

Structure of percolation channels and the metal-insulator transition in composites

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The metal-insulator transition is considered. The concept of drop structure of an infinite cluster is developed. The critical exponents for the percolation theory are found in terms of these concepts. The results agree with contemporary experiments on dilute antiferromagnets. A numerical simulation of planar anisotropic systems is carried out. It is shown that when the percolation threshold is approached the systems become rapidly isotropic. This phenomenon can be understood in terms of the drop structure of an infinite cluster.

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We consider here the conductivity of a composite consisting of a metal and an insulator. The decrease of the bulk concentration p of the metal in such a system decreases the conductivity σ . At metal concentrations lower than the critical p_c , called the percolation threshold, the conductivity of the composite is zero. Near the percolation threshold the dependence of the conductivity on the metal concentration is described by the simple power law^{1,2}

$$\sigma \sim (p - p_c)^t. \quad (1)$$

Since p_c is finite, the conductivity at the percolation threshold becomes zero because the greater part of the metal does not participate in the current flow. The current flows only along percolation channels that make up the infinite-cluster skeleton (ICS).²

An important step towards understanding how the metal-insulator transition in composites was made in Ref. 2 where the authors advanced the hypothesis that the ICS, i.e., the aggregate of percolation channels, has a structure. The ICS is a network with characteristic geometric distance between sites equal to the correlation length ξ . The ICS network sites are connected by single-conductor macrobonds (Fig. 1a) whose length \mathcal{L} is in general larger than ξ . With decreasing metal concentration the ICS network becomes less and less dense, the

values of \mathcal{L} and ξ increase and become infinite at the percolation threshold:

$$\xi \sim (p - p_c)^{-\nu}, \quad \mathcal{L} \sim (p - p_c)^{-t}. \quad (2)$$

It follows from (2) that the conductivity of the ICS, and hence of the entire composite, vanishes at the percolation threshold in accord with

$$\sigma \sim (p - p_c)^{t + \nu(d-2)}, \quad (3)$$

where d is the dimensionality of the space. It follows therefore that in the site and bond model the critical exponent t is

$$t = \xi + \nu(d-2). \quad (4)$$

It can be shown² that in this model the critical exponent ξ is independent of the dimensionality of space and is strictly equal to unity. Thus, the ICS structure model developed in Ref. 2 presents a qualitative picture of a metal-insulator junction and yields the connection between the critical exponents. In the three-dimensional case, the experimental³ and calculated^{1,4-6} values of the critical exponent t lie in the range 1.6–1.8, those of the critical exponent ν lie according to Refs. 7 and 8 in the range 0.8–0.9, and relation (4) with $\xi = 1$ is well satisfied. In the two-dimensional case, $\nu = 1.33$ (Refs. 7 and 9–12) and it follows from (2) that the distance between the ICS network

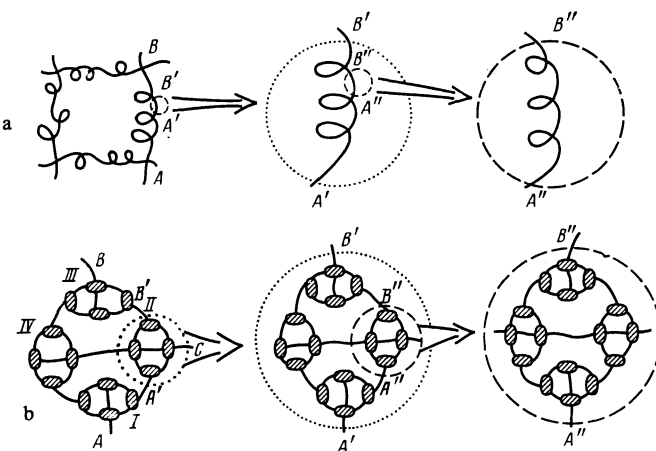


FIG. 1. a) Backbone of infinite cluster in the form of a mesh of microbonds. b) Backbone of infinite cluster with drop structure. In both cases the backbone of the infinite cluster is self-similar, inasmuch as it has the same structure at different scales. A photograph of an ICS fragment with dimension L_{AB} , taken with the minimum resolved dimension ϵL_{AB} is equivalent to a photograph of an ICS fragment having a linear dimension $L_{A'B'}$ and taken with a minimum resolved dimension $\epsilon L_{A'B'}$.

