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NONELECTRONIC PROPERTIES OF SEMICONDUCTORS (ATOMIC STRUCTURE, DIFFUSION)

On a Combined Approach to Studying the Correlation Parameters of Self-Organizing Structures

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Abstract—The correlation parameters of self-organizing structures are investigated using a combined approach, a combination of 2D detrended fluctuation analysis and the average-mutual-information method. The self-organizing structures to be investigated are model surfaces with different degrees of ordering (ordered, disordered, and mixed) and amorphous hydrogenated silicon and tetrahedral carbon films. It is demonstrated using test structures that the correlation vectors determined by kinks on the scale dependence of the fluctuation function with the use of the 2D detrended fluctuation analysis coincide with sufficient accuracy with specified periods of surface harmonic components. It is more expedient to study disordered structures using the average-mutual-information method. The physical meaning of maximum mutual information is shown to characterize the information capacity of a system. The combined approach allows the correlation parameters of combined systems to be investigated most comprehensively.

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1. INTRODUCTION

The study of self-organizing structures based on semiconductor, organic, and other compounds is one of the directions in nanoelectronics [1, 2]. The successful development of the technology of self-organizing structures is inextricably linked with methods for studying the correlation properties of nanostructures and nanomaterials.

In [3, 4], it was proposed that the correlation parameters of self-organizing structures be studied with the use of 2D detrended fluctuation analysis (2D DFA). The 2D DFA method was tested on test structures and experimental samples and its efficiency was demonstrated.

In [5], it was suggested that the self-organization of structure formation in materials be studied via estimation of the degree of surface-structure ordering, i.e., using the average-mutual-information method (AMI), which allows the random, poorly organized, or ordered structure of a material to be determined by the mutual-information value, but does not allow the correlation vector to be determined numerically. More information on the numerical value of the correlation vector is provided by the 2D DFA method.

Since different structures have different degrees of order, one should know which method is better applicable to investigations of a certain structure. Then, self-organization of the structure formation can be studied in more detail, which is necessary for solving the inverse problem, i.e., the creation of nanostructures and nanomaterials with desired properties.

The aim of this study is to investigate self-organized structures using a combined approach, which combines 2D DFA and AMI techniques. For this purpose, a number of model structures with different degrees of self-organization are created and the correlation parameters for these structures are determined. In addition, the methods are tested on experimental semiconductor samples, including amorphous hydrogenated silicon *a*-Si:H and amorphous tetrahedral carbon t*a*-C.

2. CRITERIA FOR CHOOSING TEST STRUCTURES. DESCRIPTION OF MODEL SURFACES

All surfaces can be conditionally divided into three groups: ordered, random, and mixed. Thus, to form a correlation-parameter database, it is necessary to synthesize test structures of three types and determine their correlation parameters, which characterize the self-organization of structure formation (correlation vector, scaling index, AMI, and maximum mutual information (MMI)). The 2D DFA method allows determination of the correlation vector, which indicates the periodicity of a structure, and the scaling index, which represents the structure type. AMI shows the predictability of the accuracy of coincidence of the parameters of points in a database (e.g., surface roughness heights) at a certain scanning vector, and the

