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## STUDY OF CORRELATIONS IN THE RELIEF OF COMPLEX SURFACES USING THE EXAMPLE OF AMORPHOUS HYDROGENATED SILICON

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**Abstract.** The results of a study of correlations in the surface relief of amorphous hydrogenated silicon using the methods of average mutual information and two-dimensional detrended fluctuation analysis by identifying its structural components using the scale-space technique are presented. The experimental samples were model and real surfaces of amorphous hydrogenated silicon. The model surface was formed by superimposing the surfaces "Stochastic fractal", "Particles" and "Gaussian noise". The values of the scaling index were obtained from the dependences of the fluctuation function on the scale, as well as the values of the average mutual information and the maximum mutual information were calculated. A comparative analysis of the correlation dependencies of the model and the real surfaces of amorphous hydrogenated silicon has shown that the model surface "Particles" is closest to the surface structure of the experimental sample in terms of its characteristics. It was found that particles with dimensions of  $65 \pm 10$  nm are present in the surface structure of the experimental sample.

**Keywords:** amorphous hydrogenated silicon, surface, relief, information-correlation characteristics, mutual information, fluctuation analysis, scale-space.

### 1. Introduction

Currently, materials with complex structures (composite, nanocrystalline, disordered, porous, etc.) are becoming increasingly popular in various fields of science and technology. Therefore, there is a need for special methods for studying the structure of such materials. As is known, in disordered materials there is no long-range order in the arrangement of atoms, therefore, to study the features of their structure, methods different from those used for ordered materials (for example, crystalline) are required. Most disordered materials are characterized by short- and medium-range order in the arrangement of atoms. The average order is determined by the interaction of lone pair electrons, van der Waals interaction, and is formed by atoms that are partly included in the second coordination sphere and coordination spheres of higher orders [1]. It is believed that the average order is manifested in the presence of clusters at a distance of about 10 nm. Consequently, average order can characterize the correlations of various structural elements in disordered materials. For example, the average order in amorphous hydrogenated silicon (a-Si:H) determines the order in the arrangement of silicon tetrahedra; in nanocrystalline a-Si:H, the average order determines the ordering of nanosized inclusions in the amorphous matrix. It was shown in [2] that with the formation of nanocrystalline silicon inclusions or  $(\text{Si:H}_2)_n$  chains, structural ordering of the amorphous matrix occurs.

The study of correlations in the surface topography of disordered materials helps to identify structural features at the average order level. Knowledge of the structural features allows connecting the properties of the resulting materials with the technological regimes for their production. To study correlations, it is

necessary to obtain primary information about the material in the form of data on the surface topography. Data on surface relief can be used to study the average order in the structure of disordered materials in cases where the relief is closest to the structure of the material in the bulk. The validity of using data on surface relief is associated with the theorem of F. Takens. According to the embedding method of F. Takens [3], any signal from the system contains information about all processes within it. Consequently, for disordered materials based on the surface topography, it is possible to study correlations [4] in the structure at the level of average order.

To study correlations in the surface topography of disordered materials, methods of average mutual information (AMI) [5] and two-dimensional detrended fluctuation analysis (2D DFA) were developed. The AMI method allows characterizing the structure of the material as a whole, and not individual elements of the structure, as is done by traditional methods based mainly on the calculation of statistical parameters (averaged roughness values, porosity, average grain size, etc.). The 2D DFA method allows identifying the type of correlations in the structure, as well as find correlation vectors, i.e. the scales at which the orderliness of the surface relief is manifested.

Since disordered material has a complex structure, it is also advisable to apply the scale-space technique [6]. The scale-space technique allows decomposing the surface relief into several structural components and study correlation properties for each subsurface. The use of AMI, 2D DFA and scale-space methods in combination makes it possible to study the complex structure of materials, in which both chaotic and harmonic components are present.

The purpose of this work is to study correlations in the surface topography of amorphous hydrogenated silicon using AMI and 2D DFA methods by identifying its structural components using the scale-space technique.

## 2. Description of the technique

The technique for studying surface relief correlations is as follows.

