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Local-environment model for percolation in amorphous Mo-Si alloys

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A local-environment model is employed to determine whether the chemical short-range ordering, observed in some amorphous materials, can influence the metal-nonmetal transition that occurs as a function of composition when a metal is alloyed with an insulator or semiconductor. The model uses as its basic structural unit clusters with different short-range order. Both the case of random occupancy of sites within the cluster and of chemically driven diffusion of atoms between the clusters are considered. Though the threshold for the transition is different in the two cases, the concentration dependence of the conductivity above the threshold is similar. The model provides a relationship between the volume fraction of the material that is conducting, f_v, and the concentration of conducting atoms, $1-x$. With the use of this relationship, the result from percolation theory that the conductivity is proportional to $(f_v-f_{vc})^{1.8}$ where f_{vc} is the percolation threshold can be expressed as $(x_c-x)^{t_{\text{eff}}}$ where t_{eff}, which is less than 1.8, is an effective percolation exponent. With the use of a cluster size $n=40$, the predicted concentration dependence of the conductivity is very different from the predictions of classical percolation theory and is in agreement with experimental results on amorphous Mo-Si alloys. For $n=40$, t_{eff} is approximately equal to 0.5.

I. INTRODUCTION AND MOTIVATION

The atomic arrangement of amorphous films of pure elements prepared by vacuum deposition on low-temperature substrates[1] and of some metal-metal amorphous materials[2] can be fairly well described by the dense random-packing model.[3] The atomic arrangement is less random in metal-metalloid (Si, Ge, As, B, and P) amorphous alloys because of the charge transfer between the metal and metalloid atoms which results in strong metal-metalloid bonds that tend to inhibit the metalloids from being nearest neighbors of one another. Further evidence of the importance of local environment effects is provided by the fact that theoretical models[4] of the probability distribution of the number of near neighbors in Fe-B glass alloys sharpens near the middle of the known composition range. This composition is close to the eutectic composition of 17 at. % B. This narrowing of the probability distribution suggests that a more stable structure is favored by more uniform short-range order.[5] Thus, these metal-metalloid glasses exhibit clear evidence of chemical short-range ordering effects. Such effects are even more important in amorphous insulators and semiconductors where coordination number, covalent bonding angle, and bond-length requirements are approximately satisfied. For example, the results of Raman scattering measurements on amorphous SiS_2 and $SiSe_2$ have been used[6] to suggest that these materials possess not just short-range order, but also medium-range order.

Here we shall consider how these chemical effects alter the metal-nonmetal transition that occurs in an alloy of a metal and a nonmetal when the concentration of the metallic species is reduced below a critical value called the percolation threshold. Above this threshold the electrical conductivity σ at $T=0$ K is predicted to be given by

$$\sigma=\sigma_0(p-p_c)^t,\quad p>p_c \tag{1}$$

where p is the occupation probability of conducting sites and p_c is the value of p at the percolation threshold. This equation has been applied with different values for p_c and t to very different physical situations. For example, it has been used[7] to describe the conductivity of a three-dimensional mixture of conducting and insulating spheres. In this case classical percolation theory applies[8–10] and t is approximately equal to 1.8. In classical percolation theory the value of t depends on the dimension of the system. This paper treats three-dimensional systems. On the other hand t should not depend upon such details as the lattice symmetry or even whether the material is crystalline or amorphous. This insensitivity to the details of the system near the percolation threshold is called universality and arises, as in the case of a thermodynamic phase transition, because there is a characteristic length which diverges at the transition.

