Self-organized criticality in the intermediate phase of rigidity percolation

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Experimental results for covalent glasses have highlighted the existence of a self-organized phase due to the tendency of glass networks to minimize internal stress. Recently, we have shown that an equilibrated self-organized two-dimensional lattice-based model also possesses an intermediate phase in which a percolating rigid cluster exists with a probability between zero and one, depending on the average coordination of the network. In this paper, we study the properties of this intermediate phase in more detail. We find that microscopic perturbations, such as the addition or removal of a single bond, can affect the rigidity of macroscopic regions of the network, in particular, creating or destroying percolation. This, together with a power-law distribution of rigid cluster sizes, suggests that the system is maintained in a critical state on the rigid-floppy boundary throughout the intermediate phase, a behavior similar to self-organized criticality, but, remarkably, in a thermodynamically equilibrated state. The distinction between percolating and nonpercolating networks appears physically meaningless, even though the percolating cluster, when it exists, takes up a finite fraction of the network. We point out both similarities and differences between the intermediate phase and the critical point of ordinary percolation models without self-organization. Our results are consistent with an interpretation of recent experiments on the pressure dependence of Raman frequencies in chalcogenide glasses in terms of network homogeneity.

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I. INTRODUCTION

The concept of rigidity percolation, first introduced about 25 years ago by Thorpe [1] based on work by Phillips [2], describes how an elastic network goes from floppy to rigid as constraints are added to it. This theory has been applied with success to many systems, including covalent glasses [3–5] and proteins [6]. In the last decade, however, experimental studies have shown that the rigidity phase diagram could be more complex than initially thought, uncovering the presence of an intermediate phase between the floppy phase and the stressed-rigid phase, with the system in the intermediate phase being rigid but unstressed [4,7–20].

A basic explanation for this new phase was first proposed by Thorpe *et al.* [3]. It was shown that when the network self-organizes in order to minimize the stress, the rigid but unstressed intermediate phase can indeed arise.

In the original work by Thorpe and collaborators [3,21], as well as subsequent simplified models of chalcogenide glasses by Micoulaut and Phillips [22,23], networks were constructed using an "aggregation" process, in which bonds or simple network units were added to the network without subsequent equilibration. More recently, Barré $et\ al.$ [24] have considered a thermodynamically proper model with an energy cost associated with stress and showed that in the canonical ensemble, the intermediate phase still exists. In a recent paper, we have confirmed this result for the T=0 version of the model of Barré $et\ al.$, but on a more realistic regular lattice, and also have shown that the intermediate phase is entropically feasible in actual physical systems. In

both the model by Barré *et al.* and our model, the intermediate phase has an unusual property: both percolating and nonpercolating networks coexist in the ensemble at all mean coordination numbers within the intermediate phase.

While providing a general support for self-organization, these previous studies did not look in detail at the properties of networks in the intermediate phase. Here we provide a first glimpse at some of these properties. First, we show that in both percolating and nonpercolating networks, the sizes of nonpercolating clusters have a power-law distribution. In effect, the system remains in a critical state over an extended range of mean coordinations, corresponding to a selforganized critical phase [25], but in an equilibrium system. Second, we find that adding or removing a single bond affects the rigidity of macroscopic parts of the network in the intermediate phase and in particular, can turn a nonpercolating network into a percolating one and vice versa. This property supports the interpretation by Wang and co-workers [18] of the puzzling response of vibrational frequencies to applied pressure that was observed in their experiments. Using our results, we address some intriguing questions that have to do with the unusual coexistence of percolating and nonpercolating networks in the intermediate phase. In particular, we show that percolating and nonpercolating networks can be considered identical in the intermediate phase, as both stay on the edge of percolation.

This paper is structured as follows. In the next section, we briefly review the intermediate phase. We then present our methodology. In the fourth section, we present our results on the properties of rigid clusters (both percolating and nonpercolating) in our model. In Sec. V, we look at the response of the network to local perturbations. In Sec. VI, we discuss how our results can help understand the experiments by Wang *et al.* [18]. We finish with conclusions.

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II. THE INTERMEDIATE PHASE

Using the mean-field approach first introduced by Maxwell [26] and known as Maxwell counting, we can define the rigidity of a network in terms of its number of zero-frequency motions, or *floppy modes*, F. In a d-dimensional network, each atom has d degrees of freedom. In an unconstrained network of N atoms, each degree of freedom corresponds to a floppy mode and thus F = dN. Assuming that each added constraint takes away one floppy mode, we can write

$$F = dN - N_C, \tag{1}$$

where N_c is the number of constraints in the network. As N_c increases, F decreases, until F=0 is reached, and then there are no floppy modes left and the network undergoes a rigidity transition from floppy to rigid where a percolating rigid cluster emerges in the network (rigid clusters are sets of mutually rigid atoms). Disordered networks can be conveniently characterized by the *mean coordination* $\langle r \rangle$, which is the average number of bonds connecting a site. In the mean-field approximation, the location of the rigidity transition only depends on $\langle r \rangle$ and not on other details, such as fractions of sites with a particular coordination. The transition occurs at $\langle r \rangle$ =4 for the triangular lattice of elastic springs and $\langle r \rangle$ =2.4 in chalcogenide glasses; in the latter case, it is assumed that both bond-stretching and bond-bending constraints are taken into account.

To go beyond the mean-field theory, corrections must be made. For example, adding a constraint to an already rigid region does not remove a floppy mode. Such type of constraint is called redundant. Redundant constraints introduce *stress* into the network. Such constraints do not change the number of floppy modes (and thus violate the assumption of the Maxwell counting); taking this into account, Eq. (1) becomes

$$F = dN - (N_c - N_r), \tag{2}$$

where N_r is the number of redundant constraints. Another type of correction is due to the fact that even above the rigidity transition there can still be some floppy inclusions and thus F > 0 at the transition.

To find N_r , Jacobs and co-workers have introduced a topological algorithm, the *pebble game* [27,28]. This algorithm is based on a theorem by Laman [29] which states that in two dimensions a generic network with N sites and B bonds does not have a redundant bond if and only if no subset of the network containing n sites and b bonds violates $b \le 2n-3$. A similar approach works in 3D, but in general only for bondbending networks like those used to model chalcogenide glasses.

The pebble game, described in more detail in Ref. [28], characterizes the global rigidity of a network, provides its complete decomposition into rigid clusters, and finds stressed regions. The approach uses only the topology of the network and not its exact geometry. Using the pebble game, it was possible to show that the rigidity transition occurs at $\langle r \rangle = 3.961 \pm 0.001$ [27] for the central-force triangular lattice and at $\langle r \rangle \approx 2.385$ [3] for an amorphous bond-bending 3D network.

While early measurements also identified a rigidity transition in these glasses near $\langle r \rangle = 2.4$, recent experiments have shown that there is not one but two transitions [4,7–19]. Starting at low mean coordination, the first transition seems to go from a floppy to a rigid but unstressed phase, and the second one on to a rigid and stressed phase. This rigid but unstressed phase is known as the *intermediate phase* and is believed to be related to the self-organization of the system in order to minimize the stress in the network.

This interpretation is supported by a number of theoretical works. In their pioneering work, Thorpe and collaborators demonstrated that a network constructed by adding bonds with no stress allowed until it becomes inevitable would go through three phases: a floppy, a rigid-unstressed, and a rigid-stressed [3]. The first transition, between the floppy and the rigid-unstressed phases, is the rigidity transition; the second one, between the rigid-unstressed and the rigid-stressed phases, is the stress transition and happens immediately when avoiding stress is no longer possible. The intermediate phase survives when the rigid but stress-free networks are fully equilibrated as was demonstrated by Barré and coworkers on Bethe lattices [24] and Chubynsky et al. [30] on two-dimensional triangular lattices. This is not the only approach to self-organization, however, and Micoulaut and collaborators [22,23] have shown that it is possible to recover an intermediate phase in a stressed network if this stress is localized.

III. METHODOLOGY

The model we study here is the same as the one used in our previous paper [30]. In our simulations, we use the pebble game algorithm described in the previous section; our computer code is based on the original program by Jacobs and Thorpe.

We consider two-dimensional (2D) triangular bonddiluted central-force networks with equal numbers of sites along the two lattice vector directions and folded into a rectangular box with periodic boundary conditions. While we cannot make a direct comparison with experiment, previous work has shown that the triangular lattice presents an intermediate phase similar to that of covalent glasses with angular constraints; our results should therefore be applicable, at least qualitatively, to experiments.

