Conductivity in random systems. II. Finite-size-system percolation

Juhani Kurkijärvi*

Laboratory of Atomic and Solid State Physics and School of Applied and Engineering Physics, Cornell University, Ithaca, New York 14850

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A simplified model of hopping conductivity in amorphous systems is considered. The nonexponential prefactor of the conductivity is shown to be related to the size dependence of the percolation radius, complementing the well-known picture that the infinite percolation radius determines the leading exponential factor. This leads to an extrapolation formula for the infinite-system percolation radius which involves the power of the nonexponential dependence. Computations on large numbers of small systems are used to determine the radius and the power.

I. INTRODUCTION

In a previous work[1] (hereafter to be referred to as I) a somewhat simplified preliminary model was discussed in pursuit of the temperature dependence of the Mott hopping-conductivity model.[2] It was the aim of that investigation to verify through computer simulation that the leading behavior of the conductivity is related to a percolation problem.[3] In this paper the discussion is expanded to include the nonexponential prefactor of that model. One is thus led to a slightly unusual way of looking at the percolation problem which is ordinarily approached by investigating the conditions for a germanely defined cluster to become infinite in extent.[4] In this work percolation through periodically extended finite arrays of N random points is considered. It is shown that the nonexponential prefactor of the simplified hopping-conductivity model is determined by the weak N dependence[5] of the average critical radius for percolation r_{cN}, the distance up to which points must be pairwise connected to make a continuous path across the array. A most natural extrapolation formula for r_{cN} as a function of large N is then a consequence of the nonexponential prefactor of the electrical conductivity. This formula can be used to get the infinite-system critical radius r_c starting with large numbers of systems of several finite sizes. The power of the nonexponential prefactor, which is a parameter of the extrapolation formula, is determined as a side product. This program has been carried out in the work reported here.

More specifically, the model considered in I consists of an infinite system of random points in space, pairwise connected with conductances of the form

$$g_{ij} = g_0 e^{-\alpha r_{ij}}, \tag{1.1}$$

where r_{ij} is the distance between the points i and j. In this paper the nonexponential prefactor of the asymptotic conductivity[3] σ of such a system at large values of α is shown to be related to the N dependence of the finite-system percolation radius as follows. Taking[1]

$$\sigma \propto (1/r_c\alpha)^\nu e^{-\alpha r_c}, \tag{1.2}$$

then

$$r_{cN} = r_c + A N^{-1/3\nu}. \tag{1.3}$$

The derivation in Sec. II works equally well in the other direction; Eq. (1.3) implies Eq. (1.2). It may be considered a matter of taste which one should be viewed as *a priori* better founded. For Sec. II, the case for Eq. (1.2) is discussed. On the other hand, Eq. (1.3) agrees with the idea that a finite system contains a limited amount of information on the critical point, with an error which is smaller the larger the system considered.[6] In any case, Eq. (1.3) is an extrapolation formula for the infinite system r_c and it is used here starting with Monte Carlo averages of r_{cN} over typically 150 arrays of N particles for $N = 32$, 64, 108, 256, 1000, and 2000. In this way the exponent ν of Eq. (1.2) is determined with the result $\nu = 0.6 \pm 0.25$ and the density-independent criterion for percolation $p_c = \frac{4}{3}\pi\rho(\frac{1}{2}r_c)^3$, where ρ is the density of points, is estimated to lie between $p_c = 0.337$ and $p_c = 0.358$ with the most probable value $p_c = 0.347$. There have been previous estimates of this latter quantity[7-11,1] and the somewhat conflicting results will be discussed in Sec. III. The outline of the rest of the present paper is as follows. In Sec. II the relationship between the nonexponential prefactor and the N dependence of r_{cN} is discussed. Section III addresses itself to the numerical work for the "ensemble averages" of r_{cN} and the above-mentioned comparison of results. Finally, Sec. IV includes conclusions and an Appendix follows on the prefactor of the full four-dimensional Mott model.