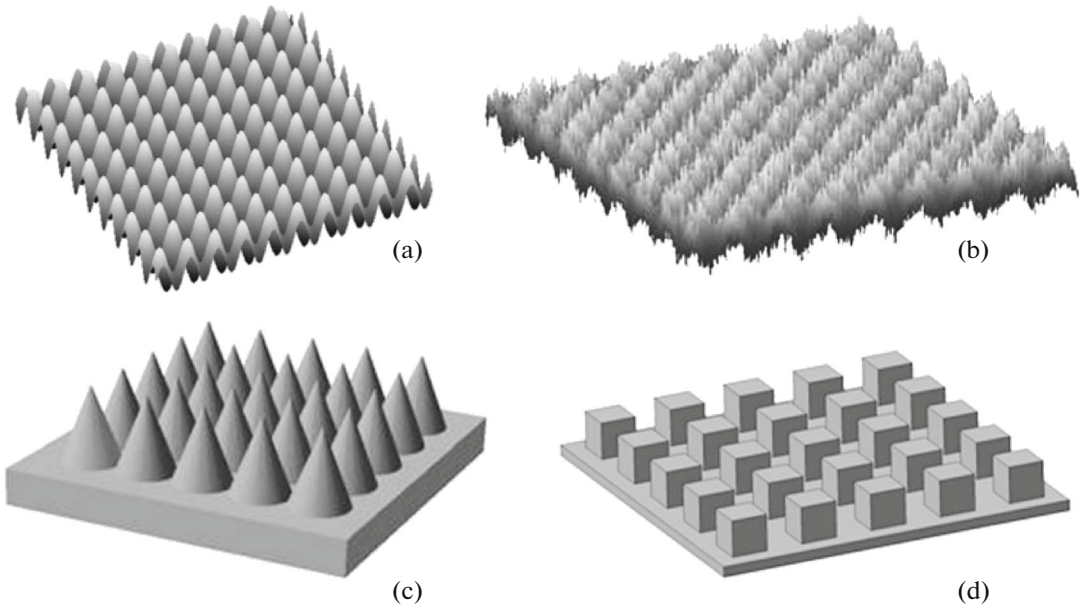

Fig. 1. Examples of model surfaces.

MMI reflects, in fact, the information capacity of a system.

The object of study was a surface structure (roughness), which can be rather easily and reliably determined in experiments at the nanoscale with the use of, e.g., scanning-electron- or atomic-force microscopy. A criterion for choosing the test structures was the presence of different degrees of structure organization. Ordered structures are characterized by the periodic arrangement of certain topographic features. As ordered structures, we chose the surfaces of "Cubes" and "Cones" and sinusoidal surfaces. The surfaces of the "Cubes" represent ordered cubic patterns on a smooth surface with different periods. They are characterized by a sharp height drop from the maximum value to zero (step function). The surfaces of the "Cones" represent ordered conic figures on a smooth surface with different periods. In this case, the height topography changes in accordance with linear law (linear step function). The sinusoidal surfaces are characterized by the nonlinear law of pattern-height variation (sinusoidal function). As disordered structures, we chose surfaces with a noise component (Gaussian or white noise). The combined surface is the superposition of the sinusoidal component and Gaussian noise. In this case, the sinusoidal component was chosen from the smooth variation in the height pattern. Examples of the surfaces are presented in Fig. 1. We specify the physical size of surface to be 1×1 μm at a pixel size of 512×512. The maximum protrusion height was chosen to be 7 nm.

Thus, the model structures were data arrays processed using the combined approach to studying self-organizing structures. The 2D DFA method allows correlations in the surface structure to be determined and the period of harmonic components (correlation vectors) in a structure to be revealed. First, the initial data array is divided into segments of different scales and fluctuations are investigated in each of them. As a result, the dependence of the fluctuation function F on scale s is obtained. The slope of the fluctuation function is the scaling index [3].

The AMI method consists in the following. Mutual information is calculated for each pair of height points on the basis of information theory. Mutual information is defined as the quantity of information about the value of the random function at point A that becomes known when its value at point B is obtained. The allowed mutual information values range from 0 to 1 and the information dimension is bits. In other words, the mutual-information values change from zero to the maximum average entropy of total chaos. Correspondingly, the higher the degree of order of a structure, the large the AMI value is [5].

3. NORMALIZATION AND CLUSTERIZATION OF MUTUAL INFORMATION

In studying real materials and structures, the number of possible surface-point heights and the number of system states can be very large and, consequently, both the entropy and information of the system can tend to infinity. To study these objects using techniques of information theory, the system is quantized, i.e., the range of values of some quantities is reduced to a finite set.

For instance, if the height of the surface profile point obtained using a scanning microscope lies in the

range [A; B], we can divide this range, e.g., into $2^8 = 256$ intervals and round the height of each point at the interval center.

After such a quantization procedure, the entropy and information become finite measurable quantities. The maximum entropy of the system from one point is equal to the logarithm of the number of states, in our case, eight bit. Mutual information is equal to the same value, if the quantity was not completely determined before measurement and became unambiguously determined after it.

The chosen quantization discreteness is a compromise between roughening of the result caused by a small number of levels and the complexity of the calculations. As was shown using the AMI method, the discretization of values comparable with that of linear coordinates in terms of the number of levels is sufficient and a further increase in their number does not significantly change the results.