sites increases more rapidly than the length of the macrobond, which is of course impossible.¹³ For all that, there are no grounds for assuming that the ICS structure is different in principle in the cases $d = 2$ and $d = 3$. Consequently, over scales smaller than ξ the ICS structure does not reduce to a single-conductor macrobond. The ICS should contain more complicated formations, which we shall call drops. Such drops were observed in computer simulation of composites¹⁴⁻¹⁶ and in an experimental investigation of island films.¹⁷

DROP STRUCTURE OF INFINITE CLUSTER

It was proposed in Refs. 15 and 18 that at scales smaller than the correlation length ξ the ICS is a self-similar structure consisting of drops joined by single-conductor microbonds. A way of calculating the critical exponents was also indicated in Ref. 15.

We assume that the ICS consists of drops joined by macrobonds (Fig. 1b). Drops of size b consist of drops of size $b/2$ joined by single-conductor macrobonds of length $b^{1/\nu}$. In turn, in a scale $2b$ drops of size b are joined by single-conductor macrobonds of length $(2b)^{1/\nu}$ into a new drop of size $2b$. As shown in Fig. 1b, the ICS structure is in this case self-similar: the law of construction of a drop of size $2b$ out of drops of size b is the same as the law of constructing a drop of size $b/2$. At first glance these assumptions seem rather arbitrary. This is not at all the case, however. Near the percolation threshold the system is scale-invariant near the percolation threshold, and the self-similarity of the ICS^{7,13-15,19,20} is a geometric expression of this fact. We shall show below that drops of scale $b/2$ combine into drops of scale b with the aid of macrobonds whose length is of order $b^{1/\nu}$.

We consider a regular lattice in which the bonds are conducting with probability p and nonconducting with probability $1 - p$. We are interested in the behavior of the system near the percolation threshold where the correlation length ξ is much larger than any microscopic dimension. In this concentration density, lattice systems should represent accurately the behavior of composite materials. We break next, with probability $1 - p_2$, the bonds belonging to the ICS. For each p there exists a maximum fraction $(1 - p_{2c})$ of bonds which can be broken without dividing the ICS into finite pieces. It is natural to call the quantity p_{2c} the threshold of percolation on the ICS. Obviously, at $p = p_c$ we have $p_{2c} = 1$. We note that this condition is not satisfied for the ICS model proposed in Ref. 20.

We consider next an ICS fragment of size b . We break the bonds with probability $1 - p_2$. There is then a probability

$p'_2(b)$ that after the bonds are broken this fragment is not divided into parts. If $p_2 > p_{2c}$, then $p'_2(b) \rightarrow 1$ as $b \rightarrow \infty$, but if $p_2 < p_{2c}$ then $p'_2(b) \rightarrow 0$ as $b \rightarrow \infty$. The critical exponent ν can be obtained in this case from the standard renormalization-group relation (see, e.g., Ref. 10)

$$\ln b / \ln [\partial p'_2(b) / \partial p_2]_{p_2 = p_{2c}} = \nu. \quad (5)$$

The left-hand side of (5) is particularly easy to calculate if $p = p_c$; then $p_{2c} = 1$ and it is necessary to find the quantity $\partial p'_2(b) / \partial p_2|_{p_2 = 1}$. It can be easily shown¹⁸ that this is simply the average total length \mathcal{L} of the unduplicated bonds over an ICS fragment of size b . Consequently

$$\ln b / \ln \mathcal{L} = \nu, \quad \mathcal{L} \propto b^{1/\nu}. \quad (6)$$

In other words, each ICS fragment must contain unduplicated bonds whose total length increases with increasing linear size b of the fragment. This circumstance was noted earlier in Ref. 18. When drops of scale $b/2$ combine to form drops of scale b the single-lead macrobonds contained in the drop of scale $b/2$ are doubled. Consequently the length of the macrobonds that join the drops of scale $b/2$ should be $b^{1/\nu}$. If $b = \xi \alpha (p - p_c)^{-\nu}$, then $\mathcal{L} \propto \xi^{1/\nu} \alpha (p - p_c)^{-1}$. This result was obtained by another method in Ref. 2.

