

## First Order Rigidity Transition in Random Rod Networks

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We study the transition from the flexible to the rigid state in a system of randomly placed rigid rods connected by free joints. At the percolation transition point the system is still flexible and the transition to the macroscopically rigid state occurs at a higher concentration of bonds. It is shown that, in contrast to the percolation transition, the rigidity transition is analogous to the first order transition in thermodynamical systems: the rigidity correlation length is finite at the transition point, there is a finite jump in the magnitude of the order parameter, there are similarities with the new phase nucleation processes, etc.

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The problem of the elastic properties of randomly diluted networks with central force interactions attracts considerable attention not only due to its wide range of applicability, but also because of remarkable inconsistencies in the results obtained by different numerical methods. The most extensively studied model is the two-dimensional randomly diluted hexagonal lattice model. For this model even the value of the critical threshold concentration of rigid bonds obtained using series expansions [1,2], transfer matrices [3,4], and finite-size scaling [5–10] is under question. The disagreement between various estimates [1–10] exceeds the accuracy of the methods used. There are also substantial disagreements about the values of critical exponents.

The numerical data in these studies were analyzed using a scaling approach, under the assumption that the rigidity transition is a second order type transition. Indeed, there is a similarity between the percolation and rigidity transitions. Randomly connected bonds can form elementary rigid clusters: in 2D it will be triangles made from three bonds, in 3D it will be tetrahedra made from four bonds. If two such clusters have a common bond, they also form a rigid construction. The concentration of elementary rigid clusters will monotonically increase with the concentration of bonds. So the size of rigid regions  $r_c$  will also monotonically increase.

If we assume for a moment that the analogy between percolation and the rigidity transition is valid, then the size of rigid regions  $r_c$  should diverge while approaching some critical concentration  $p_{\text{rig}}$ . Above  $p_{\text{rig}}$  there will be a connected rigid structure which interpenetrates the whole system—the infinite rigid cluster. The order parameter, i.e., the density of bonds contributing to the infinite cluster, is zero at  $p_{\text{rig}}$  and should grow continuously with increasing bond concentration.

We shall show below that the transition to a macroscopically rigid state looks absolutely different and is similar to a first order transition in thermodynamic systems. There is a finite jump in the magnitude of the order parameter at the transition point. The number of bonds participating in the macroscopic rigidity just above the transition point

$p \rightarrow p_{\text{rig}} + 0$  is of the order of the total number of bonds in the system. The characteristic length of the isolated rigid clusters remains finite near the transition point. There are phenomena analogous to supercooling and superheating, formation of a critical droplet, and so on. It is remarkable that the massive infinite rigid cluster is mechanically a rather fragile object because its rigidity is controlled by the few bonds which caused the transition. We shall argue that its macroscopic rigidity is proportional to  $(p - p_{\text{rig}})^{3/2}$  in the vicinity of the transition.

The main reason the analogy with percolation theory cannot be applied directly for the propagation of the rigidity is the following. Percolation describes the propagation of a “scalar” quantity; each site of the lattice either belongs to the infinite cluster or not. The rigidity is a more complex notion: In  $d$ -dimensional space each element of the rigid structure cannot be moved in any  $d$  directions with respect to the remaining structure. In addition, it cannot be rotated along  $d(d - 1)/2$  independent axes. This means that there are  $d(d + 1)/2$  independent restrictions on the possible motion of each element of the rigid structure. Each of these restrictions can be applied in its own geometric way. We can think about the propagation of rigidity as a “vector percolation” problem. This nontrivial character of propagation of rigidity can be illustrated by the simplest two-dimensional example shown in Fig. 1. It is a system of rigid clusters connected by free joints. Each cluster has three independent degrees of freedom: two translational and one rotational. Each connection with a free joint eliminates two degrees of freedom. The clusters  $A'$  and  $B'$  are part of the infinite rigid structure. Clusters which are connected to them only with a free joint can still have one degree of freedom. Clusters  $C$  and  $D$ , which are connected with clusters  $A'$  and  $B'$  and among themselves, are also part of the infinite rigid structure. The clusters next to them,  $A$  and  $B$ , which are connected with clusters  $C$  and  $D$  with joints and with each other, also become part of the infinite structure, and so on. The melting transition for this construction is very sharp: If we remove any rigid bond from the picture, all clusters to the left of this bond will fall apart from the infinite rigid structure. The

