Determination of Surface Structural Complexity of Solid-State Materials

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Abstract

In this paper we analyzed methods of investigating structure features in solid-state materials and developed a new algorithm for determining the surface structural complexity of these materials based on the two-dimensional detrended fluctuation analysis and the scale-space analysis. It is proposed to evaluate the surface structural complexity by calculating tangent of the angle of the fluctuation – spatial scale function and number of breakpoints on the fluctuation – spatial scale function. The developed algorithm was tested on the surface image of the Si layer implanted by Ag^+ ions, subjected to the pulsed laser annealing. We found that investigated surface belongs to the type of surfaces with a high structural complexity. It was also shown that the scaling index – smoothing coefficient (scale) dependence can be used to study the dynamics of changes in the correlation properties of the surface. Knowledge about surface structural complexity of solid-state materials can be used to determine the material functional purpose in different fields of science.

Keywords

Surface structural complexity, two-dimensional detrended fluctuation analysis, scale-space analysis, solid-state materials.

1. Introduction

Nowadays, there are many constantly increasing requirements for properties of solid-state materials used in various fields of science and technology. It contributes to the active synthesis of new multicomponent, nanocomposite, nanocrystalline and other materials. Structural modification of materials such as ion implantation often provides the desired properties. As a rule, such materials have a complex structure. Investigation of these structures requires special methods of data processing and analysis about their features. A surface of a structure provides data about its features, since if a surface is formed during material growth, it reflects a bulk structure. Moreover, technically it is easier to study a surface rather than a bulk.

The aim of this work is to determine criteria of determining the surface structural complexity of solid-state materials.

In order to test the algorithm, we selected Si structures implanted by metal ions, which are widely used in optoelectronics, microelectronics, solar cell technologies, sensorics [1].

The surface in these structures plays an important role. There are some investigations of their surfaces and its features. For example, the authors in [2] studied fractal characteristics of Si surfaces, amorphized by energetic Si ions. The authors of [3] investigated structural modification of a Si crystal by Kr+, Ag+ and Au+ ions.

Considering the mentioned, we think that development of a method to analyze complex surfaces of solid-state materials is a crucial task. Such technique can be used to classify the surface structural complexity. Data about surface features helps to determine the functional purpose of the material in different fields of science and technology [4].

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2. Methods for investigating structural features in solid-state materials

In a materials science, micro-, nano-, optoelectronics, solar energetic there is a problem of determining structural features of solid-state materials. Commonly, to study a material structure physical and analytical methods are used. After choosing a research method, one have to consider the spatial level, at which information about the structure is obtained, and degree of complexity of the material structure.

Diffraction and spectroscopic methods are the most common among physical research methods [5]. Diffraction methods are based on recording a spatial pattern of coherent monochromatic radiation intensity scattered by a studied object and consequent transition from the pattern to distribution of intensity in reciprocal space and calculating micro distribution of a density, using Fourier transform. For example, radiographs from a single crystal are formed by consequently spaced spots (reflexes). From polycrystals - by a system of concentric circles. From amorphous bodies, liquids and gases - by a combination of diffusion halos around the central spot. The main advantage of this method is ability to determine values of the translational vector for crystals. The significant disadvantage of the method for non-crystalline materials is impossibility of determining a similar parameter - the correlation vector. This vector is the distance, at which correlations are observed in a structure. In some cases for disordered materials, there is not any correlation vector at all. In this case, the surface structure should be evaluated by the degree of ordering [6].

Vibrational spectroscopy techniques include infrared absorption and Raman scattering spectroscopy. In both cases, details of the obtained spectra are associated with vibrations of atoms and bonds in a material structural grid. These data contain information about the forces acting within structural units.

The disadvantage of these methods is providing comprehensive information about spatial distribution of atoms in crystals only. They are uninformative for investigating complex heterogeneous materials (for example, disordered semiconductors).

We consider which analytical methods evaluate structure features of solids-state materials. These methods mathematically process the results obtained by physical methods. Since a structure of a material surface reflects a structure of a material bulk, surface images obtained by scanning probe microscopy are often used as initial data for processing and analysis of structural features. The main analytical methods to evaluate structural features of solid-state materials are: calculation of the autocorrelation function (ACF), Fourier transform, wavelet transform, calculation of surface roughness.

