Galvanomagnetic phenomena in disordered systems. Theory and simulation

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A new approach to the description of the conductivity and Hall effect in disordered systems, based on the application of a scaling hypothesis analogous to the scaling hypothesis in the theory of phase transitions, is proposed. In the framework of this approach, laws describing the variation of the conductivity and Hall constant of two-dimensional and three-dimensional systems composed of randomly located conducting and nonconducting elements near the percolation threshold are established. For the conductivity the predicted dependences agree well with the experimental data obtained previously. The Hall effect in disordered two-and three-dimensional systems is simulated for the first time by means of measurements on electrically conducting paper with randomly punched holes. The results of this experiment are also in agreement with the predictions of the theory.

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1. INTRODUCTION

Many properties of disordered systems can be understood by analyzing model systems consisting of conducting and nonconducting elements. The most interesting question here is that of the metal-insulator transition that occurs on decrease of the concentration of the conducting elements (cf., e.g., [1, 2]) and of the behavior of the electrical conductivity σ and Hall coefficient R near the transition. This paper is devoted to this question. It seems to us that the critical indices describing the behavior of σ and R should be universal in an extremely wide class of problems, including random-site problems and lattice and continuum problems [3]. However, for definiteness, in the first two sections we shall be concerned with the lattice-site problem. In this problem one considers an infinite periodic lattice of resistances. in which contact between resistances is broken at randomly chosen sites. At a certain finite fraction $x = x_c$ of "broken" sites percolation over the linked resistances disappears^[3]. A network of connected resistances over which percolation occurs for $x \le x_c$ is called an infinite cluster (IC). For $x > x_c$ the IC disappears. For $x < x_c$ we can introduce the concept of the specific electrical conductivity σ of the lattice by defining it in the following way. Out of this lattice we cut a cube (or in the twodimensional case, a square) with side l of sufficiently large size. We apply metallic contacts to opposite faces of this cube and measure the conductivity Y between them. Then,

$$\sigma = Y l / l^{d-1} = Y l^{2-d}, \tag{1}$$

where d = 3 for the three-dimensional problem and d = 2 for the two-dimensional problem.

In an analogous way we can define the Hall constant for such a lattice by performing the standard Hall measurements on a sample of sufficient size, cut out of the lattice.

The numerical calculations of [1] and model experimets of [4-6] have shown that, near the percolation threshold, σ varies with $x_c - x$ in a power-law fashion:

$$\sigma \sim (x_c - x)^t. \tag{2}$$

The index t plays a very important role, since there are reasons to hope that, unlike the percolation threshold x_c , it does not depend on the type of problem but depends

only on the dimensionality of space. Therefore, in real systems, it is this index which is compared first of all with theory ^[2]. The most detailed investigations of the index t have been carried out for lattice problems. The site problem has been studied using a cube built up of $16 \times 16 \times 16$ resistances and using a piece of standard metallic mesh with 137×137 sites. As a result it was found that

$$t=2$$
 for $d=3$,
 $t=1.38$ for $d=2$. (3)

The numerical calculation performed in ^[6] confirmed the result t = 2 for d = 3. However, the calculations of Kirkpatrick ^[1] for d = 3 gave t = 1.5 ± 0.2 for the site problem and t = 1.6 ± 0.1 for the bond problem. The reason for the discrepancy in the results is not clear.

In an analogous way one can discuss the critical properties of the Hall constant R near the percolation threshold, introducing the corresponding index g by the formula

$$R \sim 1/(x_c - x)^{s}$$
. (4)

This index has been very little discussed. In^[1] it is stated that $R \sim P^{-1}(x)$, where P(x) is the fraction of sites belonging to an infinite cluster. In the paper by Skal and Shklovskii^[7] it was postulated that an infinite cluster is a network with a characteristic length $L \sim |x_c - x|^{-\nu}$ and it was shown that, in the framework of this assumption, $g = \nu$ in the three-dimensional case.

