Uncertainty of tunnelling microscopy measurements of the field emission from multilayer nanostructures

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Abstract. The paper presents the results of scanning tunnelling microscopy investigation of electron field emission from multilayer nanostructures based on silica opal film Cr-SiO₂-Au-C. Opal film was deposited in colloidal solvent, while other films – by vacuum deposition methods. We discuss the problem of uncertainty of measurements of the field enhancement factor and carbon nano-edge sizes using the current versus applied voltage characteristics of the emission. Field enhancement factor is defined as the ratio of the electric field at the nano-edge to the field applied to the gap. We found that at well-controlled experimental conditions the combined uncertainty of field enhancement factor measurements is ~15% based on a 95% confidence interval and the combined uncertainty of nano-edge sizes measurements is ~18% based on a 95% confidence interval.

1. Introduction

The electron field emission (FE) structures (cold cathodes) can be applied in microwave, vacuum electronic and microelectronic devices, and various other devices and technologies [1,2]. The paper presents the results and metrological assurance [3] of scanning tunnelling microscopy (STM) investigation of the FE from multilayer nanostructures Cr-SiO₂-Au-C. Carbon a-C film was an emission active nanostructure. Gold film was a catalyst for the growth of carbon nanostructures. Silica SiO₂ was an opal film and had well-ordered structure. Opal film served as a matrix for formation of the topology of upper layers [4]. The Cr film provided the conductivity for the study on STM and acceptable adhesion of opal to the sitall substrate.

Opal film with diameter of silica microspheres of 220-250 nm and thickness of about 700 nm was deposited in colloidal solvent by vertical deposition method [5] on the Cr sublayer. The 100-nm thick Cr film and 80-nm-thick Au film were deposited by magnetron sputtering [6,7]. Carbon a-C structures with the thickness of about 50 nm were deposited by plasma enhanced chemical vapour deposition (PECVD). The Au and a-C films thicknesses were controlled on a witness sample – a sitall substrate. All films thicknesses were obtained by measuring the step height using atomic force microscopy (AFM). A schematic representation of the sample structure is shown in Figure 1.

In this study, we determine the uncertainties of measurements of the FE enhancement factor or gain coefficient β and carbon nano-edges radius r using the current I versus applied voltage V characteristics of the emission [8]. Factor β is defined as the ratio of the electric field at the nano-edge to the one applied...
Figure 1. Scheme of the Cr-SiO$_2$-Au-C structure on the sitall substrate.

to the gap. A large $\beta$ factor allows emitters to operate at a small applied voltage. To achieve a low turn-on field and a high current density, field emitter materials should preferably have a low work function (or electron affinity) and be nanostructured with needle-like edges to enhance significantly the field at the edges [9]. While the factor $\beta$ is inversely proportional to the radius $r$, it is important to make the edges as sharp as possible to reduce the applied external field required for turning on the electron emission [10,11].

2. Method description

The samples were studied using scanning probe microscope (SPM) Solver P-47 (NT-MDT, Russia) by STM in topographic mode by direct current method and current spectroscopy mode. Measurements were operated at a controlled temperature and humidity of 293 K and 50 %, respectively. The SPM was set on a vibration isolation system. Minimum step STM-scanning was 0.006 nm. During scanning, the voltage varied from 0.1 to 1.0 V. It was found that the relief image of multilayer nanostructures on the metal sublayer is dependent on the tunnel voltage generated in the gap as shown in the Figure 2.

This effect was most likely not related to mass transfer and was explained by the presence of emission processes. In this way, three regimes can be defined in the current $I$ versus applied voltage $V$ in a Cr-SiO$_2$-Au-C structure: (1) tunnelling regime, when the applied voltage is lower than the work function, (2) the intermediate regime, and (3) the field emission or Fowler–Nordheim regime for higher voltages.

Figure 2. The STM-images of multilayer nanostructures Cr-SiO$_2$-Au-C for various regimes of the $I$-$V$ curve: (a) tunnelling regime, 0.1 V, (b) FE regime, 1.0 V.
We measured the current-voltage characteristics of the structures and analysed them by means of a Fowler-Nordheim (FN) plot. FN theory [12] describes the electric field induced electron tunnelling from a flat perfectly conducting planar surface through a potential-energy barrier. FN plot represents $\ln(I/E_0^2)$ on $E_0^{-1}$ dependence, where $I$ is a current and $E_0$ is a macroscopic field. The field enhancement factor $\beta$ can be deduced as $E/E_0$, where $E$ is the surface field of the emitter. The curve slope $\tan \alpha$ of $\ln(I/E_0^2)$ versus $E_0^{-1}$ gives the field enhancement factor [13], which allows calculating the characteristic size of carbon nano-edge radius.

The field enhancement factor $\beta$ is expressed as a function of the electron work function of material $\varphi$ and $\tan \alpha$:

$$\beta = \frac{B \varphi^{3/2} \ln e}{\tan \alpha},$$  

(1)

where $B$ is coefficient.