In the original model of Thorpe and collaborators [3], bonds were added one by one and each checked for redundancy; redundant (stress-causing) bonds were rejected. Each new added bond was frozen in the network and was never moved nor removed. This procedure does not guarantee that stress-free networks are equiprobable, making some networks more likely than others. To eliminate this bias, we introduced a bond-equilibration scheme, allowing the system to rearrange itself by moving bonds around. Each time a new bond is added, bonds are reshuffled throughout the lattice: a bond is picked at random, removed, and then a new bond is inserted in a random place choosing among those where it would not create stress. This bond-shuffling procedure is repeated until the system is equilibrated. We find that an equilibration of ten iterations per added bond below $\langle r \rangle = 3.5$ and

100 iterations above $\langle r \rangle$ =3.5 is enough for convergence (for more details, see Chubynsky *et al.* [30]) and it is the equilibration scheme we use throughout this paper, unless stated otherwise. In equilibrium, all stress-free networks with a given number of bonds (or mean coordination) are equiprobable. This corresponds to the thermodynamic equilibrium at $T \rightarrow 0$ for any model in which all stress-free networks have equal energy, but the energies of stressed networks are higher.

As discussed in our previous paper [30], the intermediate phase in the model described above is associated with a non-trivial probability of finding a percolating network in this equilibrated model: this probability rises linearly from zero at the rigidity transition ($\langle r \rangle \approx 3.945$) to one at the stress transition ($\langle r \rangle = 4.0$), a result similar to that obtained by Barré *et al.* on the Bethe lattice [24]. Thus there are both percolating and nonpercolating networks and these two classes need to be studied separately.

All results presented in this paper (with the exception of the cluster size distributions given in Fig. 3) are obtained by running 200 independent simulations and obtaining the quantities of interest at different $\langle r \rangle$. When overall averages are presented, they are obtained by averaging over all these simulations. When, e.g., an average for percolating networks is presented, then the averaging is done over only those networks that are percolating at the given $\langle r \rangle$. Obviously, close to the rigidity transition, very few of the 200 networks are percolating, and so the corresponding quantity will be an average over a very small number of realizations and may contain a bigger error. When we report the results of insertion or removal of a single bond, only one attempt of insertion or removal per network is made, unless stated otherwise.

IV. PROPERTIES OF RIGID CLUSTERS

In this section, we look at properties of rigid clusters (both percolating and nonpercolating) that exist in selforganized networks in the intermediate phase. Some properties of rigid clusters that distinguish them from, e.g., clusters in usual connectivity percolation, need to be kept in mind. First, unlike in the connectivity case, a site can belong to more than one cluster (it then serves as a pivot joint between the clusters sharing this site). On the other hand, a bond in 2D always belongs to just one cluster (not so in 3D, where it can serve as a hinge around which several clusters can rotate). For this reason, cluster decomposition in 2D is best expressed in terms of bonds and not sites. While this is not generally so, in the case of self-organized networks in the floppy or intermediate phase, the conversion between cluster sizes expressed in terms of bonds or sites is easy: since there is no stress, there is also no redundancy, and every rigid cluster of n sites contains exactly 2n-3 bonds.

We start with properties of the percolating cluster.

A. Definition of a percolating cluster

Normally, when the probability of percolation is either zero or one in the thermodynamic limit, the exact definition of percolation does not matter. In our case, since the probability of finding a percolating cluster increases linearly with average coordination in the intermediate phase, it is not clear that we can be so cavalier.

For example, the probability that a network percolates in only one direction (no matter which one), seems to be essentially size-independent in the intermediate phase and thus should remain nonzero in the thermodynamic limit. (We define percolation in a particular direction as the presence of a rigid cluster that spans the network in that direction and connects upon itself across the boundary.) The fraction of networks percolating in one direction (1D-percolating networks for brevity) is always low, never above 10% for all $\langle r \rangle$. Close to the lower boundary of the intermediate phase, the fraction of networks percolating in two directions (2D-percolating networks) is also low and so, in fact, the fractions of networks percolating in one and two directions are comparable. Yet the fraction of 1D-percolating networks reaches a maximum at $\langle r \rangle \approx 3.95$, immediately above the lower boundary, and then decreases, approaching zero at the upper boundary of the intermediate phase at $\langle r \rangle = 4$, whereas the fraction of 2D-percolating networks increases and reaches one at the upper boundary. For this reason, the overall fraction (averaged over all $\langle r \rangle$) within the intermediate phase is only about 5% for 1D-percolating networks but is around 50% for 2Dpercolating networks.

Since the fraction of 1D-percolating networks, even though it is low, is nonzero, it is important to specify when discussing percolation if it is defined using only one direction or both. In the rest of the paper, we choose to call percolating those networks in which percolation occurs in both directions (2D-percolating), and nonpercolating those networks in which there is no percolation in either direction. We ignore 1D-percolating networks whenever we show results separately for percolating networks and for nonpercolating ones (as 1D-percolating networks do not belong to either category, according to our definition), but when overall averages are presented, all networks are taken into account, including 1D-percolating ones.

B. Size of the percolating cluster

While we have already studied the probability that a percolating cluster occurs in the intermediate phase, its size had not been characterized. Figure 1 shows the average number of bonds in the percolating cluster as a fraction of bonds actually present in the network. The averaging is done over all networks in which percolation occurs. A remarkable feature is that even at the lowest $\langle r \rangle$ at which percolation is still (rarely) observed, the size of the percolating cluster is well above zero. The smallest cluster size observed at the onset of the intermediate phase is around 40%. This behavior is different from that for both connectivity and rigidity percolation on regular lattices in the random (non-self-organized) case, where the size of the percolating cluster, considered a good order parameter, grows from zero at the transition, as expected in a second-order transition. This result in the selforganized case is reminiscent of the first-order rigidity transition, such as that observed on Bethe lattices [5,31,32].

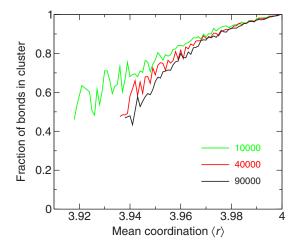


FIG. 1. (Color online) The fraction of bonds belonging to the percolating rigid cluster among all bonds in the network, averaged over all percolating self-organized networks, for different network sizes indicated in the figure. All network sizes here and in other figures in the paper are given in terms of sites.

However, our other results, as discussed below, do not support this analogy.

Figure 1 presents average sizes of percolating clusters. Given that the very existence of the percolating cluster is uncertain in the intermediate phase (since only some networks are percolating), it is reasonable to ask about the variation of the percolating cluster size. The quantity we look at is the standard deviation, or width, of the distribution of the fractions of bonds in the percolating cluster calculated as $\sqrt{(\langle F^2 \rangle - \langle F \rangle^2)} n/(n-1)$, where F is the fraction of bonds in the percolating cluster, $\langle \cdots \rangle$ denotes the average over percolating networks and n is the number of percolating networks. In ordinary percolation, this width tends to zero as the network size grows; the percolating cluster size is a selfaveraging quantity. Figure 2 shows that this is not so in our case. The width is above zero and is essentially sizeindependent. This is yet another difference from non-selforganized percolation (including that on Bethe lattices). Note

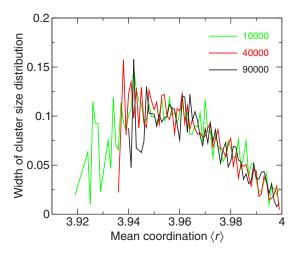


FIG. 2. (Color online) The standard deviation of the fraction of bonds in the percolating cluster for different network sizes, for selforganized networks.

at the same time that the width of the distribution of percolating cluster sizes is much smaller than the average size. In other words, an overwhelming majority of networks have either a big percolating cluster or no percolating cluster at all—there are few (if any) "intermediate cases" with small percolating clusters.