### 2.1. Scale-space technique

To study, it is necessary to have a surface height matrix, which is a square image, at each point of which with coordinates  $(x,y)$  the value of its height  $f(x,y)$  is stored. For any continuously differentiable and integrable mapping  $f: \mathbb{R}^2 \rightarrow \mathbb{R}$  there is a set  $L: \mathbb{R}^2 \times \mathbb{R}_+ \rightarrow \mathbb{R}$ , which we will call the scale representation of the image  $f$ , defined as:

$$L(x, y, \sigma) = f(x, y, \sigma) * g(x, y; \sigma) = \int_{(\xi, \eta) \in \mathbb{R}^2} f(x - \xi, y - \eta) g(\xi, \eta; \sigma) d\xi d\eta, \quad (1)$$

where  $g: \mathbb{R}^2 \times \mathbb{R}_+ \rightarrow \mathbb{R}$  – Gaussian convolution kernel on the extended plane  $g(x, y; \sigma) = \frac{1}{2\pi\sigma} e^{-\frac{x^2+y^2}{2\sigma}}$ ,  $\sigma > 0$ .

The result of the convolution  $L(x,y;\sigma)$  is a scale representation of the surface  $f(x,y)$  with a scale factor  $\sigma$ . The geometric meaning of convolving a surface  $f(x,y)$  with a Gaussian kernel is to smooth the surface with a certain scale parameter  $\sigma$ . In this case, the value  $\sigma$  is related to the result of the operation  $L(x,y;\sigma)$  in such a way that all details are removed from the original image, the linear dimensions of which do not exceed the value  $\sqrt{\sigma}$ , in other words, two local extrema will be mutually destroyed (annihilated) or one will absorb another if the distance between them is less than  $\sqrt{\sigma}$ . This allows, on the one hand, to get rid of the noise of the source material (or significantly weaken it), and on the other hand, to leave the relief elements of the linear dimension that is necessary at the current stage of processing.

Thus, the decomposition of the surface image according to scales  $\sigma$  is the result of differentiation with respect to the second derivative. The most recent image is the most smoothed and should be visually very different from the original. However, it usually contains some features that cannot be seen in images close to the original one. For example, this way can get rid of various noises. In other words, one surface can contain many subsurfaces, the properties of which can be compared with simple models.

When studying complex surfaces that contain both harmonic and chaotic components, it is advisable to isolate their frequency characteristics. Moreover, the presence of a number of surfaces with different levels of detail (smoothing scales) can provide more information about the structure compared to analyzing only the original image. A large number of surface details correspond to the presence of a high-frequency component in the selected structure; a smaller number of details (smoothed surface) correspond to a low-frequency component.

## 2.2 2D DFA method

The 2D DFA method was described in [7]. For the height matrix  $H(x,y)$  the cumulative surface  $Y(x,y)$  is calculated:

$$Y(x,y) = \sum_{n=1}^x \sum_{m=1}^y H(x,y). \quad (2)$$

The cumulative surface is divided into  $M_s \times N_s$  disjoint square segments of size  $s \times s$ , with  $M_s = M/s$  and  $N_s = N/s$ . In each segment, a surface of the type  $Y_{u,w}(x,y) = Y(l_1+x, l_2+y)$  is formed, for  $1 \leq x, y \leq s$ , where  $l_1 = (u-1)s$  and  $l_2 = (w-1)s$ , where  $u, w$  are numbers segments.

In each surface  $Y_{u,w}(x,y)$  a first-order polynomial trend function is calculated:

$$\tilde{Y}_{u,w}(x,y) = a \cdot x + b \quad (3)$$

$$\text{or } \tilde{Y}_{u,w}(x,y) = a \cdot y + b. \quad (4)$$

The remainder function is calculated:

$$\varepsilon_{u,w}(x,y) = Y_{u,w}(x,y) - \tilde{Y}_{u,w}(x,y). \quad (5)$$

The local RMS fluctuation function is calculated, bearing in mind that  $\langle \varepsilon_{u,w}(x,y) \rangle = 0$ :

$$F(s, u, w) = \sqrt{\frac{1}{s^2} \sum_{x=1}^s \sum_{y=1}^s \varepsilon_{u,w}(x,y)^2}. \quad (6)$$

The full root-mean-square fluctuation function is calculated:

$$F(s) = \frac{1}{M_s N_s} \sum_{u=1}^{M_s} \sum_{w=1}^{N_s} F(s, u, w). \quad (7)$$

For wide ranges of values  $s$ , the dependence is valid  $F(s) \sim s^a$ .

