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Simulation of dc conductance of two-dimensional heterogeneous system: application to carbon wires made by ion irradiation on polycrystalline diamond

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A percolation model of two-dimensional heterogeneous system composed of two conductive phases and a method for calculating the macroscopic electric conductance of such a system in direct current regime is proposed. The parameters of the model are its geometrical dimensions, the conductance of the two phases and the relative fractions of the phases in the system. The model satisfactorily describes the non-linear dependence on width of conductance of the carbon nano- and microwires made on polycrystalline diamond surface by focused ion beam irradiation.

1 Introduction

One of the methods describing physical properties of heterogeneous systems is the percolation theory [1]. In its standard formulation a heterogeneous system is described as a network of nodes connected with bonds. A part of the bonds is randomly removed and, at a certain critical amount of the removed bonds, a percolative phase transition takes place: the system breaks up into separate non-connected clusters and its properties change abruptly. Recently the percolation theory and its various modifications were used to solve a wide range of problems such as simulation of formation of porous materials and polymer degradation [2], description of the metal–insulator transition in systems for which the perturbation methods cannot be used [3, 4], calculations of the electric conductivity of heterogeneous systems including thin films [5].

Despite the progress made in the studies of heterogeneous systems, a number of phenomena occurring in such systems still remain obscure. One of them is the non-linear conductivity of amorphous carbon nano- and micro-wires made on the surface of polycrystalline CVD diamond films by focused 100 keV Ga^+ ion beam at irradiation doses of the order of 10^{16} cm^{-2} [6, 7].[1] The wires had equal length of $l = 10$ μm and width, which varied from $w = 0.1$ to 10 μm. The thickness of the irradiation amorphized layer (as determined by TRIM-code simulations) was about 40 nm. DC conductance of the wires was measured in as-irradiated amorphous state and after annealing at 1300 °C, when the amorphous carbon was converted

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[1] In a recent paper [8] it is shown that the conductivity of similarly fabricated nano-wires also decreases non-linearly with length for fixed width.

into graphite. The conductance of the pristine non-irradiated films was negligibly small. The ion irradiation resulted in a moderate conductance characteristic of the ion-irradiated amorphous carbon. It was found that the conductance of the as-irradiated wires depended on the wire width super-linearly.[2] After the annealing the wires conductance increased by 2 to 3 orders of magnitude manifesting the formation of graphite and the non-linear dependence on the width disappeared. No reasonable explanation or quantitative description of the phenomenon were given.

There is no common approach for comprehensive description of the conductance of two-dimensional heterogeneous systems [3]. The percolation theory in combination with the analytical methods describes the properties of the real systems under certain conditions only [4]. In particular, the "infinite" size systems are described as these having only two values of the resistance of their structural elements: 0 or 1 [10, 11]. This leads to zero-conductance for the systems below the percolation threshold.[3] As the ion irradiation of diamond may produce two type of phases distinctively different in terms of electrical conductance, namely graphitized carbon (high dose irradiation) of high electrical conductance and irradiated diamond primarily with point defects (low dose irradiation), which result in a low conductance. Thus we assume that the ion beam made carbon wires described in [7] can be considered as finite size systems composed of randomly distributed nano-islands of two conductive phases of high and low conductivity. A low conductance of the as-irradiated wires indicates that the percolation threshold in them was not reached. Thus, for the description of the system a model is needed which would consider the finite dimensions of the system, its shape, and the conductance of the low-conductive elements. Below we present a simple model that satisfies these conditions, namely a two-dimensional network of randomly distributed resistors of two types with conductance G_1 and $G_2 \gg G_1$ respectively.

2 Model

As the thickness of the ion-implanted conductive layer is comparable with the thermal wavelength of the electrons in graphite, it can be simulated by a two-dimensional conductive network. The network consists of $(m+2)$ rows, numbered from 0 to $(m+1)$ and $(n+2)$ columns, numbered from 0 to $(n+1)$. The nodes of the network are denoted as (i, j), where i is the row number, and j is the column number (Fig. 1). The conductance of the resistor, connecting the nodes (i, j) and $(i+1, j)$ is labelled as $G^{(d)}_{i,j}$, and that of the resistor connecting nodes (i, j) and $(i, j+1)$ as $G^{(r)}_{i,j}$. Each resistor may have conductance either G_1 or $G_2 \gg G_1$.

