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LOW-DIMENSIONAL SYSTEMS

Frequency Characteristics of Field Electron Emission from Long Carbon Nanofilaments/Nanotubes in a Weak AC Electric Field

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Abstract—Frequency characteristics of field electron emission from long carbon nanofilaments/nanotubes in strong dc and weak ac electric fields have been investigated. A series of narrow peaks with a quality factor of up to 1100 has been discovered in the frequency range of hundreds of kilohertz. The analysis has shown that these peaks are probably associated with mechanical oscillations of the carbon nanofilaments/nanotubes driven by the ac electric field.

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1. INTRODUCTION

The ac electric field forces mechanical oscillations of carbon nanotubes owing to the interaction of the electric field with a charge induced on the nanotube [1–4]. In the regime of field electron emission, when one end of the carbon nanotube is free and appears near the anode, to which a dc voltage is applied, the mechanical oscillations of the carbon nanotube induce oscillations of the electric field at the tip of the nanotube. As a result, an ac component at the frequency of the electric field emerges in the circuit of the emission current. If this frequency coincides with the eigen frequency of the mechanical oscillations of the carbon nanotube, the amplitude of the mechanical oscillations exhibits a maximum: the resonance occurs. Simultaneously, the frequency dependence of the emission current exhibits a sharp maximum at the eigen frequency of the oscillations of the carbon nanotube. If the high-frequency voltage is modulated at a low frequency, a signal at the modulation frequency emerges in the circuit of the emission current owing to nonlinearity of the current–voltage characteristics of the field electron emission; i.e., demodulation of the high-frequency voltage occurs. In this case, one can measure the frequency characteristics at the modulation frequency, i.e., the carrier-frequency dependence of the signal at the modulation frequency.

The effect of demodulation of the high-frequency voltage owing to the mechanical oscillations of carbon nanotubes was first demonstrated by Jensen et al. [3] and studied more thoroughly by Vinsent et al. [4]. In our previous work [5], we studied the frequency characteristics of field electron emitters composed of short carbon nanotubes and discovered a series of narrow peaks in the frequency range $f = 50–1200$ MHz. In this work, we present the results of studying the frequency characteristics of long carbon nanofilaments/nanotubes. We discovered a series of narrow peaks in the frequency range of hundreds of kilohertz. The mechanism of the emergence of these peaks is discussed.

2. EXPERIMENTAL TECHNIQUES

The samples under investigation were formed by planar layers with rare long carbon nanofilaments/nanotubes grown by chemical vapor deposition of carbon onto quartz substrates covered by iron oxalate catalyst miscoparticles [6]. The samples were fabricated by the Krestinin group at the Institute of Problems of Chemical Physics, the Russian Academy of Sciences.

Figure 1 shows the microphotograph of a surface region of one of the samples. The microphotograph was made by scanning electron microscopy by Ormont at the Institute of Radio Engineering and Electronics, the Russian Academy of Sciences. One can see in the image rare nanofilaments/nanofibers with a length of up to hundreds of micrometers. According to the data of transmission electron microscopy of similar samples [6], the nanofilaments/nanotubes in the grown layers were single-wall nanotubes with a diameter of

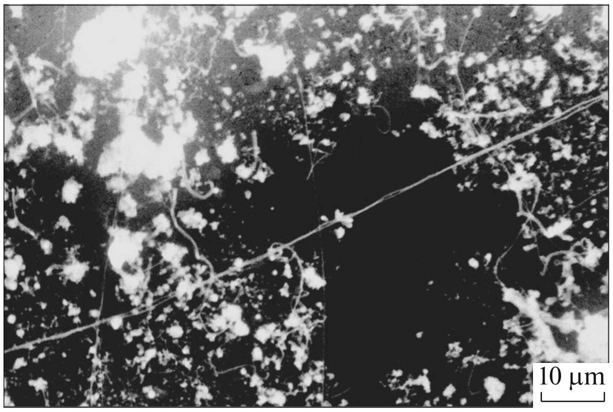

Fig. 1. Scanning electron microscopy image a region of the emitter surface with long carbon nanofilaments/nanotubes.

2–4 nm covered by a layer of disordered carbon with a diameter of 50–100 nm.

The current–voltage and frequency characteristics of carbon nanofilaments/nanotubes were studied in a high-vacuum setup at the pressure $p \sim 10^{-9}$ Torr. To measure these characteristics, a system of electrodes composed of two steel needles was fabricated and placed to the vacuum chamber. The ac electric field was applied to one of the electrodes and the other one was the anode.