The concept of a correlation length L in percolationtheory problems was introduced in ^[8]; L characterizes the scale of the non-uniformity of the infinite cluster. For $x \rightarrow x_c$ we have $L \sim |x - x_c|^{-\nu}$. In the network model this quantity coincides with the characteristic mesh size; however, as pointed out in ^[3], the network model is poor, at least in the two-dimensional case.

A scaling hypothesis relating the different "thermodynamic" indices of percolation theory to the correlationlength index was put forward, independently, in [9,10]. The numerical calculations performed in [10] corroborated this hypothesis and made it possible to determine the correlation-length index for d = 3. In the present paper, using analogous considerations, we relate the indices of the electrical conductivity and Hall coefficient to the correlation-length index.

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We also give the results of a model experiment on the measurement of the conductivity and Hall constant in two- and three-dimensional samples prepared from electrically conducting paper with randomly located holes. It seems to us that studies of the Hall effect in such a system are of special interest, inasmuch as no model experiments have been proposed up to now and there has been no computer simulation of the Hall effect in disordered systems.

2. APPLICATION OF THE SCALING HYPOTHESIS

According to ^[10], the singular part of the number of finite clusters per unit volume (the analog of the singular part of the free energy in the theory of phase transitions) vanishes like L^{-d} , where $L \sim |x - x_c|^{-\nu}$ is the correlation length and d is the number of dimensions of space. This is connected with the formation, in the entire space, of "singular" clusters whose linear dimensions become infinite like L as $x \rightarrow x_c$. On the average, in a volume L^d there is one such cluster. Our problem is to calculate the conductivity Y of a cube (d = 3) or square (d = 2) with side L. Below we introduce the electrical conductivity

$$\sigma = Y L^{2-d} \tag{5}$$

in accordance with the usual formula (1) and assume that this is the effective electrical conductivity of the medium, since the correlation length is the maximum lengthscale of nonuniformity in the system. We now need to understand how Y depends on L. It is clear that it is precisely the "singular" cluster that is responsible for the conduction in the volume of interest. In the spirit of scaling theory, we assume that the topological properties of the "singular" clusters do not change with change of $x_c - x$. Crudely speaking, we assume that photographs of clusters at different values of $x_c - x$ will coincide with each other if they are reduced by a factor of $L \sim |x_c - x|^{-\nu}$. Actually, the "singular" clusters have fine-scale "dead ends," connecting links, etc., to which the scaling hypothesis does not apply inasmuch as there exists a minimum length, equal to the lattice constant. It seems natural to us, however, that the resistivity is determined only by the large-scale properties of the clusters. Then $Y = Y_0 L^{-1}$, where Y_0 does not depend on x_c - x and is, in order of magnitude, the conductivity of one link, and

$$\sigma = Y_o L^{1-d} \sim (x_c - x)^{\nu(d-1)},$$

$$t = (d-1)\nu.$$
(6)

In an analogous way we can analyze the dependence of the Hall constant R on $x_c - x$. For this, it is also sufficient to calculate R for a singular cluster. In accordance with the scaling hypothesis, the Hall potential difference U_x for this cluster can be represented in the form

$$U_x = I \Phi H, \tag{7}$$

where I is the total current through the cluster, H is the magnetic field and Φ is a quantity determined only by the properties of the links and independent of $x_c - x$. If the resistances forming the lattice are wires with radius a and Hall constant R_0 , then Φ is of the order of R_0/a . Defining the Hall constant R in the usual way:

$$E_x = RjH, \tag{8}$$

where j is the current density, and using the relations $E_x = U_x/L$, j = I/L^{d-1} , from (7) and (8) we obtain

$$R \sim L^{d-2} \sim (x_c - x)^{\nu(2-d)}, \quad g = \nu(d-2).$$
 (9)

Thus, as the percolation threshold is approached, R be-

comes infinite like $(x_c - x)^{-\nu}$ in the three-dimensional case and has no power singularity in the two-dimensional case. In the three-dimensional case our result coincides with that of Skal and Shklovskif^[7].

According to the results of [10,11], the exponent ν is equal to 1.33 ± 0.05 for d = 2 and 0.95 ± 0.06 for d = 3. Correspondingly, according to (6) the scaling hypothesis gives t = 1.33 for d = 2 and 1.9 for d = 3, which are in very good agreement with (3).