C. Sizes of nonpercolating rigid clusters

To further characterize the intermediate phase, it is useful to look at the distribution of rigid cluster sizes. In case of a second-order phase transition, the correlation radius is finite away from the transition. As a result, at a certain cluster size there is a crossover from a power-law behavior to an exponential behavior. The correlation radius diverges as the transition is approached from either side, and so the crossover moves towards bigger sizes as the transition is approached, and exactly at the transition, the power law persists indefinitely. Since the divergence of the correlation radius (or the crossover point) is governed by the same critical exponent on both sides of the transition, it is expected that for two values of $\langle r \rangle$ at the same distance from the transition but on opposite sides, the crossover points will be the same by order of magnitude, if the transition is second-order. In case of a firstorder transition, the correlation radius does not diverge at the transition and there is always a crossover to the exponential behavior.

Figure 3 shows the distribution of rigid cluster sizes at the coordinations of $\langle r \rangle = 3.92$ (below the transition) and $\langle r \rangle$ =3.97 (above the transition). The points are chosen at about the same distance from the rigidity transition, far enough from it to make sure that the correlation radius is not too big (if the transition is second-order) yet not too far to ensure that the points are still within the critical region. All results are obtained by running 200 independent simulations on networks of 40 000 sites, each starting from an already equilibrated network and continuing for 1000 additional equilibration steps. After each equilibration step, the distribution of nonpercolating cluster sizes is obtained. While networks one equilibration step apart cannot be considered truly independent, inserting or removing a single bond often changes the rigidity of the network very significantly, explaining inclusion of the data obtained at every step. For $\langle r \rangle = 3.97$, the distributions for percolating and nonpercolating networks are plotted separately (at $\langle r \rangle = 3.92$ there are very few percolating networks and we plot the result for nonpercolating networks only). To decrease the noise in the tail, all clusters in networks of a given class (i.e., percolating and nonpercolating) are binned using a logarithmic scale.

Below the rigidity transition, in the floppy phase $[\langle r \rangle = 3.92; \text{ Fig. 3(a)}]$, there is a clear crossover between the power-law and exponential behaviors. The data are fitted using the product of a power law and an exponential:

$$f_1(x) = C_1 x^{-\alpha_1} \exp(-x/x_0).$$
 (3)

We use the data between 10^2 and $\approx 10^{4.23}$, i.e., dropping just a few data points at the tail, where noise and finite-size effects are significant, and omitting a region at the low end, as there are big deviations from the behavior described by Eq.

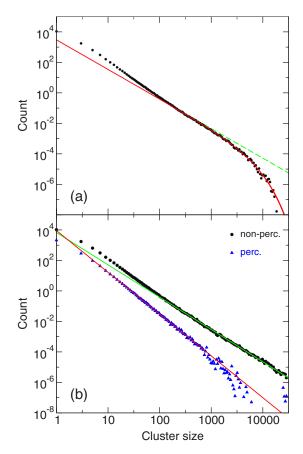


FIG. 3. (Color online) The distribution of sizes (given in terms of bonds) of nonpercolating rigid clusters in nonpercolating self-organized networks at $\langle r \rangle = 3.92$ [panel (a)] and in both percolating and nonpercolating self-organized networks at $\langle r \rangle = 3.97$ [panel (b)]. The details of the simulation and the fits (lines) are given in the text.

(3), probably due to the discreteness of the lattice. The best fit is obtained with C_1 =3000, α_1 =1.94, and α_0 =3900. The dashed line is a power law with the same C_1 and α_1 , but without the exponential factor.

Above the transition [$\langle r \rangle$ =3.97; Fig. 3(b)], however, the power-law behavior persists for nonpercolating networks, with no hint of the exponential tail, even for the largest sizes for which the data are available (around 30 000, well above the crossover observed for $\langle r \rangle$ =3.92 around x_0 =3900). For percolating nets, there is some deviation from the power law near the end, but it is likely due to finite-size effects (there is a percolating cluster taking up most of the network, so only relatively small nonpercolating clusters are possible). To fit the data, we use pure power-law functions:

$$f_{2\{n,p\}} = C_{2\{n,p\}} x^{-\alpha_{2\{n,p\}}}, \tag{4}$$

where subscripts $\{n,p\}$ refer to nonpercolating and percolating networks, respectively. We use data above 10^2 in the nonpercolating case and from $10^{1.8}$ to $10^{2.8}$ in the percolating case. The values of the parameters providing the best fits are C_{2n} =6800, α_{2n} =2.12, C_{2p} =9100, and α_{2p} =2.74. While the difference between α_1 and α_{2n} is probably due to finite size and sampling effects, the difference between α_{2n} and α_{2p} suggests that these two quantities are different.

Our results indicate that the power-law distribution of cluster sizes is observed in the whole intermediate phase, rather than at a single point (as would be the case without self-organization). In effect, the self-organization, which minimizes the stress in the network, *maintains the system in a critical state throughout the intermediate phase*.

Based on the results of this section, we see that the rigidity phase transition in our model of self-organized percolation is very different from both first- and second-order phase transitions.

V. RESPONSE TO A LOCAL PERTURBATION

Given that both percolating and nonpercolating networks coexist in the intermediate phase, it is interesting to investigate the relation between the two classes. As we show in this section, a local perturbation involving the addition or removal of even a single bond is enough to affect rigidity of huge regions of the network and often converts a nonpercolating network into a percolating one and vice versa.

This behavior is not observed in regular random networks: since the probability of percolation in these systems is always either zero or one in the thermodynamic limit, a single bond can only change the percolation property right at the transition. Since a percolating cluster at that point is fractal and involves only an infinitesimal fraction of bonds, if an infinite cluster is created or destroyed, this can only involve an infinitesimal fraction of bonds and sites; away from the transition, the size of the affected region is always finite and then the fraction is obviously infinitesimally small.

A few general comments about the consequences of addition or removal of a single bond are in order. First of all, the removal of a single bond can only break up the cluster to which this bond belongs; other clusters are not affected. This is because in 2D, all rigid clusters are always rigid by themselves, i.e., they remain rigid when taken in isolation from the rest of the network. Conversely, the addition of a bond can merge several rigid clusters into one, but will not affect the clusters outside the resulting cluster. Note that even though the self-organized networks are by definition stressfree, a newly inserted bond can be redundant and introduce stress (always confined to the cluster in which it is inserted); in this case the configuration of rigid clusters is not affected, but the created stressed region may still be macroscopic and percolate. While we will briefly consider this situation at the end of this section, we mostly concentrate on the case when the inserted bond is nonredundant and thus affects the rigidity of the network but does not create stress. For brevity, we will call the places where insertion of a bond does not create stress, as well as a bond inserted in such a place, allowed. Note that if we add an allowed bond creating a certain rigid cluster and then remove this bond and insert another allowed bond both ends of which are in the region coinciding with the cluster created by the first bond, exactly the same cluster will be created: indeed, the count of constraints within the region will be the same (2n-3) for a region of n sites), regardless of where in this region the bond is inserted.

A. Conversion between percolating and nonpercolating networks

We first look at the conversion between nonpercolating and percolating networks as a result of bond addition and

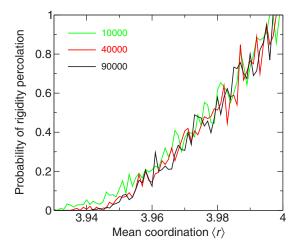


FIG. 4. (Color online) The probability that a bond added to a nonpercolating self-organized network at a random allowed place makes this network percolating.

removal. There are several related questions here.

1. Addition of a bond in an allowed position

In Fig. 4, we plot the probability that inserting a bond at a randomly chosen allowed place makes a nonpercolating network percolating. In the floppy phase (below $\langle r \rangle \approx 3.945$), this probability tends to zero in the thermodynamic limit; however, it is nonzero everywhere in the intermediate phase. This probability is close to zero just above the rigidity transition and approaches one close to the upper boundary of the intermediate phase at $\langle r \rangle = 4$.

We can also ask about the frequency of networks in which conversion from nonpercolating to percolating upon a single bond addition is possible at all. That is, for each nonpercolating network, rather than trying just one random bond and seeing if the percolating cluster emerges after its insertion (which is what we did to obtain Fig. 4), we ask if there exists any bond whose insertion would create such a cluster. Figure 5 plots the fraction of networks where such bonds exist. As is expected, this quantity is higher than that plotted in Fig. 4; but the difference is small: over most of the intermediate phase, whenever there are any allowed positions where bond insertion creates a percolating cluster, most allowed positions will do.

It is also interesting to note that the quantity in Fig. 5 is very close to linear in the intermediate phase, and is probably exactly linear, just like the probability of percolation without any bond insertions. In the Appendix, we explain why these two quantities are equal.