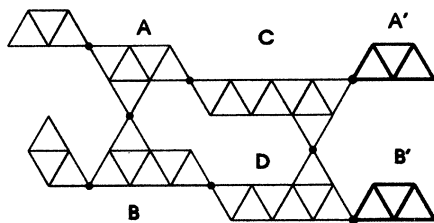


FIG. 1. If clusters  $A'$  and  $B'$  are part of the infinite rigid structure, clusters  $A$ ,  $B$ ,  $C$ , and  $D$  form a rigid construction too. The melting transition is very sharp: If we remove any rigid bond, the correlation length (to the left of this bond) will be reduced to the size of a single cluster.

correlation length of the rigidity will be reduced to the size of a single cluster. If we give up the initial assumption that clusters  $A'$  and  $B'$  are connected with the infinite rigid cluster, and assume that they are connected just with some other clusters, it is impossible to decide if the subsystem of clusters  $A', B', C, D, A, B$  is rigid or floppy. Clearly, both of these states are possible. In traditional percolation theory the situation is absolutely different: Looking at any particular realization of random clusters we can always decide if the whole system is percolating or not. The only requirement in this case is that the size of the observation region should be larger than the percolation correlation length.

For the description of the transition we shall first develop a mean field theory. For simplicity we shall discuss it in terms of a two-dimensional model: The elementary rigid object will be a triangle. The generalization to the three-dimensional case is straightforward, and the role of fluctuations will be discussed later.

Our method will be similar to the use of an external field in thermodynamic systems, or to the introduction of the "ghost" atom in percolation theory [11]. We assume that under our two-dimensional system of randomly placed bonds there is a rigid underlying surface, and that a small fraction of bonds  $h$  ( $h$  bonds) have a rigid connection with this surface. We can define the rigidity correlation function  $G(\mathbf{r}_1 - \mathbf{r}_2)$  or  $G(\mathbf{k}) = \int G(\mathbf{r})e^{i\mathbf{k}\cdot\mathbf{r}}d^2r$  in the momentum representation. This function will describe the connectivity of bonds in clusters of rigid triangles. In the initial (mean field) approximation this

$$P(\mathbf{r}) = \int_2 G(\mathbf{r} - \mathbf{r}')h(\mathbf{r}')d\mathbf{r}' - g_3 \int G(\mathbf{r} - \mathbf{r}')P(\mathbf{r}')P(\mathbf{r}'')d\mathbf{r}' + g_3 \left(\frac{t}{\tau}\right)^2 \times \int G(\mathbf{r} - \mathbf{r}')G(\mathbf{r}' - \mathbf{r}'')G(\mathbf{r}'' - \mathbf{r}''')P(\mathbf{r}''')P(\mathbf{r}''''')d\mathbf{r}'d\mathbf{r}''d\mathbf{r}''' + W(\{P(\mathbf{r}_i)\}), \quad (1)$$

where  $W(\{P(\mathbf{r}_i)\})$  describes the contribution of the higher order terms. Expanding  $P(\mathbf{r}') = P(\mathbf{r} + (\mathbf{r}' - \mathbf{r}))$  in powers of  $\mathbf{r}' - \mathbf{r}$ ,

$$P(\mathbf{r} + \mathbf{a}) = [1 + (\mathbf{a}\nabla) + \frac{1}{2}(\mathbf{a}\nabla)^2 + \dots]P(\mathbf{r}), \quad (2)$$

we obtain finally

$$(\tau + \nabla^2)P(\mathbf{r}) = h(\mathbf{r}) - g_3P^2(\mathbf{r}) + g_3\left(\frac{t}{\tau}\right)^2P^2(\mathbf{r}) + aP(\mathbf{r})\nabla^2P(\mathbf{r}) + W(P(\mathbf{r}), \nabla P(\mathbf{r})) + \dots \quad (3)$$