The situation in which one must include quantum effects as well as disorder is described as Anderson localization. In this case, if interaction effects can be neglected, t is predicted[11] to be approximately equal to 1.0. The percolation-localization crossover near the percolation threshold has been treated using scaling by Gefen *et al.*[12] and Shapiro.[13]

Experimentally one finds that different values for the parameter p_c and the exponent t are observed in different systems. For example, in experiments on quenched films of Ar-La mixtures[14] it was observed that $p_c=0.15$ and $t=1.6$. These values are in approximate agreement with the predictions of classical percolation theory. Higher values for p_c of approximately 0.5 have been observed[15,16] when a metal is codeposited with an insulator and phase separation occurs. The exponent t was measured[17] at very

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low temperatures to be 0.5 in phosphorous-doped Si and to be 1.0 in amorphous Nb-Si alloys[18] and $Gd_{3-x}v_xS_4$ (Ref. 19) where v stands for vacancies. Further t is larger than predicted by classical percolation theory in some systems, such as quenched films of certain alkali-metal alloys,[20] where values as large as 3 have been observed. Note, however, that some of the differences in the values for t discussed above may be due to the fact that t was determined in different temperature ranges.

In amorphous Mo-Si alloys[21] (a-Mo-Si), the metal-nonmetal transition may be altered by chemical effects. Figure 1 shows a comparison of the 295 and 25 K conductivity of amorphous $Mo_{1-x}Si_x$ with the prediction of Eq. (1) for $t=0.5$, 0.6, 0.8, 1.0, 1.2, 1.6, and 1.8. The theoretical curves have been normalized to agree with experiment at $1-x=0.7$. One sees that the transition is more abrupt than predicted by classical percolation theory, i.e., with $t=1.8$. The data is much better fitted using a value of 0.5. It is unlikely that this small value is due to Anderson localization. Normally Anderson localization is only observed at very low temperatures where the inelastic scattering length becomes very long. An alternative possibility is that local chemical environment effects are responsible for reducing the effective value of t.

In examining this possibility it is useful to consider the Mo-Si phase diagram.[22] Since this diagram shows a relatively deep eutectic and a very stable congruently melting compound, $MoSi_2$, it is clear that chemical effects are very important in this system. In a-Mo-Si the bonding and the number of electrons that the Si atoms contribute to the conduction band probably depend upon the local environment. Information about the Si contribution to the conduction band can be obtained from superconducting transition-temperature measurements[21] by using the results of Collver and Hammond.[23] They found that the superconducting transition temperature T_c of many amorphous superconductors could be fitted by the same function of the electron per atom ratio, e/a. Figure 2 compares the measured T_c of a-Mo-Si for $x<\frac{2}{3}$ with their function. The e/a values for a-Mo-Si were computed by assuming that the Mo and Si atoms contribute 6 and 4 electrons, respectively, to the conduction band. One sees that with this assumption, the measured T_c of a-Mo-Si agrees with their function. For $x>\frac{2}{3}$, the measured T_c drops abruptly to zero. Thus, for $x<\frac{2}{3}$ most Si atoms probably contribute several electrons to the conduction band.

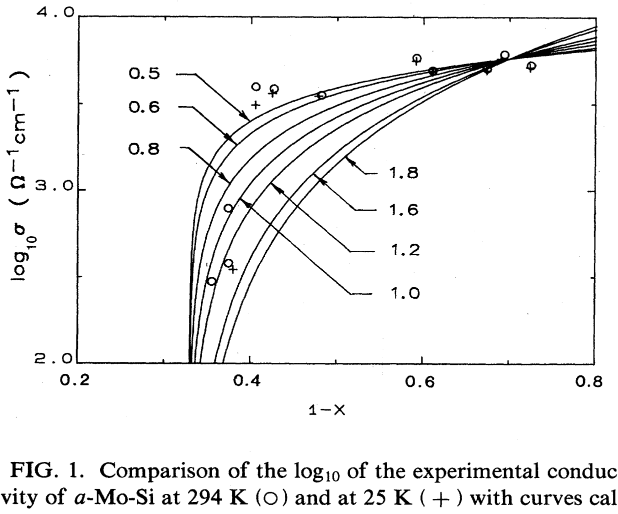

FIG. 1. Comparison of the $\log_{10}$ of the experimental conductivity of a-Mo-Si at 294 K (○) and at 25 K (+) with curves calculated using Eq. (1) with $t=0.5$, 0.6, 0.8, 1.0, 1.2, 1.6, and 1.8. The curves have been normalized to agree with experiment at $1-x=0.7$. Classical percolation predicts that $t=1.8$.