Finally, in Fig. 6, the average size of the percolating cluster arising after bond insertion is compared to the average size of the percolating cluster in those cases when it exists even without bond insertion (i.e., the quantity in Fig. 1). These values are identical: the size of the percolating cluster emerging after inserting a bond in a nonpercolating network is the same as in originally percolating networks. This suggests that a percolating network that arises after bond insertion is a typical percolating network, just like those networks that percolate without insertion. In the next subsection, we

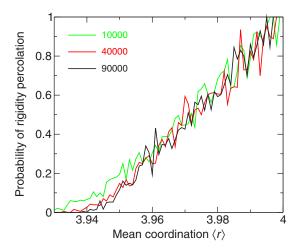


FIG. 5. (Color online) The fraction of nonpercolating selforganized networks such that there exists a bond whose insertion causes percolation.

present more evidence in favor of this. Also see a more detailed discussion of this in the Appendix.

Given the significant probability of conversion of nonpercolating networks to percolating ones (see Fig. 5), we can hypothesize that *all* nonpercolating networks can become percolating after a *finite* number of bond insertions; moreover, the average size of the percolating cluster after the minimal number of insertions needed to create it is again the same as the average size of the percolating cluster in networks that are percolating without bond insertions. This hypothesis needs to be tested in the future.

2. Removing a bond from the network

Likewise, we can consider the probability that removing a randomly chosen bond from a percolating network breaks

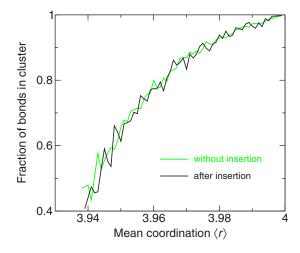


FIG. 6. (Color online) The comparison between the average size of the percolating cluster in originally percolating self-organized networks ("without insertion") and in originally nonpercolating networks that become percolating after bond insertion ("after insertion"). The latter quantity is calculated as the average over all networks that can become percolating, by using "judicious placement" of a bond, as described in the text. Networks of 90 000 sites are used.

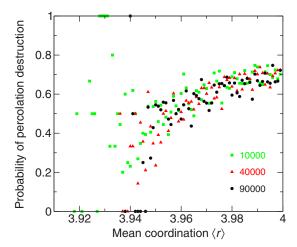


FIG. 7. (Color online) The probability that removal of a randomly chosen bond from a percolating self-organized network destroys percolation.

the percolating cluster. This quantity is shown in Fig. 7. Just like for bond addition, this probability is nonzero everywhere in the intermediate phase. Interestingly, even at the upper boundary $(\langle r \rangle \rightarrow 4)$, where almost all networks are percolating, it is still very easy to break percolation and thus create a nonpercolating network. In fact, the probability is the highest in this limit. Of course, this can be attributed to the fact that the percolating cluster is itself the biggest at this point (taking up the whole network), so there is a greater probability than elsewhere to select a bond that belongs to it (which is, of course, a necessary condition of its destruction). This effect can be factored out by dividing the quantity in Fig. 7 by the average fraction of bonds in the percolating cluster. This will then give the probability of the destruction of the percolating cluster, given that the removed bond belongs to this cluster. This is plotted in Fig. 8. It is seen that across the whole intermediate phase, this quantity is nearly constant at

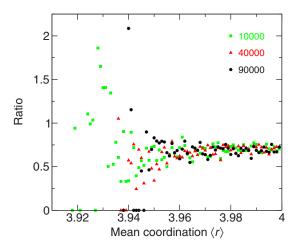


FIG. 8. (Color online) The probability that removal of a random bond destroys percolation (the quantity in Fig. 7) divided by the average fraction of bonds in the percolating cluster (the quantity in Fig. 1). This serves as an estimate of the probability that a bond chosen at random among those belonging to the percolating cluster destroys percolation.

around 70%. In other words, everywhere in the intermediate phase, the percolating cluster on average contains about 70% of bonds such that removal of any one of them will destroy percolation. This is true even close to $\langle r \rangle = 4$. This is, of course, just the average; one could ask if for some networks this quantity is zero (similar to how in the case of bond insertion, not all nonpercolating networks can be made percolating by a single bond addition, as shown in Fig. 5). To check if this is the case, we remove (and then reinsert) up to ten bonds one by one (all chosen within the percolating cluster) and see if the percolating cluster ever gets destroyed. This happened for 5398 out of 5429 90 000-site networks, or about 99.4%, including 198 out of 200 networks (99%) at $\langle r \rangle = 3.999$ (the highest mean coordination in our simulations). Since these fractions are so close to 100%, we can hypothesize that, unlike in the case of bond insertion, in the thermodynamic limit all percolating networks have some bonds whose removal destroys percolation.

To conclude this subsection, our results for the change in the percolation state of the network upon single bond addition or removal again confirm that the system remains critical in the whole intermediate phase.

B. Change of rigidity: "Rigid" and "floppy" bonds

In the previous subsection, we have found that in many cases insertion or removal of a single bond can change the percolation status of the network and affect the rigidity of its significant part. Since we were dealing with percolating clusters only, we could not study the effect of bond insertion or removal in those cases when the percolation status does not change. To do this, we introduce the concept of "rigid" and "floppy" bonds.

Below the rigidity transition, only small rigid clusters are present in the network. Interestingly, a significant fraction of them consist of just a single bond. The number of bonds belonging to these single-bond clusters decreases as the rigidity transition is approached and eventually crossed, although they are still encountered well above the transition in floppy pockets of the network. Since these bonds are associated with the floppy phase and floppy regions of the network in the rigid phase, we call such bonds *floppy*. All other bonds (i.e., those belonging to clusters consisting of more than one bond, or, in other words, rigid with respect to some other bonds) are called *rigid*. The fraction of rigid bonds as a function of $\langle r \rangle$ is plotted in Fig. 9 for both random and selforganized networks. As expected, it grows in both cases with increasing $\langle r \rangle$, but in the self-organized case it reaches 1 at $\langle r \rangle = 4$ (when the network becomes fully rigid), which in the random case does not happen until the full coordination at $\langle r \rangle$ =6. For the self-organized case, the averages over just percolating networks and over just nonpercolating networks are also shown in Fig. 9. Interestingly, the average for nonpercolating networks remains nearly constant over the whole intermediate phase at about 75%, even as $\langle r \rangle \rightarrow 4$. In this limit, the number of floppy modes per site tends to zero in all networks; yet, as these results show, in those few networks that still do not percolate, many bonds (about 25%) are still in single-bond clusters.

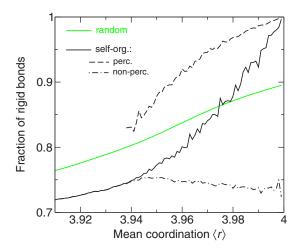


FIG. 9. (Color online) The average fraction of rigid bonds in the network in the random and self-organized cases (in the latter case, also separately for percolating and nonpercolating networks). All simulations are for networks of 90 000 sites, the overall averages are over 200 networks, the averages restricted to percolating and nonpercolating networks are over those of the 200 self-organized networks that are, respectively, percolating and nonpercolating.

Just as in the previous section we considered the width of the distribution of percolating cluster sizes, it is interesting to look at the width of the distribution of fractions of rigid bonds. Of course, since the averages are very different for percolating and nonpercolating networks, it makes sense to separate these two classes. The results are shown in Fig. 10. Note that in the intermediate phase, the width is sizeindependent and so likely remains finite in the thermodynamic limit, for both percolating and nonpercolating networks, just as we have seen for the percolating cluster size (see Fig. 2); but in the floppy phase (where, of course, only nonpercolating networks are present), the width clearly decreases fast with size (it is roughly inversely proportional to the square root of the network size). Thus the number of rigid bonds is a self-averaging quantity in the floppy phase but not in the intermediate phase, even when percolating and nonpercolating networks are considered separately. At the same time, the widths for both percolating and nonpercolating cases are much smaller than the difference between these two cases, so based on the count of rigid bonds, these two classes are clearly distinct.

We now look at the influence of insertion or removal of a bond on the number of rigid bonds and their spatial distribution.

In Fig. 11, we show the average change in the number of rigid bonds upon *insertion* of a single bond, in both the random and the self-organized cases. Note that in the random case, this change is very small on average, around ten bonds or less. On the other hand, in the self-organized case this quantity diverges very fast when the rigidity transition is approached. This is not surprising: we have seen that infinite percolating clusters can easily form in this case. For this reason, it makes sense to look at the *fraction* of bonds that undergo the change.

