Critical behavior of the conductivity of a two-component percolation system in the crossover region

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This letter examines the problem of calculating the effective conductivity of a two-component system in which the components differ markedly in conductivity, and in which the volume fraction of component x, with the higher conductivity, is close to the percolation threshold x_c. A critical exponent, which describes the x dependence of the diffusion coefficient and the conductivity of the system in the limit $x \to x_c$, is found.

In this letter we wish to examine the critical behavior of the conductivity and the diffusion coefficient in composite percolation systems near the percolation threshold. As a typical example of such a system we might cite a composite consisting of metal inclusions with a conductivity σ_1 distributed at random in a poorly conducting medium with a conductivity σ_2. If the volume fraction of the metal inclusions, x, is close to the percolation threshold x_c, and if the condition $h = \sigma_2/\sigma_1 \ll 1$ holds, then the effective conductivity of the system, σ, has the asymptotes[1]

$$\frac{\sigma(\tau,h)}{\sigma_1} \sim \begin{cases} \tau^t, & \tau>0, \quad \tau>\Delta, & \text{(1a)} \\ h^s, & \tau=0, & \text{(1b)} \\ h|\tau|^{-q}, & \tau<0, \quad |\tau|>\Delta. & \text{(1c)} \end{cases}$$

Here $\tau = x - x_c$, and $\Delta \sim h^m$ is the width of the x interval near x_c, in which the crossover from (1a) to (1c) occurs. The critical exponents t, s, q, and m are dependent quantities. A relationship among them can be established on the basis of the similarity hypothesis, which allows us to write $\sigma(\tau,h)$ in scaling form in the critical region:[1]

$$\sigma(\tau,h)/\sigma_1 \sim h^s \varphi(\tau/\Delta), \tag{2}$$

where $\varphi(z)$ is a universal function, whose form is independent of the microscopic structure of the system. From (1) and (2) we find

$$q = t(1/s - 1), \quad m = 1/(t+q). \tag{3}$$

This result allowed Efros and Shklovskii[1] to conclude that the entire set of exponents describing the critical behavior of the system can be expressed in terms of just two exponents, e.g., t and q. The exponent t is related by

$$t = \nu(d_w - 2 + d - d_f) \tag{4}$$

to the dimensionality (d_w) of a random walk on the percolation cluster.[2] The exponent q is expressed in terms of the dimensionality of the unscreened perimeter of the cluster, d_u (Ref. 3):

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$$q=\nu(d_u+2-d). \tag{5}$$

The quantity ν in (4) and (5) is the critical exponent of the correlation radius, d is the dimensionality of the system, and d_f is the fractal dimensionality of the percolation cluster.

We show below that two critical exponents are insufficient for a complete description of the behavior of the conductivity of the system near the percolation threshold. Specifically, we show that in the crossover region, i.e., at $|\tau|<\Delta$, the following asymptotic relation holds:

$$\left|\frac{\sigma(\tau,h)-\sigma(0,h)}{\sigma(0,h)}\right|\sim\left|\frac{\tau}{\Delta}\right|^{\alpha}. \tag{6}$$

Here the new exponent

$$\alpha=\nu(d-d_u) \tag{7}$$

generally cannot be expressed in terms of an algebraic combination of t and q.

We model the system by a regular lattice in which the probabilities for transitions of charge carriers between nearest sites per unit time can take on two values: w_1 and $w_2 = hw_1$, where $h\ll 1$. The links of the first type and also the clusters formed by these links are "metallic," while the set of links of the second type is the "medium." By virtue of Einstein's relation, the diffusion coefficient satisfies $D\sim\sigma$. The problem thus reduces to one of calculating D for the case in which the fraction of metallic links, x, is close to x_c.

It can be seen from (1) and (2) that in this system there is, in addition to the correlation length $\xi\sim|\tau|^{-\nu}$, a second length scale $R_h\sim\Delta^{-\nu}$ (here and below, all lengths are expressed in units of the lattice constant). Using the second scaling relation in (3), along with (4) and (5), we can write R_h as follows:

$$R_h\sim h^{-\delta},\quad \delta=\frac{1}{d_w-(d_f-d_u)}. \tag{8}$$

To see the physical meaning of R_h, we consider the following auxiliary problem. We assume that at the time $t=0$ a carrier is situated on the metal cluster. For definiteness we assume that the cluster is infinite, and that the correlation radius has the behavior $\xi\to\infty$. The diffusive displacement of a carrier along the cluster, $r(t)$, thus increases in time as $(w_1t)^{1/d_w}$ for arbitrarily large t (Ref. 2). What is the probability $f(t)$ that a carrier has not yet managed to reach another cluster by the time t? We know that nearly all the sites of the percolation cluster belong to its perimeter (more precisely, the number of perimeter sites increases with increasing size of the cluster, in proportion to the total number of sites).[4] The escape of a carrier from a cluster into the medium thus occurs at a frequency w_2. However, the probability for a transition to another cluster is small, except in the case in which the original cluster escapes through the unscreened perimeter of this cluster (otherwise, the probability for inverse trapping is high).[3] We can thus write the following equation for $f(t)$:

$$\frac{df(t)}{dt} \sim -w_2 Q(t) f(t), \tag{9}$$

where $Q(t)$ is the probability that a carrier is at the unscreened parameter of a cluster at the time t. We evidently have $Q(t) \sim N_u(t)/N(t)$, where $N_u(t) \sim r(t)^{d_u}$ and $N(t) \sim r(t)^{d_f}$ are, respectively, the numbers of sites of the unscreened parameter and the total number of sites which the carrier manages to visit by the time t as it moves through the cluster. We thus have $Q(t) \sim (w_1 t)^{-\pi}$, where $\pi = (d_f - d_u)/d_w$. From (9) we correspondingly find $f(t) \sim \exp[-h(1-\pi)^{-1}(w_1 t)^{1-\pi}]$. The time scale for the decay of the function $f(t)$ is

$$t_1 \sim w_1^{-1} h^{-\gamma}, \quad \gamma = \frac{d_w}{d_w - (d_f - d_u)}. \tag{10}$$

Noting that we have $r(t_1) \sim R_h$, we reach the conclusion that R_h is the average distance which a carrier manages to move before it finally escapes from a finite cluster with a radius $R > R_h$ or from the infinite cluster under the condition $\xi > R_h$.