ACF of any signal helps to determine a degree of difference between the signal and its time-shifted (space-shifted) copy. Therefore, ACF is a good tool at detecting the structure of linear signals.

The Fourier transform is based on the principle that almost every periodic signal can be represented by a sum of individual harmonic components. However, for irregular systems, including material surfaces, Fourier spectra are wide and continuous and decay as a degree of frequency. Therefore, it does not contain useful components in form of resonant frequencies and does not provide significant clarity to understand behavior of nonlinear systems.

The idea of the wavelet transform of an arbitrary signal is its decomposition into a series of soliton-like functions (basis wavelets). The wavelet transform is used, for example, in solving problems of filtering a signal, distorted by a noise [7]. It should be noted, that the wavelet transform is useful when it is necessary to control visually presence of the certain (given) structures on a surface. However, in the case of investigating disordered systems, the method is not informative.

Surface roughness can be determined by calculation of its various types [8]. More complete characterization of a roughness is the height-height correlation function (HHCF). HHCF graphs helps to determine the roughness coefficient, which describes a surface fractality. Also it helps to control dynamic of surface roughness changes during a film growth. However, in complex structures, it is impossible to distinguish an individual noise unit, that is a structural component of a surface. In addition, calculation of roughness parameters is based on using average data of surface heights. So the spatial scale at which certain features of a surface appear is not considered.

Thus, all real solid-state structures (surfaces) can be considered as complex structures, since there is contribution of noise components and various defects of a structure. The disadvantages of the considered analytical methods contributed to appearance of methods for evaluating structural features of solid-state

materials with different structural complexity (both ordered and disordered). The concept of "structural complexity of a surface" is highlighted in [1].

Recently, there are a lot of methods of image texture recognition becoming widespread. A texture means a surface image area with homogeneous statistical characteristics [9]. In terms of frequency features, textures can be coarse-grained, fine-grained, granular, smooth, chaotic, ruled, mottled, irregular, hilly and other. All these characteristics are expressed by a sign of tone non-derivative elements or spatial interaction between them. The Gabor filter bank is a pseudo-wavelet filter bank [10,11], in which each filter generates a virtually independent state of a local frequency content.

Statistical features of a texture are also used [12]. They are calculated as measures of homogeneity by a one-dimensional histogram of signal values (characteristics of the 1st order) and by a two-dimensional histogram of signal values (characteristics of the 2nd order). One way is to describe textures on the basis of Haralick features [13,14], who proposed to describe a texture, covering an image, by characteristics of the special occurrence matrices, calculated using the image. The matrix of joint occurrences provides calculating about twenty features. The most used of them are:

- energy (a measure of image inhomogeneity);
- contrast (a measure of local intensity changes of texture heights [15]);
- correlation (between elements of a matrix);
- dispersion;
- reverse moment of difference;
- average amount (an average height value of a texture image);
- sum of deviations (a measure of distribution showing probability of greater deviation from the average in one direction or another);
- entropy sum (a measure showing randomness in distribution of the average sum of texture heights);
- entropy (a measure showing randomness in brightness distribution of an image);
- difference dispersion;
- the difference between the entropy (measure of reduction of uncertainty);
- information measure of correlation (a measure of dependence between two random variables x and y, defined as a function of information amount in one random variable, relative to other [16]).

The next group of methods is based on calculation and analysis of information-correlation characteristics. The method of average mutual information [6,17] estimates ordering and information capacity of a structure. The method of two-dimensional detrended fluctuation analysis (2D DFA) and its application are described in [17,18]. Input information for 2D DFA method is three-dimensional data on a relief of a surface. In most cases, for real solid-state materials, the relief height fluctuations increase with increasing spatial scale (spatial scale is a number of fragments of original image dividing). Commonly, the fluctuation function F(s) of the spatial scale s is described by a power dependence, so the graph is built on a logarithmic scale. This graph is helps to find the values of s, corresponding to the breakpoints of F(s). The spatial scales are converted into values of correlation vectors, corresponding to the periods of harmonic components in a material surface structure.