In Fig. 12, we show the fraction of bonds converted from floppy to rigid when a bond is added to the network at a

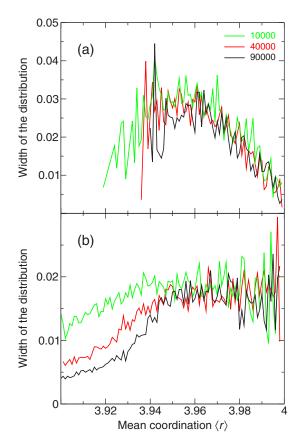


FIG. 10. (Color online) The standard deviation of the distribution of fractions of rigid bonds in percolating (a) and nonpercolating (b) self-organized networks for different network sizes.

randomly chosen allowed position, among all *floppy bonds* in the network. In panel (a), we plot the overall average, as well as partial averages restricted to those cases when the network goes from nonpercolating to percolating, remains percolating and remains nonpercolating (the curve for the latter case is barely above the baseline). In panels (b)–(d), we show these partial averages for several different sizes. Based on what we

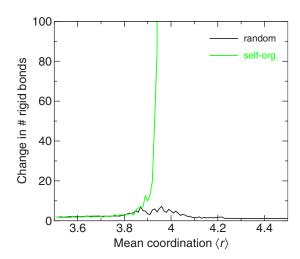


FIG. 11. (Color online) The average change in the number of rigid bonds upon addition of a single bond, for random and self-organized networks of 90 000 sites.

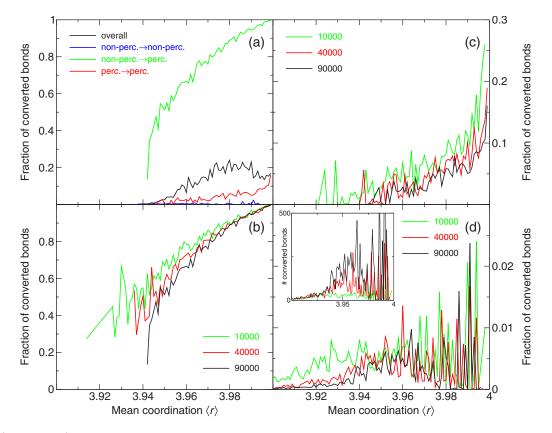
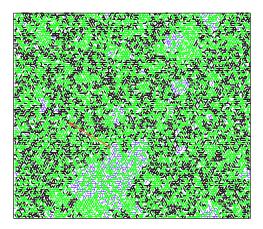


FIG. 12. (Color online) The average fraction of floppy bonds converting to rigid upon addition of a single bond to a self-organized network. (a) The overall average, as well as partial averages for the cases when the network converts from nonpercolating to percolating, remains nonpercolating, and remains percolating, for networks of 90 000 sites. (b)–(d) Partial averages in the nonpercolating \rightarrow percolating, percolating, and nonpercolating \rightarrow nonpercolating cases, respectively, for three different network sizes. The inset in (d) shows the average *number* of converted bonds in the nonpercolating \rightarrow nonpercolating case.

have already learned for networks switching from nonpercolating to percolating, it is quite natural that in this case a significant fraction of floppy bonds become rigid; this fraction approaches 1 when $\langle r \rangle \rightarrow 4$, which is again expected, since in this limit the percolating cluster takes up the whole network (see Fig. 6). The other two cases are more interesting. In the floppy phase, only the nonpercolating \rightarrow nonpercolating situation is possible, and as we see from Fig. 12(d), in this region of the phase diagram the average fraction of bonds converted from floppy to rigid decreases fast with size. In the inset, we plot the average *number* (rather than fraction) of converted bonds, and we see that this number is size-independent. So in the floppy phase, a finite number of bonds gets converted. In the intermediate phase, the situation is different. Looking first at the percolating → percolating situation [Fig. 12(c)], the fraction of converted bonds depends only weakly on the network size, and it is possible that this quantity goes to a constant in the thermodynamic limit. Even if, in fact, this fraction decreases to zero as the size N goes to infinity, it is clear that the decrease is much slower than $\propto 1/N$, and thus the mean *number* of converted bonds diverges when $N \rightarrow \infty$. In the nonpercolating \rightarrow nonpercolating case, the quality of the data is lower (there is very much noise due to a very large variation in the number of converted bonds), but still it is clear that the decrease with N (if present) is certainly much slower than $\propto 1/N$.

The fact that for the percolating \rightarrow percolating and non-percolating \rightarrow nonpercolating cases the average number of converted bonds diverges in the thermodynamic limit in the intermediate phase means that even when the percolation status does not change, the region of the network whose rigidity is affected is macroscopic *at least in some cases*. This is again consistent with the criticality of the intermediate phase. The distribution of the sizes of affected regions (or numbers of converted bonds) is likely power-law, which needs to be tested in the future.

As an illustration of effects of bond insertions we show two examples in Fig. 13. The upper panel shows an example for the case when the network switches from nonpercolating to percolating after a bond is added. The lower panel shows an event where the network remains nonpercolating, but large-scale rigidification still occurs without percolation. In both cases, the added bond is pointed with an arrow, thick green bonds (gray in print) are those that are originally rigid (and, of course, remain rigid after bond addition), thin bonds (blue online) are originally floppy and remain floppy, finally, thick black bonds are of most interest: these are the ones that switch from floppy to rigid. In the first (nonpercolating \rightarrow percolating) case, converted bonds are spread throughout the network. Many bigger rigid clusters separated by floppy "interfaces" merge into one percolating rigid cluster; in essence, the figure illustrates the rigidification of these interfaces. In the second (nonpercolating → nonpercolating) example, the



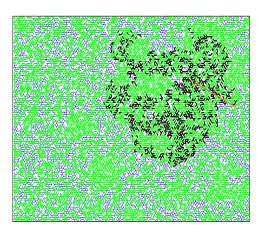


FIG. 13. (Color online) Two examples of rigidification of self-organized networks upon addition of a bond. In the top panel, the network switches from nonpercolating to percolating. In the bottom panel, it remains nonpercolating. In both cases, the added bond is pointed with an arrow, thick green bonds (gray in print) are those that are originally rigid, thin bonds (blue online) remain floppy, thick black bonds switch from floppy to rigid. Both networks contain 10 000 sites.

affected region is still large, but nonpercolating. This example is larger than average, as can be deduced from Fig. 12(d), but it is not a rare event.

We find overall similar behavior upon *removal* of a bond. The results for the fraction of rigid bonds converting to floppy are in Fig. 14. Similarly to Fig. 12, in Fig. 14(a) we have the overall average and partial averages for percolating → nonpercolating, percolating → percolating, and nonpercolating \rightarrow nonpercolating cases, for a single network size (90 000 sites). In Figs. 14(b)-14(d), we have the same partial averages, but for three different sizes. Again, the conclusions are similar to the case of bond insertion: in the percolating \rightarrow nonpercolating case, when the percolation status of the network changes [Fig. 14(b)], the fraction of bonds that switch from rigid to floppy is expectedly high. It is much lower in the other two cases, when the percolation status does not change, but still, just as for bond addition, while in the floppy phase the fraction of converting bonds falls rapidly with increasing size and, as the inset of Fig. 14(d) shows, the average number of converting bonds remains constant, in the intermediate phase the dependence of the fraction of converting bonds on the size is very slow and the number of converting bonds diverges in the thermodynamic limit—thus again, macroscopic regions of the network can be involved.

We finish this subsection with an observation: there exists a symmetry in bond conversions upon addition and upon removal. Namely, the average number of bonds converting upon bond addition in the case when the network transforms from nonpercolating to percolating is the same as the average number of bonds converting upon bond removal when the network transforms from percolating to nonpercolating. This is illustrated in Fig. 15. In this figure, we plot the ratios of the above-mentioned numbers and the total number of bonds in the network. We find that these two quantities coincide. Note that these quantities are different from those plotted in Figs. 12 and 14: in these figures, the numbers of converted bonds were divided by the number of floppy bonds and the number of rigid bonds, respectively, and not by the total number of bonds. The equality is easy to understand, if we recall that we have already seen some evidence (see Fig. 6) that networks that become percolating after bond insertion are, in fact, typical percolating networks, just like those that are originally percolating. By extension, we can assume that networks that become nonpercolating after bond removal are also typical nonpercolating networks. If so, then the average change in the number of rigid bonds should in both cases be the same as the difference in the average number of rigid bonds between percolating and nonpercolating networks. Indeed, in Fig. 15 we also plot the difference between the average fractions of rigid bonds in the nonpercolating and percolating cases (i.e., between the dashed and the dotdashed lines in Fig. 9); it is seen that this quantity coincides with the other two.