To eliminate the arbitrariness of this choice, all quantities with information dimension are normalized by the maximum entropy of the system and, in our case, are divided by eight for one point. After normalization, the value of the system entropy acquires physical meaning and changes from zero (the state is known) to unity (complete chaos with equiprobable states). The mutual information also changes from zero (the knowledge of one value provides nothing about another) to unity (the value was completely undetermined and now is exactly known).

What are the advantages and drawbacks of this normalization? For real systems, choosing normalization is good. The profile-point heights acquire all possible intermediate values and, even if these values are not equiprobable, this does not significantly decreases the entropy; therefore, the correlation in the samples is reliably established [5, 6].

The situation is different for artificial model surfaces. Let us consider a 2D surface consisting of $N \times N$ rectangular columns. It is worth noting that the heights acquire only two possible values from 256 with a probability ratio of 1:3. Then, the average entropy of such a system for an off-duty ratio of two is determined as

$$E_m = -\frac{\sum_{N^2} p(x) \cdot \log_2(p(x))}{N^2}$$
$$= -\frac{1}{4}(-2) - \frac{3}{4}(\log_2(3) - 2) \approx 0.81,$$

where $p(x)$ is the probability of their being of one of the heights at point x.

The obtained entropy is much smaller than the entropy of total chaos, which amounts to eight. Therefore, the MMI in such a system for the given normalization by chaos cannot exceed 0.1. At an off-duty ratio of four, the mutual information is even lower (no larger than 0.04.

The physical meaning of this effect is that the mutual-information value is not the degree of ordering of the system, but an estimate of the system part that is caused by the correlation of profile heights at different points. Therefore, if a system is already essentially determined, then the information obtained from the known values at other points is not large.

Nevertheless, the AMI maximum obtained in tests in the phase space of translation vectors retains its interpretation, but it is incorrect to estimate it by comparison with unity. In view of this, information can be normalized not by the average entropy of chaos, but by the average entropy of the surface under treatment.

The average mutual information calculated for all point pairs in a sample is numerically equal to the total entropy with the positive sign; therefore, normalizing the AMI by the sample entropy, we explicitly determine unity as an absolute average.

It is likely that in the case of such normalization, we can use some absolute criteria to estimate, e.g., the presence of correlation by a translation vector. This needs further investigation.

Below, we consider other types of the clusterization of mutual information. The average mutual information for a translation vector is defined as the average sum of mutual information of all pairs of points comprising this translation vector for the sample area. This can be interpreted in another way: the translation vector creates a cluster from pairs of sample points and the AMI integral is determined for these clusters. The cluster set in itself forms a phase space, two-dimensional in this case.

The presence of extrema in the cluster phase space can be interpreted as the presence of some long-range interaction, information transfer, and other structures in the sample.

Another form of clusterization already investigated by us is based on the distance between points. In this case, the cluster forms a set of pairs of points separated by the same distance from each other. Then, the phase space is linear.

To establish the presence of linear structures in a sample, we can consider a set of points lying on one straight line to be a cluster. For each such cluster, the AMI is calculated separately. In this case, the phase space is two-dimensional. This form of clusterization can reveal different linearly extended structures, including waves and lattices.

In real samples, ring structures are often observed. To identify such structures, a set of points lying on an arbitrary circle is considered to be a cluster. The phase space is three-dimensional and is difficult to reproduce, but the AMI can be averaged, e.g., for clusters over all circles of the same radius, establishing the AMI distribution over the ring structure radii.

