Critical Behavior of the Low-Field Hall Conductivity at a Percolation Threshold

David J. Bergman
Department of Physics, The Ohio State University, Columbus, Ohio 43210, and Department of Physics and Astronomy, Tel-Aviv University, 69978 Tel-Aviv, Israel, and Institute for Theoretical Physics, University of California, Santa Barbara, California 93106

and

Yacov Kantor
Department of Physics and Astronomy, Tel-Aviv University, 69978 Tel-Aviv, Israel

and

David Stroud
Department of Physics, The Ohio State University, Columbus, Ohio 43210

and

Itzhak Webman
Exxon Research and Engineering Company, Linden, New Jersey 07036
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The critical behavior of the low-field Hall effect in a three-dimensional metal-nonmetal composite near the percolation threshold has been determined for the first time from calculations on a reliable random-resistor-network model. The Hall coefficient R_e is found to diverge as $|p-p_c|^{-g}$, where $g=0.29\pm0.05$.

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The Hall effect has had a long history as a useful tool for investigating the transport of electronic charge in both good and bad conductors. In the past, it has been used extensively to investigate the metal-nonmetal transition in a variety of disordered systems.[1-3] More recently, a number of Hall measurements have been made on some granular or composite solids with a conductivity threshold in which it is clear that the disorder is macroscopic and where the threshold may be percolative in character.[4-6]

An effective-medium theory (EMT)[7] has been used to discuss the properties of the Hall effect in conductors with macroscopic disorder.[8,9] A different approach was to simulate the Hall effect on a cubic random resistor network (RRN), specific samples of which were then solved in the presence of a magnetic field H to yield an effective Hall conductivity λ_e or Hall coefficient R_e.[10,11]

A number of discussions of the critical properties of λ_e and R_e near the percolation threshold of a RRN have been given,[11-16] but there are no reliable calculations of the critical properties in three-dimensional (3D) systems. Exact results are known for the critical behavior in 2D,[14] and also in a Cayley tree.[15]

In this Letter we report on a new method for calculating these critical properties, using a RNN model, which leads for the first time to a reliable quantitative estimate of these properties in 3D systems.

Consider a two-component composite conductor with Ohmic conductivities σ_1, σ_2 and Hall conductivities $\lambda_1 \ll \sigma_1$, $\lambda_2 \ll \sigma_2$ (i.e., we are considering the low-field effect). We can show that the effective Hall conductivity of the composite λ_e is then always determined by a single function of the ratio σ_1/σ_2:

$$\frac{\lambda_2-\lambda_e}{\lambda_2-\lambda_1}=X\left(\frac{\sigma_2}{\sigma_1}\right), \tag{1}$$

where the precise form of X depends on the microgeometry.[17] For $\sigma_2=\lambda_2=0$, and for p_1 slightly above the percolation threshold p_c, we can expect λ_e and R_e to exhibit a characteristic power-law behavior, like σ_e (Ref. 18):

$$\lambda_e/\lambda_1=X(0)\propto(p_1-p_c)^{\tau};\quad \sigma_e/\sigma_1\propto(p_1-p_c)^{t};$$
$$R_e/R_1\propto(p_1-p_c)^{\tau-2t}\equiv(p_1-p_c)^{-g}. \tag{2}$$

For σ_2/σ_1 and p_1-p_c small but nonzero, $X(\sigma_2/\sigma_1)/|p_1-p_c|^{\tau}$ can be expected to depend only on the characteristic scaling parameter of the RRN $(\sigma_2/\sigma_1)/|p_1-p_c|^{t+s}$.[18]

In order to determine the critical behavior of X near the percolation threshold of an isotropic composite, we realize the Hall problem on a two-

component, discrete lattice as follows: Each element of the lattice is a triplet (doublet in 2D) of identical conductors with an Ohmic conductance σ_1 or σ_2 that lie along the coordinate axes, and which are electrically unconnected in the absence of a magnetic field H (see Fig. 1). In the presence of an H field taken to lie along the z axis, a Hall current will flow through a conductor in the x direction that depends on its Hall conductance (λ_1 or λ_2) and on the voltage across the y conductor of the same triplet. The two types of triplets (doublets in the 2D case) are placed randomly at all the sites of an $L\times L\times L$ fcc (square centered in 2D) lattice, and electrical connections are made at the centers of all the unit-cell edges as well as the body-center points (see Fig. 1; in the square-centered lattice, connections are made only at the cell-edge centers). It is easy to see that by making these connections we obtain four simple-cubic (two simple square in 2D), $L\times L\times L$ random-bond resistor networks that are electrically unconnected (for $H=0$) but are correlated with each other by virtue of the unconnected triplets (doublets) used in setting them up. A careful expansion of the equations of this lattice model of the Hall effect in powers of H up to and including $O(H)$ terms leads