C. Stress propagation

In the previous subsections, we have looked at cases when an "allowed" bond insertion is done; in other words, the bond is inserted in one of those places where it does not create stress. We now look at the opposite situation, i.e., we analyze the results of inserting a bond in one of the "disallowed" places. In this case, the inserted bond is *redundant*, which means that its insertion does not change the configuration of rigid clusters, but some bonds (including the inserted one) become stressed. We are interested in the emerging stressed region and in particular, whether it percolates or not

Similarly to the case of allowed bond insertion, we first look at the probability that the stressed region percolates. Note that the stressed region emerging upon bond insertion is always restricted to the rigid cluster containing the new bond. For this reason, the stressed region can only percolate if the original network is percolating (i.e., contains a percolating rigid cluster), and we only need to look at percolating networks. In Fig. 16, we show the probability that in a network in which rigidity percolates, a percolating stressed region emerges after insertion of a bond at a randomly chosen disallowed place (not necessarily within the percolating cluster). We note that apparently, this probability remains finite (does not go to zero) in the thermodynamic limit.

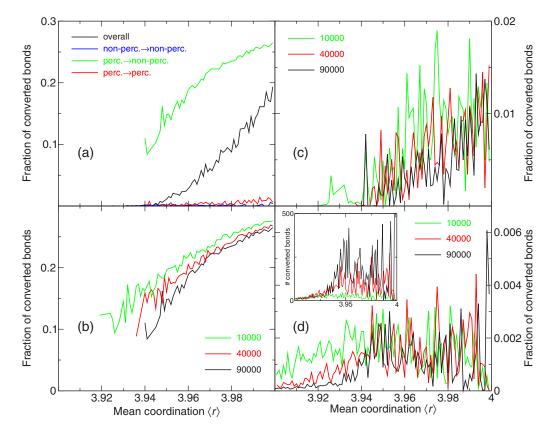


FIG. 14. (Color online) The average fraction of rigid bonds converting to floppy upon removal of a single bond from a self-organized network. (a) The overall average, as well as partial averages for the cases when the network converts from percolating to nonpercolating, remains nonpercolating and remains percolating, for networks of 90 000 sites. (b)–(d) The partial averages in the percolating \rightarrow nonpercolating, percolating \rightarrow percolating, and nonpercolating \rightarrow nonpercolating cases, respectively, for three different network sizes. The inset in (d) shows the average *number* of converted bonds in the nonpercolating \rightarrow nonpercolating case.

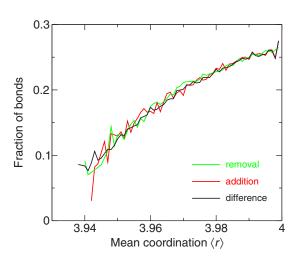


FIG. 15. (Color online) The average fractions of bonds: converting from rigid to floppy when a bond is removed and the network switches from percolating to nonpercolating (marked "removal"); converting from floppy to rigid when a bond is inserted and the network switches from nonpercolating to percolating ("addition"). These are compared to the difference between the average fractions of rigid bonds in the percolating and nonpercolating cases (marked "difference"). All quantities are for networks of 90 000 sites.

We also look at the sizes of stressed regions arising upon disallowed bond insertion. The results are in Fig. 17. Figure 17(a) shows the overall average fraction of stressed bonds and the partial averages for cases when stress does and does not percolate, for networks of 90 000 sites; Fig. 17(b) gives the partial average for the case when stress percolates; and

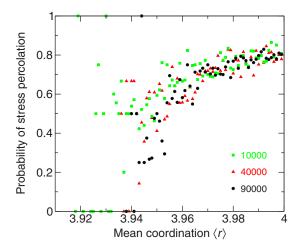


FIG. 16. (Color online) The probability that a percolating stressed region forms when a bond is inserted at a random "disallowed" place into a network with a percolating rigid cluster.

Fig. 17(c) gives the partial average for the case when stress does not percolate. We do not make distinctions based on the *rigidity* percolation status of the network. Again, we see that the size of the stressed region as a fraction of the total number of bonds is only weakly network size-dependent and thus the affected region can be macroscopic in the intermediate phase even when there is no percolation. In the floppy phase, on the other hand, the fraction of stressed bonds decays fast with increasing network size and as the inset (where the *number* of conversions, rather than the fraction, is plotted) shows, the average number of conversions is size-independent and the affected region remains finite.

VI. CONSISTENCY WITH EXPERIMENTAL EVIDENCE FOR SELF-ORGANIZATION

Many experiments underline the presence of the intermediate phase [4,7–20]. Most of these results can be understood in terms of a rigid yet unstrained network. However, in recent theoretical models of self-organization with equilibration [24,30], including the one used in this paper, the network in the intermediate phase only has a finite probability of possessing a percolating rigid cluster, raising questions as to the validity of the explanation or of these models. Here we argue based on the results of this paper that there are really no problems and our model of self-organization is consistent with experiments and their accepted interpretations.

Consider, for instance, a recent experiment by Wang *et al.* [18] on the atypical response of vibrational frequencies in glasses to applied hydrostatic pressure. In this Raman scattering experiment, the frequency of the corner-sharing (CS) tetrahedral units is followed as a function of applied pressure on the sample. Not surprisingly, the increase in external hydrostatic pressure densifies the material, eventually leading to a blueshifted CS frequency. These experimental results, published in Ref. [18], are reproduced in Fig. 18.

While in a crystal, blueshift occurs as soon as the pressure is applied, there is a pressure threshold, P_c , in glasses (see the curves labeled "Floppy" and "Stressed rigid" in Fig. 18). The explanation for this behavior proposed by Wang $et\ al.$ is that at low pressure the impact of the deformation is absorbed by inhomogeneities in the disordered network, locally under compressive or tensile stress, resulting at first in a broadening of the CS peak but without shift (see Ref. [18]); as the pressure increases, at some point the inhomogeneities can no longer shield the network, leading to a shift in the vibrational frequency.

Wang et al. observed, however, that while the pressure threshold P_c is finite in the floppy and the stressed-rigid phases, it vanishes in the intermediate phase. The original explanation for this unexpected result was that since in the intermediate phase the network has percolating rigidity without stress, it is uniformly rigid, with no floppy or overconstrained regions, so it has no inhomogeneities to absorb the deformation and should therefore behave like a crystal. However, in our model a finite fraction of the networks in the intermediate phase are floppy; these networks consist of many clusters of all sizes and thus seem very inhomogeneous. But as we have shown in this paper, even when there

is no percolating cluster, such a cluster has a good chance of appearing after a single bond insertion; we have also argued that a finite number of bond insertions is always enough, even in an infinite network, for a percolating cluster to appear. Such a percolating cluster has the same properties as in originally percolating networks, in particular, it always spans a *finite fraction* of the network. Of course, insertion of just a few bonds changes nothing physically, so percolating and nonpercolating networks, despite appearing very different at first glance in reality have the same physical properties.

One can then repeat the same arguments that led to the original conclusion of homogeneity. In the intermediate phase, there is a percolating region that is either rigid or becomes rigid after a small number of bond additions. This percolating region is also stress-free. These properties can only be achieved by a precise balancing between constraints and degrees of freedom not just on average, but on all length scales, in other words, by the network being homogeneous because of self-organized criticality. We note also that the way rigidity and stress propagate through a macroscopic fraction of the network in our simulations upon a perturbation is analogous to how stress propagates uniformly when external pressure is applied in experiments.

Thus the conclusion is that while the relation between P_c and inhomogeneity still needs to be fully established, assuming that relation, self-organized criticality provides a solid explanation for the absence of a pressure threshold in the vibrational frequency of the CS tetrahedral units in the intermediate phase.

