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Registration of the time dependence of the number of emission sites as a tool for analyzing field cathodes current fluctuations

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A new tool for obtaining the frequency characteristics of the interaction of emission sites with adsorbates was developed. The technique is based on the use of a computerized field projector with online processing of patterns of the distribution of emission sites over the cathode surface. The resulting dependences reflect the frequency of fluctuations in the activity of individual sites. The influence of the choice of the threshold brightness parameter, the proximity of the sites to each other and the stability of the total emission current on the shape of these curves is considered. The coincidence of the shapes of curves constructed by various methods (in online mode, in post-processing mode and in the mode of random processes modeling) is analyzed.

Keywords: field emission, emission current fluctuations, carbon nanotubes, registration of emission sites, adsorption processes on the surface.

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1. Introduction

Free electron sources are an integral part of modern vacuum electronics. Multi-tip nanostructured field cathodes based on the effect of field emission are currently the most promising. Conductive nanosized particles make it possible to distribute the current load over the cathode surface thus increasing the total current per unit area. At the same time, a high degree of the electric field focusing at the tops of these structures significantly decreases the threshold voltages. The best results in this regard are demonstrated by carbon nanotubes [1]. Their attachment on the cathode surface using a polymer matrix (the use of nanocomposites) makes it possible to simplify the technology, and at the same time increases the characteristics stability [2].

The main methods for properties assessment of field cathodes are: registration of current-voltage characteristics (IVC) with the calculation of effective emission parameters (field enhancement factor γ_{eff} and emission area A_{eff}) [3], registration and assessment of the current stability level at constant voltage over a long time period (time test) [4], registration of the glow sites distribution over the cathode surface (glow patterns on the luminescent anode [5], the emitter scanning with an anode-needle or anode with a hole — field emission scanning microscope [6], as well as observation of microscopic emission sites in a Mueller field microscope [7,8]).

Registration and analysis of the IVC hysteresis [9,10], calculation of the effective work of the output [11], stability analysis of the current level during voltage surges (transient processes) [9,12], registration of mass spectrometric data on composition of volatile products in the interelectrode space (source of adsorbates and ion current) [13], registration of distribution spectra of free electron energy [14], as well as registration and analysis of emission current fluctuations [15,16].

As an example of rare methods, one can name the registration of light and infrared radiation of field emitters [17,18], registration of the emission sites distribution using a resistive anode coating [19], as well as registration of threshold triggering voltages as individual emission sites burn out on the glow pattern [20].

This paper will present a new method for fluctuations registration of the emission activity of multi-tip field cathodes.

The study of fluctuations in the emission current is associated with the determination of the reasons of the field cathodes instability at high currents. The most common cause of these fluctuations is the particles of the residual atmosphere in the interelectrode gap, which, being adsorbed on nanosize emission sites, lead to a change in their emission current [7,21,22]. The adsorption fluctuations can disappear if current increases [22].

The method presented by us is based on the analysis of the so-called „collection curves“. It was obtained as a result of the development of another, more complex method, which is aimed at registration and online analysis of the characteristics of individual emission sites on the surface of multi-tip field cathode [23–25].

2. Features of plotting the collection curve in stable mode

The study of individual sites is based on the use of a computerized field projector with a luminiferous anode, on

