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LETTER TO THE EDITOR

Dynamic *in situ* field emission of a nanotube at electromechanical resonance

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Abstract

The dynamic field emission of a carbon nanotube at mechanical resonance has been studied by *in situ* transmission electron microscopy. The coupling between the field emission (FE) of a nanotube under a dc voltage applied longitudinally and its mechanical resonance stimulated transversely by an ac field shows that the frequency of the FE oscillating current is twice that of the mechanical resonance. This result has been modelled using the Fowler–Nordheim equation. The nanoscale configuration of the nanoelectromechanical system makes it possible to create novel devices that could correlate the mechanical behaviour with their sensitive FE, and the FE current could provide feedback signals for controlling the devices.

(Some figures in this article are in colour only in the electronic version)

Field emission (FE) of carbon nanotubes (CNTs) has attracted considerable attention in view of the scientific importance and technological applications [1]. A stable emitting current is one of the hotly pursued aims for flat-panel displays and point electron sources in high-resolution electron-beam instruments [2–5]. Many of the studies have been focused on the properties of static field emission at dc voltages, while little work has been done on observing the dynamic response of the field emission of a nanotube to the perturbation of an external field, which is important for understanding its stability and the feedback mechanism for controlling its behaviour and performance. Moreover, dynamic carbon nanotube oscillators have been proposed for possible use in future nanoelectromechanical systems (NEMS) [6, 7]. Combination of the environmentally sensitive field emission and the electromechanical behaviours of a carbon nanotube in NEMS could be of potential use in new kinds of nanosensors [8] and nanodetectors [9]. Further, the alternating emitting electrons could inspire novel nanoscale oscillating circuits and RF signal processing as well as providing a model system for exploring quantum phenomena in coherent electron systems [10].

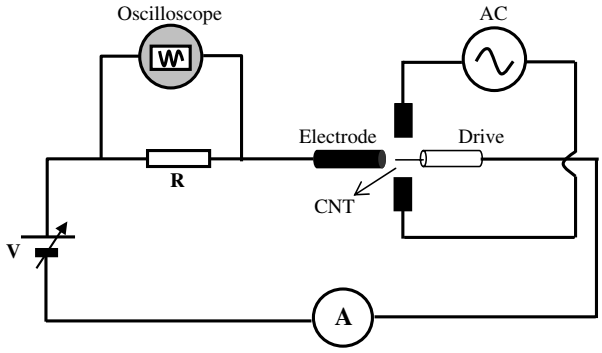

Figure 1. Schematic diagram of the experimental set-up and electronic circuit. The specially designed TEM holder allows the field emission measurement to be performed at mechanical resonance of a carbon nanotube.

The excitation of mechanical resonance, as an important phenomenon in nanomechanics, has been realized for CNTs by applying an external electric field [11, 12]. The electromechanical response and microstructures were directly imaged *in situ* with a transmission electron microscope (TEM); the field emission of a nanotube can also be studied in this way [13–16]. Purcell *et al* [17] found that the field emission of a nanotube can be used to directly observe the mechanical resonances of CNTs in field electron microscopy and field ion microscopy, and they showed that the resonance frequency of a nanotube can be tuned using the tension created by an electric field. Recently, Scheible *et al* [18] have demonstrated periodic field emission from a single nanomachined gold island that was brought mechanically into an electric field.

In this letter, we present studies in which we directly observe the dynamic field emission of a CNT while it is being resonantly excited by an external ac field inside a TEM. The field emission current shows a periodic oscillation component with a frequency that is twice the mechanical resonance frequency. This result is explained using the Fowler–Nordheim equation with a modulated field enhancement factor.