Likewise, the self-organized criticality of the network is also in agreement with other experimental observations in the intermediate phase. One might ask, for instance, why the absence of aging of glasses in the intermediate phase [15] is consistent with our results, given that the configuration of rigid clusters can change so easily with a single bond addition or removal so the network appears very "unstable." Again, the answer is that changes caused by moving one bond in or out of the network cannot be significant physically, no matter what formal rigid cluster decomposition might suggest.

VII. CONCLUSION

We have studied the structural and response properties of the intermediate phase in the phase diagram of rigidity percolation using a model of self-organization on a 2D triangular network.

We had shown previously that the probability of rigidity percolation in the intermediate phase increases linearly from zero to one as a function of the mean coordination $\langle r \rangle$. At any $\langle r \rangle$, there are both percolating and nonpercolating networks in the ensemble. In this paper, we have looked at the properties of both percolating and nonpercolating clusters, the latter separately for percolating and nonpercolating networks. It turns out that at the point at which the percolating networks first emerge (the rigidity transition), the percolating cluster takes up about 40% of the network, unlike the case of the usual second-order phase transition, where the emerging cluster is fractal and thus the fraction of bonds belonging to

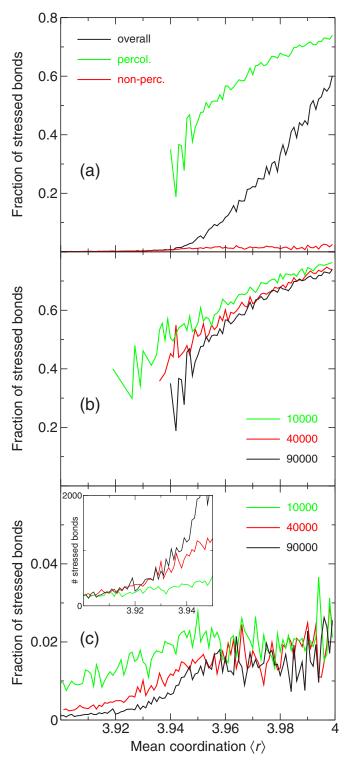


FIG. 17. (Color online) The average fraction of stressed bonds in the self-organized network after a single "disallowed" bond is inserted. (a) The overall average, as well as the partial averages for cases when stress does and does not percolate, for networks of 90 000 sites. (b) The partial average for the case when stress percolates, for three different network sizes. (c) The partial average when stress does not percolate, again, for three different network sizes; in the inset the average *number* of stressed bonds is shown for the same case.

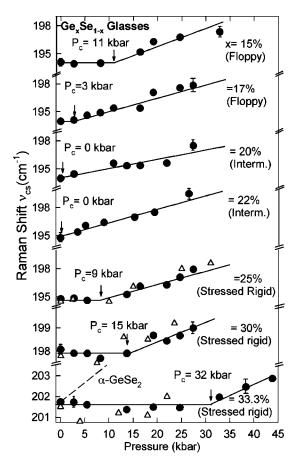


FIG. 18. Variations in the vibrational frequency of CS tetrahedral units as a function of pressure for different Ge_xSe_{1-x} glasses. Triangles are results taken from the work of Murase and Fukunaga [33] and filled circles are results of the work of Wang *et al.* [18]. The figure is taken from Ref. [18].

it is zero in the thermodynamic limit. The size of the percolating cluster and some other characteristics are not selfaveraging, but the distributions are rather narrow, if percolating and nonpercolating networks are considered separately. The distribution of sizes of nonpercolating clusters is exponential at large sizes in the floppy phase, but power law for arbitrarily big cluster sizes in the whole intermediate phase and not just at the transition. The power-law exponents are different for nonpercolating and percolating networks.

We have also looked at the changes in the rigidity of the network due to a microscopic perturbation in the form of insertion or removal of a single bond. It turns out that one bond is often enough to convert a nonpercolating network into a percolating one and vice versa; moreover, when a nonpercolating network is turned into a percolating one, the average size of the resulting percolating cluster is the same as that for the initially percolating networks. In a sense, the percolating cluster is "hidden" in the nonpercolating network and is revealed upon addition of a single bond. It appears, further, that *all* nonpercolating networks can be converted into percolating ones with a finite number of bond additions, with the same size of the resulting cluster as in networks that percolate without bond addition. This implies that in the thermodynamic limit, there is no difference between percolating

and nonpercolating networks. The intermediate phase can then be thought of as being the region of the phase diagram where all networks possess a percolating region that is *nearly isostatic*. That is, this region can technically be floppy or stressed, but the number of floppy modes or redundant constraints causing stress is negligible in the thermodynamic limit. The lower boundary of the intermediate phase is then, strictly speaking, not the rigidity percolation transition, but rather the "near-isostaticity" percolation transition.

These unusual properties indicate that the intermediate phase is a self-organized critical phase, with the system staying at the rigidity percolation threshold for the whole range of mean coordinations. Our results are consistent with the interpretation of recent pressure experiments [18] in terms of network homogeneity. Since rigidity percolation occurs on a finite fraction of the network under any microscropic perturbation, the strain associated with the external pressure is immediately transferred to a macroscopic fraction of the sample, leading to a shift in the Raman spectrum.

We can certainly expect other surprises associated with this intermediate phase in the rigidity phase diagram, but for that, we will likely need to create more realistic models of the experimental systems.

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APPENDIX: RELATION BETWEEN THE PROBABILITY OF PERCOLATION IN THE INTERMEDIATE PHASE AND THE PROBABILITY OF INDUCING PERCOLATION BY SINGLE BOND INSERTION

Numerically, we observe that the probability that there exists a place in a nonpercolating network such that insertion of a bond at that place causes percolation is the same linear function in the intermediate phase as the probability of rigidity percolation (see Fig. 5). Here we show that this equality is a necessary consequence of the latter quantity being finite (between zero and one) and continuous throughout the intermediate phase. In the proof, we also rely on our observation that percolating networks obtained from nonpercolating ones by bond insertion are typical percolating networks (see the discussion of Figs. 6 and 15). In particular, we assume that averages of various quantities for such networks are the same as these averages over the ensemble of stress-free percolating networks where all such networks are equiprobable (which is the ensemble generated by our equilibration procedure). Since this assumption is based on numerical evidence, it is, strictly speaking, only a conjecture, but even if it is only approximate, the accuracy is very good. More on this assumption below.

Given the probability of percolation among networks with B bonds, we can calculate this probability for networks with B+1 bonds. The method is similar to that used to calculate the bond-configurational entropy of self-organized networks in our previous paper [30]. Suppose there are $N_{\rm bc}(B)$ networks (or bond configurations) with B bonds that are stressfree. If the probability of percolation is s(B), then $N_p(B)$ $=s(B)N_{bc}(B)$ of these are percolating and the rest, $N_n(B)$ = $[1-s(B)]N_{bc}(B)$, nonpercolating. Also suppose there are on average $N_{ap}(B)$ allowed places to insert a bond in percolating networks and $N_{an}(B)$ in nonpercolating networks and the probability that a nonpercolating network becomes percolating upon a random bond insertion is $\pi(B)$. Since all percolating networks will remain percolating upon bond insertion, then an average percolating network with B bonds will produce $N_{\rm ap}(B)$ different percolating networks. On the other hand, only a fraction $\pi(B)$ of nonpercolating networks will become percolating, so an average nonpercolating network will produce $\pi(B)N_{an}(B)$ percolating networks and [1 $-\pi(B)]N_{\rm an}(B)$ nonpercolating networks. If we simply multiply these numbers by the number of networks with B bonds of each kind, this will not produce the correct count of percolating and nonpercolating networks with B+1 bonds because each such network can be obtained in many different ways. Specifically, each network with B+1 bonds can be produced by bond insertion from as many different networks with B bonds as there are the latter that can be obtained by bond *removal* from the former. This number is always B+1, since removal of every bond will produce a distinct network and all of them are stress-free. So the count has to be divided by B+1, and we get for the number of percolating networks with B+1 bonds,

$$N_p(B+1) = \frac{N_p(B)N_{\rm ap}(B) + N_n(B)\pi(B)N_{\rm an}(B)}{B+1}, \quad (A1)$$

and for the number of nonpercolating networks with B+1 bonds,

$$N_n(B+1) = \frac{N_n(B)[1 - \pi(B)]N_{an}(B)}{B+1}.$$
 (A2)