In general, by plotting the dependence of  $F$  on  $s$  on a double logarithmic scale, a scaling index characterizing the type of correlations in the structure can be calculated from the tangent of the slope angle, and a correlation vector showing the scale of manifestation of these correlations can be calculated from the inflection of the fluctuation function.

## 2.3 AMI method

Mutual information [5] is defined as the amount of information that becomes known about the value of a random function at point  $A$  when its value becomes known at point  $B$ . If  $D$  is the domain of definition,  $Z$  is the domain of a random function,  $P_X$  is the probability distribution density at point  $X$  (as a function on  $Z$ ),  $P_{XY}$  is the joint distribution density at points  $X$  and  $Y$  (as a function on  $Z_2$ ), then the mutual information  $I_{AB}(z_1, z_2)$  for a pair of known and predicted values is calculated according to the following formula:

$$I_{AB}(z_1, z_2) = \log_2 \left[ \frac{P_{AB}(z_1, z_2)}{P_A(z_1) \cdot P_B(z_2)} \right]. \quad (8)$$

AMI is defined as the average expected value of mutual information for a given pair of points by integrating over  $Z^2$ :

$$I(A, B) = \iint_{Z^2} P_{AB}(z_1, z_2) \log_2 \left[ \frac{P_{AB}(z_1, z_2)}{P_A(z_1) \cdot P_B(z_2)} \right] dz. \quad (9)$$

For practical tasks, so-called "convolutions" (transformed AMI functions) are calculated:

$$I_s(s) = \iint_{S(d_1, d_2)=s} \frac{I(d_1, d_2)}{M_{S_D}(s)} d(d_1) d(d_2), \quad (10)$$

where  $d_1, d_2$  are generalized parameters of points in the space  $D^2$  ( $d_1$  corresponds to two coordinates  $x_1, y_1$ , and  $d_2 - x_2, y_2$ ). For each value of parameter  $s$  integration is carried out over all pairs of points that this convolution maps to a new space  $S_D$ ,  $I$  is the average mutual information between points characterized by parameter  $s$ , and  $M_{S_D}$  is a measure of the new space  $S_D$  displayed by the selected convolution by parameter  $s$  in space  $D^2$ .

Ultimately, the AMI graph is a surface in a Cartesian coordinate system, the AMI values for any vector are in the range from 0 to 1. The criterion for the presence of correlation between the points of the sample under study is the presence of maxima on the AMI distribution graph.

The AMI method allows calculating directly the AMI value ( $\Psi_{OR}$ ), which characterizes the degree of ordering of the structure, and the value of maximum mutual information (MMI,  $\Psi_{IC}$ ), which shows the

spread of data (the number of points of different heights). Knowing the value of the MMI and the number of identical elements in the structure, it is possible to calculate the information capacity of the structure (relief).

The analysis of the results obtained using the scale-space, 2D DFA and AMI methods is carried out taking into account the criteria for determining the structural complexity of the surface of solid-state materials, classification of the values of  $\alpha$ ,  $\Psi_{OR}$  and  $\Psi_{IC}$  [8].