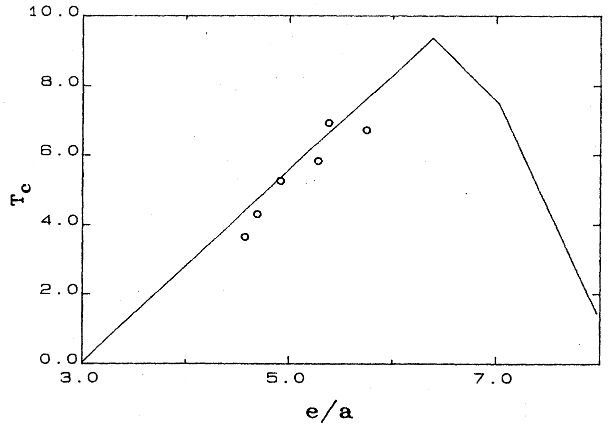

FIG. 2. Comparison of the experimental superconducting transition temperatures T_c of a-Mo-Si with the electron per atom e/a dependence (solid curve) that is satisfied by many amorphous superconductors. (See Ref. 23.) The e/a values were obtained by assuming that Mo and Si contribute 6 and 4 electrons, respectively, to the conduction band.

The model uses a more specific assumption about local environment effects. It assumes that local regions in which $\frac{1}{3}$ or more of the atoms are Mo atoms are metallic. Further evidence for the assumptions made concerning local environment effects is provided by x-ray measurements[21] in the metallic regime of amorphous $Mo_{1-x}Si_x$. These measurements show that the local environment smoothly evolves with increasing x until $x=0.63$. Since this is nearly the Si concentration in the metallic compound $MoSi_2$, it is possible that the local environment evolves in the metallic regime with increasing x until it resembles crystalline $MoSi_2$. If this occurs, it is relevant that photoemission spectroscopy[24] on crystalline $MoSi_2$ shows that Si p states are deep in the conduction band and the Fermi level lies on the high-energy side of a Mo d-band peak. Band-structure calculations[24] on refractory metal silicides show that Mo d states hybridize with lower-lying Si p states to form bonding complexes.

Here we include chemical effects in a model treating the metal-nonmetal transition in a-Mo-Si to test whether these effects can give rise to the small effective value of t, t_{eff}, discussed above. The model incorporates local environment effects into classical percolation theory by assuming that the basic structural units in the material are clusters of atoms. The basic entities entering into the conduction process are the clusters and not the individual atoms. Hence the percolation threshold is reached when a sufficient fraction of clusters are conducting. Whether a given cluster is conducting or not is assumed to depend upon its composition and thus the local environment. Using such clusters is very reasonable if phase separation occurs. There is evidence,[21] based upon transmission electron micrographs, that a-Mo-Si does phase separate for $x \gtrsim \frac{2}{3}$ into two amorphous phases, one metallic and the other resembling amorphous Si. In the absence of phase separation the clusters can be viewed as a way of representing regions with different local environment. It should be noted that cluster models have been used previously[25,26] to describe amorphous materials.

The three main conclusions of this study are as follows. (1) The metal-nonmetal transition that occurs as a function of composition can become more abrupt, as discussed above in the case of a-Mo-Si, when chemical effects are important. (2) The abruptness of the transition increases as the range of the short-range order increases. (3) For the cluster size which best fits the data, the model's results are equivalent to a small value of approximately 0.5 for the effective exponent t_{eff}. To test whether these conclusions are sensitive to the specific form of the model, two versions are presented. In one the composition of the clusters is assumed to be completely random. In the other chemically driven diffusion of atoms between the clusters is included. Since the composition dependence of the conductivity above the percolation threshold is approximately the same for both forms of the model, all three main conclusions hold for either form of the model.

