Ultra-low frequency spectra for Li$^+$ ion thermoemission noise

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The surface diffusion of Li on polycrystalline tungsten and ionic thermal desorption are studied by a method based on the Li ion current noise arising from the fluctuation of the work function as a result of random fluctuations of the Li adsorbate density. The activation energy for surface diffusion of Li has been determined by measuring the spectral density functions and their parameters depending on temperature.

1. Introduction

This paper is a continuation of the author's earlier studies [1] − [3] in which a model for generating ion thermoemission current noises was described.

According to the model used, fluctuations of ionic current intensity are formed due to the fluctuations of local work function of ion emitter lead. These fluctuations result from the surface diffusion of adsorbate and the statistical process of ionic and atomic desorption processes.

The mean square of the ion current fluctuations is given by [3]:

$$\langle \mathrm{d}i^2 \rangle = i_0^2 \left(\frac{\pi e \mu n}{\varepsilon_0 k l} \right)^2 T^2 D \tau_0 \tag{1}$$

where i_0 is the DC ion current, μ − the dipole moment of the adsorbed atoms, ε_0 − the permittivity of vacuum, l − the geometrical parameter of the tungsten ribbon, D − the diffusion coefficient, n − the concentration of atoms adsorbed on tungsten emitter, k − the Boltzmann's constant, T − the temperature of emitter, and τ_0 − the mean residence time of lithium atoms on tungsten, measured by computer fitting the spectral density function to the experimental data.

The operation of the thermal emission ion source is based on the fact that when atoms impinge onto a hot metallic surface, some of them are evaporated as positive ions.

Our earlier studies of spectral density of fluctuation (noise) of ion currents emitted thermally from clean tungsten surfaces showed that the spectral course is of the Lorentzian character in the frequency range from 10 Hz to 10 kHz

$$S(f) = \frac{S_0}{1 + (\omega \tau_0)^2} \tag{2}$$

where $\omega = 2\pi f$, but τ_0 is the average life-time of atoms in the state of adsorption on the tungsten surface.

Such a function form indicates that in a wide range of frequencies at $\omega\tau_0 \ll 1$ function $S = S_0 = \text{const.}$ At $\omega\tau_0 \gg 1$, when value of f increases to infinity, the spectrum drops according to the function $S = a/f^2$ reaching the shot noise level of no interest $S = 2ei_0$, where i_0 is the average value of ion current intensity, *i.e.*, the so-called constant intensity component. The turnover (3 dB) frequency is: $f_0 = 1/(2\pi\tau_0)$.

We determine parameter τ_0 by fitting the $S(f)$ curves to the experimental data.

2. Experimental

The measurements were performed in a stainless steel vacuum chamber allowing a high vacuum of 10^{-9} torr. The ion current fluctuations were amplified by a wide-band DC electrometer. The spectral density functions were investigated with the Unipan-237 selective nanovoltmeter with relative selectivity $\Delta f/f = 0.014$.

For very small frequencies (important in the case of emission from oxidized tungsten) we applied the method of sampling and computer calculation of the spectral density function.

The lower range of frequencies of the spectrum under investigation is determined from the measurement time, *i.e.*, the length of the sample from the statistical process. However, the measurement time is limited by the finite amount of the element deposited on the emitter which while evaporating slowly makes the emission process non-stationary. For such a process procedures of statistical studies are quite complex.

In our case a non-stationary character appeared in the form of exponential, slow decrease in ion current intensity and level of spectral power density

$$S = A \exp\left[\frac{-t}{\tau}\right]. \tag{3}$$

For the temperature range from 1200 K to 2100 K the time constant τ was from 5 to 2 hours, respectively.

We applied the method for removal of the trend of non-stationary condition by computer subtraction of the exponential function earlier entered into the actual process. As a result, for the Fourier analysis the pure statistical process was left with the average zero value and the Gaussian distribution of the probability of occurrence of fluctuation with a given amplitude.