Then the probability of percolation for a network with B+1 bonds is

$$s(B+1) = \frac{N_p(B+1)}{N_p(B+1) + N_n(B+1)}$$

$$= \frac{N_p(B)N_{\rm ap}(B) + N_n(B)\pi(B)N_{\rm an}(B)}{N_p(B)N_{\rm ap}(B) + N_n(B)N_{\rm an}(B)}.$$
 (A3)

Note that since the probability of percolation changes continuously, the difference between s(B+1) and s(B) is O(1/N), where N is the network size. Neglecting terms that are O(1/N), we should equate s(B+1)=s(B) and then, omitting the argument B for brevity and using $N_n/(N_n+N_n)=s$,

$$s = \frac{sN_{\rm ap} + \pi(1 - s)N_{\rm an}}{sN_{\rm ap} + (1 - s)N_{\rm an}},$$
 (A4)

or

$$s = \pi \frac{N_{\rm an}}{N_{\rm an} - N_{\rm ap}}.\tag{A5}$$

At this point, we must use our conjecture that averages for networks obtained by bond insertion are the same as for the ensemble of equiprobable networks. Note that if all quantities associated with self-organized networks of a particular class (percolating or nonpercolating) were self-averaging, this would be fully expected. Indeed, in this case, atypical networks with parameters different from the average are infinitely rare in the thermodynamic limit; for such networks to be produced with a nonzero probability they need to be infinitely more likely to be produced by bond insertion than typical networks, in other words, there needs to be an infinitely strong bias towards atypical networks. But this cannot be true: as shown above, each network can be produced in exactly B+1 ways, and networks with B bonds with probability 1 can produce either $N_{\rm ap}$ (if percolating) or $N_{\rm an}$ (if nonpercolating) different networks [in any case, O(N)], so infinite bias is impossible. However, note that in our case, at least some properties of self-organized networks are not selfaveraging (see Figs. 2 and 10). In this case, any finite bias can produce an ensemble of networks with average parameters slightly deviating from the averages over the ensemble where all networks are equiprobable. Note, however, that the widths we observe in Figs. 2 and 10 are very small, much smaller than the averages. This explains why the deviations should at least be very small. The fact that we see no deviations may mean that they are so small as to be undetectable; or there may be some subtler reasons for there to be no deviations at all. In what follows, we conjecture that there are no deviations.

Applying our conjecture means that on average, when a bond is inserted into a nonpercolating network and it becomes percolating, the change in the number of allowed bonds is $N_{\rm an}-N_{\rm ap}$. But bonds that become disallowed are exactly those that are within the percolating cluster. According to our argument at the end of Sec. V, insertion of any of these bonds will also create the same percolating cluster and thus make the network percolating. On the other hand, insertion of any of the other allowed bonds will not cause percolation. Indeed, since these bonds are not within the original percolating cluster, the cluster that insertion of such a bond would create cannot overlap with the original percolating cluster over any of the allowed bonds and it is extremely unlikely (probably impossible in the thermodynamic limit) that a region that does not overlap with a percolating region over any allowed bonds is also percolating. So, if for a given network making it percolating after one bond insertion is at all possible, then the probability that percolation will occur after random insertion is equal to the fraction of allowed bonds within the percolating cluster, or $(N_{\rm an}-N_{\rm ap})/N_{\rm an}$, and then the right-hand side of Eq. (A5) is exactly the probability that percolation after one bond insertion is possible, and the proof is complete.

- [1] M. F. Thorpe, J. Non-Cryst. Solids 57, 355 (1983).
- [2] J. C. Phillips, J. Non-Cryst. Solids 34, 153 (1979).
- [3] M. F. Thorpe, D. J. Jacobs, M. V. Chubynsky, and J. C. Phillips, J. Non-Cryst. Solids 266-269, 859 (2000).
- [4] P. Boolchand, X. Feng, D. Selvanathan, and W. J. Bresser, in *Rigidity Theory and Applications*, edited by M. F. Thorpe and P. M. Duxbury (Kluwer Academic/Plenum Publishers, New York, 1999), p. 279.
- [5] M. F. Thorpe, D. J. Jacobs, N. V. Chubynsky, and A. J. Rader, in *Rigidity Theory and Applications*, edited by M. F. Thorpe and P. M. Duxbury (Kluwer Academic/Plenum Publishers, New York, 1999), p. 239.
- [6] D. J. Jacobs, A. J. Rader, L. A. Kuhn, and M. F. Thorpe, Proteins 44, 150 (2001).
- [7] D. Selvanathan, W. J. Bresser, and P. Boolchand, Phys. Rev. B 61, 15061 (2000).
- [8] P. Boolchand, D. G. Georgiev, and B. Goodman, J. Optoelectron. Adv. Mater. 3, 703 (2001).
- [9] P. Boolchand, X. Feng, and W. J. Bresser, J. Non-Cryst. Solids 293, 348 (2001).
- [10] P. Boolchand, D. G. Georgiev, and M. Micoulaut, J. Optoelectron. Adv. Mater. 4, 823 (2002).
- [11] P. Boolchand, D. G. Georgiev, T. Qu, F. Wang, L. Cai, and S. Chakravarty, C. R. Chim. 5, 713 (2002).
- [12] D. G. Georgiev, P. Boolchand, H. Eckert, M. Micoulaut, and K. Jackson, Europhys. Lett. 62, 49 (2003).
- [13] T. Qu, D. G. Georgiev, P. Boolchand, and M. Micoulaut, in

- Materials Research Society Symposium Proceedings Vol. 754, edited by T. Egami, A. L. Greer, A. Inoue, and S. Ranganathan (Materials Research Society, Pittsburgh, 2003), p. CC8.1.
- [14] U. Vempati and P. Boolchand, J. Phys.: Condens. Matter 16, S5121 (2004).
- [15] S. Chakravarty, D. G. Georgiev, P. Boolchand, and M. Micoulaut, J. Phys.: Condens. Matter 17, L1 (2005).
- [16] E. Černošková, T. Qu, S. Mamedov, Z. Černošek, J. Holubová, and P. Boolchand, J. Phys. Chem. Solids 66, 185 (2005).
- [17] T. Qu and P. Boolchand, Philos. Mag. 66, 875 (2005).
- [18] F. Wang, S. Mamedov, P. Boolchand, B. Goodman, and M. Chandrasekhar, Phys. Rev. B 71, 174201 (2005).
- [19] Y. Vaills, T. Qu, M. Micoulaut, F. Chaimbault, and P. Boolchand, J. Phys.: Condens. Matter 17, 4889 (2005).
- [20] G. Lucovsky and J. C. Phillips, Appl. Phys. A: Mater. Sci. Process. 78, 453 (2004).
- [21] M. F. Thorpe and M. V. Chubynsky, in *Properties and Applications of Amorphous Materials*, edited by M. F. Thorpe and L. Tichý, NATO Science Series, II. Mathematics, Physics and Chemistry (Kluwer Academic, Dordrecht, 2001), Vol. 9, p. 61.
- [22] M. Micoulaut, Europhys. Lett. **58**, 830 (2002).
- [23] M. Micoulaut and J. C. Phillips, Phys. Rev. B 67, 104204 (2003).
- [24] J. Barré, A. R. Bishop, T. Lookman, and A. Saxena, Phys. Rev. Lett. 94, 208701 (2005).
- [25] P. Bak, C. Tang, and K. Wiesenfeld, Phys. Rev. A 38, 364 (1988).

- [26] J. C. Maxwell, Philos. Mag. 27, 294 (1864).
- [27] D. J. Jacobs and M. F. Thorpe, Phys. Rev. Lett. 75, 4051 (1995).
- [28] D. J. Jacobs and B. Hendrickson, J. Comput. Phys. 137, 346 (1997).
- [29] G. Laman, J. Eng. Math. 4, 331 (1970).
- [30] M. V. Chubynsky, M.-A. Brière, and N. Mousseau, Phys. Rev. E 74, 016116 (2006).
- [31] C. Moukarzel, P. M. Duxbury, and P. L. Leath, Phys. Rev. E **55**, 5800 (1997).
- [32] P. M. Duxbury, D. J. Jacobs, M. F. Thorpe, and C. Moukarzel, Phys. Rev. E **59**, 2084 (1999).
- [33] K. Murase and T. Fukunaga, in *Optical Effects in Amorphous Semiconductors*, edited by P. C. Taylor and S. G. Bishop (American Institute of Physics, New York, 1984), p. 449.