Our experiments were carried out in a JEOL 2010F TEM fitted with a home-built sample holder, at room temperature and in a 10^{-5} Pa vacuum. This set-up simultaneously acquires the mechanical resonance phenomena, the corresponding field emission properties, and the microstructures of the CNT on which the measurements were performed, as schematized in figure 1. There are two pairs of electrodes oriented perpendicularly with each other. One pair is for the field emission measurements, in which an electrochemically etched tungsten needle acts as a movable cathode, and the opposite gold panel is the anode. The distance between the two electrodes can be adjusted from several hundreds to several tens of nanometres, and a dc voltage is applied across the electrodes for inducing field emission. Another pair is used to apply an ac voltage across its two counter-electrodes, so that the mechanical resonance of the nanotube can be induced when the applied frequencies match the harmonic frequencies of the CNT. In this experiment, the aligned multiwalled CNTs were grown by the microwave plasma enhanced chemical vapour deposition (MPCVD) method [19, 20]. An individual nanotube is mounted on the tungsten needle by a piezo-driven nanomanipulator; this procedure was carried out first inside a scanning electron microscope to allow precise control of the geometrical configuration. The front of the needle with the CNT and its counter-electrode are directly observed by the TEM.

We first studied the field emission properties of a static CNT. The I–V characteristics were stable, as shown in figure 2. There was no obvious changes in the field emission behaviour

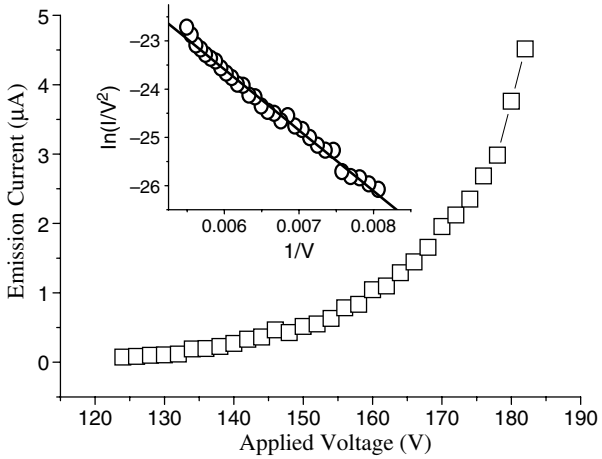

Figure 2. The field emission behaviour of a static carbon nanotube. The inset shows the corresponding FN plots.

after several measurements. The Fowler–Nordheim (FN) plot, $\ln(I/V^2)$ versus $1/V$, where I is the emission current (μ in A) and V is the applied electric field (V), can be fitted as a straight line, in agreement with a Fowler–Nordheim tunnelling mechanism.

As an ac oscillating voltage was applied to the pair of transverse electrodes across the CNT so that the mechanical resonance was excited by adjusting the frequency [11, 12], a dc voltage was applied across the longitudinal electrodes for stimulating field emission of the nanotube. In the meantime, the nanotube mechanical resonance frequency was shifted by the tension created by the dc field [17]; thus, the new resonance frequency had to be found according to the applied dc field. Figure 3(a) shows the resonance of a CNT at $f = 1.473$ MHz under a dc voltage $V = 130$ V. It should be noted that the excitation amplitude of the ac voltage was 3 V, but the true amplitude at MHz frequencies was much smaller because of the narrow band pass of the driving transmission line, so the influence of the ac excitation voltage on the FE current is negligible. An Agilent 54852A oscilloscope was used to trace the field emission current, as shown in figure 3(b), which indicates ac oscillating signals at $V = 130$ and 0 V. Ideally, there should be no field emission at $V = 0$ if there is no influence from the transversely applied ac, but in practice there is an oscillating signal component at the oscilloscope that is due to the coupling with the ac source. Therefore, at $V = 130$ V, the output of the circuit current signal is composed of two parts: one from the FE current of the CNT, and the other from the coupling with the ac excitation source. To separate the two components, the Fourier transformation of the circuit signal shown in figure 3(b) was carried out and the results are given in figure 3(c). If we take the data recorded at $V = 0$ as the reference for the ac coupling signal, the signal recorded at $V = 130$ V at the resonance frequency $f = 1.473$ MHz has a $(5/6)\pi$ phase shift compared to the coupling signal. At double the fundamental frequency of excitation source, $f = 2.946$ MHz, the amplitude of the total signal is distinctly different from the coupling signal, possibly indicating that the FE signal is an oscillating current with a frequency that is twice the mechanical resonance frequency, but it is close to being out of phase with the coupling signal, in agreement with the $(5/6)\pi$ phase shift measured from the Fourier analysis.