In certain cases, the field enhancement factor $\beta$ can be expressed as:

$$\beta = \frac{1}{\sqrt{2Rr}}$$  

(2)

where $R$ is the distance between nano-edge and the probe. Consequently, nano-edge radius $r$ is expressed as a function of the electron work function $\varphi$, the slope $\tan \alpha$, and the distance $R$:

$$r = \frac{R \tan^2 \alpha}{2B^2 \varphi^{3/2} \ln^2 e}$$  

(3)

In this case, equations (1) and (3) are the measurement models [14]. According to the Guide to the Expression of Uncertainty in Measurement (GUM) [15], a measurement results in a value of a well-defined physical quantity – the measurand – and a corresponding uncertainty, which characterizes the dispersion of the values attributed to the measurand [15]. It is widely recognized that any measurement result has an uncertainty range, within which the true value may be found with a certain probability [16]. The measurement uncertainty is determined by identifying, assessing, and combining all the sources of uncertainty associated with the quantity value.

We identified two types of uncertainty evaluations for inputs [15]. For equation (1) of type B, evaluation includes uncertainty in the electron work function $\varphi$. For equation (3) of type B, evaluation includes uncertainties in the electron work function $\varphi$ and the distance between nano-edge and the probe $R$. For both equations (1) and (3) of type A, evaluation is derived from repeated observations of the slope $\tan \alpha$. Based on these uncertainty evaluations, an uncertainty calculation scheme [15] is developed in this work.

3. Results

Fifteen observations of the current $I$ versus applied voltage $V$ characteristics were carried out for various dots on three samples. Typical $I$-$V$ curve is shown in Figure 3. The FE regime was observed for higher applied voltage.

The relative combined standard uncertainty $U_{rc}$ was calculated by collecting all aforementioned uncertainties. Finally, the relative expanded uncertainty $U_{exp}$ for the coverage factor $k$ at the confidence level of 95% was obtained as follows:

$$U_{exp} = kU_{rc} = 2U_{rc}$$  

(4)

To investigate the influence of the inputs on the uncertainty of the measurement, uncertainty budgets for the factor $\beta$ and the nano-edges radius $r$ were calculated. The uncertainties of the contributing variables for the factor $\beta$ are showed in Table 1. When the factor $\beta$ is 19.30 and the radius $r$ is 1.85 nm, the total results are reported in Table 2.
Figure 3. I-V curve of the Cr-SiO₂-Au-C structure on the sitall substrate.

Table 1. Uncertainty analysis.

| Uncertainty evaluations type | Probability distribution | Degrees of freedom | Relative standard uncertainty (%) |
|------------------------------|--------------------------|--------------------|-----------------------------------|
| φ B                          | normal                   | ∞                  | 2.2                               |
| tan α A                     | normal                   | 14                 | 8.4                               |

Table 2. Uncertainties of the factor β and the nano-edge radius r measurements.

| Measurand | The relative combined standard uncertainty (%) | The effective degrees of freedom | The coverage factor | The relative expanded uncertainty (%) |
|-----------|-----------------------------------------------|---------------------------------|---------------------|--------------------------------------|
| β         | 7.4                                           | ∞                               | 2                   | 14.8                                 |
| r         | 9.1                                           | ∞                               | 2                   | 18.2                                 |

Examination of tan α measurements shows remarkable influence of this contamination on the detected factor β and radius r.

4. Discussion
The reproducibility of the samples properties has the largest influence on the uncertainty of the I-V curve and tan α. Multilayer film deposition is a very complex process. Study of films growth kinetics revealed that many factors were critical for reproducible deposition of the structure.

A fine tuning of structure properties was difficult, because growth parameters were very interdependent. It was shown that the opal film packing depended significantly on substrate pretreatment, roughness of Cr film, colloid properties, and lifting velocity. The Au precursor resulting roughness depended on packing density of microspheres and was a dominant factor affecting a-C film growth. So it was hard to repeat structure growth process for several samples and various dots on the samples surface and reproducibility of the samples properties sometimes was not sufficient. More precise technologies and equipment could be further employed to reduce the uncertainty.
5. Conclusion
In this paper, the procedure and analysis of the uncertainty of tunnelling microscopy measurements of the electron field emission from multilayer nanostructures Cr-SiO$_2$-Au-C were given. The relative expanded uncertainties ($k=2$) of field enhancement factor measurements and of nano-edge sizes measurements were $\sim 15\%$ and $\sim 18\%$, respectively. The result of our estimation of uncertainty is associated with determination of the factors influencing on the detected factor $\beta$ and radius $r$ in the measurement models (1) and (3). The standard uncertainty of the tan$\alpha$ of a Fowler-Nordheim plot has the largest influence on the measurement results uncertainty. The tan$\alpha$ uncertainty is strongly influenced by the reproducibility of the sample properties. Thereby, the phenomenon of uncertainty of STM measurements of factor $\beta$ and radius $r$ is mainly related to the reproducibility of the film deposition process and unevenness of multilayer structure. The main application of this methodology is laboratory testing of the emission nanostructures.

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