II. PREFACTOR AND N DEPENDENCE OF r_{cN}

Assume a finite system of N points randomly distributed in space and every pair of points connected with conductances g_{ij} as given in Eq. (1.1). Then, when α tends to infinity, the electrical current I_N across such a system subjected to a voltage V varies as

$$I_N = V g_0 e^{-\alpha r_{cN}} . \tag{2.1}$$

This is intuitively clear as discussed in I, since the whole voltage finally concentrates on a single conductance, the one implied by the longest r_{ij} the current has to face to get through the system. All other conductances become exponentially better when α tends to infinity. If V and g_0 are taken to be unit voltage and conductance, respectively, the plot of the logarithm of Eq. (2.1),

$$\ln I_N = -\alpha r_{cN} , \tag{2.2}$$

crosses the $\alpha = 0$ axes at $\ln I_N = 0$. For finite α, the current approaches this asymptote from above. This is so irrespective of the size of the system as long as a unit voltage is imposed across it. To get the conductivity one has to divide the linear size of the system by a length which is proportional to $N^{1/3}$ at constant density. Therefore, the logarithm of the conductivity in the asymptotic region is

$$\ln\sigma_N = \ln(N^{-1/3}) - \alpha r_{cN} , \tag{2.3}$$

i.e., the asymptote now cuts the $\alpha = 0$ axes at points that lie lower for larger N. For each N, the finite α conductivity approaches the asymptote from above.

To complete the picture it is necessary to have an idea of how r_{cN} will behave as a function of N. It is intuitively suggestive, and confirmed by actual computation, that percolation is slightly easier in a large system than in a small system of a given shape, as the distance to go grows slower than the area over which one may try; in other words, more and more complicated paths become possible. It should be noted that one must take I_N and r_{cN} in Eqs. (2.1)–(2.3) as ensemble averages over all systems of a given number of points.

Assume now that a finite system adequately describes the conductivity of the infinite system as long as α is small enough not to force the conductivity closer to the asymptote [Eq. (2.3)] than say a factor of 10. Then the picture of Fig. 1 emerges. For small α the conductivity curves of all sizes of system coincide. One by one, in the order of growing N, the conductivity curves of the finite systems fall below[12] the mainstream which represents the infinite-system conductivity as a function of α. The logarithm of the infinite system conductivity then has no asymptote but a continuous curvature. Equation (1.2) has this feature justifying the choice of the particular functional form.

It is now a simple matter to derive Eq. (1.3). Unless the infinite-limit conductivity and the support curve formed by the successive asymptotes [Eq. (2.3)] drift apart, the two curves will have the same curvature at large α. Let $x = x(N)$ denote the coordinate of the point at which the asymptote belonging to systems of N points cuts the $\alpha = 0$ axis in Fig. 2 and call the slope of the asymptote $k = k(N) = k(N(x))$. Take another asymptote, lower by Δx on the $\alpha = 0$ axis. If α is now the point where the two asymptotes intersect

$$\Delta x = \Delta k \alpha = \frac{dk}{dx} \Delta x \alpha$$

or (2.4)

$$\frac{dk}{dx} = \frac{1}{\alpha} ,$$

i.e., the intersection point has a finite limit when $\Delta x \to 0$. On the other hand, from Eq. (1.2),

$$\frac{d^2}{d\alpha^2} \ln\sigma = \frac{\nu}{\alpha^2} \tag{2.5}$$

and this curvature is equal to $dk/d\alpha$. Consequently,

$$\frac{dk}{d\alpha} = \frac{dk}{dx}\frac{dx}{d\alpha} = \frac{1}{\alpha}\frac{dx}{d\alpha} = \frac{\nu}{\alpha^2} . \tag{2.6}$$

Thus, the following differential equation has been established:

$$\frac{dx}{d\alpha} = \frac{\nu}{\alpha} , \tag{2.7}$$

whose solution is

$$\alpha = C_1 e^{x/\nu} , \tag{2.8}$$

and from Eq. (2.4)

$$k = -(\nu/C_1) e^{-x/\nu} + C_2 . \tag{2.9}$$

To get Eq. (1.3) from this, one just has to observe that $-r_{cN}$ is the slope of the asymptote and $x = x_0$

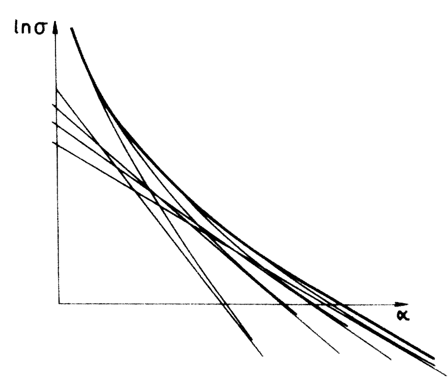

FIG. 1. Infinite-limit conductivity plotted against α with successive asymptotes of the conductivities of growing finite systems. Each finite system would correctly describe the infinite system up to some value of α and then depart downward toward its asymptote. Four such finite-system curves are depicted in the figure.