In the three dimensional case $\nu = 0.8 - 0.9$ and the relative length of the macrobond $\mathcal{L}/b \propto b^{1/\nu - 1}$ increases with increasing scale. Therefore the site and bond model² describes well the behavior of many quantities, including the conductivity, near the percolation threshold. We note that an ICS with such a structure is self-similar if it is assumed that it can be represented, in a scale $b < \xi$, as a helix with a pitch b/k . The self-similarity of the ICS means then that k does not depend on b (Fig. 1a). When the scale is doubled the macrobond length is then increased by k times (Fig. 2a). On the n -th step, when $b = 2^n a_0 < \xi$, we have

$$\mathcal{L}_n = k \mathcal{L}_{n-1} = k^n a_0 \sim b^{1/n} / \ln 2, \quad (7)$$

where a_0 is the characteristic microscopic dimension. Inasmuch as $\mathcal{L} \sim b^{1/\nu}$ in this model, we can uniquely determine $k = 2^{1/\nu}$. It might seem that the assumption that a single characteristic length exists contradicts the sinuous behavior of the macrobond ($k \neq 2$), since one more scale, the macrobond length \mathcal{L} , appears. Actually, however, \mathcal{L} appears as the total order parameter. The fact that the natural dimensionality of \mathcal{L} coincides with the dimensionality of length does not mean at all that the critical dimensionality of \mathcal{L} must be the same as that of the correlation length ξ .

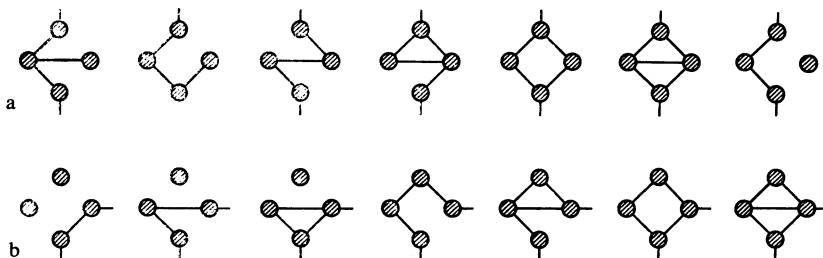


FIG. 2. All sorts of configurations that arise when four drops of scale b are joined to form a drop of scale $2b$. The drops of scale b are joined into drops of scale $2b$ by single-conductor macrobonds of length $\mathcal{L} \propto (2b)^{1/\nu}$.

The critical exponent β' that determines the ICS density is equal in the site and bond model to $\nu d - \zeta \approx 1.7$, which at $d = 3$ is double the value $\beta' = 0.9$ obtained from the numerical calculations.¹⁴ This result can be understood by assuming that an appreciable part of the metal contained in the ICS is concentrated in drops. It is not yet clear at present, however, the behavior of which macroscopic quantities can be affected by this circumstance in the three-dimensional case.

In the two-dimensional case we have^{7,9-12} $\nu = 1.33$ and the relative distance $b^{1/\nu}/b$ between the drops decreases with increasing scale b . The drop structure of the ICS becomes important.

In a real system, drops of scale b combine into drops of scale $2b$ by various methods, so that a set of $2b$ -drop configurations exists. It follows from the self-similarity property of the ICS that these configurations are identical at any scale b . Such a hierarchical structure of ICS can yield, in principle, all the critical exponents exactly, but in this case it is necessary to take full count of the entire set of drops. For the actual calculations we have confined ourselves to a set of configurations in which a drop of size b consists of not more than four drops of size $b/2$ (see Fig. 2). This is a Wheatstone bridge in which the macrobonds are present with a certain probability p_3 , assumed to equal 0.5. These premises agree well with the results of computer simulation of a percolation system.¹⁵

When drops of size b are joined into a drop of size $2b$, some of the b -drops move along and some across the drop (Fig. 1b) in the directions AB and AC , respectively. We introduce therefore the conductivity σ along the drop and the conductivity σ_1 across the drop. When calculating the conductivity of a $2b$ -drop we consider only configurations that have a percolation channel in the corresponding direction. Knowing $\sigma(b)$ and $\sigma_1(b)$ we can find $\sigma(2b)$. To this end we calculate the conductivity between the points A and B (Fig. 1b) in the drops shown in Fig. 2a. For example, the conductivity along the first or second drop of Fig. 2b is equal to

$$\sigma(2b) = \sigma(b) [1 + 2\sigma(b)/\sigma_b(b) + 2\sigma(b)/\sigma_1(b)]^{-1}. \quad (8)$$