II. LOCAL ENVIRONMENT MODEL WITHOUT DIFFUSION

The present model is, in part, based upon the ideas of Gaskell[25] and Gilman.[26] Instead of using the dense random-packing model mentioned above in which the basic constituents are spherical atoms, they independently suggested that the way to incorporate chemical short-range order in transition-element—metalloid glasses was to consider them composed of new structural units, which are polyhedron with definite short-range order and bonding arrangements. Here clusters of atoms, which are analogous to their polyhedron, are used as the basic building blocks in a model for treating a-Mo-Si. The motivations for the model's assumptions incorporating local environment effects were discussed above. The assumptions of the first form of the model are as follows.

(1) The amorphous material is composed of clusters of atoms in which each cluster contains the same number of atoms, denoted by N.

(2) Individual clusters are either primarily metallically bonded and are treated as metallic, or primarily covalently bonded and treated as insulators.

(3) A cluster is metallically bonded if and only if $m/N \geq \frac{1}{3}$, where m is the number of Mo atoms in a cluster. If $m/N < \frac{1}{3}$ a cluster is covalently bonded. This assumption is based on the idea that if $(N-m)/N = \frac{2}{3}$ is the highest local Si concentration which supports metallic bonding, then the replacement of a single Mo atom by a Si atom in such a cluster will remove most of its metallic character.

(4) Random occupancy is assumed, i.e., the probability $P(m,N,x)$ that a cluster of size N has m Mo atoms is

$$P(m,N,x) = \frac{N!}{m!(N-m)!} x^{(N-m)} (1-x)^m \,. \tag{2}$$

(5) The volume $V_c(m,N)$ of a conducting cluster is obtained from a linear interpolation between the end points $x=0$ and $x=\frac{2}{3}$ assuming that their densities are 95% that of crystalline Mo and $MoSi_2$. The volume of an insulating cluster $V_i(N)$ is that of N-Si atoms having a density 95% that of crystalline Si.

(6) Classical percolation theory can be used to calculate the conductivity σ, i.e., σ is given by Eq. (1) where we chose $t=1.8$ and replace the variable p, the occupation probability, with the volume fraction of the material that is conducting f_v. These two variables are equivalent in the case of classical percolation if the metallic and insulating atoms are the same size and one assumes that they occupy all of the material. It should be noted that Scher and Zallen found[8] that the fraction of space occupied by conducting atoms was the same at the percolation threshold for different lattices and equal to 0.15, whereas the percolation thresholds, expressed in terms of p, are very different.

From these assumptions it follows that the conducting volume fraction is given by

$$f_v(N,x) = \frac{\sum\limits_m V_c(m,N) P(m,N,x)}{\sum\limits_m V_c(m,N) P(m,N,x) + V_i(N) \sum\limits_{m'} P(m',N,x)} \tag{3}$$

where $N/3 \leq m \leq N$ and $0 \leq m' < N/3$. The conducting volume fraction $f_v(N,x)$ is plotted in Fig. 3 versus the atomic concentration of Mo, $1-x$, for various values of N. There is a large step in f_v at $x=\frac{2}{3}$ that sharpens with increasing N which is a consequence of assumption (3) and the fact that large deviations from the average composition x becomes less likely with increasing N. Since the step in f_v occurs near $x=\frac{2}{3}$, it is reasonable to choose the percolation threshold, x_c, equal to 0.67. Making this