Table 1. Test-structure parameters determined using AMI and 2D DFA methods

No.	Description	MMI	AMI	α	*d*, nm
		Ordered surface			
		Sinusoidal surfaces			
1	Period $T = 333$ nm	0.727	0.126	2; 0	332
2	Period $T/2 = 167$ nm	0.731	0.127	2; 0	168
3	Period $T/4 = 84$ nm	0.730	0.129	2; 0	83
4	Period $T/8 = 42$ nm	0.718	0.123	2; 0	41
5	Periods $T/2 + T/4$	0.672	0.079	2; 0.7; 0	170; 84
6	Periods $T + T/8$	0.651	0.070	2; 0.6; 0	333; 41
		Surfaces of "Cones"			
7	Period 104 nm	0.243	0.20	1.75; 0	111
8	Period 78 nm	0.355	0.035	1.75; 0	84
9	Period 62 nm	0.553	0.058	1.63; 0.13	59
10	Period 48 nm	0.640	0.66	1.63; 0.10	53
		Surfaces of "Cubes"			
11	Period 115 nm	0.101	0.004	1.64; 0	106
12	Period 80 nm	0.167	0.008	1.75; 0	78
13	Period 58 nm	0.278	0.014	1.9; 0	59
14	Period 35 nm	0.484	0.019	1.84; 0.4	32
		Random surfaces			
15	"Gaussian noise"	0.542	0.002	0.5	–
16	"White noise"	0.752	0.001	0.5	–
		Combined surface			
17	"Gaussian noise + sinusoid" with a period of $T/2$	0.542	0.009	1.1; 0.1	195

4. RESULTS AND DISCUSSION

The calculated AMI and MMI, scaling index α, and correlation vector *d* for the test structures are listed in Table 1.

We first analyze the 2D DFA data. When test structures with one harmonic component are studied by the 2D DFA technique, the scale dependence of the fluctuation function should contain one kink and the slope (scaling index) should be two before the kink and zero after it. When the surface structure contains several harmonic components, then the number of kinks in the 2D DFA plot should correspond to the number of these components. The 2D DFA data on the sinusoidal model surfaces confirm this statement. The correlation vector decreases with the period of the sinusoid [3, 4].

For the second group of surfaces ("Cones"), the scale dependence of the fluctuation function also contains one kink, which is indicative of the presence of a harmonic component in the structure. Thus, using the 2D DFA technique, it was established that the investigated structures of the "Cones" are ordered. The correlation vector decreases with the period.

Similar results were obtained using the 2D DFA method for the model surfaces of "Cubes".

For the random surfaces of "Gaussian noise" and "White noise", the scale dependence of the fluctuation function contains no kinks and the scaling index is 0.5. Upon superposition of the sinusoidal component and "Gaussian noise", the 2D DFA method yielded a kink, which confirms the existence of a harmonic component in the structure. A slope (scaling index) of 1.1 before the kink and 0.1 after it corresponds to the presence of a noise component.

It can be seen from Table 1 that the correlation vectors determined according to kinks agree fairly well with the model surface periods. The most exact values of the correlation vectors were obtained for the sinusoidal model surfaces. For the profiles of "Cones" and "Cubes", a small error in the correlation vector arises. Upon the superposition of the sinusoid and "Gaussian noise", the error is somewhat larger (the correlation vector was found to be 195 nm at an initial period of the sinusoid of 167 nm), since the kink in the fluctuation function curve was blurred. Thus, the 2D DFA method allows harmonic components in the structure to be revealed and the specific values of the correlation