3. Results

Our investigations show that the way in which atoms are supplied on the ionizing surface is not essential. This can be done by filling the ionizer surrounding with vapours of the studied element. It is also possible to direct a colimated stream of

atoms or spread them earlier by means of capillary. Shapes of statistical functions and their parameters are always the same. The only difference is that in the case of macroscopic, manual covering of the metal surface we do not know much about work function of the adsorption system: metal (wolfram) with the macroscopic layer of ionized and desorbed atoms covering it.

The way of preparing stable emitters in the form of oxidized tungsten with the work function $e\varphi = 6.3$ eV was described in paper [4].

The applied clean tungsten emitters were characterized by dominant texture (001) with the work function of 4.61 ± 0.05 eV [5].

3.1. Emission of Li ions from clean tungsten

Spectral density function for the wide range of temperatures is of the Lorentzian form (Fig. 1).

The value of atom life-time in the adsorption state read from the Lorentzians computer inscribed between measuring points changes exponentially with the temperature of the emitter

$$\tau_0 = \tau_0^0 \exp\left[\frac{Q}{kT}\right]. \tag{4}$$

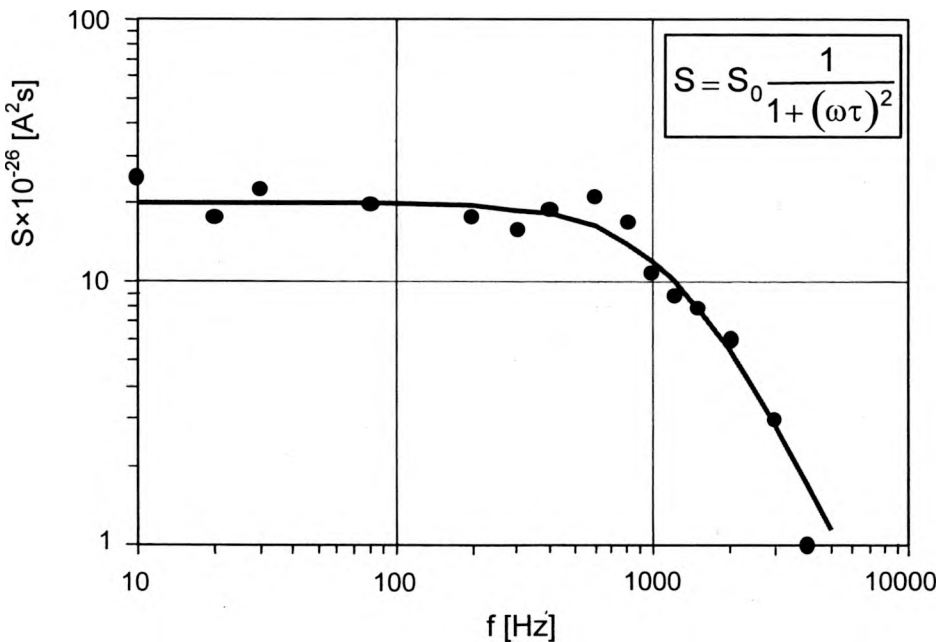The atomic desorption energy Q was calculated from these relations.

Fig. 1. Spectral density function for Li on tungsten ($T = 1980$ K, clean tungsten).

Though ions are a source of information about desorption, due to high ionization potential of Li ($V = 5.39$ volt) the lithium ionization coefficient is small and the atomic processes on the surface decide about fluctuation phenomena. Thus the calculated value $Q = 2.35 \pm 0.05$ eV is the heat of desorption of lithium atoms but not ions.

The normalized mean square fluctuation $m = \langle \mathrm{d}i^2 \rangle T^2 / i_0^2 \tau_0 \sim \exp[-E/kT]$ was applied to evaluate the dependence of the diffusion coefficient D on temperature, whereas from the slope $\log m$ versus $1/T$ the value for Li equal to $E = 0.60 \pm 0.02$ eV was found.