function can be written in the form  $G(\mathbf{k}) = 1/(\tau + k^2)$ , where  $\tau \sim \rho^* - \rho$  and  $\rho^*$  is the density of bonds at which there is a percolation transition over the connected triangles. Note that  $G(k=0) = \tau^{-1}$  is the average number of bonds in rigid clusters. If any bond of the rigid cluster is connected with the underlying substrate, so is the entire cluster. To the first approximation the density  $P$  of bonds which belong to the infinite rigid structure is  $P = G(k=0)h = h/\tau$ . This equation overestimates  $P$ , because if one rigid cluster contains two or three  $h$  bonds which are connected with the underlying substrate, then this cluster gives a double or a triple contribution to  $P$ . Proper corrections can be represented by the second and third terms in the right hand side (rhs) of Fig. 2(a). The next order corrections are also shown in Fig. 2(a) in the first row. These diagrams are the same as in percolation theory [11].

Besides these terms, there are contributions to  $P$  shown in the second row of Fig. 2(a) which are absent in percolation. They represent additional contributions to  $P$  from configurations in which the cluster is connected to two or more other clusters containing  $h$  bonds through free joints.

For example, the diagram in the second row of Fig. 2(a) describes the contribution from configurations where the given cluster is connected to two others through free joints. These two other clusters already belong to the infinite rigid structure, because each of them contains  $h$  bonds. The corresponding contribution to  $P$  from this diagram is  $(1/\tau)^3g_3(t/\tau)^2h^2$ , where  $g_3$  is the statistical weight of configurations containing a triple vertex. We should note that all these extra diagrams describe additional rigid configurations and contribute with a "plus" sign, in contrast to the oscillating sign behavior of regular percolation diagrams.

Resummation of the diagrams in the rhs of Fig. 2(a) gives us the graphic equation, Fig. 2(b). The higher order terms in  $P$  are not shown here. If the external field  $h$  is a function of space coordinates  $h = h(\mathbf{r})$ , then the order parameter also becomes coordinate dependent:  $P \rightarrow P(\mathbf{r})$ . The graphic equation of state, Fig. 2(b), can be written in a coordinate dependent form

In the limit of a large system and far from the boundaries,  $P(\mathbf{r}) = \text{const}$  and Eq. (3) can be simplified as follows:

$$\tau P = h - g_3P^2 + g_3\left(\frac{t}{\tau}\right)^2P^2 + W(P). \quad (4)$$

This equation is similar to the mean field equation of state for the order parameter in percolation theory. The only difference is that Eq. (4) has an additional term  $g_3(t/\tau)^2P^2$  which diverges at small  $\tau$ , and the sum of the two terms in Eq. (4) containing  $P^2$  can change its sign. In

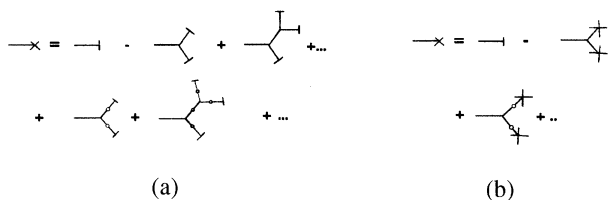


FIG. 2. (a) Diagrammatic expansion for the density of the infinite rigid structure  $P$ , denoted by symbol  $\times$ , each symbol  $|$  represents  $h$  bonds and contributes a factor  $h$ , each open circle gives an additional factor  $t$ , proportional to the density of free joints in the system, each line gives a factor  $\tau^{-1}$ , triple vertex factor  $g_3$ .

percolation we have a second order phase transition, but now, when the sign of the leading nonlinearity is changed, we can expect a first order transition.

The solution to this equation can be found graphically [see Figs. 3(a)–3(d)]. Here the straight line is the left hand side of Eq. (4), and the curve is given by the rhs of this equation. At small  $P$  the shape of the curve is determined by the first three terms in the rhs of Eq. (4); at large  $P$  the high order terms become important. At large  $P$  the curve representing the rhs of Eq. (4) is going down [12].