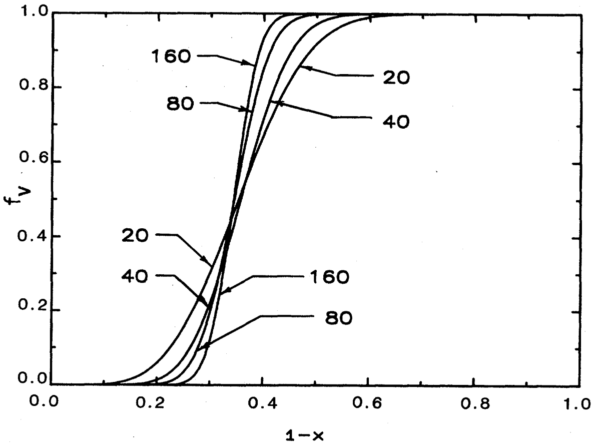

FIG. 3. Conducting volume fraction f_v predicted by the first version of the model, i.e., without diffusion, vs the Mo concentration $1-x$ for $N=20,40,80,160$.

choice and using Eqs. (1)—(3), the conductivity σ has been calculated and plotted in Fig. 4 versus $1-x$ for $N=20$, 40, 80, and 160. The curves have been normalized to agree with the experimental conductivity of amorphous $\mathrm{Mo}_{1-x}\mathrm{Si}_x$ at $1-x=0.7$, $T=295$ K. As seen in Fig. 4, the experimental results for the metal-nonmetal transition in the high-conductivity region of interest here are only slightly different at lower temperatures. The calculated conductivity agrees approximately with the experimental conductivity[21] at $T=295$ K for $20<N<80$. These bounds on N correspond to amorphous Si clusters which are between 9.3 and 15 Å in diameter. One can use these results to obtain a value for an effective exponent t, t_{eff}. Suppose one considers using the variable $1-x$ instead of the variable f_v in an equation of the form of Eq. (1) with t replaced by t^{eff}. By comparing the curves presented in Figs. 1 and 4, one sees that for $N=40$, t_{eff} is approximately equal to 0.5.

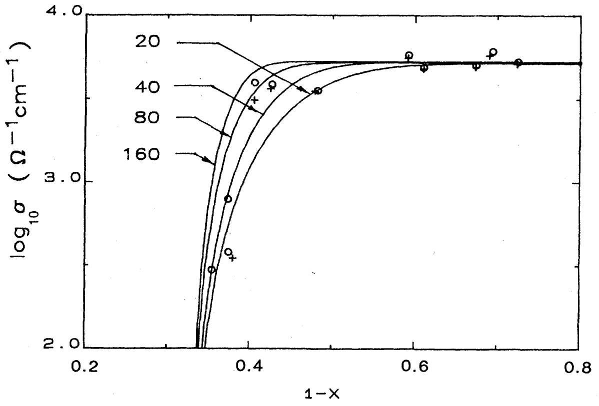

FIG. 4. Comparison of the model's predictions for the $\log_{10}$ of the conductivity without diffusion with $N=20,40,80,160$ as a function of $1-x$ with experimental data on a-Mo-Si at $T=294$ K, denoted by the symbol (○) and at $T=25$ K, denoted by the symbol (+).

III. LOCAL ENVIRONMENT MODEL WITH DIFFUSION

There are two reasons for considering an alternative form of the model. First, one would like to investigate the sensitivity of the predicted conductivity to the specific form of the model. Second, in the first form of the model, it is assumed that Mo atoms can exist in the insulating phase in which the Si atoms are presumably covalently bonded in a tetrahedral arrangement. It is not clear how one can incorporate Mo atoms in such a structure. For these reasons an alternative form of the model is considered in which Mo atoms diffuse from covalently bonded clusters. In this form the model's assumptions for the probability distribution are the following.

(1) Before diffusion occurs each cluster contains N atoms and their occupancy is random, i.e., it is given by Eq. (2).

(2) Only Mo atoms from covalently bonded clusters diffuse.

(3) Mo atoms diffuse from covalently bonded clusters containing m Mo atoms and n Si plus Mo atoms if the probability of finding such a cluster $P(m,n,x)$ is greater than some allowed probability A.