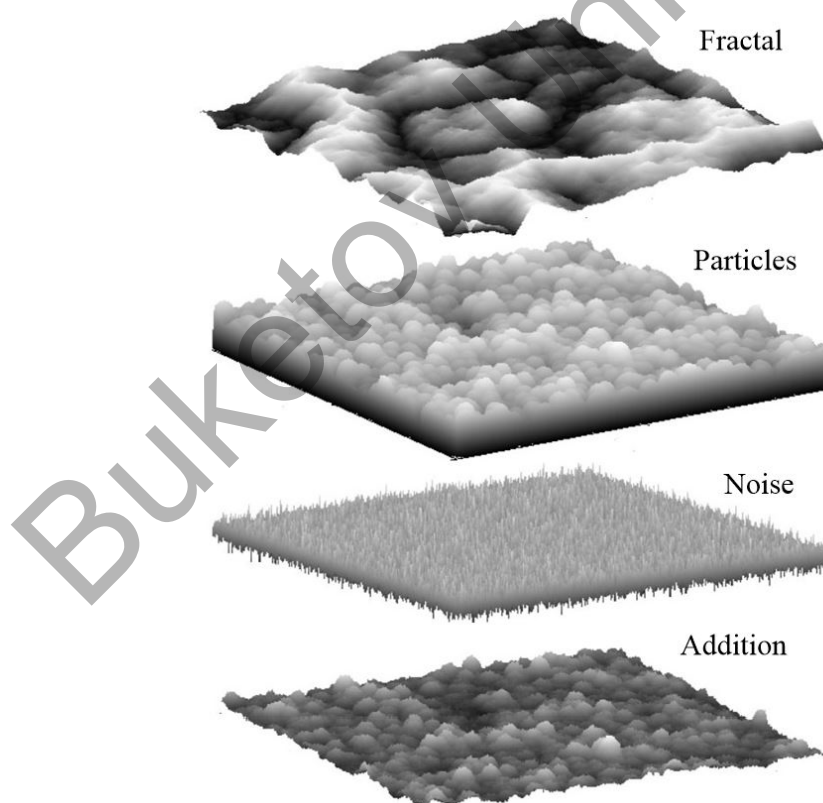
At  $0.5 < \alpha < 2$ , long-range or non-power correlations may be present in the system. In particular,  $\alpha=1$  is characteristic of flicker noise ( $1/f$  noise) and regular fractal,  $\alpha=1.5$  - for Brownian noise. The value  $\alpha=2$  is characteristic of strictly ordered structures.

For the cases of  $\Psi_{OR} < 0.02$ , there is a low orderliness of the structure,  $0.02 < \Psi_{OR} < 0.1$  - average orderliness of the structure,  $\Psi_{OR} > 0.1$  - high orderliness of the structure.

For the cases of  $\Psi_{IC} < 0.5$ , there is a low information capacity and entropy of the structure,  $0.5 < \Psi_{IC} < 0.7$  - average information capacity and entropy of the structure,  $\Psi_{IC} > 0.7$  - high information capacity and entropy of the structure.

### 3. Investigation of correlations in a-Si:H surface relief

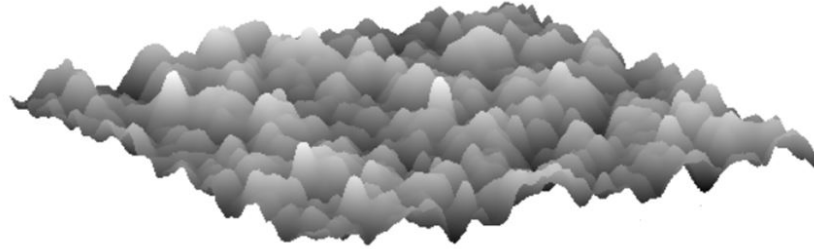
To study correlations in the relief of the a-Si:H surface, a model surface was synthesized, and an experimental a-Si:H sample was selected. The model surface was created based on the presence of several components in the a-Si:H structure. Firstly, amorphous silicon has fractal properties [9]. Secondly, there is an average order in amorphous silicon [2, 10], which is manifested in the presence of clusters on the surface. And thirdly, amorphous silicon is a disordered material, so there must be a chaotic component in its structure. Based on these considerations, the model surface a-Si:H was created by superimposing three initial model surfaces on each other: "Stochastic fractal", "Particles", "Gaussian noise" (Fig. 1).