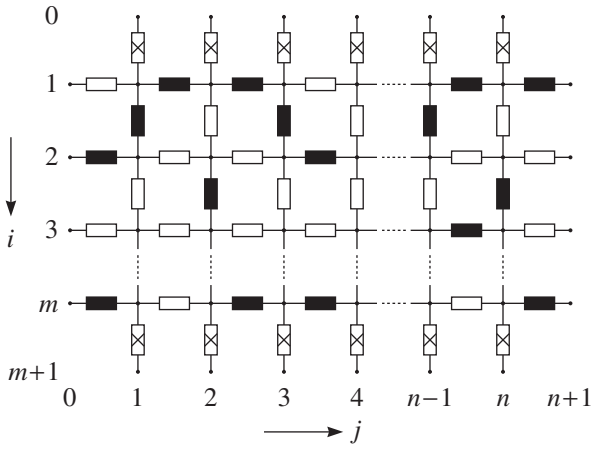

Fig. 1 Two-dimensional resistor network: white and black resistors are low and high conductive resistors respectively. The resistors labelled with crosses have zero conductance (no electrical connection).

[2] It is obvious, that near the percolation threshold the dependence of local electrical conductivity on temperature, caused by the Joule–Lentz heat, as well as its dependence on the local electric field strength [9] can be significant. But in papers [6, 7] these effects, were most likely not deciding due to the large thermal conductivity of diamond.

[3] In [12] an infinite three-dimensional resistor network with a broad distribution of the values of conductances is considered.

3 Calculation algorithm

The following electric potentials are assumed to be applied to the nodes on the network interface:

$$\varphi_{0,j} = P_j\,, \qquad \varphi_{m+1,j} = Q_j\,, \qquad \varphi_{i,0} = S_i\,, \qquad \varphi_{i,n+1} = T_i\,,$$

where $j = 1, \ldots, n$ and $i = 1, \ldots, m$. For each node the total current flowing into the node equals the total current flowing out of it. The top-to-bottom and left-to-right directions of the current flow are taken as positive (Fig. 1). Then the current balance in the nodes is described by the system of linear equations:

$$(\varphi_{i-1,j} - \varphi_{i,j})\, G^{(\mathrm{d})}_{i-1,j} + (\varphi_{i,j-1} - \varphi_{i,j})\, G^{(\mathrm{r})}_{i,j-1} = (\varphi_{i,j} - \varphi_{i+1,j})\, G^{(\mathrm{d})}_{i,j} + (\varphi_{i,j} - \varphi_{i,j+1})\, G^{(\mathrm{r})}_{i,j}\,, \tag{1}$$

where $\varphi_{i,j}$ is the potential of the node (i, j). To solve Eq. (1) all the unknown potentials $\varphi_{i,j}$ are written into a column X_ν as follows:

$$X_{(i-1)n+j} = \varphi_{i,j}\,, \qquad i = 1, \ldots, m\,, \qquad j = 1, \ldots, n\,. \tag{2}$$

Using Eq. (2) the system Eq. (1) can be rewritten in matrix form:

$$\sum_{\nu=1}^{m\times n} A_{\mu,\nu} X_\nu = B_\mu\,, \qquad \mu, \nu = 1, ..., m \times n\,, \tag{3}$$

where the square matrix $A = (A_{\mu,\nu})$ is given in block form of $m \times m$ blocks:

$$A = \begin{pmatrix} M_1 & L_1 & 0 & 0 & \cdots & 0 & 0 \\ L_1 & M_2 & L_2 & 0 & \cdots & 0 & 0 \\ 0 & L_2 & M_3 & L_3 & \cdots & 0 & 0 \\ 0 & 0 & L_3 & M_4 & \ddots & 0 & 0 \\ \vdots & \vdots & \vdots & \ddots & \ddots & \ddots & \vdots \\ 0 & 0 & 0 & 0 & \ddots & M_{m-1} & L_{m-1} \\ 0 & 0 & 0 & 0 & \cdots & L_{m-1} & M_m \end{pmatrix}. \tag{4}$$