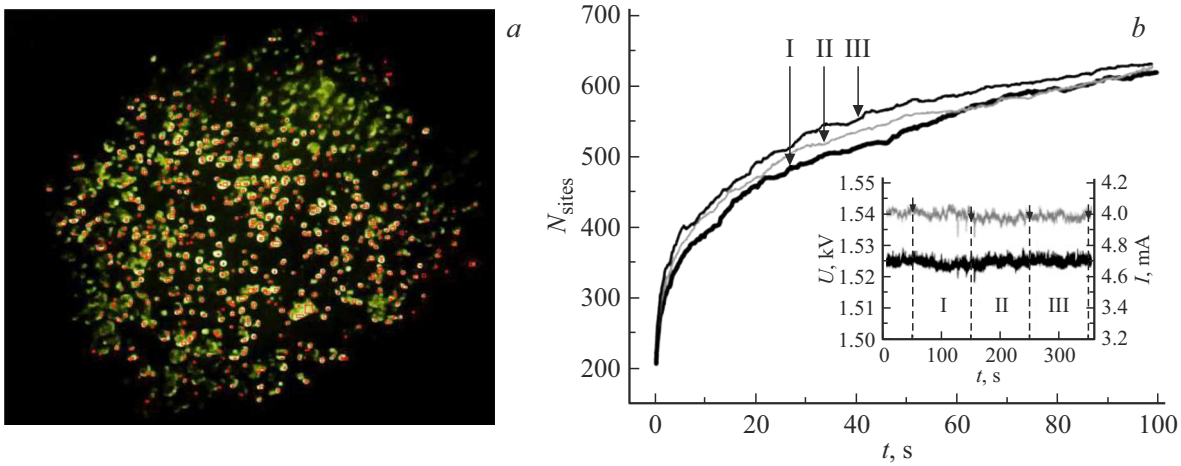

Figure 1. Determination of the emission sites location: *a*) glow pattern with superimposed fluctuation zones of individual emission sites (red squares), *b*) three collection curves — are the time characteristics of number of registered emission sites obtained for three consecutive time intervals I, II, III (the inset shows the time relationship of the levels of the total emission current and voltage). Parameter $Y_{th} = 150$.

which flows of electrons form the glow patterns (analog is the method developed by Kopelevsky [5]). Each of the registered glow patterns is processed in real time. The information is gathered in several stages: 1) position registration of the emission sites, 2) registration of the current load on each emission site (the sites brightness is used as a weighting factor in the total current distribution over the sites), 3) obtaining the maximum values of the local current load at a stable voltage level, 4) evaluating the uniformity of the emission sites distribution in space and with by the current loads, 5) plotting a histogram of the local field enhancement factors at a fixed emission area, 6) plotting local current-voltage characteristics and assessment of the corresponding effective parameters, 7) plotting group current characteristics (the sum of the currents of a group of sites), which allow analyzing the effect of the value of the field enhancement factor of sites field on their interaction with adsorbates and participation in transient processes associated with adsorption. A software tool, that implements these procedures, including multichannel registration of experimental data streams and automatic recording of research results into files, was created using the LabVIEW graphical programming platform. In addition to it, a complex was developed for recording and replaying the experiment in the emulation mode [26].

All of the above procedures are based on the results of information collection on the location and number of emission sites. This collection is complicated by the fact that the images of the emission sites on the luminescent screen continuously fluctuate due to stochastic adsorption processes (they go out one second, they go up the next due to the precipitation and release of adsorbates, they shift next due to the adsorbate movement over the surface) [7,8,27].

The developed technique for finding the sites is based on the superimposition of patterns of the brightness peaks location (white pixels on black background) found for each frame. The emission sites position is calculated by automatically combination of adjacent pixels on the resulting collection diagram into rectangular zones (Fig. 1, *a*). The number of such zones is determined online, and time characteristic of this number is plotted (collection curve). When the relationship curve reaches a relatively horizontal level, the emission sites search can be considered completed.

When collection emission sites of various field cathodes (with an emission surface made of carbon nanotubes, graphene, silicon nanostructures, etc.), we noticed the relative constant shape of the collection curve, which indicates the link between this relationship and physical processes occurring on the cathode surface (see Fig. 1, *b*).

The key feature of the experimental collection curve is the threshold brightness level Y_{th}, which is set by the researchers in the program and automatically cuts off digital noise and various glare effects such as the halo effects [28]. These effects lead to the appearance of false emission sites in the black-and-white diagram, as well as to displacement and merging of images of real sites. At the same time, some of the low-current sites with low brightness are also cut off. Thus, Y_{th} value is selected according to two criteria: registration of the maximum number of sites and at the same time obtaining the minimum number of overlaps of the fluctuation zones.