The measurements of the current–voltage characteristics of the field electron emission were carried out with the use of a Keithley 248 high-voltage source and a Keithley 6485 picoammeter. The ac electric field was provided by a R&S SMB100A high-frequency signal generator. The operating range of high frequencies was bounded by a band of 100 kHz to 1 MHz. A high-frequency signal with the amplitude $V_{ac} = 1.5$ V was modulated at a frequency of 1 kHz and the modulation depth $m = 0.7$. To prevent the dc voltage and the low-frequency voltage from the generator output from coming to the sample capacitive decoupling and a system of high-pass filters were implemented. The frequency characteristics was measured at the modulation frequency with the use of the Stanford Research Systems SR830 lock-in detector connected to the sample via the system of low-pass filters. A Lecroy HDO6104 high-resolution digital oscilloscope was used to control the amplitudes and estimate frequency distortions of high-frequency signals. To perform the measurements of the amplitude–phase–frequency characteristics with data processing in complex numbers, we elaborated a LabView-based software, which swept automatically the generator frequency and synchronized data acquisition from the lock-in detector and oscilloscope. Our software/hardware setup based on a personal computer and the above devices allowed measuring the frequency spectra of carbon nanofilaments/nanotubes with a high accuracy and resolution (up to a few hertz).

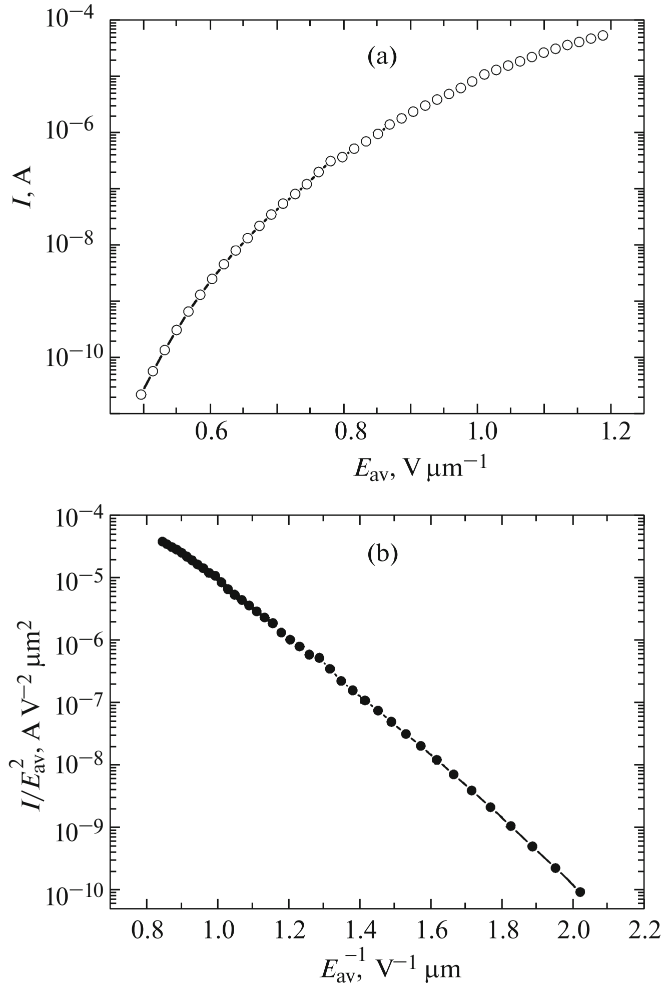

Fig. 2. Typical current–voltage characteristics of the emission current from long carbon nanofilaments/nanotubes as the (a) $\log I$–E_{av} and (b) Fowler–Nordheim plots.

3. RESULTS AND ANALYSIS

A typical current–voltage characteristics of the emission current from one of the emitters is shown in Fig. 2 as the (a) $\log I$–E_{av} and (b) Fowler–Nordheim plots. Here, I is the emission current and E_{av} is the average electric field in the anode–emitter gap. As is seen in Fig. 2, the current–voltage characteristics is linear in the Fowler–Nordheim coordinates, which indicates the field-induced electron emission mechanism of the emission current.

We studied the frequency characteristics of the emission current, i.e., the carrier-frequency dependence of the signal V_s at the modulation frequency in the circuit of the emission current. During the measurements of the frequency characteristics, the magni-

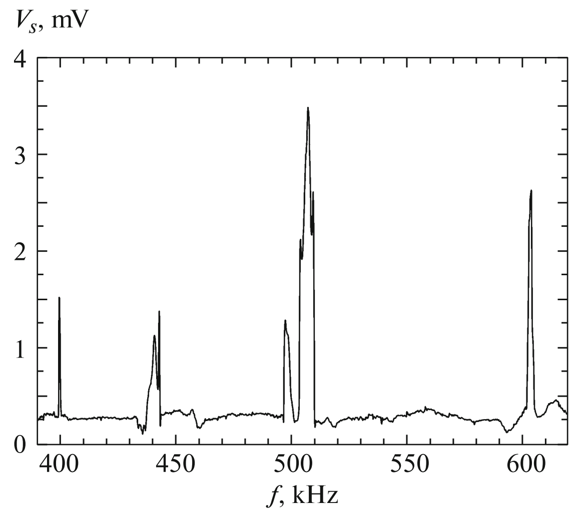

Fig. 3. Frequency characteristics of an emitter with long carbon nanofilaments/nanotubes in the frequency range of 390–620 kHz.