The following sections of the article are devoted to an experimental investigation of the index g, which has not been determined before.

3. EXPERIMENTAL RESULTS

For the measurements, conducting graphite paper with sheet resistance of the order of 250 ohm and thickness 60 μ m was used. Samples of the shape and size shown in Fig. 1 were cut from it. The maximum size of the samples was determined by the diameter of the pole pieces of the magnet (100 mm). The ratio of the sides, equal to three, was necessary for measurements in the regime of zero Hall current^[12]. The holes were punched at sites of a square lattice with a lattice spacing of 1 mm. The coordinates of the random sites at which the holes were punched were generated on a computer by a random-number generator. The paper used had a negative quadratic magnetoresistance. For H = 18 kG the quantity $\Delta \rho / \rho$ amounted to 8×10^{-4} .

1. <u>Two-dimensional case</u>. In the two-dimensional case the size of the holes was chosen so that holes punched at neighboring sites positioned along a side or along a diagonal of a unit square of the network (cf. Fig. 1a) overlapped. Simple topological arguments show that, in this case, the percolation threshold, i.e., tearing of the paper, should set in at a concentration of punched holes equal to the critical concentration of broken sites in the site problem ($x_c = 0.41$). The holes were punched at the same sites on opposite sides of the sample; this corresponded to periodic boundary conditions.

Two samples, with different arrangements of random holes (two realizations), were investigated. The results of the measurements of the dependences of the Hall constant (cf. Fig. 2b) and relative conductivity σ/σ_0 (cf. Fig. 2a) of the samples on the hole concentration x are practically the same for both realizations (σ_0 is the electrical conductivity at x = 0). The dependence $\sigma(x)/\sigma_0$ measured in the paper ^[4] is also given in Fig. 2a. As can be seen from Fig. 2a, the agreement between the results of ^[4] and our measurements is good. It can be seen from Fig. 2b that, as follows from the arguments cited above based on the scaling hypothesis, the Hall coefficient does not depend on the concentration of punched holes in the planar case.



FIG. 1. a) Two-dimensional samples in which 390 holes have been punched; 1, 2-current contacts, 3, 4 and 5, 6-two pairs of Hall contacts; b) photograph of one of the fifteen planar layers composing the three-dimensional sample (x = 0.6).



FIG. 2. Dependence on the hole concentration x (x = n/1200, where n is the number of punched holes) of (a) the relative conductivity σ/σ_0 and (b) the Hall constant R, for the two-dimensional case. \triangle -realization 1, \bullet -realization 2. For both samples the last point corresponds to the situation in which the next hole interrupted the percolation by cleaving the sample into two parts. The solid curve in Fig. 2a shows the results of measurements of σ/σ_0 from the data of [⁴].

It is interesting to compare the results obtained with the conclusions of the effective-medium theory. To interpret the results of the measurements we have modified the three-dimensional effective-medium theory described in ^[13] to the case of two dimensions. The results of the calculation show that the Hall constant does not depend on the concentration of punched holes. Thus, in the two-dimensional case the effective-medium theory correctly describes the dependence of the Hall constant on the concentration of holes.

As can be seen from Fig. 2b, the results obtained contradict the hypothesis of Kirkpatrick mentioned in the Introduction, according to which the Hall constant is inversely proportional to the fraction of sites belonging to an infinite cluster.