Note that the zones merging in the diagram („the effect of sites merging“) can also be useful if the same site appears on the screen in different pixels (for example, due to the adsorbate movement over the emitter surface or due to the position change of the emitter itself, if it has mobility). On the collection curve the effect of merging is reflected as the number of sites found decreasing.

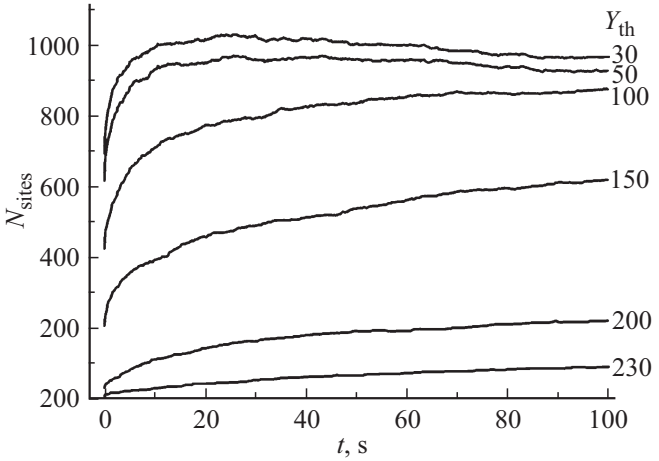

Figure 2. Gathering curves for values $Y_{th} = 30$, 50, 100, 150, 200, 230.

Fig. 2 shows the collection curves obtained for different levels of threshold brightness at the same time interval (section I in Fig. 1, *b*). The curve with $Y_{th} = 50$ starts to fall down over time due to the „effect of sites merging", and the curve with $Y_{th} = 230$ does not fall almost everywhere. Note that the more emission sites are recorded, the smoother the collection curve is.

3. Unstable regime effect on the shape of collection curve

Long-term studies of multipoint field cathodes reveal another problem of collection and analyzing the emission sites — the instability of the emission surface. First of all, it is associated with adsorption-desorption processes, and secondly, — with microscopic vacuum discharges, which can deform the cathode surface: the discharge can destroy the highest emission sites and at the same time generate new sites, or by cleaning the surface of adsorbates, or by adding new protrusions to the surface, for example, by an explosion. The collection curve can reflect four types of emitting surface instability. The first type is observed at the very beginning of the emission sites collection and is reflected in the form of an exponential relationship (section *1* in Fig. 3). It is associated with fast fluctuations in the brightness of the sites.

In a stable mode of cathode operation the shape of the collection curve shall have exactly this form with an exit to a relatively horizontal level. However, if the characteristic on-off times of the sites have a rather wide scattering, the curve can continue its exponential growth indefinitely.

The second type of instability leads to the presence of an almost linear slope in the shape of the collection curve, which becomes noticeable after its exponential growth (section *2* in Fig. 3). It is associated with slow brightness fluctuations of the sites, which can be caused by adsorption processes with a long lifetime of a particle on the emission site.

If the turn-on speed of the sites is equal to the turn-off speed, then the total current remains constant (as shown in Fig. 3, *b*). Note that if the total current increases, the cathode is unstable, and new sites turn on faster than old ones turn out (this is especially noticeable during transient processes [25]).

The third type of instability — is a change in the growth rate of the number of sites, which can be associated with change in vacuum conditions, with small changes in the level of the applied voltage, as well as with barely noticeable (not affecting the total current level) abrupt processes on the emitter surface, when whole groups of emission sites burn out or abruptly activate. On the collection curve these processes are reflected in the form of kinks (point *3* in Fig. 3).