**Fig. 1.** Obtaining a model surface a-Si:H from the components "Stochastic fractal", "Particles", "Gaussian noise"

The experimental sample a-Si:H obtained by the glow discharge method of silane-containing mixtures was also used in the work. The frequency of the exciting generator was 13.56 MHz, the discharge power was maintained at 200 watts, a mixture of silane and hydrogen (10 % SiH<sub>4</sub>+90 % H<sub>2</sub>) was used as the supplied

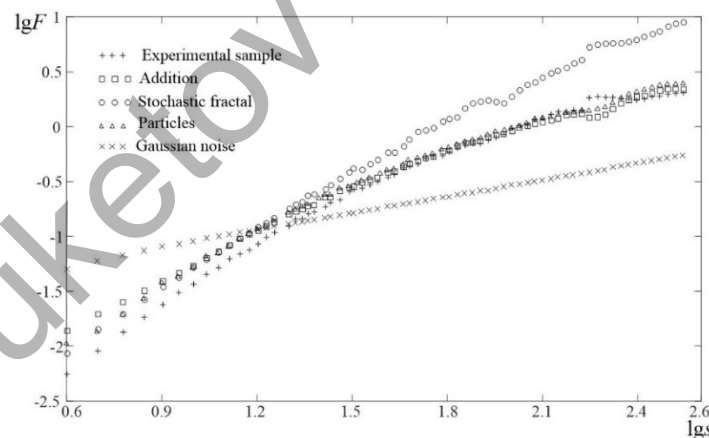
gas. The substrate was *n*-type epitaxial silicon doped with antimony. The substrate temperature was 170 °C, the film thickness a-Si:H – 3 microns. The surface images of the experimental sample were obtained using an atomic force microscope (AFM) NTegra Aura in semi-contact mode (the diameter of the cantilever probe was 30 nm, the scan size was 5×5 microns). Enlarged for better visualization, a specific AFM image of the a-Si:H film in in three-dimensional representation is shown in Fig. 2.



**Fig. 2.** The surface relief of the experimental sample a-Si:H (the dimensions of the presented area are 2.5×2.5 microns)

The initial model surfaces and the image of the surface of the experimental sample a-Si:H were investigated by 2D DFA and AMI methods. Graphs of the dependence of the fluctuation function  $F$  on the spatial scale  $s$  are shown in Fig. 3. It can be seen from the graphs that the model surfaces "Particles" and "Addition", as well as the surface of the sample a-Si:H have similar dependencies. On the one hand, it can be concluded that the structure of the surface of the experimental sample is close to the structure of the model surface of the "Particle", that is, it contains randomly arranged particles of different diameters. On the other hand, on the scales  $\lg s = 2.2-2.5$ , the type of dependence  $F(s)$  of the experimental sample is similar to the addition of the dependencies "Stochastic fractal" and "Gaussian noise".

Table 1 shows the values of the scaling index  $\alpha$  calculated from the dependencies  $F(s)$  (Fig. 3) up to the first inflection of the fluctuation function, as well as the values of AMI ( $\Psi_{OR}$ ) and MMII ( $\Psi_{IC}$ ) obtained from the distribution of mutual information.