In the matrix (4) only the blocks on the main diagonal and those adjacent to the main diagonal are non-zero matrices. Each of the non-zero blocks in the matrix A is a square symmetric matrix of the size $n \times n$. The matrices L_i are diagonal ones with elements:

$$L_i = \mathrm{diag}\,(G^{(\mathrm{d})}_{i,1}, G^{(\mathrm{d})}_{i,2}, G^{(\mathrm{d})}_{i,3}, ..., G^{(\mathrm{d})}_{i,n})\,. \tag{5}$$

The matrices M_i are three-diagonal ones:

$$M_i = \begin{pmatrix} -W_{i,1} & G^{(\mathrm{r})}_{i,1} & 0 & 0 & \cdots & 0 & 0 \\ G^{(\mathrm{r})}_{i,1} & -W_{i,2} & G^{(\mathrm{r})}_{i,2} & 0 & \cdots & 0 & 0 \\ 0 & G^{(\mathrm{r})}_{i,2} & -W_{i,3} & G^{(\mathrm{r})}_{i,3} & \cdots & 0 & 0 \\ 0 & 0 & G^{(\mathrm{r})}_{i,3} & -W_{i,4} & \ddots & 0 & 0 \\ \vdots & \vdots & \vdots & \ddots & \ddots & \ddots & \vdots \\ 0 & 0 & 0 & 0 & \ddots & -W_{i,n-1} & G^{(\mathrm{r})}_{i,n-1} \\ 0 & 0 & 0 & 0 & \cdots & G^{(\mathrm{r})}_{i,n-1} & -W_{i,n} \end{pmatrix}, \tag{6}$$

where

$$W_{i,j} = G^{(\mathrm{d})}_{i-1,j} + G^{(\mathrm{r})}_{i,j-1} + G^{(\mathrm{d})}_{i,j} + G^{(\mathrm{r})}_{i,j} \,. \tag{7}$$

The free-term column $B = (B_\mu)$ in Eq. (3) can also be written in block form:

$$B = \begin{pmatrix} b_1 \\ b_2 \\ \vdots \\ b_m \end{pmatrix}, \tag{8}$$

with m block-columns of n elements each. The first and the last blocks have the form:

$$b_1 = \begin{pmatrix} -P_1 G^{(\mathrm{d})}_{0,1} - S_1 G^{(\mathrm{r})}_{1,0} \\ -P_2 G^{(\mathrm{d})}_{0,2} \\ -P_3 G^{(\mathrm{d})}_{0,3} \\ \vdots \\ -P_{n-1} G^{(\mathrm{d})}_{0,n-1} \\ -P_n G^{(\mathrm{d})}_{0,n} - T_1 G^{(\mathrm{r})}_{1,n} \end{pmatrix}, \qquad b_m = \begin{pmatrix} -Q_1 G^{(\mathrm{d})}_{m,1} - S_m G^{(\mathrm{r})}_{m,0} \\ -Q_2 G^{(\mathrm{d})}_{m,2} \\ -Q_3 G^{(\mathrm{d})}_{m,3} \\ \vdots \\ -Q_{n-1} G^{(\mathrm{d})}_{m,n-1} \\ -Q_n G^{(\mathrm{d})}_{m,n} - T_m G^{(\mathrm{r})}_{m,n} \end{pmatrix}. \tag{9}$$

All other blocks have the form:

$$b_i = \begin{pmatrix} -S_i G^{(\mathrm{r})}_{i,0} \\ 0 \\ 0 \\ \vdots \\ 0 \\ -T_i G^{(\mathrm{r})}_{i,n} \end{pmatrix}, \qquad i = 2, \ldots, m-1 \,. \tag{10}$$

To find the macroscopic conductance G of the network the system of $m \times n$ linear Eqs. (3) has to be solved.