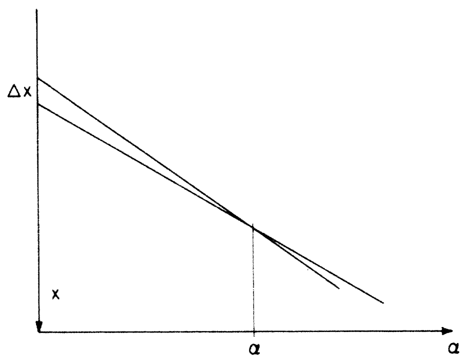

FIG. 2. Two asymptotes for two nearly equal sizes of system. The intersection point of the asymptotes is marked as α. This α tends to a finite limit when Δx approaches zero as described by Eq. (2.8).

$+\ln(N^{1/3})$. Finally, the slope at $N=\infty$ is equal to $-r_c$, which fixes the constant C_2, and Eq. (1.3) follows with $A=+\nu e^{-x_0/\nu}/C_1$.

The key assumption in this derivation is that the support curve and the infinite-system conductivity curve have the same asymptotic curvature. The physical assumption that each size of system correctly describes the infinite limit conductivity to a fixed distance from its asymptote is sufficient to ensure this. Actually, the numerical study gives the value 0.6 for the quantity ν suggesting that the intersection point of the asymptotes, α in Eq. (2.8), moves out faster than the point at which one would expect each size system to settle on its asymptote. The latter one guesses to go as $N^{1/3}$ does, since $N^{1/3}$ is roughly proportional to the number of links in the critical path and therefore its inverse is proportional to the difference between the worst and the second-worst links in the best chain. To resolve such a difference, α has to grow accordingly. Therefore a value $\nu<1$ increases ones confidence in the above derivation as the support curve and the infinite-limit conductivity merge at large α.

Because the above construction is independent of exactly how the finite system is subjected to the electrical voltage, i.e., the exact boundary conditions used, the derivation should hold as long as percolation in the finite system is so defined that it tends to the correct limit when $N\to\infty$. All boundary conditions properly defined in this sense should lead to the same value of ν. What is meant by different boundary conditions is periodic or nonperiodic.

III. NUMERICAL STUDIES

An extensive computer study of percolation in random arrays was carried out to determine the quantity r_{cN} as a function of N. Here two methods were used, the second vastly more efficient than the first. The first method was directly inspired by the previous work in I. The idea was to locate the most-power-dissipating conductor in the system with periodic boundary conditions and a voltage imposed accordingly, namely so that a point and its forward periodic image were separated by unit voltage. The network equations were iterated for the potentials of the points and the process was stopped if the most-power-dissipating resistor no more changed when α was increased. In spite of rather elaborate tricks to improve its convergence, this method remains costly and somewhat untrustworthy, since the most-power-dissipating resistor for finite α may not be the same as for $\alpha=\infty$. Therefore, an algorithm was adopted that just locates the first continuous path across the system when the linking radius was stepwise increased. The same periodic boundary conditions were used as in the first method, now in the sense that percolation was considered taking place when any point and its periodic image in the z-direction got linked with each other. Where both approaches were applied, perfectly accordant percolation radii were obtained.[13]