There are cases, when the investigated structure has a lot of harmonic components, and it is difficult to determine the breakpoints on the fluctuation – spatial scale dependence due to their merging. For this case, a technique for studying self-organizing structures on the basis of the scale space method (scale-Space DFA [19]) was developed. The term "scale-space" was introduced by A.P. Witkin, when he proposed a method for processing one-dimensional signals by convolution with the Gaussian kernel [20]. Further development of the scale space method led to demonstrating of its application for the two-dimensional case, in particular, for processing raster images [21].

By the Scale-Space DFA an image of a complex structure can be decomposed into simpler components using a smoothing filter at different smoothing coefficients. This results in several dependencies of fluctuation – spatial scale. These dependencies provide simple determination of all breakpoints in the fluctuation function, corresponding to the periods of harmonic components in a surface structure of an investigated material.

3. Algorithm for determining the surface structural complexity of solid-state materials

In this paper, we developed an algorithm for determining the surface structural complexity of solid-state materials, based on the Scale-Space DFA method (Figure 1).

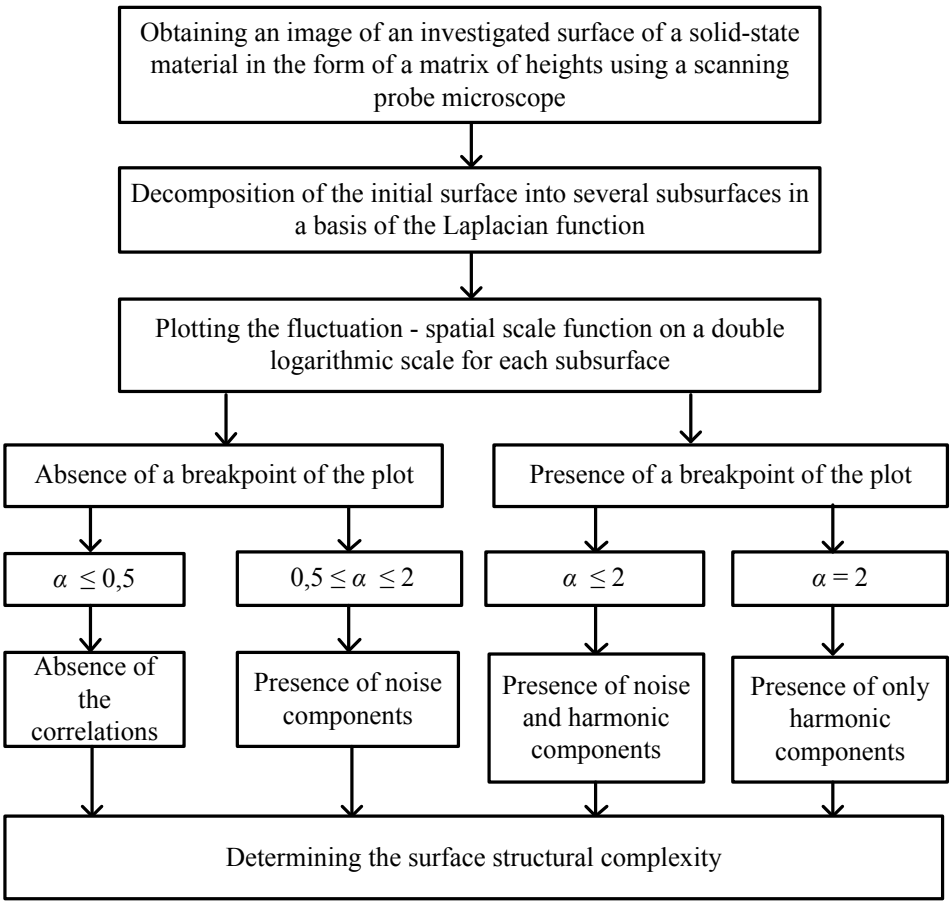

Figure 1: Algorithm for determining the surface structural complexity of solid-state materials

The steps of the algorithm are:

