===== **SUPERCONDUCTIVITY** =====

Fractal Dimension of Cluster Boundaries in Porous Polycrystalline HTSC Materials

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Abstract—The fractal dimension of the boundaries of clusters formed by pores and granules in polycrystalline materials is shown to be determined by the sample density and crystallite sizes. The dependence of the fractal dimension on the density has a maximum. It is shown that the maximum diamagnetic response can be obtained in a porous high-temperature superconductor with a porosity of 50−60% and small crystallite sizes.

1. INTRODUCTION

The presence of various defects (cracks, pores, vacancies, and caverns) influences the transport characteristics of polycrystalline materials. In terms of the percolation model [1−5], this influence in porous structures and composite materials was studied in many works [6−11]. The percolation theory holds that the transport properties of a material are influenced by the particle shape and size along with the physical density [12]. A series of experiments [6−11] were performed to construct the dependences of the transport parameters on the density that can be easily controlled. There are also works in which structures with both the random Gaussian and fractal distributions of material particles over the sizes were studied by computer simulation [13]. Based on satisfactory correlation with the experiments, the authors of [13] showed that the computer simulation can be an effective tool for predicting the transport properties of various materials. In all the aforementioned works, an analysis was made of model isotropic materials consisting of particles with a simple configuration (ball and cube) and a certain particle size.

The specific features of the structure of many disordered natural materials force to use the fractal geometry for their description [14]. The geometry was designed by Mandelbrot to describe complex natural objects, and its peculiarity is the topological dimension that differs from the dimension in Euclidean geometry in that it can be a fractional number. In two-dimensional systems, the relationship between the area and perimeter $S \propto P^D$ is valid [14]. The fractal dimension D is a numerical parameter characterizing the degree of boundary kinking. Kuzmin [15, 16] described the pinning on fractal boundaries of the normal phase clusters in two-dimensional superconducting films. In polycrystalline superconductors, both the current percolation over superconducting clusters and percolation of the Abrikosov vortices over non-superconducting phase clusters (normal inclusions, pores, etc.) take place. In real samples, a spatially complicated arrangement of normal and superconducting clusters can form. However, the percolation over such chaotic systems is adequately described in the simple two-dimensional model of percolation over parallel chains [17]. Thus, the Kuzmin model is applicable to bulk samples as well. The transport properties of high-temperature superconductors (HTSCs) with a porous structure are adequately described in the framework of the model of pinning on normal phase clusters with fractal boundaries [18]. The current−voltage characteristics of the materials under consideration are found to be dependent on the fractal dimension of projections of intergranular boundaries.

The transport parameters of a superconductor are dependent on the internal structure of a real sample, and the parameters of a foamed state are substantially different from those of densely compacted HTSCs [19]. It is evident that the effective cross section of a sample with a porous structure is less than the cross-section area of a dense sample, and the trajectory of passing the electric current is, conversely, larger, since the trajectory is not a straight line in the Euclidean space [17].

In this work, we simulate the dependence of the fractal dimension on the structure of porous materials with variable density and average size of plane rectangular-shaped crystallites (which qualitatively corresponds to real microcrystals). This allows us to study the influence of the internal geometric parameters on the transport properties of porous polycrystals.
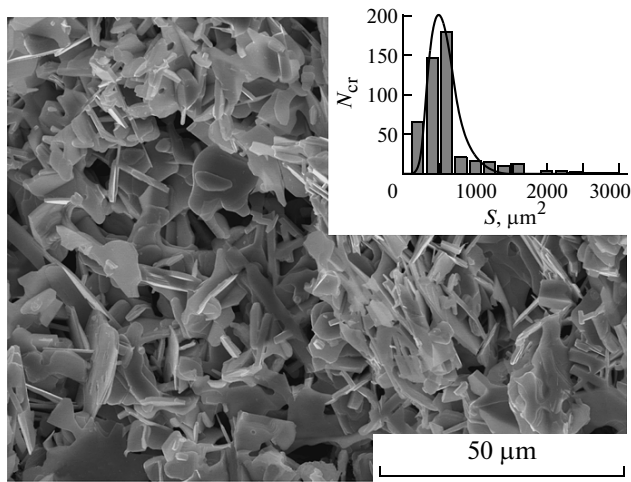
Fig. 1. Micrograph of the structure of the porous HTSC Bi2223 with the density accounting for 22% of the theoretical density. The inset shows the area distribution of crystallites of the porous HTSC Bi2223.