The results of the computation are shown in Fig. 3. Typically 150 arrays were run for each value of N, only 50 however for $N=2000$. The optimum values of the parameters in Eq. (1.3) were determined by minimizing χ^2 with the result $p_c=0.347$, $\nu=0.6$, and $A=0.047$. To see how uncertain this prediction may be, two constrained minimizations were also performed. In one case the points $N=32$ and $N=2000$ were left out, as the rest of the points superficially seem to imply a higher p_c. The result is shown as the lowest curve in Fig. 3, $p_c=0.358$, $\nu=0.333$, and $A=0.24$. This looks like a fairly firm upper limit for p_c since the exponent ν is just at the value $\frac{1}{3}$ which is its lower bound.[1] For a lower limit $\nu=0.833$ was taken. Then the result is $p_c=0.337$, $A=0.028$. The value of χ^2 in this case was 6.3 for five degrees of freedom, whereas it was 4.5 for four degrees of freedom in the best fit giving $p_c=0.347$. Thus, this lower limit is not nearly as firm as the upper limit for p_c. The number $\nu=0.833$ was chosen for no better reason than the fact that it makes the plot of $\ln(r_{cN}-r_c)$ in Fig. 3 parallel with the best straight-line fit for the logarithm of the width w_N (the symbol σ is used for conductivity) of the distribution of r_{cN} in different arrays of N points. The fact that the optimum slope of $\ln(r_{cN}-r_c)$ is larger than the latter seems to imply that the width of the uncertainty in r_{cN} decreases slower than its mean value moves to the infinite limit r_c.

Up until I, the history of the number p_c, to the knowledge of the author, is as follows: Roberts

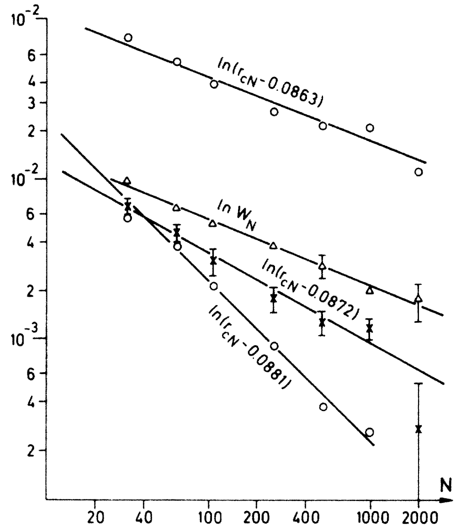

FIG. 3. Topmost and the two bottommost curves depict three different straight-line fits to the quantity $\ln(r_{cN}-r_c)$. The second line from the bottom is the best fit with standard deviations marked on the computed points. The lowest and highest curves are the upper- and lower-bound estimations for r_c and hence p_c. [Although the highest curve looks like a good fit, one must keep in mind that the standard deviations on the logarithmic scale are smaller when $(r_{cN}-r_c)$ is larger. Also the point $N=2000$ does not have the same weight as the others.] The second line from the top, marked with triangles, shows the logarithm of the width of the distribution of r_{cN} for arrays of N points with two typical uncertainties. The scales and the r_c marked in the figure refer to a density of 1000 points in a unit cube, the higher scale is for the topmost curve and the lower for the rest. The unit of length is the edge of the box.

and Storey,[7] $p_c = 0.37 \pm 0.015$; Holcomb and Rehr,[8] $p_c = 0.29 \pm 0.03$; Holcomb, Iwasawa, and Roberts,[9] $p_c = 0.29$; Pike and Seager,[10] $p_c = 0.34$, with unspecified confidence limits; Dalton, Domb, and Sykes or Domb and Dalton,[11] $p_c = 0.34$, by extrapolating to long-range interaction from series expansions on ordered lattice as opposed to the Monte-Carlo-like computer methods used in the other references. Only the last two of these are in fair agreement with the present estimate $p_c = 0.347$. As far as the work reported in I is concerned, $p_c = 0.30 \pm 0.015$, the probably-too-low result, arises from the difficult convergence of the iteration process. It seems to be a property of the scheme used in that study that the badly converged currents were always too high, thus warping the result. Further iterations not only improved the convergence but also pulled down the currents. It should be stressed that in spite of this numerical error, all the conclusions reached in that work remain correct. Holcomb and Rehr[8] have recently considered some nonlinear extrapolation procedures that indicate that p_c may in fact be higher than their previous estimate, based upon a linear extrapolation. It may be worth pointing out that any error in r_c is magnified in the dimensionally invariant number p_c and it is usually r_c which is determined by computation. Also one should keep in mind that p_c probably, at least if determined by the procedure used here, is inordinately sensitive to the quality of the random numbers used as discussed in Appendix D of I. In the present investigation the extra correlation removing device described in I was used with both the IBM generator RANDU and the CDC generator RANF.