The fourth type of instability is associated with strong vacuum discharges, which are visible on the time characteristic of total current. In this case, the emitter surface undergoes a

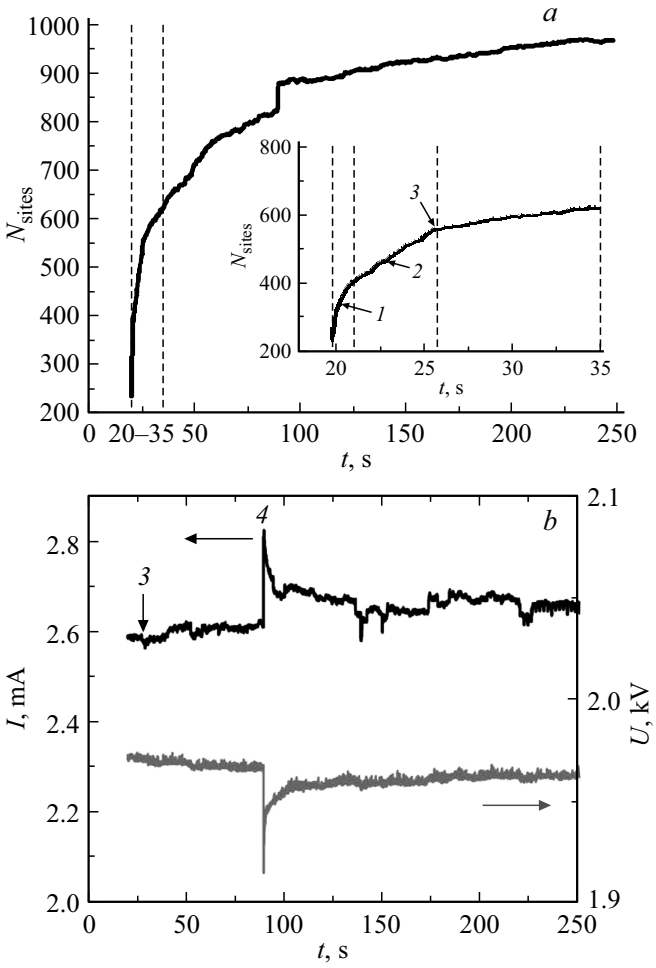

Figure 3. *a*) Different types of sections on the collection curve (description of sections (1)–(4) in the text). *b*) Time characteristic of current with a characteristic peak of a vacuum discharge.