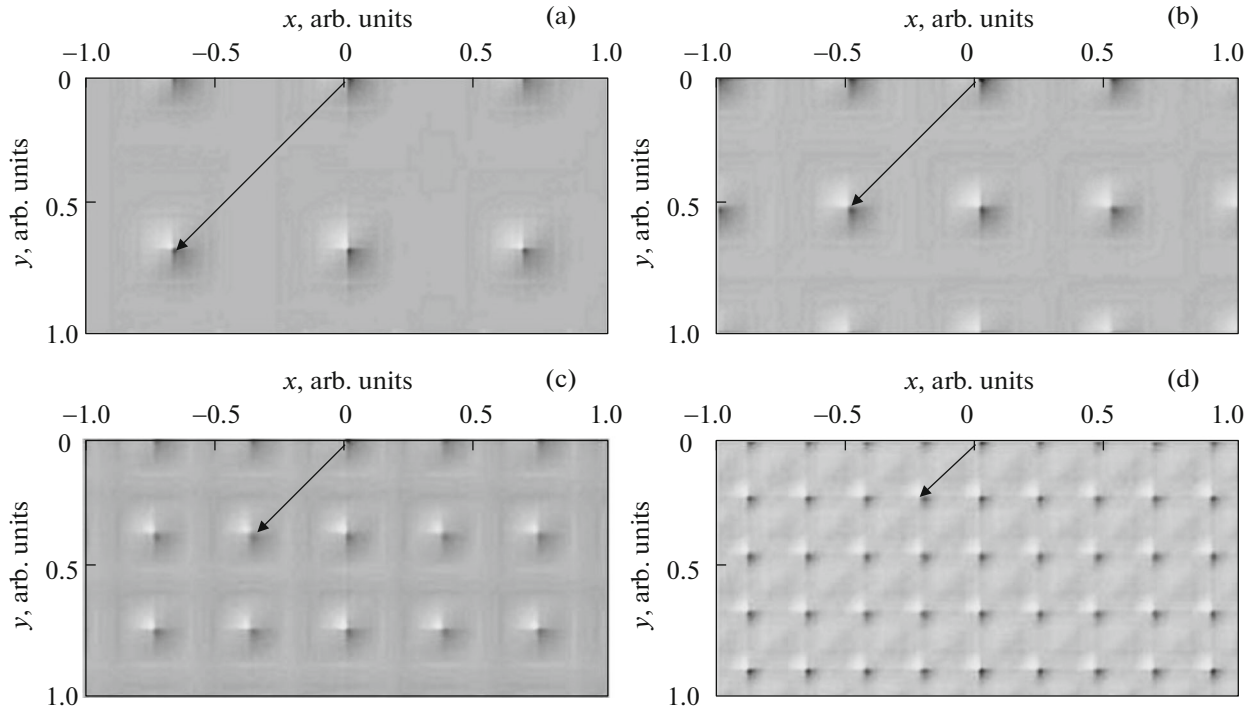

Fig. 2. Average mutual information in the phase space for an undirected vector for the surfaces of "Cubes" at periods of (a) 115, (b) 80, (c) 58, and (d) 35 nm.

vector to be determined. Below, we consider the results obtained using the AMI method.

The largest MMI value (0.752) was obtained for "White noise". "White noise" is an example of a completely random system without correlations between its points. Consequently, the entropy of such a system is at maximum. Since, according to the Shannon formula, entropy is directly proportional to the natural logarithm of information, the MMI (in fact, the information capacity of a system) tends to its maximum.

For the sinusoidal model surfaces, the MMI values appeared somewhat smaller than for "white noise". The AMI values were maximum among all test surfaces.

For the surface of "Gaussian noise", the MMI is 0.542, which corresponds to a poorly organized structure. In fact, according to construction principles, the structure is random. In our case, this is confirmed by the AMI value. The same result (0.542) was obtained for the surface of "Gaussian noise + sinusoid" and the AMI value corresponds to the category of random structures.

For the surfaces of "Cubes", the AMI values were the smallest. In fact, these structures are ordered and have a certain period. In this case, the degree of order for the structure can be determined from the mutual-information values and the presence of AMI-distribution peaks in phase space. For all four surfaces, one can see pronounced peaks in the AMI distribution (Fig. 2). These features are indicative of the presence of long-range correlations in the structure, which, in turn, can bear witness to structure ordering and result from self-organization processes.

Analysis of the AMI spatial distribution for the surfaces of "Cones" yielded results similar to data on the surfaces of "Cubes". The AMI field also included pronounced peaks; therefore, the surface of "Cones" is an ordered structure.

5. EXAMPLES OF STUDYING THE CORRELATION PARAMETERS OF THE STRUCTURFE OF SEMICONDUCTOR MATERIALS USING THE COMBINED METHOD

Since semiconductor materials have very diverse structures, the correlation parameters of the surface structure are interesting to study. The model surfaces studied here have semiconductor analogs. In [3–7], disordered semiconductors were mainly studied using the 2D DFA and AMI methods. Disordered semiconductors are formed under nonequilibrium conditions with the violation of symmetry in a thermodynamically open nonlinear system, which will allow the creation of self-organized systems on their basis in the future. Examples are *a*-Si:H and t*a*-C films (Fig. 3). These samples were obtained by pulsed laser sputter-

