presented. The obtained amplitude of several millimetre and the corresponding value of DC voltage of 1 kV indicate a reasonable agreement with the experiment.

The processes which take place during the oscillations may be described as follows. When the electric field is applied to the yarn, it begins to deflect towards the anode under the action of the Coulomb force. This deflection is accompanied by an increase of the charge and the field strength on the oscillator surface. When the field reaches the threshold value, the FE current starts to flow from the emitter surface resulting in a rapid decrease of the charge. The Coulomb force is therefore also reduced and the deflection of the yarn oscillator decreases until the Coulomb force exceeds the elastic force. The duration of the downward motion is characterized by the capacity recharge time $\tau = RC$. After that, the process repeats and the oscillator again deflects. If τ is small the oscillations will decay due to the mechanical friction in the system. If τ is high enough and the value of the FE current will be limited by the rate of the charge drag to the emitter surface, then it will be possible for the oscillator to exceed the deflection of the previous period. An important role plays a shape of the field strength dependence on the

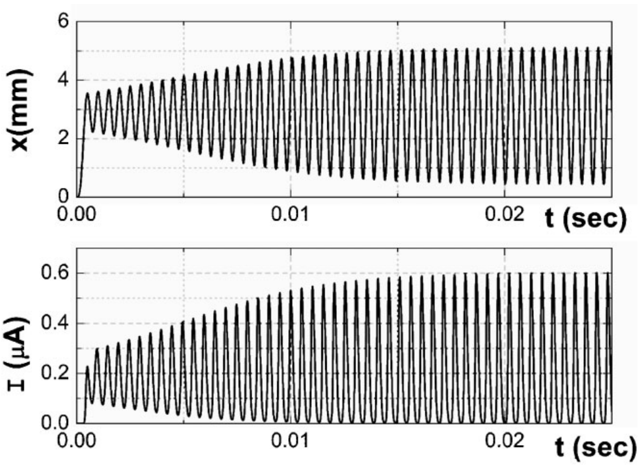

Figure 4 A solution of the system (3) at $V_0 = 1000$ V, corresponding to the self-oscillating regime of the MWNT yarn. The parameters used for simulations were estimated by the electrostatic calculations or directly from the experimental data: $\omega = 2\pi \times 2000$; $Q = 10$; $R = 7 \times 10^8\,\Omega$; $m = 2 \times 10^{-7}$ kg; $k_{c1} = 3 \times 10^{-13}$ F; $k_{c2} = 3 \times 10^{-12}$ F/m; $k_{E1} = 3 \times 10^5$ V/m; $k_{E2} = 3 \times 10^7$ V/m^2; $\beta = 10$; $C_1 = 2.8 \times 10^{-17}$ A; $C_2 = 7.3 \times 10^{10}$ V/m.

coordinate $E(x)$, which determines the rate of charge outflow due to the FE current. An increase of the amplitude will continue until the energy transferred to the oscillator by the electric force coincides the friction force work for the period. In this point, the oscillations became self-sustained. The FE current in this model plays a role of a nonlinear limiting element, which is a component of all self-oscillating systems.

This model is equally correct for the macro- and nanoscale systems. In case of MWNT yarn, the frequency of self-oscillations ω_{macro} is of the order of kilohertz. In the first approximation, the frequency should be inverse proportional to the characteristic dimension of the system L. Assuming for the yarn $L = 1$ mm for the case of single nanotube emitter, we obtain proportionally $[\omega_{\mathrm{nano}}] =$ kHz $\times$ mm/nm $=$ GHz. Thus, the nanoscale flexible emitters, such as CNTs, should generate electromagnetic waves in gigahertz range of frequencies in the self-oscillating regime.

This phenomenon opens a way to develop a new type of electronic devices. For example, using arrays of numerous oscillating nanoemitters it is possible to obtain an efficient generation of the high frequency electromagnetic waves for application in telecommunications. Single self-oscillating nanoemitters may be used in the new type of nanoelectromechanical system (NEMS) which converts a constant voltage into electromagnetic waves and transmits the information to the other parts of NEMS.

4 Conclusion

FE properties of MWNT twisted yarns have been explored in parallel to the anode geometry. The mechanical deflection of the yarn due to the electrostatic force has been observed during FE. At certain values of the applied DC voltage FE current became oscillating in time with the frequency of several kilohertz. The phenomenon was ascribed to the electromechanical self-oscillations of the flexible MWNT yarn. A model of the system based on the classical Newton's and Kirchhoff's laws has been developed. The simulations have shown a possibility of the self-sustained regime at certain voltages. A reasonable agreement between theory and experiment has been demonstrated. The developed model can be applied to any flexible field emitter including nanoemitters like individual CNTs, for which a generation of GHz oscillations is expected. This phenomenon may be used to develop new types of electromechanical devices on the basis of efficient conversion of DC voltage into GHz electromagnetic waves.

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