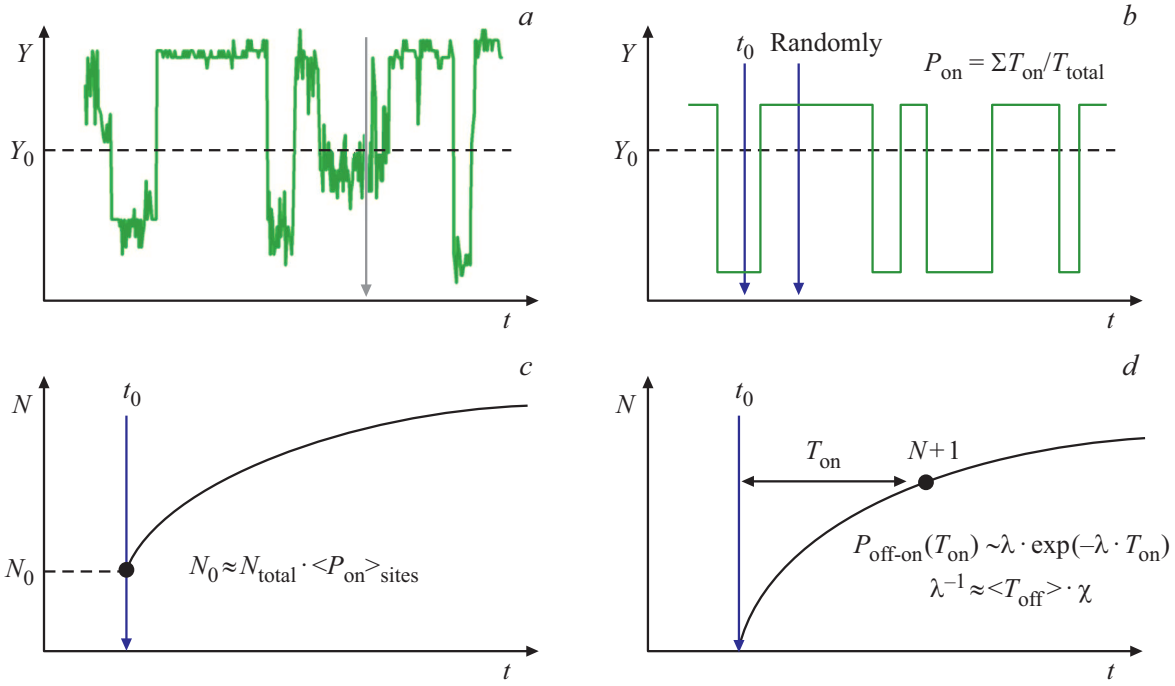

Figure 4. A number of assumptions allowing to link the experimental collection curve to the ideal curve at a given threshold brightness Y_0: *a*) experimental time characteristic of brightness of one emission site; *b*) „on/off" idealization of two states of the site (t_0 — is the moment of the sites collection beginning); *c*) the contribution of the sites, which were in the state „on" at the moment t_0, to the initial value of the collection curve N_0; *d*) the contribution of the sites that were „off" at the time t_0 to the shape of the collection curve at the time they were turned on. All formulas in the graphs are described in the text.

significant rearrangement: a large number of new emission sites appear simultaneously (point *4* in Fig. 3).

4. Collection curve simulation

To get an idea of the collection curve nature, we developed a theoretical model based on stochastic adsorption-desorption processes. The model considers adsorbates that increase the output work of electrons of emission sites (for example, oxygen on CNTs [29,30]).

Fig. 4, *a* shows the experimental time relationship of one of the emission sites, recorded during the experiment, which demonstrates the characteristic jumps in the site activity associated with the adsorption-desorption process. The transition from a real system to a model one is carried out through a number of assumptions.

The first assumption is that fluctuations in the brightness value Y of each emission site have „on/off" nature relative to the general threshold brightness level Y_0 established by the researcher. I.e., each emission site can exist exclusively in one of two states: „on" — without adsorbate on the surface, when $Y = Y_{on} > Y_0$, and „off" — with adsorbate when $Y = Y_{off} < Y_0$ (Fig. 4, *b*).

There is an objection to this statement associated with the fact that there can be several adsorbates on one emission site; then the number of states increases by several times. Moreover, the experimental system has a resolution limit and can take several closely located sites as one (a bundle of nanotubes, for example).

The second assumption is that at the moment when the collection of emission sites starts, the number of sites in the state „on" (the starting value of the collection curve N_0) is related to the probability of each site to be in the state „on" and can be expressed as the sum of binary random values $X_{on/off}$, taking the value 0 or 1 (Fig. 4, *c*):

$$N_0 = \Sigma X_{on/off}. \quad (1)$$

Experimentally, for each site, the probability of being in the state „on" (P_{on}) at time t_0 can be estimated as the ratio of the sum of the times when the site is in the state „on" to the total site tracking time (i. e. fill factor) (Fig. 4, *b*):

$$P_{on} = \Sigma T_{on}/T_{total}. \quad (2)$$

This is approximately equal to the ratio of the corresponding time-average values

$$P_{on} \approx \langle T_{on}\rangle/(\langle T_{on}\rangle + \langle T_{off}\rangle). \quad (3)$$

If the starting number of found sites N_0 is estimated as the sum of math. expectations $M(X_{on/off})$, then it will be proportional to the fill factor averaged over the number of sites ($\langle P_{on}\rangle_{sites}$) (share of the total number of sites N_{total}):

$$N_0 \approx \Sigma M(X_{on/off}) = \Sigma P_{on} = N_{total} \cdot \langle P_{on}\rangle_{sites}. \quad (4)$$

Thus, the starting value of the collection curve N_0 shall be related to the value $\langle T_{\text{on}}\rangle/T_{\text{total}}$, averaged over sites.