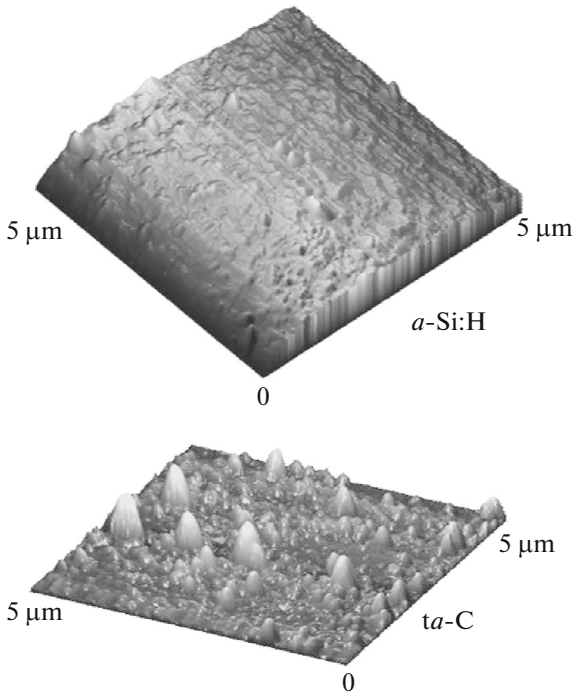

Fig. 3. Atomic-force-microscopy images of the *a*-Si:H and t*a*-C film surface.

ing. The *a*-Si:H film was deposited onto a glassceramic substrate over 7 min at a laser-pulse repetition rate of 10 Hz, a laser-radiation energy density of 11.8 J/cm^2, and a substrate temperature of 200°C. The t*a*-C film was deposited onto a [100] silicon substrate doped with phosphorous over 36 min 40 s at a laser-pulse repetition rate of 10 Hz, a laser-radiation energy density of 14.4 J/cm^2, and room temperature of the substrate.

In atomic-force-microscopy images of the experimental samples, one can see conic and sinusoidal structures with a noise component on the surface.

The AMI and 2D DFA surface parameters of the experimental samples are listed in Table 2. The combined use of the 2D DFA and AMI methods showed that the investigated *a*-Si:H and t*a*-C samples have a correlation vector and, judging by the AMI value, relate to random structures. The MMI value for the *a*-Si:H film was found to be fairly small. If we compare it with the MMI values given in Table 1, then it is similar to the value for the surfaces of "Cones" with a high off-duty ratio of conic structures. Thus, the *a*-Si:H sample studied here belongs to structures with a low information capacity of the surface. The MMI value for the t*a*-C film is much larger and similar to the value for the model surface of "Cones" and combined surface of "sinusoid + noise". The t*a*-C film has a high information capacity of the surface.

Table 2. Surface parameters of the experimental samples as obtained using the AMI and 2D DFA methods

Sample	MMI	AMI	*d*, nm
a-Si:H	0.146	0.017	234
t*a*-C	0.447	0.010	490

It is worth noting that the combined method for studying the correlation parameters of self-organizing structures on the basis of 2D DFA and AMI methods is not only informative for disordered materials. There are many interesting objects for investigations among crystalline semiconductors as well, e.g., textured crystal silicon with pyramidal surface structures, which is currently used in solar cells.

6. CONCLUSIONS

We demonstrated that the 2D DFA method is more informative for studying structures with a harmonic component. When random structures are investigated, it is sufficient to use the AMI method, since it allows the degree of disorder to be estimated. The MMI technique is not always suitable for calculations, either. It was established that when the surface is a structure where maximum (minimum) points cannot be distinguished, then it is incorrect to compare the MMI with unity. In addition, it is incorrect to estimate the properties of a structure using the MMI value in the case of random surfaces, which have no determined correlation vector. In this case, it is expedient to use AMI. In studying complex self-organizing surfaces with an unknown structure, which can contain both harmonic and random components, the combined use of 2D DFA and AMI methods is the most effective.

Using test structures, it was shown that the 2D DFA method allows the period of harmonic components to be determined with sufficient accuracy by kinks on the scale dependence of the fluctuation function. For the AMI method, it was established that the mutual information characterizes the information capacity of a system: the larger the MMI, the higher the information capacity.

The combined use of the 2D DFA and AMI methods allows the correlation parameters of a material structure to be studied most comprehensively. The obtained knowledge makes it possible to control the information capacity and degree of order of the developed self-organizing structures, which, in turn, will allow complex structures with the desired properties to be synthesized.

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