At small  $h$  and  $\tau > t$  the order parameter is proportional to the external field:  $P = h/\tau$  [see Fig. 3(a)]. As  $h \rightarrow 0$  the order parameter goes to zero. At  $\tau < t$  the sum of the two terms with  $P^2$  in Eq. (4) becomes negative and there appears a bump on the curved line [Fig. 3(b)]. This bump will grow when  $\tau \rightarrow 0$ , and at the same time the straight line  $\tau P$  will go down. Below some critical value  $\tau < \tau^*$  a straight line should cross this bump. This means that two additional solutions of Eq. (4) appear [see Fig. 3(c)]. One of them, shown by the heavy dot, is stable and the magnitude of the order parameter  $P$  given by this solution is finite even at zero field  $h = 0$ . At  $\tau < 0$  we have only one solution [Fig. 3(d)].

This situation is exactly the same as for the first order phase transition in thermodynamic systems. The only dif-

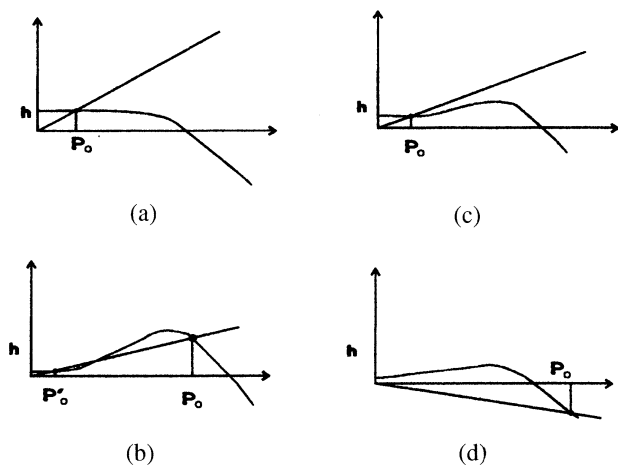


FIG. 3. Graphical solution of Eq. (4) for the density of the infinite rigid cluster  $P_0$ ; (a), (b) one solution; the density goes to zero if  $h \rightarrow 0$ ; (c) there are three solutions, two of them stable; (d) one solution.

ference is that we obtained Eqs. (3) and (4) directly by diagrammatic expansion, while for traditional thermodynamic systems a similar equation of state can be obtained through the minimization of a free energy functional. Unfortunately, reconstruction of the “free energy” for our problem is not possible because of the terms in Eq. (3) which cannot be directly integrated, i.e.,  $P \nabla^2 P$ .

Two different solutions in the region  $\tau^* > \tau > 0$  correspond to the completely flexible system ( $P = 0$  at  $h = 0$ ) and to a system in which a finite fraction  $P = P_0$  of bonds form a rigid skeleton. In traditional thermodynamics the selection of one of the solutions is straightforward: The phase with the lowest free energy should be dominant in large systems. In our case this is impossible because the free energy does not exist. The only regular way to select one of the two possible states of a large system is by finding the stable solution of the integral equation Eq. (1), or its simplified version Eq. (3). We can choose free boundary conditions on the right boundary (for example), rigid boundary conditions on the left boundary, and periodic boundary conditions in other directions. The solution should include a rigid region starting from the left boundary, a flexible region starting from the right, and the domain wall between these two regions. In a large system the domain wall will be close either to the right or to the left boundary. This will indicate which state, rigid or flexible, dominates in the thermodynamic limit. If all boundaries are of the same type, all rigid or all free, a phenomenon analogous to superheating (supercooling) can be obtained. The phase induced by the boundary can prevail in the bulk, if large enough droplets of another phase are not present. Such droplets can be formed due to fluctuation of local density of rigid bonds, and the probability of such fluctuation can be exponentially small.

Similar considerations can be carried out for the three-dimensional case. The total number of degrees of freedom of a rigid object in 3D is six and each junction eliminates three of them. We can write the same diagrams as for the 2D problem and equations similar to Eqs. (2)–(4) [13].

All of the above results were obtained in the mean field approximation. It is useful to check how these results will be affected by fluctuations. The simplest one-loop diagrams contributing to the correlation function  $G(\mathbf{r}_1 - \mathbf{r}_2)$  and to the triple interaction vertex  $g_3$  are shown in Fig. 4.