**Fig. 3.** Dependence of the fluctuation function  $F$  on the scale  $s$

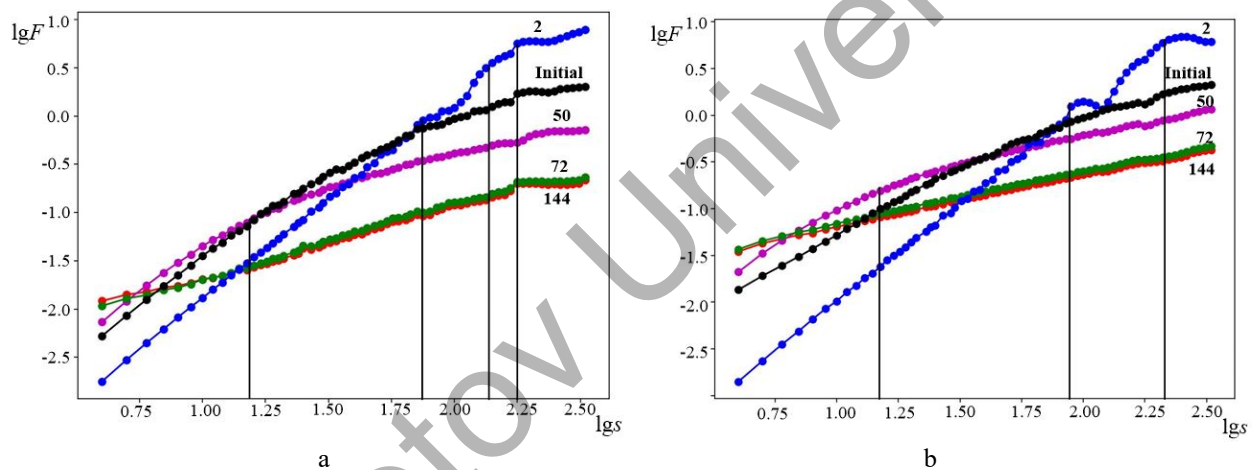
According to Table 1, it can be seen that the values of  $\Psi_{IC}$  fall into various categories of information capacity [8]: the "Stochastic fractal" surface refers to structures with high information capacity ( $\Psi_{IC} > 0.7$ ), the surface of the experimental sample – with low information capacity ( $\Psi_{IC} < 0.5$ ), and the surfaces "Particles", "Gaussian noise" and "Addition" – with average information capacity ( $0.5 < \Psi_{IC} < 0.7$ ). The value of the information capacity of the surface of the experimental sample is closest to the  $\Psi_{IC}$  surface "Addition". By the value of  $\Psi_{OR}$ , the "Stochastic fractal" surface falls into the category of [8] structures with average ordering ( $0.02 < \Psi_{OR} < 0.1$ ), and the rest – with low ordering. In this case, the value of the degree of ordering at the surface of the experimental sample is closest to the value  $\Psi_{OR}$  of the surface "Particles" than the surface "Addition". This can be attributed to the peculiarities of obtaining an image of the surface of the experimental sample. Since AFM was used, the surface image is a "convolution" of the probe and therefore the chaotic component is suppressed to some extent.

Therefore, to study correlations in the surface relief of disordered materials, it is advisable to use data obtained not only by AFM, but also by scanning electron microscopy (SEM). This conclusion is also confirmed by the results of calculating the scaling index, since the value of  $\alpha$  at the surface of the experimental sample is very close to the value of  $\alpha$  for strictly periodic signals ( $\alpha=2$ ), while for other surfaces the values of  $\alpha$  are lower.

**Table 1.** Information and correlation characteristics of the studied surfaces.

Surface	$\Psi_{IC}$	$\Psi_{OR}$	$\alpha$
Stochastic fractal	0.73	0.034	1.91
Particles	0.60	0.008	1.72
Gaussian noise	0.54	0.002	0.50
Addition	0.51	0.006	1.50
Experimental sample	0.46	0.013	1.98

Fig. 4 shows the results of the study of surface images using the scale-space technique, namely the dependencies of  $F(s)$  at different smoothing scales  $n$  for the experimental sample (Fig. 4, a) and the "Addition" surface (Fig. 4, b).



**Fig. 4.**  $F(s)$  dependences at different smoothing scales  $n$  for the experimental sample (a) and the "Addition" surface (b)