To understand the oscillating FE current, we interpret the above FE process on the basis of the Fowler–Nordheim equation, which gives a relation between the emitting current I (A) and the local field at the emitter surface F (V m^{-1}). F is usually related to the applied voltage

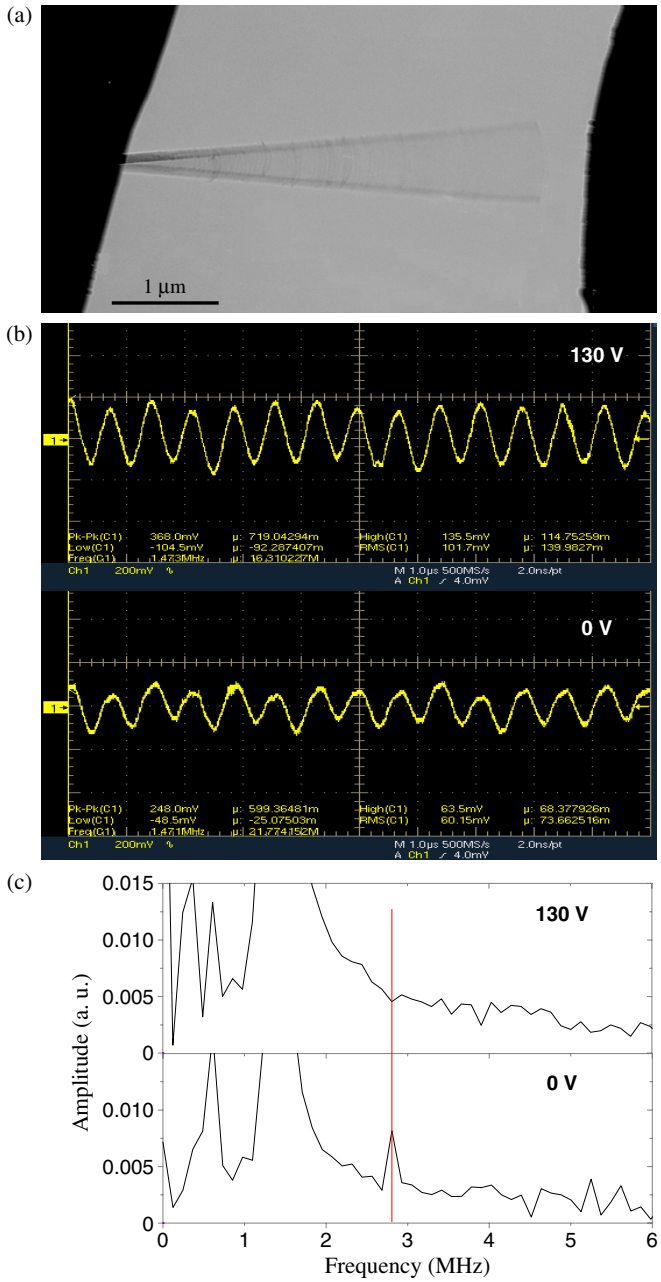

Figure 3. (a) A resonant carbon nanotube at $f = 1.473$ MHz. (b) The circuit currents recorded by the oscilloscope shown in figure 1 at the applied dc voltage $V = 130$ V (above) and $V = 0$ (below). (c) Fourier transform analysis of the current signals presented in (b).

V by $F = \beta V/d$, where d is the interelectrode distance and β characterizes the ability of the emitter to amplify the applied field V/d and is named the field enhancement factor. The FN equation is expressed as