The third assumption says that the probability of the emission site transition from the state „off" to the state „on" is described by some exponential law, related to the exponential distribution (Fig. 4, *d*):

$$P(T_{\text{off}\to\text{on}}) = \lambda \cdot \exp(-\lambda \cdot T_{\text{off}\to\text{on}}), \quad (5)$$

where λ — is parameter associated with the structure of the emission site and its ability to interact with adsorbates. Note that exponential relationship is a standard physical principle that is used to describe random desorption processes [31].

The turn-on time $T_{\text{off}\to\text{on}}$ of each of the sites, which were in the state „off" at the moment of the collection beginning, can be compared to the mathematical expectation of the turn-on time $\text{M}(T_{\text{off}\to\text{on}})$, which according to the exponential distribution:

$$\text{M}(T_{\text{off}\to\text{on}}) = \lambda^{-1}. \quad (6)$$

By experiment the mathematical expectation $\text{M}(T_{\text{off}\to\text{on}})$ can be estimated as the average time when the site is in the state „off":

$$\text{M}(T_{\text{off}\to\text{on}}) \approx \langle T_{\text{off}}\rangle \cdot \chi. \quad (7)$$

The additional factor χ is associated with possible changes in the conditions of emission sites interaction with adsorbates: heating of the cathode material, change in the ions concentration in the interelectrode space, etc.

The collection curve — is the time dependence of the number of activated emission sites, which is the integral of the sites distribution over time $T_{\text{off}\to\text{on}}$. At a sufficiently large set of sites, the shape of the real curve shall be close to the shape of the sites distribution function by mean time $\text{M}(T_{\text{off}\to\text{on}})$.

The fourth assumption concerns fast fluctuations in the brightness of the sites (see noise at the levels „on" and „off" in Fig. 4, *a*), which have a significant effect on the shape of the collection curve. They can be associated with halo effects (reflections of neighboring emission sites in the glow pattern) [28], with the adsorbates movement along the tops of the sites [8], with change in the sites shape (for example, elastic vibrations of nanotubes [32]), with conductivity fluctuations (in nanotubes it can be caused by the presence of a two-level tunneling system or the percolation effect in conductivity [33]), with voltage fluctuations in the supply network, etc. The assumption assumes that all these fluctuations can be considered by choosing a random value χ, which should change the idealized turn-on time of each site $T_{\text{off}\to\text{on}}$.

Fifth assumption is that the effect of sites merging in the process of plotting the collection curve in real time does not lead to significant change in the curve shape, leaving it within the limits of natural (adsorption) fluctuations. An objection to this statement is the strong dependence of this effect on the value of the threshold brightness, which cannot be chosen unambiguously.

These five assumptions allow the actual collection curve comparison with the characteristics of the model emission system. It follows from them that the shape of the collection curve reflects the emission sites distribution by the average $\langle T_{\text{off}}\rangle$, and its starting value is related to the value of $\langle T_{\text{on}}\rangle/T_{\text{total}}$.

5. Theory and experiment comparison

The shape of the experimental collection curve is affected by two „parasitic" effects: the effect of sites merging on the gathering diagram (error of the registration system) and the effect of stochasticity of sites switching on-off (randomness of adsorption processes). We used the experiment recording and multiple emulation tool to plot the gathering curve with minimal effect of these effects [26]. In the first pass the optimal threshold brightness was selected, in the second pass, emission sites were found (here the merging effect occurred), in the third pass — the brightness of these sites as function of time was obtained (there is no merging effect in these functions, since we monitor the already found zones). Based on the results of the third pass the collection curves were plotted (generally speaking, model ones, since they were plotted using the data post-processing method). Fig. 5, *a* shows the time characteristic of the number of sites *N*, whose brightness at least once exceeded the threshold brightness by the time $T_{\text{off}\to\text{on}}$ (curve Model 1). The gray lines show the ranges of curve variation at a cyclical shift of the start time of the gathering, as well as its average.