(4) Mo atoms diffuse first from the most unstable (lowest value of m/n) clusters with $P(m,n,x)$ greater than A to any of the more chemically stable clusters with an equal probability. Once the process of diffusion from a given type of cluster begins, it continues to completion through the sequence $m\rightarrow(m-1)\rightarrow(m-2)\rightarrow(m-3)\rightarrow\dots\rightarrow 0$.

(5) Diffusion occurs from all clusters with a given m before it begins on the next higher value of m.

To compute the conductivity from the distribution after diffusion has occured, assumption (6) in the first form of the model is used and assumption (5) for the volume of conducting and insulating clusters is generalized to incorporate the variable cluster size. Assumption (3) is made to prevent the matrix $P(m,n,x)$, which is used to describe the probabilities at fixed x, from becoming too large. This difficulty does not arise for small values of N. Physically one can interpret A as allowing Mo to exist in covalently bonded clusters. This situation, which is a higher level of metastability, would be important if the chemical driving energy is less than the diffusion barrier. In the limit $A\rightarrow 1$, no diffusion occurs. In calculating all the results presented below, A was kept equal to 0.04. Figure 5 shows the region in the m,n plane where diffusion occurs. In this figure the clusters diffuse along diagonal trajectories. The region that is labeled as the stable region indicates the region containing the metallically bonded clusters which never lose Mo atoms. In this region, the trajectories are only in one direction. A single step in the diffusion process, which reduces the probability of occupancy of the cluster m,n to 0, affects the probabilities as follows:

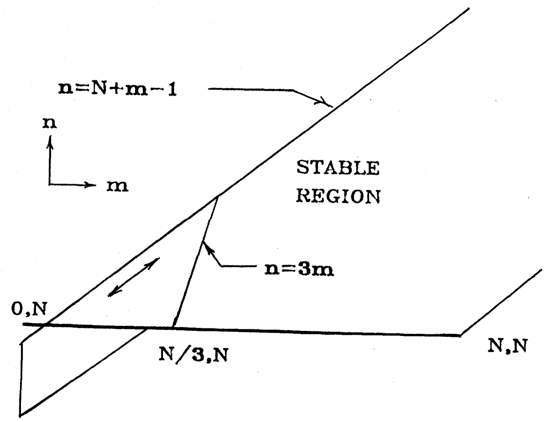

FIG. 5. Region in the m,n plane where diffusion occurs. The clusters are distributed along the horizontal line $n=N$ between the points $(0,N)$ and (N,N) before diffusion occurs. The clusters in the stable region can only gain Mo atoms. The cluster's move is along diagonal trajectories.

$$
\begin{aligned}
&P(m-1,n-1,x)\rightarrow P(m-1,n-1,x)+P(m,n,x)\ ,\\
&P(m',n',x)\rightarrow P(m',n',x)+P(m'-1,n'-1,x)P(m,n,x,)/Z\ ,\\
&P(m'-1,n'-1,x)\rightarrow P(m'-1,n'-1,x)-P(m'-1,n'-1,x)P(m,n,x)/Z\ ,\\
&\vdots\\
&P(m,n,x)\rightarrow 0\ ,
\end{aligned}
\tag{4}
$$

where Eq. (4) includes all m' and n' such that $(m'-1)/(n'-1)$ is greater than m/n, i.e., diffusion of Mo atoms can occur to any more stable cluster. This is done to satisfy the assumption that the Mo atoms diffuse to all more stable clusters with an equal probability. The quantity Z is equal to the fraction of the clusters which are more stable than the cluster m,n and is given by

