shifts and therefore does not account for the 0.13-eV reduction in E_g that Donovan and co-workers[4,10] observed when they decreased T_s from 300 to 20°C. This downward shift with decreasing T_s may instead be due to dilation of the amorphous material with diminishing island size, associated with the enhanced proximity of all atoms to surfaces of the islands. Using pressure data for crystalline Ge,[21] the 0.13-eV downshift corresponds to a 1.3% increase in the nn distance. This adds a density deficiency of ~4% to the 5% assigned to voids and yields a total deficiency of ~9%, in good agreement with that measured directly by DAS.[4]

Films formed under different conditions of substrate temperature, atom flux, and gaseous contamination will perforce have differing void structures. Moreover, the voids existent after annealing at a temperature $T_a > T_s$ may not have the same character as those obtained by direct growth on a substrate at temperature T_a. Connected networks of submicroscopic cracks will strongly affect many other physical properties, as will be discussed elsewhere.

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Percolation Theory and Electrical Conductivity

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We report the results of an experiment to determine the bulk conductivity of a sheet of colloidal graphite paper with holes randomly punched in it. The behavior of the conductivity is found to be quite different from that of the percolation probability.

It has been argued by Ziman[1] that the mobility of an electron in a disordered semiconductor should be given by a solution of the classical percolation problem. This idea has been explored by a number of workers, and Eggarter and Cohen[2] in particular have assumed that the mobility should be proportional to the percolation probability $P(p)$.[3] Percolation theory has also been used for the theory of hopping conductivity in semiconductors,[4] and for a model of switching in amorphous semiconductors.[5] There is, however, no theory of the electrical conductivity of a classical percolation system, and so we have measured the conductivity of a two-dimensional system of this sort by measuring the resistance of a sheet of conducting paper with holes randomly punched in it. It is found that the conductivity drops to zero much less sharply than $P(p)$ near the critical concentration of the holes. This is because near p_c a lot of the paths available for conduction either are dead ends or become very constricted in places, and thus contribute very little to the electrical conductivity.

The system consists of a sheet of conducting colloidal graphite paper measuring 5 in. (127 mm) square, covered by a square grid of spacing 0.1 in. (2.54 mm). This represents a square lattice of 2500 sites, each of which may be "open" or "closed." A hole punched in the paper represents a "closed" site. The holes are approximately 4 mm in diameter and thus first- and second-nearest neighbors overlap, as shown in Fig. 1. The reason for making the hole size larger than the size of a "site" was to avoid problems due to two nearest-neighbor holes not completely blocking off the "bond" between them. As far as the critical concentration is concerned, the system may be considered to be equivalent to a site percolation problem on a square lattice with nearest-neighbor bonds.

The conductance of a square of paper 1×1 cm was $42.69 \times 10^{-5}\ \Omega^{-1}$ so that the resistance of the sheet varied during the experiment from about 2 to 500 kΩ at the critical concentration of holes.

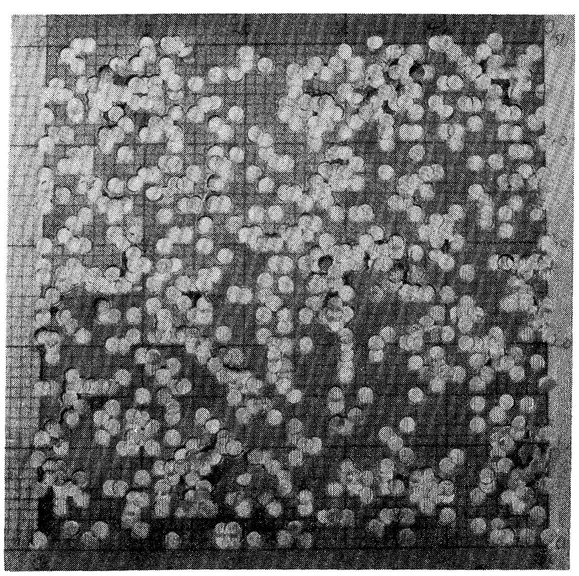

FIG. 1. Photograph of the sheet of conducting paper at the stage where the concentration of holes is 0.268.

The holes were punched randomly in the paper with the aid of a table of random numbers to determine the two coordinates. The resistance of the sheet was measured after every 25 pairs of numbers (i.e., approximately every 1% holes, allowing for repeated pairs) by comparison with a standard resistance. High-accuracy and high-input-impedance digital voltmeters were used, and the measurements were corrected for the effect of the small currents taken by the meters.