IV. CONCLUSION

In this work the nonexponential prefactor of the electrical conductivity in an array of random points linked with conductances varying exponentially with separation distance has been considered. The prefactor has been shown related to the size dependence of the percolation threshold in random arrays. Exploiting the thus established extrapolation formula for the infinite system percolation threshold, the nonexponential prefactor exponent ν [Eq. (1.2)], and the dimensional invariant p_c for percolation have been determined. The results for p_c are higher than most previously published, probably larger than 0.337, and fairly certainly smaller than 0.358. For ν the value 0.6 is found with an estimated uncertainty ± 0.25. The temperature-dependent prefactor of the four-dimensional Mott random-hopping model is discussed in the Appendix.

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APPENDIX

The result

$$\sigma \propto (1/r_c\alpha)^{0.6} e^{-\alpha r_c} \tag{A1}$$

can be used to sharpen the bound suggested for the prefactor of the full four-dimensional Mott model in Appendix C of I. In the percolation construction[3] the quantity E_{max} varies as $T^{3/4}$ changing the effective density of traps in the same fashion. Hence there is a purely geometrical factor $T^{1/4}$ in front of the main exponential dependence of the conductivity. In Appendix C of I it is suggested that an additional factor $T^{\nu/4}$ should follow from (A1) since the prefactor depends on $\rho^{\nu/3}$, or on T as $T^{\nu/4}$. The resulting nonexponential factor would be $T^{(1+0.6)/4} = T^{0.4}$. However, the fourth dimension has been neglected in this analysis. The temperature-dependent contribution should not affect the prefactor were it not that E_{max} decreases slower than linearly with the temperature. The rate at which tolerable upward deviations from E_{max} decrease is linearly with T (since the exponent depends on E/kT). Therefore, the lowering of the temperature leads to a stricter limit E_{max} and a more constrained situation and therefore presumably a smaller current. Hence one expects the exponent $\frac{1}{4}(1+\nu)$ to be a lower bound.

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*Present address: IFF/Kilogram Frederick Air (KFA), 517 Jülich 1, Germany.

[1]V. Ambegaokar, S. Cochran, and J. Kurkijärvi, Phys. Rev. B 8, 3682 (1973).

[2]N. F. Mott, Philos. Mag. 19, 835 (1969).

[3]V. Ambegaokar, B. I. Halperin, and J. S. Langer, Phys. Rev. B 4, 2612 (1971).

[4]See, for instance, H. L. Frisch and J. M. Hammersley, J. Soc. Indust. Appl. Math. 11, 894 (1963); or, for a more recent review, V. K. S. Shante and S. Kirkpatric, Adv. Phys. 20, 325 (1971).

[5]P. Dean, Proc. Camb. Philos. Soc. 59, 397 (1963).

[6]M. F. Sykes and J. W. Essam, Phys. Rev. 133, A310 (1964).

[7]F. D. K. Roberts and S. H. Storey, Biometrika 55, 258 (1968).

[8]D. F. Holcomb and J. J. Rehr Jr., Phys. Rev. 183, 773 (1969); and private communication.

[9]D. F. Holcomb, M. Iwasawa, and F. D. K. Roberts, Biometrika 59, 207 (1972).

[10]G. E. Pike and C. H. Seager, Bull. Am. Phys. Soc. 18, 307 (1973).

[11]N. W. Dalton, C. Domb, and M. F. Sykes, Proc. Phys. Soc. Lond. 83, 496 (1964); C. Domb and N. W. Dalton, Proc. Phys. Soc. Lond. 89, 859 (1966).

[12]Below, since a larger system is not constrained to conduct through a particular part of it but can look for others *better than average*.

[13]Strictly speaking, the boundary condition used here is not quite periodic. Any infinite cluster which needs more than one box to hook up with itself, or loops back at some point more than one and a half boxes, will go unrecognized in one of the three spatial directions but will be recognized if it occurs in the other two. This should not change the infinite volume result as any cluster of the type described implies a double percolation, in some sense, without the presence of single percolation and is therefore of small statistical weight.