Fig. 2. Simulated structure imitating the cross section of the polycrystal with the filling density corresponding to the physical density of 22%.

2. ANALYSIS OF THE DIMENSION OF THE BOUNDARIES OF A POROUS HTSC

Figure 1 shows the electron microscopy image of the porous $Bi_2Sr_2Ca_2Cu_3O_x$ (Bi2223) HTSC sample with plate-like particles and a density of 22%.

It is seen from the figure that the structure of the porous HTSC is a set of randomly oriented microcrystals contacting each other. All the microcrystals have the thickness of almost 2 μm, and their extent can vary over rather wide limits. Intergranular boundaries are identified as zones of a high contrast as the micrograph contrast is varied (Fig. 1). We choose the contrast at which the summary area of crystallites mapped after using threshold filtrations was maximal. It, in turn, allows the identification of microcrystals themselves and determination of their size distribution function.

The inset to Fig. 1 shows the area distribution of the porous HTSC crystallites. The size distribution of crystallites is described by a lognormal law (solid line in the inset), and the average crystallite size is 25 μm. Using the micrographs, we determine the fractal dimension of the intergranular boundary projections from the known relationship between the area and perimeter of a figure with an irregular contour $S \propto P^D$. The micrographs were covered by a network of squares; in this case, we computed the number of squares having the boundaries in the interior. The fractal size is equal to the slope of the dependence of the number of intersections on the square size in the double logarithmic coordinates [14]. As a result, the dimension of the intergranular boundary projections was obtained to be $D = 1.67$.
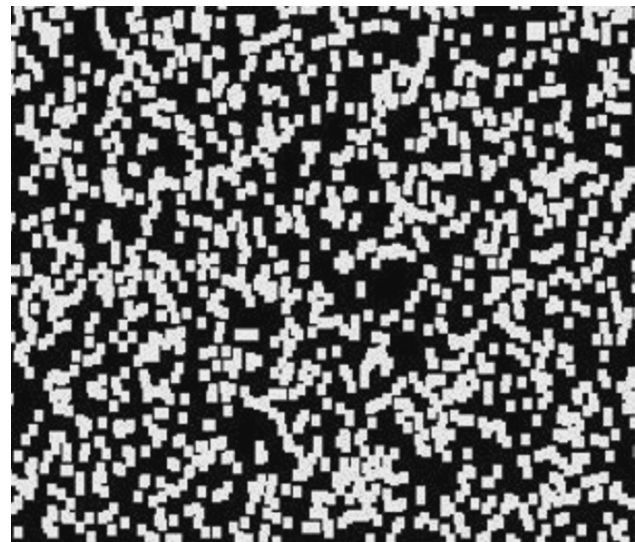
3. DESCRIPTION OF THE MODEL

The process of simulating consists of a random arrangement in a two-dimensional discrete finite space of squares imitating crystallites of a material with given sizes. The arrangement was performed by a uniform white noise (Fig. 2), assuming the isotropy of the density in the material cross section, which reflects the absence of the gradient of the material concentration in the real sample.

The figure obtained is a rectangular field containing 1000×1000 points that is filled with squares of given sizes imitating crystallites with increasing density. The fractal dimension of the boundaries in the structure prepared was analyzed by the method of covering with squares of various scales described in Section 2. Since the boundary between individual noncomplanar two-dimensional crystallites is one-dimensional ($D = 1$), the fractal dimension of the whole boundary increases with an increase in the disorder and reaches the value $D = 2$ in the limiting case.

Varying the crystallite density and size, we calculated the fractal dimension of the boundaries of the simulated porous structure of a discrete space of the array (Fig. 2). The relative size of the squares is related to the real crystallite size through the scale of SEM micrographs of the real structure of the sample. The dependences obtained are depicted in Fig. 3 as the $D(\rho, L/L^*)$ diagram. The fractal dimension $D = 1$ at $\rho = 0$ or 100%. The densities ρ close to zero are physically abstract, and they are presented for completeness of the picture.