The diagrams (a) and (c) are the same as in percolation theory. Their signs are negative. Diagrams (b) and (d) are specific for the rigidity problem. These diagrams are more divergent than the percolation diagrams when  $\tau$  is small. They produce a positive contribution to the renormalized correlation function and to the triple vertex. The situation is the same with other leading higher order corrections. All this indicates a first order instability in a system with a finite correlation length when  $t \approx \tau$ . Qualitatively, they are the same criteria as for the mean field transition.

So far only the geometrical aspect of the rigidity transition was considered. The rigidity order parameter

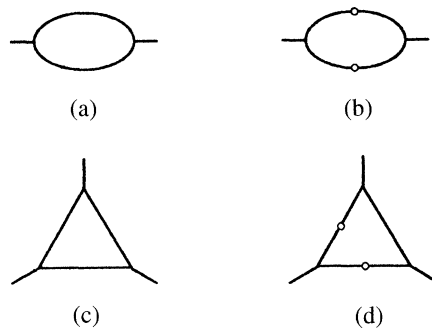


FIG. 4. One-loop corrections to the correlation function  $G(\mathbf{r}_1 - \mathbf{r}_2)$  and to the triple vertex  $g_3$ .

$P$  is the analog of the order parameter in percolation theory. It indicates the fraction of bonds participating in the infinite rigid cluster. But it does not provide insight into the stress distribution in the random rigid network, nor does it give the value of the macroscopic rigidity moduli. The rigidity transition is first order, so that the addition of a few bonds can "solidify" the whole system. This means that a small number of bonds carry all stress applied to the system. If each bond has a finite rigidity, the macroscopic rigidity moduli appear to be small (zero in the thermodynamic limit).

We know already that in the vicinity of the transition the size of rigid clusters is finite. If this transition is characterized by a single correlation length, the critical exponent characterizing the growth of the macroscopic elastic moduli can be trivial and should not depend on the details of the model on smaller scales. We calculated this exponent using a simplified model of an elastic network, similar to that introduced recently by Rubinstein, Leibler, and Bastide [14] to describe fluctuations in a gel. The 2D version of this model is just a tetrafunctional network of freely joined rigid rods [see Fig. 5(a)]. This network is flexible and can be folded along every row or along every column of plaquets [Fig. 5(b)], but the addition of a few more constraints makes it rigid. In Fig. 5(c) the same network is shown with a few new bonds added. Each new bond (diagonal in the elementary plaquet) eliminates the flexibility along the corresponding vertical and horizontal rows of plaquets. In the large  $L \times L$  system containing  $L^2$  bonds it is sufficient to add randomly of order  $L$  new bonds in order to make the system completely rigid. In the thermodynamic limit the fraction of these new bonds  $\Delta\rho \approx 1/L$  goes to zero. Thus a transition from the floppy to the rigid state in this system is first order.

A straightforward calculation shows that the macroscopic elastic modulus in a large system grows as a

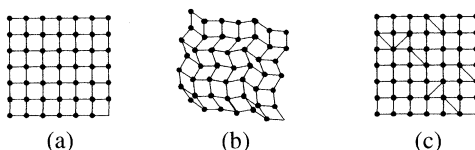


FIG. 5. The floppy two-dimensional structure becomes completely rigid after addition of a few bonds.

$\mu \approx (\rho - \rho_c)^{3/2}$  (in both 2D and 3D). Combining the above two dependences, we can get the finite size scaling formula for the elastic modulus:

$$\mu \approx (\rho - \rho_c)^f F((\rho - \rho_c)L^\nu), \quad (5)$$

with  $f = \frac{3}{2}$ ,  $\nu = 1$ , and  $F(x)$  some universal function. This is in remarkable agreement with experimental values  $f = 1.4$  and  $\nu = 1.0$  obtained by Lemieux, Breton, and Trembley [3] for the elasticity of random 2D networks.

In conclusion, we have shown that the rigidity transition in a rigid rod network is a first order transition. The correlation length is finite at the transition point, there are phenomena similar to nucleation, etc. The density of bonds which belong to the infinite rigid cluster has a finite jump at the transition point. Nevertheless, the infinite rigid cluster is very fragile mechanically near the transition point, and the elasticity modulus is a continuous function of bond concentration  $\mu \approx (\rho - \rho_c)^f$ ,  $f = 3/2$ .

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