

# Exact Relations Between Elastic and Electrical Response of $d$ -Dimensional Percolating Networks with Angle-Bending Forces

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Arguments are presented to demonstrate that exact equality relations exist between the critical exponents which characterize the macroscopic conductivity  $\sigma_e$  and the macroscopic elastic stiffness moduli  $C_e$  of percolating systems of any dimensionality. Using the notation  $\sigma_e \propto \Delta p^t$ ,  $C_e \propto \Delta p^T$  for the critical behavior of a randomly diluted system slightly above the percolation threshold  $p_c$ , ( $\Delta p \equiv p - p_c > 0$ ) and  $\sigma_e \propto |\Delta p|^{-s}$ ,  $C_e \propto |\Delta p|^{-S}$  for the critical behavior of a random mixture of normal and perfectly conducting or normal and perfectly rigid constituents slightly below that threshold, ( $\Delta p \equiv p - p_c < 0$ ) we show that  $T = t + 2\nu$  and  $S = s$ , where  $\nu$  is the percolation correlation length critical exponent  $\xi \propto |\Delta p|^{-\nu}$  ( $\Delta p \geq 0$ ).

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**KEY WORDS:** Elastic percolation; exact results; critical exponents; composite media.

## 1. INTRODUCTION

It was recently shown that, in two-dimensional (2D) percolating networks, there exist exact equality relations between the critical exponents which characterize the macroscopic elastic response and those which characterize the macroscopic electrical conductivity.<sup>(1)</sup> In a diluted network, where  $\Delta p \equiv p - p_c > 0$  measures how far the fraction  $p$  of occupied bonds or sites lies above the percolation threshold  $p_c$ , the macroscopic conductivity  $\sigma_e$  and any of the macroscopic elastic stiffnesses  $C_e$  tend to 0 when  $\Delta p \rightarrow 0^+$  as

$$\sigma_e \propto \Delta p^t, \quad C_e \propto \Delta p^T.$$

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In a network made of normal electric or elastic bonds mixed with perfectly conducting (superconducting for brevity) or totally rigid bonds, (we will call such a mixture a “superelastic” network) where  $\Delta p \equiv p - p_c < 0$  measures how far the fraction  $p$  of perfectly rigid or perfectly conducting bonds or sites lies below its percolation threshold  $p_c$ , those same macroscopic moduli tend to  $\infty$  when  $\Delta p \rightarrow 0^-$  as

$$\sigma_e \propto |\Delta p|^{-s}, \quad C_e \propto |\Delta p|^{-S}.$$

In the case of 2D networks,  $s = t$  exactly,<sup>(2,3)</sup> and the following exact equalities also hold<sup>(1)</sup>:

$$T = t + 2\nu, \quad S = s, \tag{1}$$

where  $\nu$  is the critical exponent which characterizes the divergence of the percolation correlation length  $\xi$  as  $\Delta p \rightarrow 0$  from either side

$$\xi \propto |\Delta p|^{-\nu}. \tag{2}$$

The precise definition I have in mind for  $\xi$  is explained in Section 5.1 later, where it is used in a derivation.

In this article I argue that the equalities of Eq. (1) hold also for three-dimensional (3D) percolating networks, and indicate the elements of similar arguments for percolating networks of any dimensionality  $d$ . The arguments, though not couched in rigorous mathematical language, actually constitute proofs, but they are conditioned upon various assumptions, many of which are unproven though widely believed. Some of the assumptions involve stability of the universality class of critical behavior of standard network models under quite drastic alterations. Those assumptions need to be verified before the proofs presented here can be considered conclusive. It is hoped that the informal description of these proofs will motivate other researchers to look carefully and critically at those assumptions in order to determine the validity and reliability of the results which are being claimed.