For further considerations we shall assume the absence of conductance between some nodes: the bonds between the 0-th and $(m+1)$-st rows and the rest of the network are broken (marked with crosses in Fig. 1). This corresponds to the absence of contacts on the sides of conductive carbon wires described in [7]. In this case, all the potentials P_j and Q_j do not influence the current flow in the network and they can be set to zero for $j = 1, \ldots, n$. Then all B_μ (see Eqs. (8)–(10)) equal zero, except

$$B_{(i-1)n+1} = -S_i G^{(\mathrm{r})}_{i,0} \,, \qquad i = 1, \ldots, m \,, \tag{11}$$

$$B_{i \times n} = -T_i G^{(\mathrm{r})}_{i,n} \,, \qquad i = 1, \ldots, m \,. \tag{12}$$

If the conductances G_1 and G_2 are non-zero a single solution of the system (3) exists. It means, that the matrix A is non-degenerate. In this case [13]:

$$X_\nu = \frac{D_\nu}{D} \,, \tag{13}$$

where $\nu = 1, \ldots, m \times n$; D_ν is a determinant obtained from $D = \det A$ by exchanging the ν-th column by the free-term column.

Of some interest is the case, when the conductance $G_1 = 0$. Then, the potential of a node surrounded by the zero-conductance resistors is undetermined, which means that $\det A = 0$. There can be several iso-

lated nodes, however, the part of the system (3) describing the non-isolated nodes of the network can be solved. The potentials of the isolated nodes need not be calculated, as the current does not flow through these nodes. A formal approach to solving the problem in this case is to replace the diagonal zero elements of the rows of the matrix A that consist of all zeros with 1 and to solve the modified system of equations. The calculations in the case $G_1 \neq 0$ are similar to the discussed above with the difference that it is not necessary to replace zeros on the diagonal of matrix A with ones as the matrix is non-degenerate.

To obtain the total current in the network (from left to right) it is sufficient to know the potentials on the nodes of the first or the last column. We shall calculate the potentials for the nodes of the last column. The calculations for the first column are similar. The potential of a node at the intersection of the last column and the i-th row of the network is by definition $\varphi_{i,n} = X_{i\times n}$, where $i = 1, \ldots, m$. The current flowing out of this node is given by the expression:

$$I_i^{\text{out}} = G_{i,n}^{(\text{r})}[X_{i\times n} - T_i], \tag{14}$$

$X_{i\times n}$ is calculated using Eq. (3). The potentials, applied to all nodes on the left and the right sides of the network are assumed to be equal:

$$S_i = S, \qquad T_i = T, \qquad i = 1, \ldots, m.$$

Then the total current flowing through the network is

$$I = \sum_{i=1}^{m} I_i^{\text{out}}.$$

The relation between the current I and the conductance of the network G is:

$$I = (S - T)\, G,$$

from which the final expression for the total conductance G is found as:

$$G = \frac{1}{S - T} \sum_{i=1}^{m} G_{i,n}^{(\text{r})} \left[\frac{D_{i\times n}}{D} - T \right]. \tag{15}$$

4 Description of the program

To numerically calculate the expression (15) we have developed an algorithm for calculating the determinants D and D_v and a computer program taking into account the specific properties of the matrix A. (For the techniques of calculations using specific properties of matrix A see e.g. [13].) The program generates a random resistor network. If p is the fraction of the resistors with high conductance G_2, then the fraction of the resistors with low conductance G_1 is $1 - p$. The calculation of the total conductance of the network consists of the following steps:

(i) the matrix A and the free-term column B are formed in accordance with the Eqs. (4)–(7) and Eqs. (11), (12), where it is assumed that $S_i = 1$, $T_i = 0$ for $i = 1, \ldots, m$;

(ii) all diagonal zero elements in the matrix A are replaced by ones;

(iii) the matrix A is transformed to upper-triangular form using the Gaussian elimination. Corresponding transformations of the free-term column B are performed simultaneously;

(iv) the free-term column B is substituted for each $i \times n$ column of the matrix A. The obtained matrix is transformed to upper-triangular form;

(v) determinants of the matrices obtained in steps (iii) and (iv) are calculated. The determinant of the upper-triangular matrix is a product of all terms of the matrix diagonal;

(vi) the network conductance is found using Eq. (15).

The calculations are carried out for networks of different m and fixed n, what corresponds to the carbon wires of different width and equal length. Steps from (i) to (vi) are executed 50 times for each m value. The final value of the conductance is found for each m as an average over 50 calculations.