1. Obtaining a surface relief of a material, grown in technological process under specified modes.
2. Acquisition of surface data by scanning probe microscopy in form of a matrix of surface heights. Size of the matrix equals size of the scanned area.
3. Filtering of the height matrix by a smoothing filter in several passes at different smoothing coefficients, which determine scale of the initial surface specification in the basis of the Laplace function. The result is multiple surfaces with different degrees of specification. Each subsequent image from the original one represents the result of differentiation by the second derivative. The last image is the most smoothed and visually different from the original one. However, it usually contains some features, that images close to the original one represent. This way helps, for example, to eliminate various noises. Thus, a single surface can contain many subsurfaces, whose properties can be compared to simple models.
4. Calculation of the fluctuation – spatial scale function for each subsurface by the 2D DFA method.
5. Finding breakpoints on the fluctuation function – spatial scale function. These points determine stable correlation properties for a given scale, by linking the position of the breakpoints along the scale axis with the periods of harmonic components in a surface structure.

6. Calculating tangent of the angle of the fluctuation function (scaling index α) before and after a breakpoint, to estimate type of correlation in a given scale range. In the absence of breakpoints, only a slope of the fluctuation function in all scale ranges is calculated.

7. Analysis of the surface structural complexity by values of the scaling index, number and positions of the breakpoints.

Thus, the surface structural complexity can be classified according to values of tangent of the angle of the fluctuation – spatial scale function and number of breakpoints on this function.

The surface parameters and characteristics obtained using the presented method will be primarily useful for analysis of complex solid-state materials. The structure of such materials contains a large number of harmonic and noise components. Visually (for example, by analyzing the image of the surface obtained using an atomic force microscope) in such materials, it is difficult to reveal features of the structure. Therefore, the proposed algorithm will be useful for analyzing complex surfaces. The advantage of this technique is that it takes into account the spatial scale at which correlations exist in the structure.

Also the Scale-Space DFA method in combination with the average mutual information (AMI) method [17] helps to:

- evaluate degree of ordering of a surface structure;
- calculate information capacity of systems;
- detect local defects in a surface structure;
- fix a change in periodicity in a surface structure;
- calculate correlation vectors (periods of harmonic components) in a surface structure.

4. Experimental results

To test the developed algorithm we used the images of surface obtained by the authors [1]. These structures are Si layers implanted with Ag+ ions subjected to pulsed laser annealing. The surface morphology of the samples was studied using atomic force and scanning electron microscopes. Pulsed laser annealing led to formation of the complicated surface relief. There are small (5-15 nm) and large (40-60 nm) silver nanoparticles on the surface [1]. Therefore, such structures are appropriate for a research by the proposed algorithm for determining the surface structural complexity.

Figure 2 shows the results of surface image processing using the Scale-Space DFA technique. The fluctuation function – spatial scale dependences are presented for the original and the filtered image at some smoothing coefficients. Coefficient 160 corresponds to the minimum smoothing, coefficient 2 corresponds to the maximum smoothing.

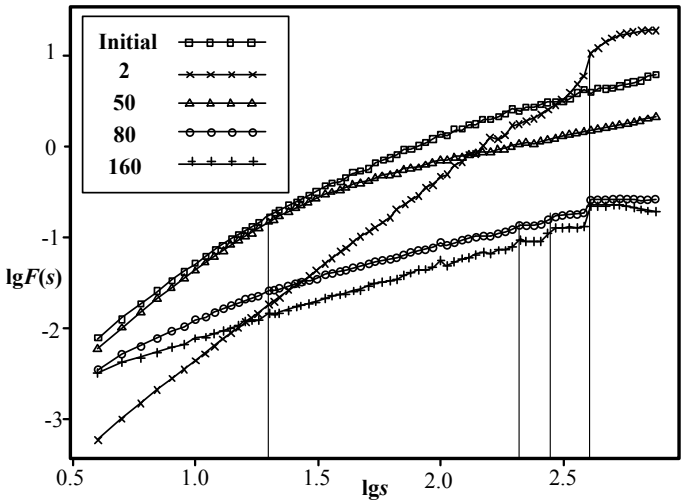

Figure 2: The fluctuation functions – spatial scale dependences for different smoothing coefficients

For the initial surface image, it is quite difficult to distinguish the breakpoints on the fluctuation function – spatial scale dependence. Filtering the image with a smoothing filter resulted in appearance of breakpoints on the charts. The breakpoints of the fluctuation function correspond to the presence of harmonic components in the surface structure. The observed breakpoints at different spatial scales. These scales were recalculated to the values of correlation vectors d, that reflect the values of periods of harmonic components in the structure. We obtained the following d values: 19±5 nm; 200±5 nm; 266±5 nm; 398±5 nm. According to the surface image of the sample, one can conclude that d=19±5 nm corresponds to the average period in the arrangement of small Ag nanoparticles. Values d=200±5 nm; 266±5 nm; 398±5 nm correspond to the periods in the arrangement of large Ag nanoparticles.