tude of the emission current was typically within 5–10 μA and the distance between the anode and the emitter was 200–400 μm. Figure 3 shows the frequency characteristics for one of the emitters under investigation in the frequency range of 390–620 kHz. The majority of peaks for this sample were situated within this particular region. The frequency characteristics was composed of a series of narrow peaks. During the measurements of the frequency characteristics, the dc anode voltage was $V_{dc} = 460$ V, the emission current was $I \approx 5$ μA and the ac voltage was $V_{ac} = 1.5$ V.

A large number of peaks in the frequency characteristics can be associated with resonances of the fundamental modes of mechanical oscillations of various carbon nanofilaments/nanotubes involved in the emission process. Different carbon nanofilaments/nanotubes have dissimilar parameters, primarily length, and hence the frequencies of the fundamental modes of their mechanical oscillations differ. According to [1, 7], the eigen frequency f_i of various harmonics of mechanical oscillations of a carbon nanotube is

$$f_i = \frac{\gamma_i^2}{8\pi}\frac{1}{L^2}\sqrt{(D^2 + D_1^2)}\sqrt{\frac{E_b}{\rho}}. \quad (1)$$

Here, L is the nanotube length, D and D_1 are the outer and inner diameter of the nanotube, E_b is the elastic modulus, ρ is the density of the nanotube, i is the harmonic number, $\gamma_1 = 1.875$, $\gamma_2 = 4.694$ [1, 5]. To estimate the average frequency f_1 of the fundamental harmonic of mechanical oscillations of the nanofilaments/nanotubes under investigation we take $L = 10$ μm, $D = 50$ nm, $D_1 \approx 2$ nm [6], $E_b = 10^{11}$ Pa, $\rho = 2.26 \times 10^3$ kg/m^3 [2] (the same as for graphite). Substituting these values into Eq. (1) for f_i we find $f_1 \approx 4.7 \times 10^5$ Hz. Thus, the fundamental frequency of mechanical oscillations of the nanofilaments/nanotubes with the parameters $L = 10$ μm, $D = 50$ nm, $E_b \approx 10^{11}$ Pa lies within the frequency range of the peaks observed in our experiment (Fig. 3). This coincidence confirms our assumption that the peaks seen in the frequency characteristics are associated with mechanical oscillations of carbon nanofilaments/nanotubes. The parameters of various nanofilaments/nanotubes are seemingly slightly different, which results in a large number of close peaks.

Additional information on the properties of carbon nanotubes can be gained from the analysis of the shape of the peaks near the resonance. By such analysis, we found the quality factor of the nanofilaments/nanotubes. According to [4, 7], the amplitude of driven oscillations of a tip of a carbon nanotube near the resonance is given by the formula

$$y_m = \frac{qE_{sc}}{m_{eff}\sqrt{(\omega_0^2 - \omega^2)^2 + \left(\frac{\omega_0\omega}{Q}\right)^2}}. \quad (2)$$

Here, q is a change at the tip of the carbon nanotube, E_{ac} is the ac electric field near the tip, Q is the quality factor of the nanotube, m_{eff} is the effective mass of the carbon nanotube, ω_0 is the eigen frequency of its mechanical oscillations, ω is the frequency of the electric field.

To find the Q value from the frequency characteristics, we fitted the experimental peaks $V_s(f)$ to a linear combination (sum) of Lorentzian functions with the use of the Mathcad software package. The $V_s(f)$ curve was fitted to the function

$$V_s(f) = \sum_{k=1}^{K}\left(\frac{2A_k}{\pi}\frac{\Delta f_k}{4(f - f_{0k})^2 + \Delta f_k^2}\right). \quad (3)$$

Here, K is the number of the used Lorentzian peaks, whose shapes are described by a set of independent variables (model parameters): the central frequency f_{0k}, the weight factor A_k and the width Δf_k of the kth peak.