2. The three-dimensional case. A three-dimensional sample was built up from 15 layers of the electrically conducting paper described above. In its shape and size, each layer was a two-dimensional sample as described above. Each of the punched holes was characterized by three random coordinates: the layer label z (1-15), the coordinate x (1-20) and the coordinate y (1-60). Thus, the three-dimensional sample contained 18,000 sites. A random number b(x, y, z), with probability density uniformly distributed in the interval from zero to unity, was assigned to each site, on a computer. If the condition $b \le x$ was fulfilled, where x was the specified concentration of holes, a hole was punched at the site. In the three-dimensional case the diameter of the holes was such that only holes punched at neighboring sites overlapped. Holes located at sites along a diagonal of a unit square of the planar lattice, unlike in the two-dimensional case, did not overlap. After all 15 layers had been punched they were placed in a special cassette and tightly compressed. At large values of x, when finite punchedhole clusters of appreciable sizes formed in the layers, in order to prevent spurious electrical contacts of layers through intermediate layers nonconducting paper of the same thickness as the conducting paper was placed in the voids. In order to monitor this process, and also to position the isolated conducting clusters correctly within each layer, besides the coordinates of the punched holes the pattern of punched holes was printed out for each value of x with the aid of the computer.

Measurements were performed for the values x = 0, 0.2, 0.4, 0.5 and 0.6. To trace the transition from the planar to the three-dimensional case for all the investigated values of x, the conductivity of "samples" composed of one, two, three and fifteen layers with random FIG. 3. Dependence of the relative conductivity of the samples on the number of layers composing them, for different values of x. The points are the results of measurements when the cassette was compressed by a weight of 20 kg; the points in circles are the results of measurements at the higher compression at which the measurements in a magnetic field were performed.



hole-concentration x was measured. The samples were placed in a cassette and compressed by a weight of 20 kg. The dependences of the relative specific conductivity σ/σ_0 of these samples on the number of layers are shown in Fig. 3. The small deviations from unity of the dependence of σ/σ_0 on n at x = 0 are associated with the scatter in the specific conductivity in the individual unpunched paper layers, which did not exceed 10%. For all values of x, saturation of the dependence of σ/σ_0 on n was already reached for n = 4-5, and this, evidently, corresponded to the transition to the three-dimensional case¹⁾. We note that even for x = 0.4 some of the planar layers (in particular, layer no. 1) were cleaved. At x = 0.6 all the planar layers were cleaved (cf. Fig. 1b).

Figure 4 shows the dependences of the relative conductivity σ/σ_0 (Fig. 4a) and of the relative Hall constant R/R_0 (Fig. 4b). Also shown in Fig. 4a, by a dashed line, is the dependence of the relative conductivity σ/σ_0 on the concentration x of "broken" sites, calculated in ^[1] for a simple cubic lattice for a sample with dimensions $20 \times 20 \times 20$. Although simple topological arguments show that the problem of the overlap of punched holes in the three-dimensional case is not equivalent to the site problem, a comparison of the dependences of σ/σ_0 on x given in Fig. 4a shows that these dependences are fairly similar.

We do not know the exact value of the percolation threshold (i.e., the critical concentration of holes at which a three-dimensional sample is cleaved). Experimentally, the following difficulties prevent a closer approach to the threshold. First, at large values of x each of the planar layers constituting the bulk sample breaks down into a large number of small isolated clusters which are practically impossible to position in the composite sample, even when the punched-hole patterns printed out by the computer are available. Secondly, already at x = 0.6 the measurements were hindered by fluctuations of the current in time, which were not connected with instability of the power supply. It is probable that these fluctuations are explained by the absence of reliable contact between the edges of a small fraction of the many thousands of punched holes.

Since we do not know the exact value of the percolation threshold, we can check qualitatively the theoretical arguments put forward in Sec. 2 by constructing the dependence of the quantity $(\sigma/\sigma_0)(R/R_0)^2$ on x (cf. Fig. 5). As can be seen from this dependence, as the percolation threshold is approached the quantity $(\sigma/\sigma_0)(R/R_0)^2$ tends to a constant value, as follows from the theoretical arguments based on the scaling hypothesis.



FIG. 4. Dependence of the relative conductivity (Fig. a) and relative Hall constant (Fig. b) on x. The dashed line shows σ/σ_0 from the data of [¹].



A calculation of the Hall constant was carried out in the framework of the effective-medium theory in [¹³], for an isotropic nonuniform medium. According to [¹³], as x changes from zero to $x = x_c$ the Hall constant increases only by a factor of two. Our experiments point to a considerably sharper increase of the Hall constant (by a factor of approximately 2.5) even far from the threshold, and thus, in the three-dimensional case, our results contradict the theory expounded in [¹³].