For finite clusters with radius $R < R_h$, the distance which a carrier moves away before the time of the final escape from the cluster is on the order of R. We thus have $Q \sim R^{d_u - d_f}$, and for the time scale for the escape from the cluster we have, in place of (10),

$$t_2 \sim w_2^{-1} R^{d_f - d_u}. \tag{11}$$

Finally, we consider the kinetics of the escape of a carrier from an infinite cluster, under the condition $\xi < R_h$. In this case we have $N_u(t) \sim \xi^{d_u}[r(t)/\xi]^d$ and $N(t) \sim \xi^{d_f}[r(t)/\xi]^d$ and thus $Q \sim \xi^{d_u - d_f}$. The time scale for escape from the cluster is therefore

$$t_3 \sim w_2^{-1} \xi^{d_f - d_u}. \tag{12}$$

Over scales greater than ξ, the diffusive displacement along the cluster is $r(t) \sim (D_0(\xi) t)^{1/2}$, where $D_0 \sim w_1 \xi^{2-d_w}$ (Ref. 2). Consequently, before it escapes from the cluster, the carrier manages to undergo a displacement over a distance

$$\tilde{R}_h \sim r(t_3) \sim h^{-1/2} \xi^{\phi}, \quad \phi = (d_f - d_u + 2 - d_w)/2. \tag{13}$$

It can be seen from (10)–(12) that for large clusters the escape time is much greater than the time w_2^{-1}, which is the average time the carrier spends in the medium before trapping. Accordingly, the motion of a carrier from cluster to cluster can be thought of as a diffusion with a random hopping length: An escape of a carrier from a finite cluster of radius R corresponds to a hop of length R_h (if $R > R_h$) or R (if $R < R_h$), while escape from an infinite cluster corresponds to R_h (if $\xi > R_h$) or $\tilde{R}_h$ (if $\xi < R_h$). In the crossover region we have $\xi > R_h$, so we can write the following asymptotic expression for the mean square diffusive displacement $\langle R^2(t) \rangle$:

$$\langle R^2(t) \rangle \sim \int_1^{R_h^{d_f}} ds\, \mu(s,t) R^2(s) + R_h^2 \int_{R_h^{d_f}}^{\xi^{d_f}} ds\, \mu(s,t) + R_h^2 \mu(\infty,t) \vartheta(\tau). \tag{14}$$

Here $\vartheta(x)$ is the unit step function, $R(s) \sim s^{1/d_f}$ is the radius of the finite clusters expressed as a function of the number of sites s, and $\mu(s,t)$ and $\mu(\infty,t)$ are the numbers of visits, over a time t, at finite clusters of s sites and at an infinite cluster, respectively. In the long-time limit, the quantity μ is proportional to $w_2 t$, and the concentrations of sites of the unscreened parameter of the corresponding clusters are

$$\mu(s,t) \sim w_2 t n(s) R^{d_u}(s), \quad \mu(\infty,t) \sim w_2 t \xi^{d_u - d}. \tag{15}$$

Here $n(s) \sim s^{-\zeta}$ is the concentration of clusters of s sites. Using the hyperscaling relation $\zeta = 1 + d/d_f$, we find the following expression from (14) and (15):

$$\sigma, D \sim \frac{d\langle R^2(t)\rangle}{dt} \sim w_1 h^s \left[1 \pm A_\pm \left| \frac{\tau}{\Delta} \right|^\alpha \right], \tag{16}$$

where A_+ and A_- are unknown constants for the regions above and below the threshold, respectively, the exponent α is given by (7), while the exponent

$$s = \frac{d_w - 2 + d - d_f}{d_w - d_f + d_u} \tag{17}$$

satisfies the first scaling law in (3).

The following expressions were derived for d_u in the mean-field approximation in Ref. 3:

$$d_u \begin{cases} (d+d_f)/2 - 1, & d_f \geqslant 2, \\ d/d_f + d_f - 2, & d_f \leqslant 2. \end{cases} \tag{18}$$

Using known estimates[5] for ν and d_f, we find $\alpha \simeq 1.4$, 1.1, 0.9, and 0.9 for $d = 2$, 3, 4, and 5 from (7) and (18). For $d = 6$, expressions (7) and (18) lead to the value $\alpha = 1$.

Outside the crossover region, i.e., at $|\tau| > \Delta$, the approach outlined above agrees with existing results. In this case we have $\xi < R_h$, and in place of (14) we should write

$$\langle R^2(t)\rangle \sim \int_1^{\xi^{d_f}} ds \, \mu(s,t) R^2(s) + \tilde{R}_h^2 \mu(\infty,t) \vartheta(\tau). \tag{19}$$

In the case $\tau < 0$, we find (1c) from (19), where the exponent q is given by (5). At $\tau > 0$ we find σ, $D \sim w_1 \tau^t (1 + h \tau^{-q/t})$, i.e., the leading term of asymptote (1a) along with a correction term.

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