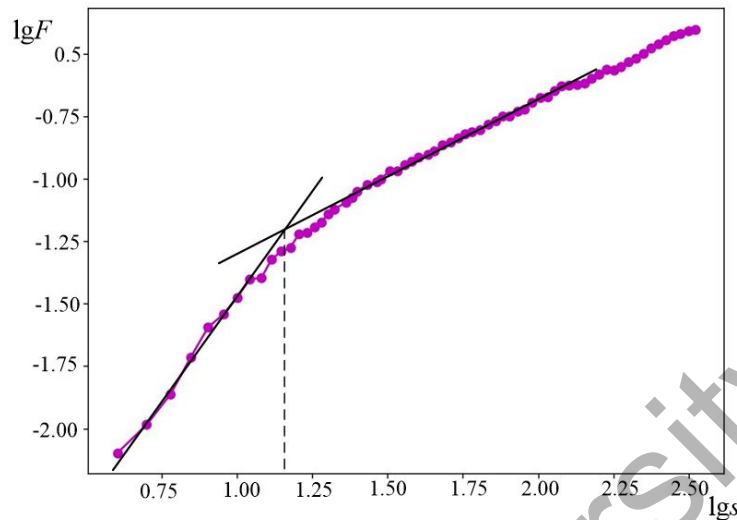
At large smoothing scales (72, 144 in Fig. 4), where high-frequency components of surfaces are manifested, there are practically no inflections on the dependencies  $F(s)$ . This means that no harmonic components have been detected in the structure. The scaling index  $\alpha$  has values of 0.6 and 0.55 for the experimental sample and the "Addition" surface, respectively. Such a value of  $\alpha$  is close to the surface of "Gaussian noise". Thus, at large smoothing scales, the noise component prevails.

On the average smoothing scales (50), areas close to linear were approximated on the dependencies  $F(s)$ , as a result of which an inflection of the function on a spatial scale of the order  $lgs = 1.18$  was revealed. The presence of an inflection indicates that there is a harmonic component in the surface structure. To determine the period, use the formula,  $x = (b \cdot 10^{lgs})/a$  where  $b$  is the image size in microns (in this case 1.5 microns),  $a$  is the image size in pixels (in this case 350 dots). The period of the harmonic component was  $65 \pm 10$  nm. A similar inflection was found on the dependence  $F(s)$  for the surface "Particles" (Fig. 5). Thus, particles with characteristic dimensions of  $65 \pm 10$  nm are present in the structure of the surface of the experimental sample.

On small smoothing scales (2 in Fig. 4), where low-frequency components of surfaces are manifested, several inflections are observed on the dependencies  $F(s)$  in the range of spatial scales  $lgs = 1,8-2,5$ . The values of the spatial scales corresponding to these inflections were  $320 \pm 10$ ,  $580 \pm 10$  and  $760 \pm 10$  nm for the experimental sample; for the "Addition" surface –  $380 \pm 10$  and  $900 \pm 10$  nm.

On the one hand, such scale values can be explained by the frequency of repetition of particles, for example,  $380 \text{ nm} / 65 \text{ nm} \approx 6$ ,  $900 \text{ nm} / 65 \text{ nm} \approx 14$ , taking into account the error associated with the initial

image resolution and the calculation method. On the other hand, the appearance of additional inflections in the dependence of the fluctuation function may be associated with fractal properties.



**Fig. 5.** Example of determining the correlation vector by the inflection of the fluctuation function for the model surface "Particles"

#### 4. Conclusion

In this work, the use of the scale-space technique allowed identifying the structural components of the surface relief of amorphous hydrogenated silicon and investigating information-correlation characteristics at each structural level. The selection of low-frequency components allows studying the features of the harmonic components of the structure. The isolation of high-frequency components allows studying the features of chaotic components of the structure.

A comparative analysis of the information-correlation characteristics of the model surfaces and the experimental sample of amorphous hydrogenated silicon showed that the model surface "Particles" is closest to the structure of the experimental sample surface in its characteristics. This may be due to the peculiarities of obtaining an image of the surface by atomic force microscopy.

The study of the information-correlation characteristics of the a-Si:H film surface showed that the a-Si:H structure at different spatial scales exhibits the properties of all surface components used in the "Addition" model surface. Thus, any complex surface using the methods of AMI, 2D DFA, scale-space can be studied by decomposing into simpler components.

#### Conflict of interest statement

The authors declare that they have no conflict of interest in relation to this research, whether financial, personal, authorship or otherwise, that could affect the research and its results presented in this paper.

#### CRedit author statement

Rybina N.V.: Conceptualization, Data Curation, Writing - Original Draft; Rybin N.B.: Methodology, Investigation; Litvinov V.G.: Writing - Review & Editing, Supervision. The final manuscript was read and approved by all authors.

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