We average next over all the configurations shown in Fig. 2a. Similarly, to find $\sigma_1(2b)$ we calculate the conductivity between the points A and C (Fig. 1b) for the configuration on Fig. 2b and average. Assuming that the conductivity σ_b of the macrobond is inversely proportional to its length, we obtain for the variables $a = \sigma(b)/\sigma_b(b)$ and $k = \sigma(b)/\sigma_1(b)$ the following relations:

$$\begin{aligned} a(2b) &= a(b) 2^{1/\nu} Z^{-1}(a(b), k(b)), \\ k(2b) &= [(a+2k)^{16} (1+2k+2a)^6 (2+2k+3a)^2 (2+6k+7a+3k^2+8ak \\ &+ 5a^2)]^{1/21} \{ [(1+2a+2k)(1+3a+4k)^3 (2+12k+10a+7k^2 \\ &+ 16ak+8a^2)(3+3a+2k)]^{1/21} Z(a, k) \}^{-1}, \\ Z(a, k) &= [(1+2k+2a)^5 (1,5a+2k) \\ &\times (4k+3a+7k^2+12ak+5a^2)^2 (1+3a+4k)^{-2}]^{1/8}. \end{aligned} \quad (9)$$

In (9), just as in Ref. 21, we used in the averaging the geometric mean. Applying the doubling procedure n times we arrive at larger scales $L \sim 2^n b$. The quantities a and k tend simultaneously with the increase of L to values corresponding to the stable stationary point of the system (9): $a^* = 0$ and $k^* \approx 0.7$. Linearizing the system (9) in the vicinity of this point we easily obtain the asymptotic behavior of $\sigma(L)$ at large L :

$$\sigma(L) \propto L^{t'}, \quad t' = \ln Z(a^*, k^*) / \ln 2 \approx 1.03. \quad (10)$$

A system with dimension larger than the correlation length has macroscopic properties, and consequently

$$\sigma(p) \sim \sigma(\xi) \sim \xi^{1.03} \sim (p-p_c)^t, \quad (11)$$

i.e., $t/\nu = 1.03$. The obtained value of the exponent t agrees well with latest calculated values $t/\nu = 1 \pm 0.05$.^{15,22}

We can determine similarly the following: the critical behavior of the ICS density $B(p) \propto (p-p_c)^{\beta'}$; the length of the shortest path through a fragment of the system of size $b < \xi$, namely $\mathcal{L}_{sh} \propto b^{\xi'}$, and the average number N of percolation channels through such a fragment. The values of β' , ξ' and N are listed in Table I.

Although we have previously dealt with the backbone of an infinite cluster, the model proposed is in fact valid for the infinite cluster itself. An infinite cluster includes, besides the ICS, drops that are attached to the ICS at one point. No current flows through such channels, and they make no contribution to the conductivity. These drops are usually called "dead ends."¹³ On a scale $b < \xi$ we cannot determine whether we are dealing with part of the ICS or with part of a dead end of scale $b' > b$, $b' < \xi$. Consequently the dead ends have the same structure as the ICS, i.e., a dead end of scale b is a drop consisting of drops of scale $b/2$, etc. Self-similarity is therefore a property not only of the ICS, but also of an infinite cluster. Repeating the reasoning used in the calculation of t , we can find an exponent that describes the critical behavior of the specific volume of an infinite cluster, $P(p) = (p-p_c)^\beta$ (see Table I).

If the critical exponents β and ν are known, the other exponents (α , γ , δ) can be determined from the scaling relations.¹³ Thus the hypothesis that an infinite cluster is made up of drops has enabled us to obtain from a single viewpoint the critical exponents that determine the ICS topology (β' , ξ'), the transport properties (t , λ , ...), and the "thermodynamics" (β , ...) of percolation systems near the percolation threshold.