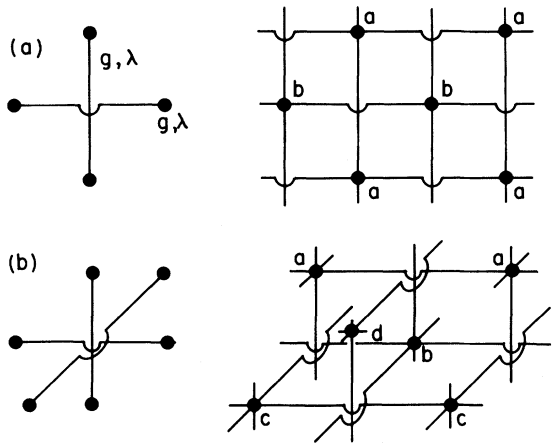

FIG. 1. Schematic drawing of portions of the RRN's used to realize the Hall effect in a discrete system in 3D (an fcc lattice of identical but unconnected mutually perpendicular triplets), and in 2D (a square-centered lattice of identical but unconnected mutually perpendicular doublets). Electrical connection points that lie on the same connected portion of the network are labeled by identical letters. Thus the 2D network is composed of two unconnected (but correlated) simple-square resistor networks, while the 3D network is composed of four unconnected (but correlated) simple-cubic resistor networks.

to the conclusion that it is not necessary to actually solve the network in the presence of a magnetic field. It is sufficient to solve it (twice) to find the potential distribution when an external electric field only is applied along the x and the y directions. The function X is then given by[19]

$$X\left(\frac{\sigma_2}{\sigma_1}\right)=\frac{1}{L^3}\sum_a V_a^{(y)}\theta_a V_{a\times H}^{(x)}, \tag{3}$$

where the sum is over all bonds of the network, and $V_a^{(y)}$ is the voltage across the bond a when a total voltage of magnitude L is applied across the network in the y direction. The other voltage appearing in the sum, $V_{a\times H}^{(x)}$, is the voltage across another bond from the same triplet (or doublet)—the one that lies along the vector product $\vec{a}\times\vec{H}$, with a minus sign if it points in the opposite direction—that results when a voltage L is applied across the network in the x direction. Finally, $\theta_a=1$ if a is a bond of type σ_1, and $\theta_a=0$ otherwise.

The triplet (doublet in 2D) elements chosen above to represent the Hall problem in a discrete form were arrived at by considering a six-terminal (four-terminal in 2D) linear circuit element —two terminals along each coordinate axis—and requiring that when there is a potential difference only along, say, the x axis, and a magnetic field along the z axis, a purely Ohmic current flows along the x axis, while a pure Hall current flows along the y axis. The resulting 2D doublet element, when used in constructing a random square-centered array, is the only way known to us of obtaining a random array that is also self-dual. The exact results on 2D systems require this self-duality in order to be valid (continuous systems are automatically self-dual), so that the RRN's used in Refs. 11 and 15 are inadequate from that point of view. In fact, we have found that the uncorrelated version of the simple-cubic RRN of Ref. 11 has a critical behavior that is very different from that of our special form of RRN in both 3D and 2D, and thus they belong to different universality classes of critical properties.[19]

We also note that our basic Hall element has the feature that a Hall current will flow through a conducting bond only if there is a potential difference along a conducting bond perpendicular to it. This property is essential for a correct representation of a continuous random material of either two or three dimensions. By contrast, in the uncorrelated RRN model that has been widely used for the simulation of the conductivity near