Due to the absence of the merging effect, it does not coincide in some places with the experimental curve and passes under it. Note that for other, lower threshold brightness, the discrepancy increases significantly.

To understand how much this model collection curve can change its shape when the time of the sites collection beginning changes, we looped the corresponding time interval (300 s of the experiment) and plotted all possible collection curves for this cyclic interval (without changing the length of the collection curve over time, but displacing its beginning). Fig. 5, *a* shows the maximum and minimum values of this variety, as well as its mean, which has almost no stochastic form. The experimental collection curve was within this range, i.e. its deviation from the Model 1 curve associated with the merging effect is within the natural stochasticity of the process.

The inset in Fig. 5, *a* shows the histograms of the distribution of emission sites over the on-time $\Delta N(T_{\text{off}\to\text{on}})$, plotted from the data of experimental and model collection curves (see Fig. 5, *a* and *b*). In general, the relationship has a descending exponential form.

To get rid of the stochasticity effect, we plotted another type of model curve. In it, we used the mean periods $\langle T_{\text{off}}\rangle$, found by the method of computer analysis of the time characteristics of the sites brightness (Fig. 5, *b*). This curve has a much sharper rise than the experimental curve,

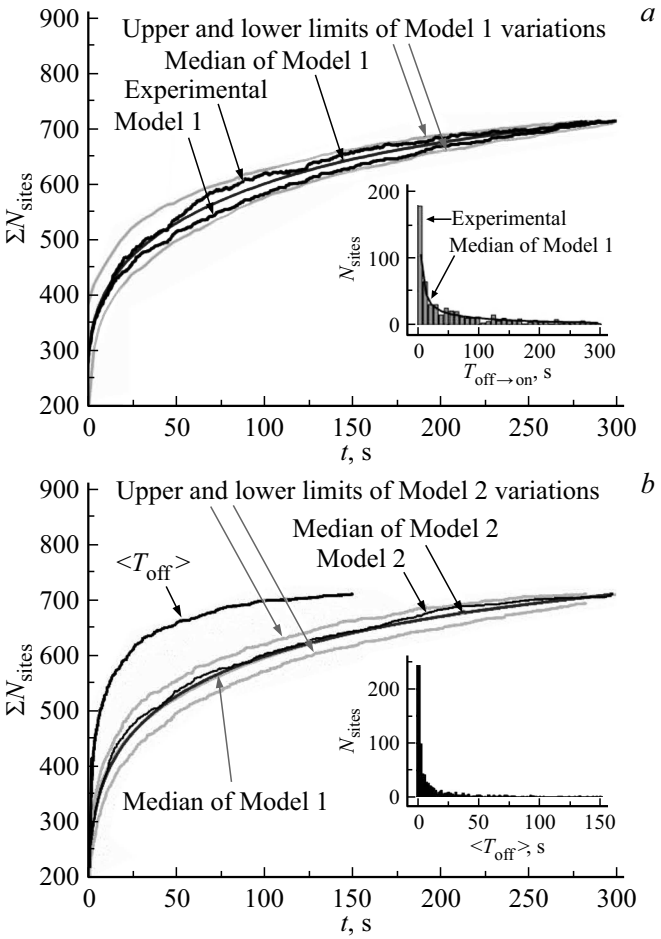

Figure 5. Comparison of experimental and model collection curves. *a*) Model curve obtained as a result of tracking the individual emission sites. In inset: histogram of the sites distribution over the time when the threshold brightness $T_{\text{off}\to\text{on}}$ is exceeded (experimental and averaged from Model 1). *b*) Model curve obtained from the sites distribution by the mean time spent in the state „off" and model curve with exponential stochastic. In inset: histogram of sites distribution by value $\langle T_{\text{off}}\rangle$.

which says that the mean periods of $\langle T_{\text{off}}\rangle$ do not directly determine the turn-on times of the sites.