We call attention to the extreme behavior of the dependence of the fractal dimension on the density at almost any values of the relative crystallite sizes. It was shown experimentally [20] that the dependence of the
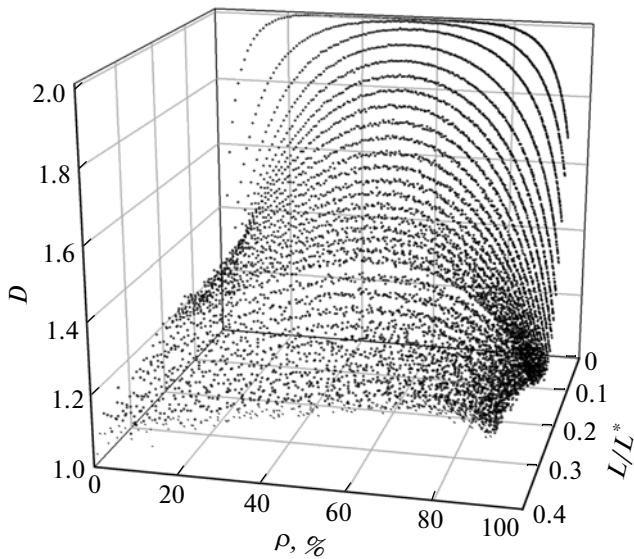
Fig. 3. Model dependence of the fractal dimension D on the density ρ and average crystallite size L.
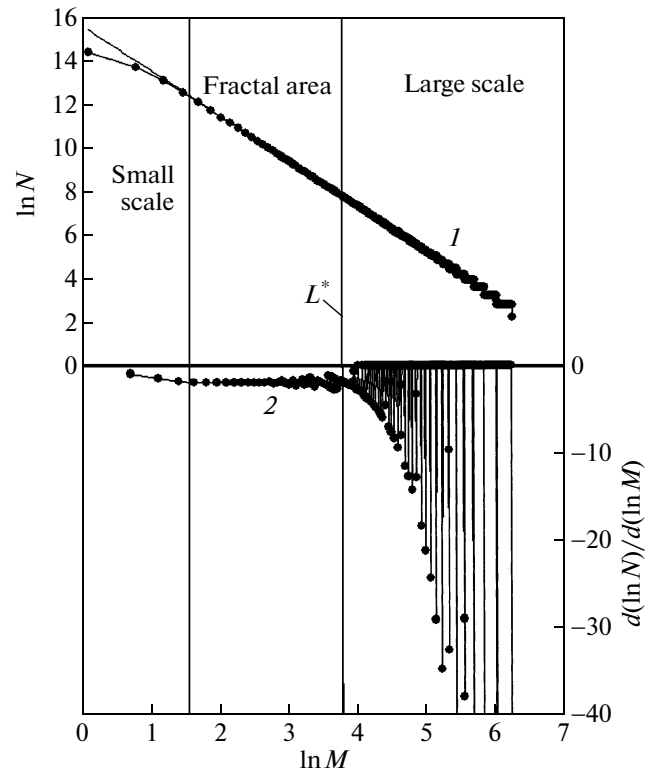

Fig. 4. (*1*) Dependence of the number of intersections of the boundary projections and squares plotted in the double logarithmic coordinates and (*2*) its derivative $d(\ln N)/d(\ln M)$ that allows the determination of the region in which the self-similarity takes place. The self-similarity region is between the regions of large scales and small scales in which the dependence of $\ln N$ on $\ln M$ ceases to be linear.

fractal dimension also has a maximum over wide range of porosities of the ceramics, and this fact confirms the model proposed.

In Fig. 3, the size is normalized to L^* that is the effective scale of the microstructure at which the fractality effects begin to appear. It reflects the fact that all natural porous structures exhibit the self-similarity properties in a limited range of scales of observations, unlike the mathematical fractal. At $L \geq L^*$, the boundary dimension D is unity independently of the parameters. Figure 4 shows the results of determination of the cluster dimension from the micrograph (Fig. 1) using the method described in Section 2. Figure 4 allows us to determine the range of observation scales in which the structure self-similarity law is valid and to understand the physical sense of the parameter L^*.