$$Z=\sum_{m',n'}P(m',n',x)\ , \tag{5}$$

where the sum over m' and n' includes all clusters that are more stable than the cluster m,n. Steps of this kind are continued in the order described above until all probabilities, $P(m,n,x)$, of finding a Mo atom in a covalently bonded cluster ($m/n<\frac{1}{3}$) are less than A. The resulting probability distribution $P(m,n,x)$ for $N=45$, $x_c=0.67$, and $A=0.04$ is shown in Fig. 6. One sees, as expected, that there is a large probability of finding smaller clusters without any Mo atoms. For $N=45$, the volume fraction of stable clusters at the percolation threshold, $x=0.67$, computed by generalizing Eq. (3) to incorporate the variable-cluster size, is increased from 0.439 to 0.609 by diffusion. Since diffusion can convert an unstable cluster into a stable one, this result is just an obvious consequence of the model for diffusion. To illustrate how this increase in f_v changes with N, the volume fraction of conducting clusters f_v at the percolation threshold $x=0.67$ is plotted in Fig. 7 with and without diffusion for various values of N. One sees that there is a tendency for the increase of f_v caused by diffusion to increases as N increases. This tendency would be even larger if the parameter A were decreased, since the values of $P(m,n,x)$ decrease with increasing N. In the absence of an experimental determination of $f_v(x_c)$, we shall examine the concentration dependence of the conductivity above the percolation threshold by using the same value for the threshold x_c used in the first form of the model, i.e., x_c is kept equal to the approximate experimental value 0.67. This shifts the value of f_v at the percolation threshold. The conductivity computed in this way with and without diffusion is shown in Fig. 8 for $N=15$ and 45. One sees that despite the large increase in the volume fraction of stable conducting clusters, incorporating diffusion as done above does not greatly affect the concentration dependence of the conductivity above the threshold.

IV. CONCLUSION AND DISCUSSION

The present work demonstrates that the local environment can have a large effect on the conductivity in a metal-nonmetal transition. The size of the effect increases as the range of the short-range order increases. The model fits the experimental data for amorphous Mo-Si alloys for clusters containing about 40 atoms. This corresponds to a cluster size of approximately 10 Å and a value for the effective percolation exponent of approximately 0.5. The fact that the concentration dependence of the conductivity above the percolation threshold is very similar for both forms of the model suggests that the concen-

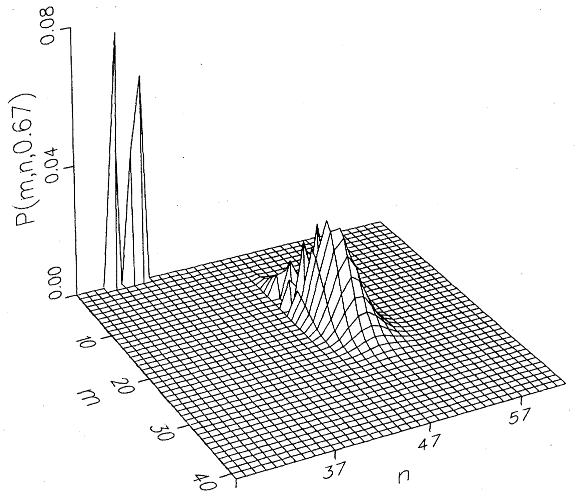

FIG. 6. Probability distribution $P(m,n,0.67)$ at threshold with $N=45$, and $A=0.04$ after diffusion has occurred.

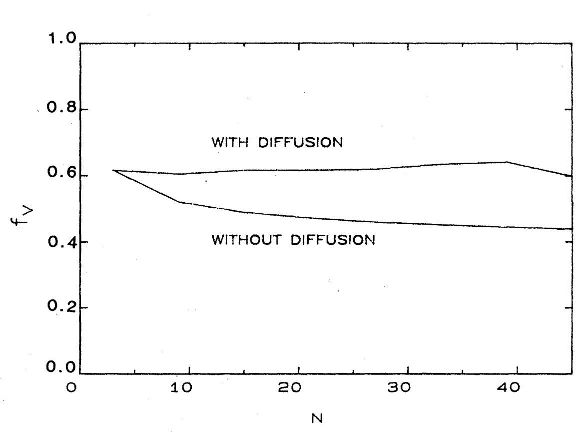

FIG. 7. Volume fraction of conducting clusters f_v at threshold, i.e., $x=0.67$ before and after diffusion vs N for $N=3$, 9, 15, 21, 27, 33, 39, and 45.