In order to show that  $S = s$  and  $T = t + 2\nu$ , we will demonstrate the following four inequalities:

$$S \leq s, \tag{3}$$

$$T \leq t + 2\nu, \tag{4}$$

$$S \geq s, \tag{5}$$

$$T \geq t + 2\nu. \tag{6}$$

These have been listed in order of increasing difficulty of proof. The first inequality follows from a discussion based on some classical variational principles of continuum elasticity and conductivity.<sup>(4)</sup> The second inequality was first announced in ref. 5, though the derivation presented there appears somewhat less than convincing. An actual proof of that inequality was first given in ref. 6 for 2D networks. The last two inequalities were demonstrated for 2D systems only very recently.<sup>(1)</sup> In this article we extend the proofs of Eqs. (4)–(6) to systems of higher dimensionality. This is done in full detail for the case  $d = 3$ , and only in outline for higher values of  $d$ . We also review the proof of the first inequality for general dimensionality. Because these proofs and the associated discussions are sometimes technical and intricate, I first give a brief overview of the essential ideas which underlie those discussions.

All the proofs are based on variational principles, which are introduced and exploited in the subsequent sections. Elastic behavior is described by mathematical quantities that differ in essential aspects from those used to describe electrical behavior, e.g., elastic vector displacement field vs. scalar electric potential field. In spite of this, the variational principles are sufficiently similar that it is sometimes possible to use an exact solution of one problem as trial values for the other problem. This is relatively easy to do in the case of the classical continuum variational principles, which are exploited in Section 2 to derive Eq. (3). In order to derive the other inequalities, Eqs. (4)–(6), it is necessary to base the discussion on discrete network models, where a different basic set of mathematical quantities are used to characterize the detailed state of the system—bond lengths and interbond angles for the elastic network vs. site potentials for the electrical network. Here, too, there exist variational principles for the two types of network problems. Again, a correspondence is found between the different descriptions of those two types of networks, which allows us to use a solution of one problem as a set of trial values for the other problem. However, in order to achieve this correspondence, we need to consider a conducting network that differs in detail from the elastic network, but is obtained from it by a simple transformation. Moreover, we often need to modify these networks, sometimes in a quite drastic fashion, in order to achieve the desired close correspondence. This is especially true when we tackle the proofs of Eqs. (4)–(6) in dimensionalities greater than 2, where we need to replace a finite fraction of the “normal bonds” by bonds of the “third type.” This means that, in a diluted network ( $p > p_c$ ), a finite fraction of the present bonds are reset to be perfectly rigid or perfectly conducting, while in a superelastic network ( $p < p_c$ ) a finite fraction of the normal bonds are deleted. The derivation presented here needs to assume that these drastic alterations do not change the critical behavior, i.e., that

the universality class remains unaffected. In addition to this, more drastic measures are needed in the derivation of Eq. (6), where we use an exact solution of the electrical network as trial values for the elastic network: The variational properties of the elastic network involve constraints which must be satisfied by all trial values of the bond variables, but these constraints are usually not satisfied by the exact solution of the electrical network. This difficulty appears already at  $d = 2$ , and only becomes more serious with increasing  $d$ .

The variational principles, with trial values chosen assiduously, lead to inequalities between the macroscopic responses of the two types of network. These inequalities must be applied to a macroscopic continuum description of the system, in order to obtain inequalities between the macroscopic or bulk effective elastic moduli and conductivity. That is achieved by considering sample networks whose size is of the order of the percolation correlation length  $\xi$ —that is the smallest size which can still justify invoking a continuum description of the system. The choice of these macroscopic samples is somewhat different for  $p > p_c$  and  $p < p_c$ , and takes into account some important exact properties of the percolating cluster or nearly percolating cluster when  $|p - p_c|$  is very small.