3.2. Emission of Li ions from oxidized tungsten

The spectrum of "$1/f^\varepsilon$"-type with the average slope $\varepsilon = 0.93 \pm 0.07$ was observed in the very wide range of frequencies from 0.01 Hz to 2 kHz (see Fig. 2).

A detailed analysis of the spectrum will be made using the model of the "$1/f$"-type spectrum as the sum of Lorentzians in which a continuous distribution of time constants occurs

$$S(f) = a \int_0^\infty \frac{1}{1 + (\omega \tau_0)^2} \, g(\tau) \mathrm{d}\tau$$

where $g(\tau) = \ln[\tau_2/\tau_1]^{-1} \tau^{-1}$.

Using this time-constant g distribution, interpreted as the average life-time of atoms on the complex, oxidized tungsten surface we obtain

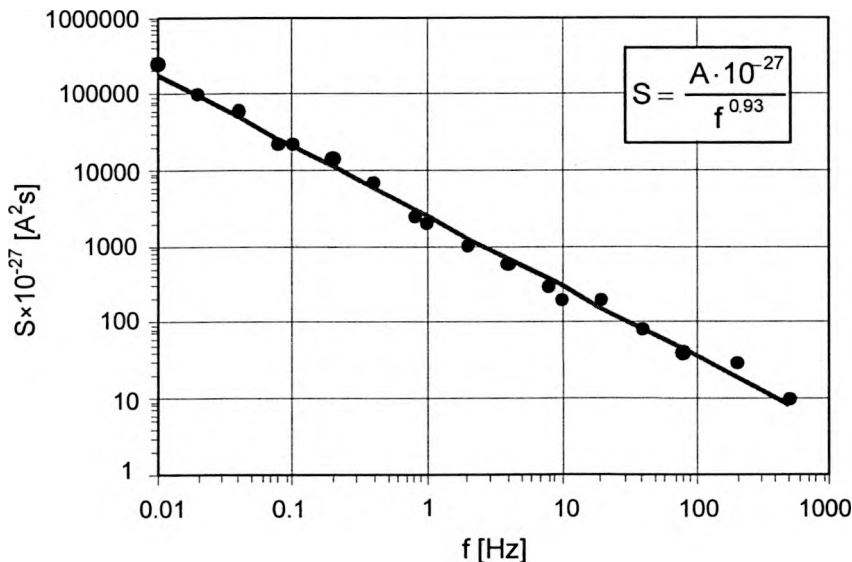$$S(f) = a \left[\ln \frac{\tau_2}{\tau_1} \right]^{-1} f^{-1}. \tag{5}$$

Fig. 2. Spectral density function for Li on oxidized tungsten ($T = 1230$ K).

A very wide range of life-time of adsorbed atoms is required to reproduce the whole spectrum. Our measurements were limited on the side of the smallest frequencies 0.01 Hz by prolonged measuring time and the influence of non -stationary condition. The measurable values of spectral density reached 2 kHz. Such a range of spectrum requires assuming the value range of τ: $10^{-4}\,\text{s} < \tau < 10\,\text{s}$.

This wide time range could be real if the desorption energies Q related to dependence (4) occupied the range ΔQ

$$\frac{\tau_2}{\tau_1} = \exp\left[\frac{\Delta Q}{kT}\right], \quad \Delta Q = 1.2 \text{ eV.}$$

This is the real value for the complex surface of oxidized tungsten.

Lithium has the smallest ionic and atomic radius of the elements whose emission can be studied using the described methods ($R_i = 0.60$ Å, $R_a = 1.55$ Å, respectively). It was assumed that lithium atoms and ions diffusing on the surface of oxidized tungsten will undergo interactions of various energy values. This is confirmed by a relatively high value of noise amplitude and a large range of the value ΔQ.

References

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