CONDUCTIVITY OF ANISOTROPIC SYSTEMS NEAR THE PERCOLATION THRESHOLD

An important criterion of the validity of any premise concerning the organization of an ICS is the behavior of anisotropic systems near the percolation threshold.²³ Let us consider the problem of conductivity of a regular lattice, in which conducting and nonconducting bonds are distributed with respective to probabilities p and $(1-p)$. The conducting bonds have along the zx and y directions respective conductivities σ_{0x} and σ_{0y} . The degree of an-

TABLE I.

Exponent	Computer calculations	Experiment	Drop model
t/ν	1±0.01 [15] 0.98±0.02 [22]	1.17±0.11 [28]	1.03
β'/ν	0.4±0.05 [8]	0.35±0.06 [17]	0.46
ξ'/ν	1.12±0.02 [16]		1.25
β/ν	0.104±0.005 [29]	0.1±0.02 [17]	0.15
λ/ν	0.75±0.15 *		
N	0.65±0.1 [24] 1.1±0.1 [30]	— —	1.27 1.05

*Present work.

isotropy of the conductivity is defined as

$$\alpha(p) = \sigma_x(p) / \sigma_y(p) - 1,$$

where $\sigma_x(p)$ is the specific conductivity of the entire lattice along the x direction, and $\sigma_y(p)$ is the specific conductivity in the y direction. Near the percolation threshold the behavior of both the conductivity and of the degree of anisotropy of the entire system is determined by the topology of the ICS. It can be shown²³ that in the site and bond model the behavior of $\alpha(p)$ near the percolation threshold is described by the power-law

$$\alpha(p) \propto (p - p_c)^\lambda, \quad \lambda = \xi - \nu. \quad (12)$$

In the three-dimensional case $\nu = 0.8-0.9$, hence $\lambda = 0.1-0.2$. In the two-dimensional case $\nu = 1.333$ and relation (13) no longer holds, since the exponent λ turns out to be negative. At any rate, by starting from model premises^{2,23} one might expect the degree of anisotropy to decrease more slowly at $d = 2$ than at $d = 3$. The conductivity of anisotropic systems was first investigated numerically in Ref. 6. The exponent found for $d = 3$ is $\lambda = 0.3 \pm 0.1$ and agrees with (12). In the case $d = 2$ it was found that the degree of anisotropy decreases rapidly as the percolation threshold is approached, but the small size 50×50 of the investigated systems could not lead to any conclusion concerning the value of the exponent λ at $d = 2$.

In Ref. 24 the value of λ/ν was obtained by numerically solving the renormalization-group equations for large cells. It was found that the exponent λ depends significantly on the form of the renormalization-group equations. For the simplest transformation²¹ we have $\lambda/\nu = 0.18$ and as the form of the transformation becomes more complicated λ/ν increases to 0.69. We note that the accuracy of the transformations in Ref. 24 is still a moot point. The very value $\lambda/\nu = 0.69$ of the critical exponent cannot be explained within the framework of the simple model of an ICS in the form of sites and bonds.

We examine now how the degree of anisotropy ($d = 2$) changes near the percolation threshold in the drop model of the ICS. On scales $b < \xi$ the percolation channel consists mainly of segments of a percolation channel that pass over drops, since the length of the macrobonds between the drops is $\mathcal{L} \sim b^{1/\nu} \ll b$. On the average one percolation channel passes through a drop, and

depending on its direction the drops can be grouped into z -drops (Fig. 1b, drops I and III) and y -drops (Fig. 1b, drops II and IV). In an anisotropic system this separation is significant, since the conductivities of the x - and y -drops are different. Each x - or y -drop of scale $2b$ consists of x - as well as y -drops of scale b . We denote the conductivity along the x drop by σ_x and across it by σ_{1x} , and analogously for the y -drop. The degree of anisotropy of an ICS fragment of size b is then

$$\alpha(b) = \sigma_x(b) / \sigma_y(b) - 1. \quad (13)$$

In analogy with the procedure used in the isotropic case (9), we can determine how the conductivity and the degree of anisotropy are transformed on going from the scale b to the scale $2b$. These transformations have a stable fixed point at which $\alpha^* = 0$, $a^* = 0$, $k^* = 0.7$. Linearizing the transformations in the vicinity of this point, we obtain the asymptotic behavior of the quantities at large scales, and then also the dependence on the concentration

$$\alpha(p) \sim (p - p_c)^\lambda, \quad \lambda/\nu = 1.27. \quad (14)$$

We note that in the three-dimensional case the ICS conductivity, as indicated earlier, is apparently determined by the conductivity of the macrobonds, and then the conclusions of Ref. 23 are valid.