The rest of this article is organized as follows: In Section 2 we review the classical continuum variational principles and use them to prove Eq. (3) for systems of arbitrary dimensionality  $d$ . In Section 3 we discuss representations of 3D elastic and electrical networks and establish a correspondence between these two types of networks. In order to establish such a correspondence, it is necessary to consider the electrical problem on a network that differs from the elastic network, but is obtained from it in a simple, well defined manner, and is called the “covering network.” In Section 4 we apply some network variational principles in order to derive inequalities between the macroscopic responses of corresponding 3D elastic and electrical networks. This is achieved by using an exact solution of an elastic problem in order to provide trial values for the electrical problem on the covering network, which is a quite straightforward matter, and also by using an exact solution of an electrical problem on the covering network in order to provide trial values for the elastic problem on the original network. The latter application of a network variational principle is, however, far from straightforward, due to constraints which appear in the elastic problem but are absent from the electrical problem. These constraints are dealt with in various ways, including drastic alterations of the original network models. We present arguments, not proofs, to explain why we expect that the universality class of critical behavior remains unchanged despite those alterations. The resulting inequalities are applied, in Section 5, to macroscopic samples of networks, of size sufficiently large (i.e., at least

equal to  $\xi$ ) that they can be treated as homogeneous samples, in order to derive Eqs. (4)–(6) for 3D systems. In Section 6 indications are given on how to extend the derivation of these inequalities to systems of arbitrary  $d$ . Section 7 presents a summary and discussion of the results obtained, and indicates avenues for further research.

## 2. THE “CLASSICAL” VARIATIONAL PRINCIPLES

The classical variational principles of continuous media include the following two, for electrical conductivity and elastic behavior, respectively,

$$\frac{W_L}{V} = \min_{\phi} \left( \frac{1}{V} \int_V dV \nabla \phi \cdot \hat{\sigma} \cdot \nabla \phi \right) \equiv \langle \nabla \phi \rangle \cdot \hat{\sigma}_e \cdot \langle \nabla \phi \rangle, \quad (7)$$

$$\frac{2E_L}{V} = \min_{\mathbf{u}} \left( \frac{1}{V} \int_V dV \hat{\epsilon} \cdot \hat{C} \cdot \hat{\epsilon} \right) \equiv \langle \hat{\epsilon} \rangle \cdot \hat{C}_e \cdot \langle \hat{\epsilon} \rangle, \quad \epsilon_{\alpha\beta} \equiv \frac{1}{2} \left( \frac{\partial u_{\alpha}}{\partial r_{\beta}} + \frac{\partial u_{\beta}}{\partial r_{\alpha}} \right). \quad (8)$$

Here  $V$  is the total volume of the system,  $W_L$  is the total rate of production of Joule heat,  $E_L$  is the total elastic potential energy,  $\phi(\mathbf{r})$  is the local electrical potential,  $\mathbf{u}(\mathbf{r})$  is the local elastic displacement field,  $\hat{\sigma}(\mathbf{r})$  is the local electrical conductivity, which is usually a second rank tensor quantity, and  $\hat{C}(\mathbf{r})$  is the local elastic stiffness, which is always a fourth rank tensor quantity. In the minimization of the first quadratic functional,  $\phi(\mathbf{r})$  is allowed to range over all scalar functions of  $\mathbf{r}$  that have prescribed values  $\phi_0(\mathbf{r}) = -(\mathbf{E}_0 \cdot \mathbf{r})$  at the external boundary, as a consequence of which the volume averaged electric field is equal to  $\mathbf{E}_0$  for all of those potential fields  $\langle -\nabla \phi \rangle = \mathbf{E}_0$ . Similarly, in the minimization of the second quadratic functional,  $\mathbf{u}(\mathbf{r})$  is allowed to range over all vector functions of  $\mathbf{r}$  that have prescribed values  $\mathbf{u}_0(\mathbf{r}) = \hat{\epsilon}_0 \cdot \mathbf{r}$  at the external boundary, as a consequence of which the volume averaged strain field is equal to  $\hat{\epsilon}_0$  for all of those elastic displacement fields  $\langle \hat{\epsilon} \rangle = \hat{\epsilon}_0$ . The functions  $\phi(\mathbf{r})$ ,  $\mathbf{u}(\mathbf{r})$  which achieve those minima are unique, and they describe the physical state of the system subject to the specified boundary conditions. The minimum values of the two functionals are the correct physical values of  $W_L/V$  and  $2E_L/V$ , and they can also serve to define the macroscopic or bulk effective moduli  $\hat{\sigma}_e$ ,  $\hat{C}_e$  of the medium, as done in the above equations. These variational principles apply to systems of *arbitrary integer dimensionality*  $d$ .