The values of the scaling index are also calculated for the original image left to the breakpoint with the smallest scale value and right to the breakpoint with the largest scale value. Accordingly, the following values of α are 0.6 and 1.8.

Thus, by analyzing number of the breakpoints and the values of α, we can conclude that the studied surface belongs to the type of surfaces with a high structural complexity.

In addition, the scaling index – smoothing coefficient dependence was obtained (Figure 3). As the figure shows the type of the dependence is not monotonous. When the coefficients are of 0-37 α is in the range of 1.6-2.1. These high values are due to the fact that maximum smoothing occurs at small scales. In this case, the harmonic component of the structure prevails. Then there is a decline of α to the scale 95. This means the transition from the harmonic component to the noise component. Then the value α becomes stable (at 0.8). This indicates that the noise component prevails at these scales.

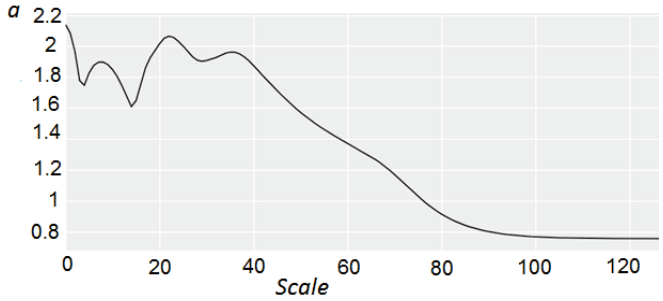

Figure 3: The scaling index – smoothing coefficient (scale) dependence

Using this plot allows studying the dynamics of changes in the correlation properties of the surface. In particular, it is possible to identify features of noise and harmonic components in the structure.

5. Conclusion

Analysis of research methods of structure features of solid-state materials showed that development of complex surfaces processing techniques of solid-state materials is an actual task.

The algorithm for determining the surface structural complexity of solid-state materials based on the Scale-Space DFA method is developed. It is proposed to evaluate the surface structural complexity by tangent of the angle of the fluctuation – spatial scale function and determining number of breakpoints on the fluctuation – spatial scale function. Surfaces can be classified by the type of the structural complexity. The advantage of the developed algorithm is that it can be used to determine the spatial scale at which correlations exist in a structure.

The developed algorithm was tested on the surface image of Si layer implanted with Ag+ ions, subjected to pulsed laser annealing. The investigated surface belongs to type of surfaces with a high structural complexity.

It was also shown that the scaling index – smoothing coefficient (scale) dependence can be used to study the dynamics of changes in the correlation properties of the surface.

Knowledge about surface structural complexity of solid-state materials can be used to determine the material functional purpose in different fields of science. The developed algorithm can be used in materials science, micro-and nanoelectronics, optoelectronics, etc. First of all, the Scale-Space DFA technique is useful for analyzing complex surfaces with a large number of harmonic and noise

components. Moreover, the Scale-Space DFA method in combination with the AMI method provides solving a wide range of scientific and practical problems, such as evaluating degree of ordering of a surface structure; calculating information capacity of systems; detecting local defects in a surface structure; detecting changes in periodicity in a surface structure; calculation of correlation vectors in a surface structure.

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7. References

[1] R.I. Batalov, V.V. Vorobev, V.I. Nuzhdin, V.F. Valeev, D.A. Bizyaev, A.A. Bukharaev, R.M. Bayazitov, Yu.N. Osin, G.D. Ivlev, and A.L. Stepanov, "The effect of pulsed laser radiation on a Si layer with a high dose of implanted Ag+ ions", Optics and Spectroscopy, vol. 125, No. 4, 2018, pp. 571-577.