As the mesh size is small, the crystallites are large with respect to the square size, and a larger part of the image is occupied by the surface of the crystallites themselves. We do not observe any kinks of the boundary projections. On the other hand, as the observation scale is larger than L^*, the mesh size significantly exceeds the crystallite size, the boundaries are smeared, and an excess noise appears in the dependence of $d(\ln N)/d(\ln M)$ on $\ln(M)$ that determines the fractal region boundary.

4. DISCUSSION OF THE RESULTS

Based on the model proposed, the polycrystal structure was simulated with the parameters similar to those of the real porous HTSC considered in Section 2. The density of 22% and average crystallite size of 25 μm correspond to the fractal dimension of the boundaries $D = 1.65$ that is close to the value obtained

from an analysis of the micrographs of the porous HTSC. Possible discrepancies can be due to the absence of the contribution from the change in the actual size of granules under simulation during the arrangement when fine particles are "sintered" into coarse conglomerates with increasing sample density.

The intergranular boundary dimension influences the transport properties of a superconducting material with fractal boundaries of normal clusters. According the Kuzmin theory, the current−voltage characteristics of superconducting films are determined by values of the flow resistance R_f, critical current I_c, and fractal dimension D [3].

Figure 5 shows the complete diagram calculated by the Kuzmin model that demonstrates the influence of the fractal dimension on the transport properties of HTSC.

It is seen from Fig. 5 that, as the fractal dimension increases, the voltage drop decreases with increasing transport current; i.e., the stronger the intergranular boundary is developed, the stronger the pinning and the higher the current-carrying ability of a material, which coincides with the theory statements [16].
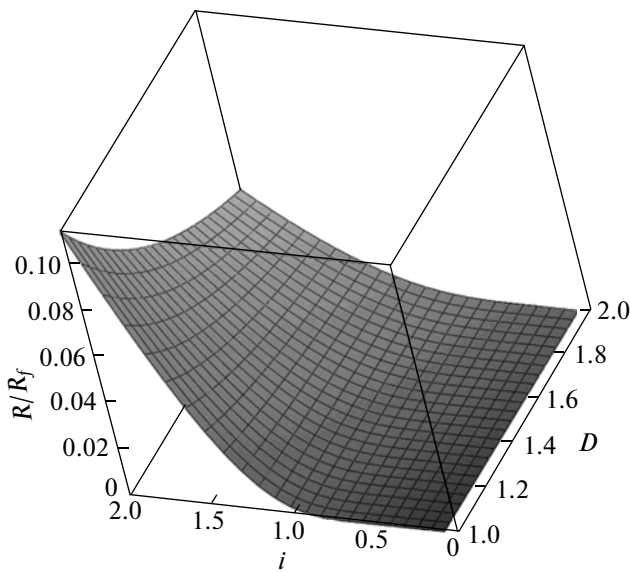
Fig. 5. $D(R/R_f, i)$ diagram. Designations: R_f is the flow resistance [15, 16]; R is the resistance; $i = I/I_c$ is the transport current normalized to the critical current; and D is the fractal dimension.

Thus, prescribing the porosity of materials and the average crystallite size, we can obtain the fractal dimension of the intergranular boundary that immediately influences the transport properties of a superconductor. According to the diagram (Fig. 3) the maximum fractal dimension is observed at the minimal crystallite size in the density range 20−80%. The density range with high fractal dimensions ($D > 1.8$) is sharply narrowed to 50−60% with increasing the crystallite size.

5. CONCLUSIONS

The fractal dimension of the cluster boundaries that determines the development of the intergranular boundaries is dependent on the physical density and granule sizes in polycrystalline materials. The dependence of the fractal dimension on the physical density of a material has a maximum at any values of the average crystallite size. To obtain the maximum critical current in a porous HTSC, it is necessary that its porosity would be 50−60%, and the crystallite sizes would be minimal.

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