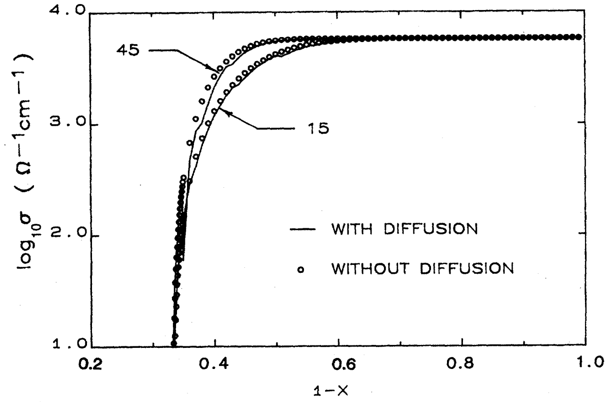

FIG. 8. Comparison of the predicted composition dependence of the $\log_{10}$ of the conductivity σ above percolation threshold with (solid curve) and without (symbol ○) including diffusion for $N=15$ and 45. For both cases the threshold is maintained at $x=0.67$.

tration dependence above threshold may be insensitive to the details of the model and may depend primarily on the size of the clusters. As discussed in the Introduction, it is natural to use such a cluster model if phase separation occurs. There is evidence[21] that for $x>0.67$ amorphous Mo-Si phase separates into an amorphous metallic phase and a covalently bonded phase. In the absence of phase separation the clusters can be considered as regions with different local environment. It should be noted that a related system, amorphous Mo-Ge exhibits a very different metal-nonmetal transition from the one described here. Specifically, in amorphous Mo-Ge alloys the metal-to-nonmetal transition is more gradual and occurs at low concentrations.[27] This is consistent with chemical effects being less important in this system. That these effects are smaller is not surprising since the Mo-Ge phase diagram[28] shows only peritectic compounds, none of which are as stable as $MoSi_2$. Furthermore this system has no eutectic. Another difference between these two systems is the theoretical heat of formation of alloys based upon the Miedema model.[29] Figure 9 shows a plot of this theoretical latent heat for the Mo-Si and Mo-Ge systems. One sees that, as expected, the negative heat of formation for $1-x$ near 0.6 is much larger for Mo-Si than it is for Mo-Ge, but not as large as the heat of formation[30] of $MoSi_2$. In addition for both systems, the heat of formation is predicted to be large and positive for dilute Mo concentrations. Thus there is a tendency for phase separation in this concentration region. Local chemical environment effects could be important in the quenched films of alkali metal films[20] discussed in the Introduction, but it is difficult to see, using the present model, how one can obtain values for the effective exponent t_{eff} which are greater than 1.8.

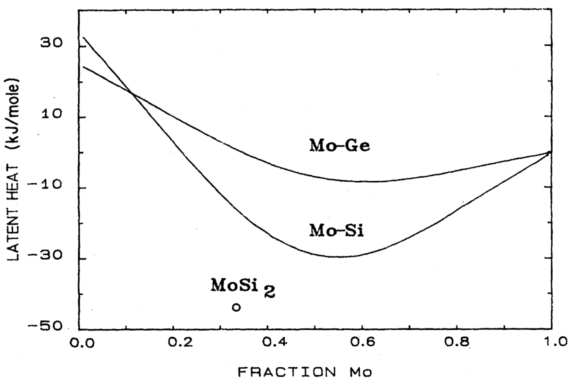

FIG. 9. Theoretical heat of formation vs Mo concentration $1-x$ for Mo-Si and Mo-Ge. Also shown is the heat of formation of $MoSi_2$.

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