[2] G. Maity, S. Ojha, I. Sulania, K. Devrani, and P. Patel Shiv "Fractal characterizations of energetic Si ions irradiated amorphized-Si surfaces", Surface and Interface Analysis, vol. 51, 2019, pp. 817-825.

[3] R. Mikšová, P. Horák, V. Holý, and A. Macková, "Study of thermal recrystallisation in Si implanted by 0.4 MeV heavy ions", Surface and Interface Analysis, vol. 51, 2019, pp. 1113-1120.

[4] D.G. Castner, "Surface analysis: From single crystals to biomaterials", Surface and Interface Analysis, vol. 5, 2018, pp. 981-990.

[5] Yu.K. Egorov-Tismenko,Kristallografiya i kristallohimiya, M.: KDU; 2005.

[6] S.P. Vikhrov, T.G. Avacheva, N.V. Bodyagin, N.V. Grishankina, and A.P.Avachev, "Determination of the degree of ordering of materials' structure by calculating the information-correlation characteristics", Semiconductors, vol. 46, No. 4, 2012, pp. 415-421.

[7] S.Sh. Rekhviashvili, "Wavelet-transform processing of images in atomic force microscopy", Technical Physics Letters, vol. 28, No. 3, 2002, pp. 237–238.

[8] GOST 25142-82 SHerohovatost' poverhnosti. Terminy i opredeleniya. http://vsegost.com/Catalog/43/43751.shtml.

[9] A.A. Potapov, "Novye informacionnye tekhnologii na osnove veroyatnostnyh teksturnyh i fraktal'nyh priznakov v radiolokacionnom obnaruzhenii malokontrastnyh celej", Radiotekhnika i elektronika, vol. 48, No. 9, 2003, pp. 1101-1119.

[10] B.S. Manjunath, and W.Y. Ma, "Texture features for browsing and retrieval of image data', IEEE TPAMI, vol. 18, No. 8, 1996, pp. 837-842.

[11] M. Idrissa, and M. Acheroy, "Texture classification using Gabor filters", Pattern Recognition Letters, vol. 23, No. 9, 2002, pp. 1095-1102.

[12] R.M. Haralick, "Statistical and structural approaches to texture", Proceedings of the IEEE, vol. 67, No. 5, 1979, pp. 786-804.

[13] Yu.V. Konkin, and A.N. Kolesenkov, "Raspoznavanie izobrazhenij na osnove teksturnyh priznakov Haralika i iskusstvennyh nejronnyh setej", Izvestiya TulGU, vol. 2, 2016, pp. 117-123.

[14] R.M. Haralik, K. Snanmugam, and I.H. Dinsten, "Textural features for image classification", IEEE TSMC, vol. 6, 1973, pp. 610-621.

[15] F. Albregtsen, "Statistical texture measures computed from gray level cooccurence matrices", Image Processing Laboratory Department of Informatics University of Oslo, 2008.

[16] Sposob raspoznavaniya izobrazheniya tekstury kletok. www.freepatent.ru/patents/2385494.

[17] A.V. Alpatov, S.P. Vikhrov, N.V. Vishnyakov, S.M. Mursalov, N.B. Rybin, and N.V. Rybina, "On a combined approach to studying the correlation parameters of self-organizing structures", Semiconductors, vol. 50, No. 1, 2016, pp. 22-28.

[18] A.V. Alpatov, S.P. Vikhrov, A.G. Kazanskii, V.L. Lyaskovskii, N.B. Rybin, N.V. Rybina, and P.A. Forsh, “Study of the correlation properties of the surface structure of nc-Si/a-Si:H films with different fractions of the crystalline phase”, Semiconductors, vol. 50, No. 5, 2016, pp. 590-595.
[19] A.V. Alpatov, and N.V. Rybina, “Application of the Scale-Space method in studying self-organizing structures”, N.V. J. Synch. Investig., vol. 13, No. 4, pp. 652-657.
[20] A.P. Witkin, “Scale-space filtering”, Proc. 8th Int. Joint Conf. Art. Intell., vol. 2, 1983, pp. 1019-1022.
[21] J.J. Koenderink, “The structure of images”, Biol. Cyb., vol. 50, 1984, pp. 363-370.