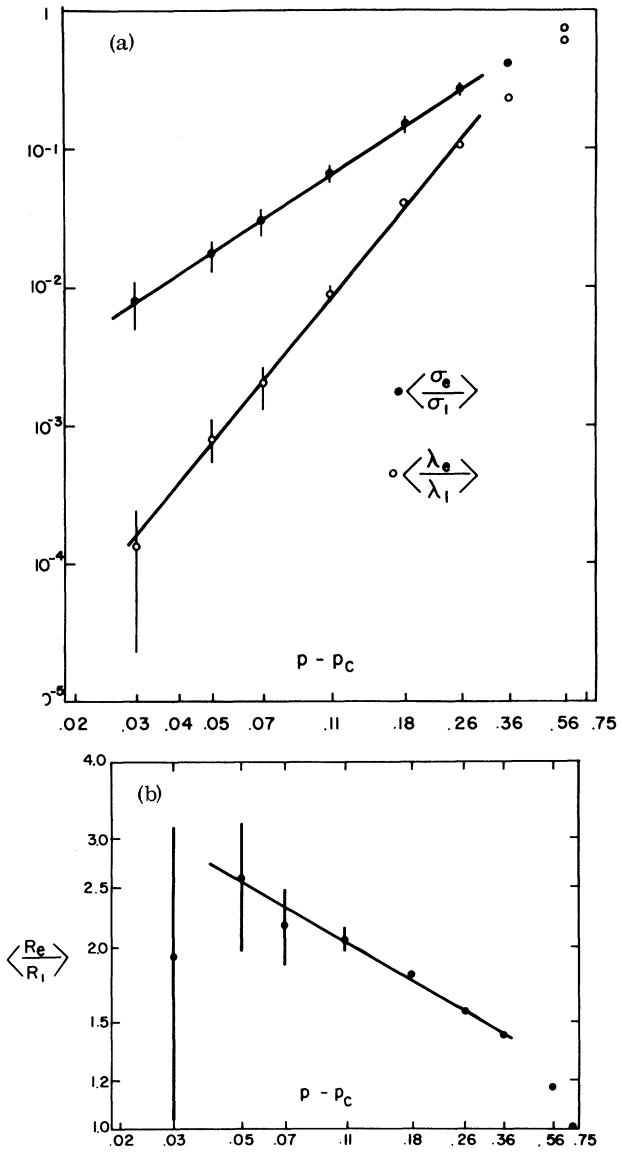

FIG. 2. Log-log plots of (a) the ensemble-average bulk effective Ohmic conductivity $\langle\sigma_e\rangle$ and Hall conductivity $\langle\lambda_e\rangle$ vs the fraction p_1 of conducting bonds of the random-resistor network and (b) the Hall coefficient $\langle R_e\rangle = \langle\lambda_e/H\sigma_e^2\rangle$ vs p_1. The abscissa axis is actually $\ln(p_1 - p_c)$, where the percolation threshold is taken to be $p_c = 0.2465$ (Ref. 24). For each value of p_1, a certain number n of $15\times15\times15$ random fcc networks of conducting and nonconducting triplets, constructed as described in Fig. 1, were solved, with an applied electric field first in the x direction, and then in the y direction. Equations (3) and (2) were then used to evaluate λ_e, while Eq. (4) was used to evaluate two candidates for σ_e, σ_{ex} and σ_{ey}, for each network. The averages $\langle\sigma_e\rangle$, $\langle\lambda_e\rangle$, and $\langle R_e\rangle$, respectively, represent $\frac{1}{2}(\sigma_{ex}+\sigma_{ey})$, λ_e, and $4\lambda_e/H(\sigma_{ex}+\sigma_{ey})^2$ averaged over the n (usually, $n=12$ except for $p>0.5$, where the statistics allow a smaller n to be used) different networks at each value of p_1. The error bars in (a) represent the statistically determined standard deviations of λ_e/λ_1 and σ_e/σ_1 for a *single* network, while the error bars in (b) represent the similarly determined standard deviation of R_e/R_1. The straight lines in (a) are visual best fits to all the points with $0.28 \le p_1 \le 0.51$, while the straight line in (b) is a visual best fit to all the points with $0.30 \le p_1 \le 0.61$.

a percolation threshold, a Hall current can flow in a conducting bond as a result of a voltage along insulating bonds. This property is clearly unphysical, and can be shown to lead to the drastically different critical behavior of the Hall effect in that model.[19]

We note that an expression analogous to Eq. (3) exists also for the effective Ohmic conductivity σ_e,[20] namely

$$\frac{\sigma_e}{\sigma_1} \simeq \frac{\sigma_2-\sigma_e}{\sigma_2-\sigma_1} = \frac{1}{L^3}\sum_{a\parallel x} V_a^{(x)}\theta_a, \tag{4}$$

where the sum is now only over bonds a that lie in the x direction.

We have solved for the detailed voltage distribution in a number of 3D RRN's of size $15\times15\times15$ and $p_1 - p_c$ as low as 0.03, and used the results to obtain $\langle\sigma_e\rangle$, $\langle\lambda_e\rangle$, and $\langle R_e\rangle = \langle\lambda_e/H\sigma_e^2\rangle$ above and near the percolation threshold p_c.[21] The results are plotted in Fig. 2, and they lead to the conclusion that

$$\tau = 3.0\pm0.1; \quad t = 1.64\pm0.04;$$
$$g = 0.29\pm0.05. \tag{5}$$

Note that while g was evaluated separately from τ and t, its value obviously satisfies the relation $g = 2t - \tau$, as it should. Since the results for t are in good agreement with other, more detailed analyses,[18] we are confident in the new results for τ and for g.

Our result for g may be compared with Straley's estimate $g = 0.5$–0.6,[16] which is based on the nodes-links model of the percolating backbone, and with an early conjecture of Holcomb and Rehr, $g = 0.4$, based on an identification of the critical behavior of $1/R_e$ with that of the size of the infinite cluster.[22] It disagrees rather strongly with Skal and Shklovskii's estimate $g = 0.8$ that is based on a different use of the nodes-links model.[12] Clearly, our result is more reliable than any of the above and its accuracy can and will be improved upon by further calculations. It will then be able to serve as a new test of the nodes-links model and other models of percolat-

ing systems.

It would now be of interest to do experiments on metal-nonmetal composites near the percolation threshold of the metal in order to check our result. Although it seems that Bandyopadhay *et al.*[6] may have already observed critical behavior in Hall measurements near p_c, they were not able to measure the metal fraction p_1 in their samples, so that a quantitative comparison with our theory is impossible. We note that for a microscopic metal-nonmetal transition by electron localization, theory predicts that the Hall coefficient remains finite.[23] This is very different from the behavior that we have found, and thus measurements of the Hall effect might help to distinguish between microscopic and macroscopic metal-insulator transitions.

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