We have also performed a direct numerical experiment. We considered a quadratic lattice in which the concentration of the conducting bonds corresponded to the percolation threshold p_c . In the actual realization the potential was first applied along the x direction, while periodic boundary conditions were imposed along y and the conductivity $\sigma_x(L)$ of the entire system was determined. In the second step of the same realization of the system, the potential was applied along the y direction, while periodic boundary conditions were applied along x . The conductivity $\sigma_y(L)$ was determined, followed by the degree of anisotropy $\alpha(L)$. For each dimension L , the value of $\alpha(L)$ was averaged not less than over 100 different realizations of the system.

The algorithm used to calculate the conductivity was an elaboration of the algorithms proposed in Refs. 15 and 22. It was based on the fact that the ICS has a drop structure. Before solving the Kirchhoff equations the investi-

gated equation was subjected to an identity transformation: individual sites were eliminated, and the conductivities of their nearest neighbors were renormalized in accord with the formulas

$$\sigma_{ik}' = \sigma_{ik} + \sigma_{il}\sigma_{ik}/\Sigma\sigma_{il},$$

where the summation is over the sites closest to l , where l is the number of the eliminated site, and i and k are sites to which the site l was directly bound. The real distance between the nearest neighbors increases in this case. Generally speaking the sites i and k need not be previously bound, i.e., $\sigma_{ik} = 0$, and then the number of bonds at the sites i and k increases in the course of elimination of the site l , and in final analysis this does not make the numerical calculations easier. But on scales smaller than the correlation length the algorithm is more effective. In a 400×400 system at $p = p_c$ there are left after the described procedure about 150 sites with not more than seven bonds per site. In the initial lattice the maximum number of bonds per site was four. Provision was made in the algorithm for increasing the number of the bonds at the site to seven, thereby increasing the efficiency of the algorithm of Refs. 15 and 22 by one order. The remaining sites cannot be eliminated because of the presence of a limit to which the indicated procedure cannot be extended. This nontrivial behavior of the algorithm can be understood within the framework of the ICS drop model. Indeed, the gist of the described algorithm can be thought of as the following: the first to be eliminated are only the sites inside the drop of scale $b/2$. This transforms the drop simply into one site, and the drop of scale b consists no longer of drops of scale $b/2$, but simply of sites connected by macrobonds. The site elimination procedure can now be carried out in a drop of scale b , by replacing it thus by a site, etc. Consequently, the fact that the algorithm permits the exclusion of almost all the sites at the percolation threshold is connected with the drop structure of the ICS.

The value obtained for the critical exponent λ/ν in the numerical experiment, $\lambda/\nu = 0.7 \pm 0.15$, is less than that predicted by the drop model, but one can nevertheless speak of approximate agreement.

CONCLUSION

We have discussed above questions connected with the conductivity of composite materials. But everything said concerning the structure of an infinite cluster is valid for any system close to the percolation threshold, for example for dilute ferro- and antiferromagnets. As the percolation threshold is approached from above, the Curie temperature in such systems is lowered and becomes equal to zero at the percolation threshold. At concentrations below critical the system does not go over at any temperature to the ordered state, inasmuch as the ferromagnetic atoms are connected in this case only to form finite clusters. For Heisenberg systems the Curie tem-

perature $T_C \propto (p - p_c)^\varphi$ (Refs. 25, 26). It was shown in Ref. 27 that in the two-dimensional case the exponent φ coincides with the exponent t for the conductivity. The exponent φ was determined experimentally in Ref. 28 for a planar $\text{Rb}_2\text{Mn}_p\text{Mg}_{1-p}\text{F}_4$ system and the ratio $\varphi/\nu = 1.17 \pm 0.11$ was obtained, in satisfactory agreement with the results of the drop model (see Table I).

In our opinion, it would be of interest to investigate, near the percolation threshold, a dilute ferromagnet with an essentially anisotropic interaction. In all likelihood, the susceptibility of such a system should become rapidly isotropic in the two-dimensional case as the percolation threshold is approached.

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