The Levenberg–Marquardt optimization algorithm yielded the optimal values of the model parameters of the peaks in the frequency ranges $f_1 = 398.5$–401 kHz and $f_2 = 600$–607 kHz. The results of the analysis are presented in Figs. 4a and 4b, which show the resonance peaks and fitting Lorentzian functions. The resonance peak in the frequency range $f_1 = 398.5$–401 kHz (Fig. 4a) was fitted to two Lorentzian functions with the central frequencies $f_{0k} = 399.67$, 399.94 kHz, whereas the resonance peak in the frequency range $f_2 = 600$–607 kHz (Fig. 4b) was fitted to four Lorentzian functions with the central frequencies

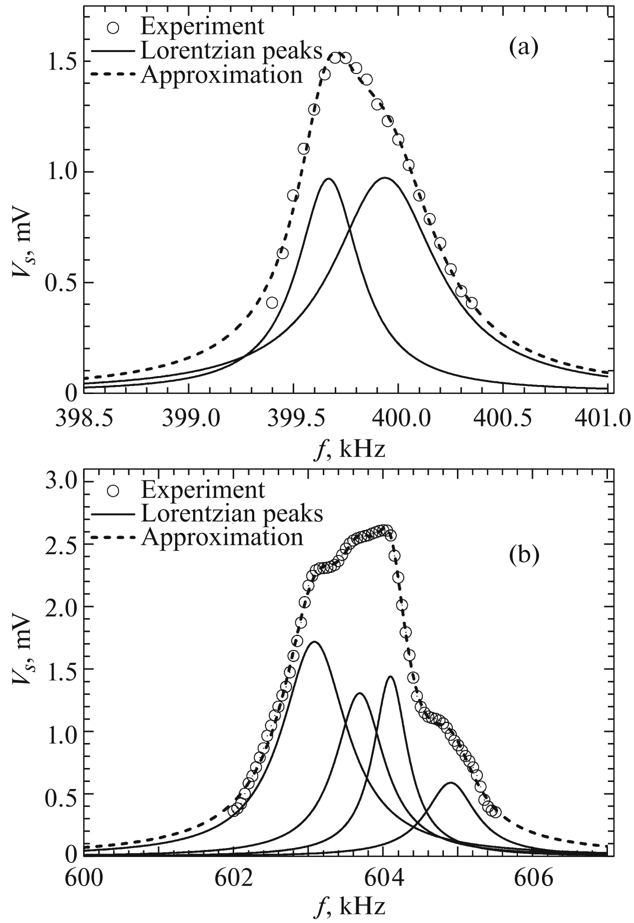

Fig. 4. Shape of two peaks in the frequency characteristics of an emitter with long carbon nanofilaments/nanotubes (see Fig. 3) in the frequency ranges (a) f_1 = 398.5–401 kHz and (b) f_2 = 600–607 kHz. The peaks are fitted to the sum of Lorentzian functions with the central frequencies (a) f_{0k} = 399.67, 399.94 kHz and (b) f_{0k} = 603.10, 603.69, 604.10, 604.90 kHz.

f_{0k} = 603.10, 603.69, 604.10, 604.90 kHz. To calculate the quality factors we used the expression $Q = f_0/\Delta f$, where Δf is the FWHM of the peak. We found Q_{1k} = [1105, 663] (Fig. 4a) and Q_{2k} = [566, 759, 1085, 742] (Fig. 4b) in the first and second cases, respectively.

Thus, the emitters under investigation contain nanofilaments/nanotubes with very close resonance frequencies and hence their resonance peaks are not resolved in the frequency characteristics and the resulting envelopes have complicated shapes, which can be fitted well to a linear combination of Lorentzian functions.

4. CONCLUSIONS

A series of narrow peaks in the frequency range of hundreds of kilohertz was discovered in the frequency characteristics of field electron emission from long carbon nanofilaments/nanotubes in the ac electric field. The analysis leads to a conclusion that these peaks are associated with a resonance of driven mechanical oscillations of individual nanofilaments/nanotubes in the ac electric field. The quality factor of the nanofilaments/nanotubes can be as high as 1100.

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REFERENCES

1. P. Poncharal, Z. L. Wang, D. Ugarte, and W. A. de Heer, Science (Washington) **283**, 1513 (1999).
2. S. T. Purcell, P. Vincent, C. Journet, and V. T. Binh. Phys. Rev. Lett. **89**, 276103 (2002).
3. K. Jensen, J. Weldon, H. Garcia, and A. Zettl, Nano Lett. **7**, 3508 (2007).
4. P. Vincent, P. Poncharal, T. Barois, S. Perisanu, V. Gouttenoire, H. Frachon, A. Lazarus, E. De Langre, E. Minoux, M. Gharles, A. Ziaei, D. Guillot, M. Choueib, A. Ayari, and S. T. Purcell, Phys. Rev. B: Condens. Matter **83**, 155446 (2011).
5. A. L. Musatov, K. R. Izrael'yants, and E. V. Blagov, JETP Lett. **99** (4), 224 (2014).
6. N. A. Kiselev, A. V. Krestinin, fli A. V. Raevskii, O. M. Zhigalina, G. I. Zvereva, M. B. Kislov, V. V. Artemov, Yu. V. Grigoriev, and J. L. Hutchinson, Carbon **44**, 2289 (2006).
7. S. P. Strelkov, *Introduction to the Theory of Oscillations* (Lan', St. Petersburg, 2005) [in Russian].

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