The results of the experiment are shown in Fig. 2(a), which is a plot of the ratio of the conductance of the paper to its initial value against the concentration of holes. The conductance becomes zero when there are no conducting channels across the paper, and this occurred at a concentration

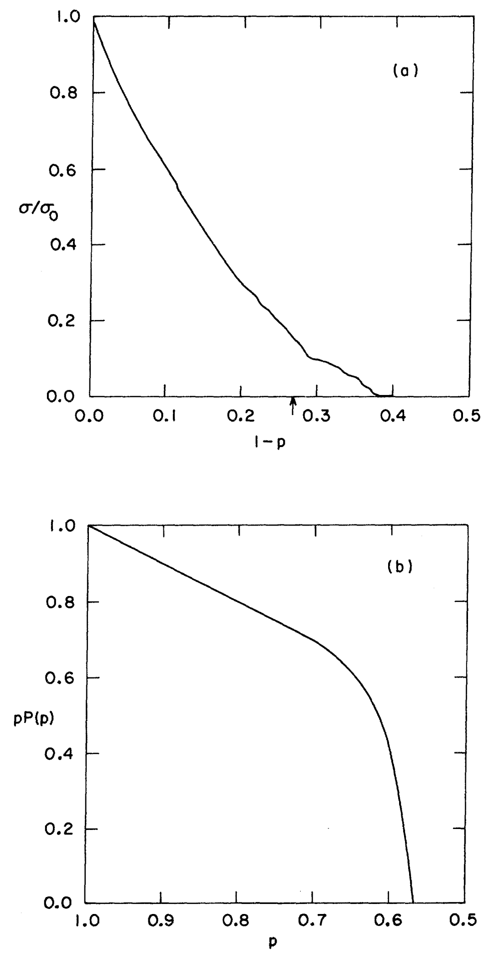

FIG. 2. (a) Graph of the conductivity as a function of the concentration of holes $(1-p)$. The bulk conductivity of the conducting paper is σ_0. The arrow shows the point at which the photograph of Fig. 1 was taken. (b) Graph of $pP(p)$ for the site problem on the square lattice, where p is the concentration of "open" sites (from Ref. 3).

of holes of 0.40, i.e., a concentration of open sites of 0.60, which is close to the estimate of 0.59 ± 0.01 for the critical proportion of open sites for this problem.[6] This curve has curvature opposite to that of the curve for $pP(p)$, the proportion of sites belonging to an infinite cluster, as can be seen from Fig. 2(b). We account for the difference between these two curves by observing that, for p close to its critical value, much of the material contributes very little to the conductivity, because, although it is connected to the rest of the material, channels through it are tortuous and constricted. In Fig. 1 it can be seen that although almost all the material is connected, the resistance is quite high because of the many constrictions of the channels. Dead ends appear in greater and greater numbers as the proportion of holes is increased.

Our results for a low concentration of holes follow a reasonably smooth curve which can be accounted for to a first approximation by an effective medium theory in which the loss of conductivity is proportional to the area of paper removed.[7] Such a smooth curve would go to zero at a concentration of holes considerably less than the critical concentration, and the conductivity in the critical region lies above this smooth curve. Our results contain too large a statistical error for us to be able to determine the shape of this tail of the conductivity curve, but from these results, and from the results of a preliminary run, it seems that the limiting slope is zero, but the approach to zero is more rapid than that of a parabolic curve.

For most applications of the theory[1,2,4] we are interested in a network of resistances which do not all have the same value, as they do in this problem or in the standard percolation problem, but which vary continuously in some range. One approximation which is used[4] is to replace those resistances less than some value R_C by a resistance R_C, and to remove those resistances greater than R_C, so that a percolation problem is obtained as a variational bound on the problem. It has been argued that R_C should be chosen so that the network formed is critically connected. The results obtained here show that in this case the resistance of the network would be very large. A better bound on the resistance would be obtained by choosing a larger value of R_C so that the network is better connected. With this larger value of R_C the network should no longer be in the critically connected region, and an effective medium theory of the network might be appropriate. The theoretical value of the critical concentration for percolation is therefore likely to be irrelevant in such problems, and should be replaced by the higher concentration of carriers (lower concentration of holes) at which the smooth curve extrapolates to zero.

We do not have experimental results for three-dimensional systems, but arguments similar to those used here suggest that the conductivity should go to zero less abruptly than the percolation probability in that case also. We believe that it is more appropriate to compare the mobility of an electron in a disordered system with the conductivity than with the percolation probability.

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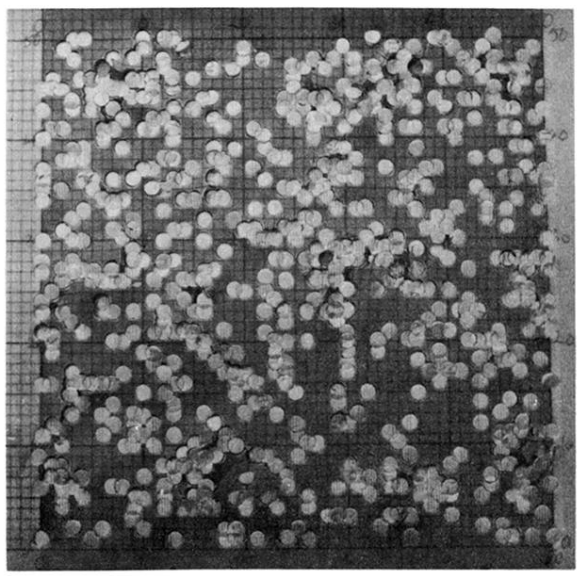

FIG. 1. Photograph of the sheet of conducting paper at the stage where the concentration of holes is 0.268.