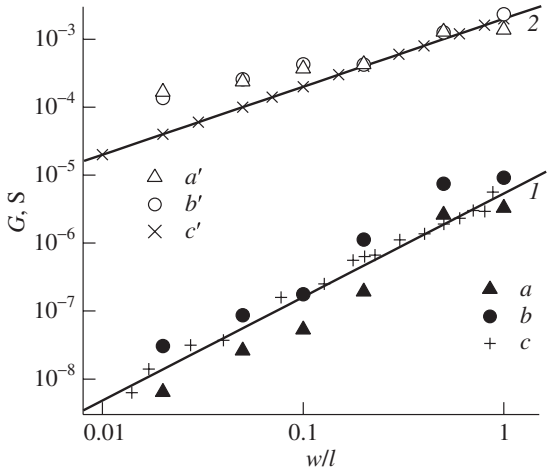

Fig. 2 Conductance G of carbon wires made by Ga^+ ion irradiation on surface of polycrystalline diamond film versus the ratio w/l of their width and length (based on the data from [7]): a, a' – irradiation dose 10^{16} cm^{-2}, b, b' – irradiation dose 2×10^{16} cm^{-2}; a, b – as-irradiated, a', b' – after annealing at 1300 °C. Calculations using the proposed model for $G_1 = 10^{-7}$ S, $G_2 = 2\times10^{-3}$ S: c – $p = 0.48$; c' – $p = 1$. The solid lines approximate the calculated data c, c' with power function (16): 1 – for $p = 0.48$ ($q = 1.55 \pm 0.03$), 2 – for $p = 1$ ($q = 1$).

5 Results

The experimental dependence of the wire conductance on its width taken from [7] is compared with the results of the calculations obtained on the resistor networks with different m. The wire length l and width w correspond to the numbers n and m in the network. The p value is taken as a fitting parameter to get the best agreement between experimental and calculated data. For the as-irradiated wires the best fit has been found for $p = 0.48$, which is below the percolation threshold $p_c = 0.5$ for the bond percolation on the infinite two-dimensional square lattice [11, 14].

For finite-size systems, the percolation threshold is a random number, its average value is equal to the percolation threshold of the infinite system [15]. We have found that for a sufficiently large n (over 300) the conductance G of the network depends only on the ratio (m/n), or the ratio of the width w to the length l of the film.[4]

Figure 2 compares the experimental and calculated dependences of the conductance G on the ratios w/l and m/n respectively. It is seen, that the calculated dependence $G(m/n)$, for $G_1 = 10^{-7}$ S, $G_2 = 2\times10^{-3}$ S, and $p = 0.48$, on the whole agrees with the experimental dependence $G(w/l)$. The wire conductance depends on its width as a power function [16]:

$$G(w) \propto w^q \,, \tag{16}$$

where the coefficient q depends on the conductivity ratio G_1/G_2 of the resistors and on their fraction ratio $p/(1-p)$ in the network. The coefficient q is calculated as a fitting parameter by least-square adjustment over a large number of randomly generated networks. The dependence (16) corresponds to the behaviour of a network below the percolation threshold. Above the percolation threshold the model gives a spasmodic increase of the conductance. Based on this result it can be assumed, that the conductance of the as-irradiated carbon wires is controlled by a percolation mechanism below the percolation threshold. If this assumption is correct, then the annealing of the sample can be simulated by increasing p up to 1. The physical meaning of this increase is the formation of a homogenous conductive graphite layer. With the increase in p a percolative phase transition occurs and the dependence $G(m/n) = G(w/l)$ becomes linear.

In conclusion, a percolation model is proposed, which allows calculations of the electrical conductance of two-dimensional system composed of two randomly distributed conductive phases. The model takes into account finite dimensions of the system. The model has been successfully applied to explain the non-linear conductance of carbon wires made by ion irradiation on surface of polycrystalline dia-

[4] The considered system shows a transition from a two-dimensional square lattice ($w/l = 1$) to a one-dimensional chain ($w/l \to 0$). It is clear, that the mean value of the percolation threshold will increase for such a transition, i.e. $p_c > 0.5$ [11].

mond films. Apart from the ion-irradiated diamond the model can be adopted for the description of heterogeneous condensed systems, which can be described by resistor networks.

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