$$I = KF^2/\phi \exp(-B\phi^{3/2}/F) \tag{1}$$

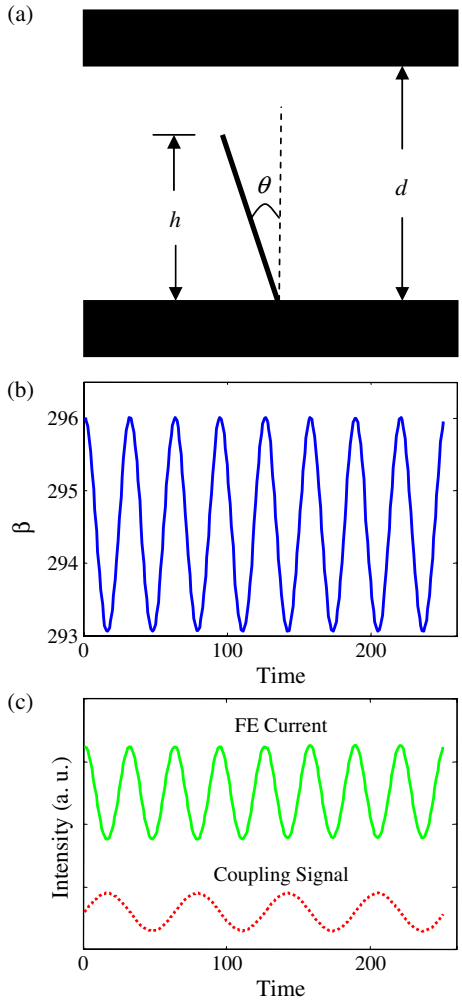

Figure 4. (a) Simplified geometry of the dynamic field emission process of a nanotube at harmonic vibration. (b) The field enhancement factor β–time curve of a resonant carbon nanotube. (c) The oscillating field emission current (solid) and the coupling signal of the ac excitation source (dotted).

where K, B, and ϕ are constants. Figure 4(a) is a schematic drawing of the FE experiment. Here the distance between the emitting tip and its counter-electrode is very small, so β is rather large and is sensitive to the variation in the distance [21]. For a resonant CNT with a sharp tip, the effect of changing the gap distance between the CNT tip and its counter-electrode on the field enhancement factor β is significant. In this study, we use the result from Bonard *et al* [22, 23]:

$$\beta = \beta_0 \times \left[1 + 0.013 \times \left(\frac{d-h}{d}\right)^{-1} - 0.033 \times \left(\frac{d-h}{d}\right)\right] \tag{2}$$

where $d - h = d - l|\cos 2\pi f t|$, $\beta_0 = 1.2 \times (2.5 + \frac{l}{r})^{0.9}$, f is the CNT resonance frequency, i.e. the applied frequency of the ac excitation source; l and r are the length and radius of the CNT, respectively, which can be directly measured inside the TEM. Taking the measurement parameters in figure 2, the β–time curve is obtained and this is presented in figure 4(b), which indicates that β is a periodic oscillating function and its value is nearly consistent with the

experimental data obtained from the FN plot. Taking into account the oscillating β, the FE current is an oscillating function versus time according to equation (1), as displayed in figure 4(c) (solid curve). In comparison to the ac coupling signal of the CNT as shown by the dotted curve in figure 4(c), the frequency of the FE current is twice that of the ac oscillating signal, in agreement with the experimental results.

The *in situ* TEM method demonstrated here has the advantage of driving the CNT very close to the counter-electrode, and the physical environment can be designed according to experimental needs, so that the oscillation component in the FE as stimulated by the ac coupling is detectable experimentally. This method allows the observation, manipulation, and measurement to be performed simultaneously, which also allows the structure and properties of individual CNTs or nanowires to be defined, providing a powerful technique for investigating the nanoscale electromechanical coupling in NEMS.

In summary, the dynamic field emission of a CNT at mechanical resonance has been studied by an *in situ* TEM using a newly home-built experimental set-up. The coupling between the field emission of a nanotube under a dc voltage applied longitudinally and its mechanical resonance stimulated transversely by an ac field shows that the frequency of the FE current is twice that of the mechanical resonance. The oscillating FE current has been explained theoretically using the Fowler–Nordheim equation. The nanoscale configuration of NEMS makes it possible to create a novel device that could correlate the mechanical behaviour with its sensitive FE, and the FE current could provide feedback signals for controlling the device. The oscillating FE current could also have potential applications in nano-oscillating circuits, low-power-loss radio-frequency filters in NMES, etc.

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