We simulated the stochastic effect (Model 2 curve in Fig. 5, *b*) using the average $\langle T_{\text{off}}\rangle$, $\langle T_{\text{on}}\rangle$ and three sets of random numbers.

The first set specifies the initial state of the sites („on" or „off" at the time t_0), determining which sites will participate in the formation of the curve shape, and which will determine the height of its first point. These numbers were generated by uniform distribution and compared with the mean times $\langle T_{\text{off}}\rangle$ and $\langle T_{\text{on}}\rangle$.

The second set of numbers determined the turn-on time of each site t. It was generated exponentially with the value $\lambda^{-1} = \langle T_{\text{off}}\rangle \cdot \chi$, where χ was initially equal to 1.

Note that one of the indispensable conditions of the model was that by the end of the time T_{total} (the time of the entire experiment) all emission sites shall be found, so that random numbers t were generated until each of them was in the range $[t_0, T_{\text{total}}]$.

The third set of random numbers introduced stochasticity into the parameter χ, and was generated as the square of the normal distribution $N(\mu = 1, \sigma^2)$. This form of distribution is selected due to several reasons. On average over time, the parameter χ shall remain equal to 1. Normal distribution is expected when recording the total current characteristics [34]. The square of the current flowing through the emission site, in the first approximation, determines its temperature. As a result, this type of statistics gave the best fit between the model and experimental collection curves, including the Model 1 curve, with a wide variation of the value σ.

Multiple repetition (~ 5000 times) of the introduced stochastics made it possible to obtain the range of scattering of the model collection curve and its average value (shown by gray lines in Fig. 5, *b*). The best fit of the Model 2 curve average with the Model 1 curve average was obtained with $\sigma = 5$. The stochastic shape of the parameter χ and the value of σ, in particular, can be associated with various physical processes, such as fluctuations of the emission current, which affect the probability of adsorption processes. However, separate studies are necessary to plot the appropriate model.

6. Conclusion

We presented a new method for recording the frequencies of fluctuations of emission activity on the surface of multipoint field cathodes online. It is shown that the time characteristic of the number of found emission sites (collection curve) has a rather stable shape at a stable level of emission current.

The dependence of the shape of the collection curve on the selected threshold brightness level is shown. The criteria for its selection are: the achievement of the maximum number of found emission sites with the minimum effect of the found sites merging.

The effect on the shape of the collection curve of the current instability of the emission system was reviewed. The characteristic sections on the curve are marked, corresponding to vacuum discharges and migration of emission activity over the emitter surface.

A theoretical approach to understanding and modeling the collection curve is presented, which is based on the representation of the field cathode as a set of emission sites with two adsorption states „on" and „off". The sites switch over exponentially with additional stochastics in the form of the square of the normal distribution.

The relationship between the experimental collection curve and the model curves, which were plotted by method of post-processing the tracking results of individual emission sites, is shown. The model plotted by the time of the first exceedance of the threshold brightness for each site (Model 1) showed good agreement with the experiment.

The model based on the exponential law of sites switching over „off → on“ (Model 2) also gave good agreement with the experiment, but subject to the additional contribution of noise processes in the form of the square of the normal distribution. This contribution can be associated with fluctuations in the emission current.

The experimental curve is fully within the limits of permissible deviations of both types of model curves. This confirms its representativeness in the analysis of the fluctuation activity of the LAFE emission sites.

Conflict of interest

The authors declare that they have no conflict of interest.

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