4. MAGNETORESISTANCE OF THE THREE-DIMENSIONAL SAMPLES

Figure 6 shows the dependence of the magnetoresistance of the sample on the magnitude of the magnetic field H for different values of x. As can be seen from the Figure, for x = 0 the three-dimensional sample possesses a negative quadratic magnetoresistance-the same as in the two-dimensional samples. The magnitude of the magnetoresistance did not depend on the magnitude and polarity of the current or on the magnetic-field direction. At x = 0.2 the magnitude of the magnetoresistance is slightly decreased. At x = 0.4 (curve 3 in Fig. 6) the magnetoresistance is negative in weak magnetic fields but at H = 11 kG it changes sign and increases sharply on further increase of the field. The dependence $\Delta \rho / \rho$ for x = 0.4 was plotted for ten values of the current from i = 5 to 30 ma. It was established that $\Delta \rho / \rho$ depends neither on the magnitude or polarity of the current, nor on the direction of the applied magnetic field. Next we broke down this bulk sample into its constituent planar sheets and measured the dependence of the magnetoresistance $\Delta \rho(\mathbf{H})/\rho$ for two planar sheets that had still not cleaved at x = 0.4. In both planar samples the dependence $\Delta \rho(H)/\rho$ practically coincided with the same dependence in the unpunched samples (curve 1 in Fig. 6). We then assembled a new three-dimensional sample by arranging the same fifteen sheets in a different order. The results of the measurements coincided with curve 3 of Fig. 6. The magnitude of the electrical conductivity of

FIG. 6. Dependence of the magnetoresistance $\Delta \rho / \rho$ on the magnetic field H for different values of x. The scale for negative values of $\Delta \rho / \rho$ is 10 times smaller than for positive values: curve 1) x = 0, 2) x = 0.2, 3) x = 0.4.



this second sample and the value of the Hall constant differed from the corresponding values for the first sample by not more than 10%. For x = 0.5 the dependence $\Delta\rho/\rho$ for the newly assembled first sample practically coincides with the curve for x = 0.2, i.e., the magnetoresistance again becomes quadratic and negative in the entire range of magnetic fields investigated. For the value x = 0.5 we also constructed a second sample by arranging the punched planar sheets in a different order. The results of the measurements were practically the same for both samples. For x = 0.6 the dependence $\Delta\rho(H)/\rho$ was also practically the same as the analogous dependence for x = 0.

In order to check that the existence of the positive magnetoresistance for x = 0.4 was not connected with anisotropy of the magnetoresistance in the direction parallel to the magnetic field, i.e., in the direction perpendicular to the plane of the layers, we measured the magnetoresistance for 15, 30, 40 and 50 layers of paper in the case when the current flowed in a direction perpendicular to the layers and the magnetic field was parallel to the current. The measurements showed that the resistance in the direction perpendicular to the plane of the layers is approximately 20 times greater than in a direction parallel to the layers. However, the longitudinal magnetoresistance $(\Delta \rho / \rho)_{\parallel}$ in the direction perpendicular to the layers is also negative and is equal, in order of magnitude, to the transverse magnetoresistance in a magnetic field perpendicular to a current flowing parallel to the planes of the layers.

The results of our Hall measurements show that the effective mobility of the conducting paper in the plane of a layer is $\mu = R\sigma \approx 1 \text{ cm}^2/\text{V} \cdot \text{sec.}$ Thus, for H = 18 kG we have $\mu \text{H/c} = 2 \times 10^{-4}$. Unfortunately, we could not devise anything to explain such a significant variation of the magnetoresistance with x.

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¹⁾The small decrease in the quantity σ/σ_0 for $n \ge 10$, which is especially noticeable for x = 0.2, is apparently connected with the fact that the force of 20 kg was insufficient for tight compression of the samples. This is confirmed by the fact that, with stronger compression of the sample before the principal measurements are performed, the conductivity is slightly raised. The values of σ/σ_0 for n = 15 with tight compression of the cassette are also shown in Fig. 3.

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