We will assume that the local response is isotropic, therefore  $\hat{\sigma}(\mathbf{r}) = \sigma(\mathbf{r})$  is a position dependent scalar and  $\hat{C}(\mathbf{r})$  depends on just two position dependent Lamé coefficients  $\lambda$  and  $\mu$

$$C_{\alpha\beta\gamma\omega} = \lambda \delta_{\alpha\beta} \delta_{\gamma\omega} + \mu (\delta_{\alpha\gamma} \delta_{\beta\omega} + \delta_{\alpha\omega} \delta_{\beta\gamma}). \quad (9)$$

These coefficients satisfy the following relations

$$\begin{aligned} C_{\alpha\beta\alpha\beta} &\equiv \mu > 0, & \text{for } \alpha \neq \beta, \\ C_{\alpha\alpha\alpha\alpha} &\equiv C_{11} \equiv \lambda + 2\mu = \rho v_s^2 > 0, \\ \kappa &\equiv \lambda + \frac{2}{d}\mu = C_{11} - 2\frac{d-1}{d}\mu > 0, \end{aligned}$$

where  $\kappa$  is the bulk modulus,  $\mu$  is the shear modulus,  $C_{11}$  is another one of the elastic stiffness moduli, written in Voigt's notation,  $v_s$  is the longitudinal sound velocity, and  $\rho$  is the mass density of the solid.

Denoting by  $\phi_\alpha(\mathbf{r})$  the electrical potential which results when  $\mathbf{E}_0 = \mathbf{e}_\alpha$ , where  $\mathbf{e}_\alpha$  is the unit vector along  $r_\alpha$ , we use the trial function  $\mathbf{u}(\mathbf{r}) = \phi_\alpha(\mathbf{r}) \mathbf{e}_\beta$  to get

$$\hat{\epsilon} \cdot \hat{C} \cdot \hat{\epsilon} = (\lambda + 2\mu) \left( \frac{\partial \phi_\alpha}{\partial r_\beta} \right)^2 + \mu \sum_{\gamma \neq \beta} \left( \frac{\partial \phi_\alpha}{\partial r_\gamma} \right)^2 < (\lambda + 2\mu) (\nabla \phi_\alpha)^2 = C_{11} (\nabla \phi_\alpha)^2.$$

Applying this inequality to the integrand of Eq. (8), we get the following results for  $\hat{\epsilon}_0$  and  $\hat{C}_e$

$$\epsilon_{\beta\alpha}^{(0)} = \epsilon_{\alpha\beta}^{(0)} = -\frac{1}{2} (1 + \delta_{\alpha\beta}), \quad \text{all the other } \epsilon_{\gamma\omega}^{(0)} = 0,$$

$$C_{\alpha\beta\alpha\beta}^{(e)} \leq \frac{1}{V} \int_V dV C_{11} (\nabla \phi_\alpha)^2 = \sigma_{\alpha\alpha}^{(e)} [\sigma(\mathbf{r})],$$

$$\sigma(\mathbf{r}) \equiv C_{11}(\mathbf{r}) \equiv \lambda(\mathbf{r}) + 2\mu(\mathbf{r}) = \kappa(\mathbf{r}) + 2\frac{d-1}{d}\mu(\mathbf{r}).$$

This means that the macroscopic conductivities of a heterogeneous medium, where the local conductivity  $\sigma(\mathbf{r})$  is equal to the local stiffness coefficient  $C_{11}(\mathbf{r})$ , provide *upper bounds* for some of the macroscopic stiffness moduli. In particular, if we are dealing with a composite medium that has an isotropic microstructure, where  $\hat{\sigma}_e \equiv \sigma_e$  is a scalar and  $\hat{C}_e$  has the form of Eq. (9), then

$$\mu_e \leq \sigma_e [\sigma(\mathbf{r})], \quad C_{11}^{(e)} = \lambda_e + 2\mu_e \leq \sigma_e [\sigma(\mathbf{r})], \quad \kappa_e = \lambda_e + \frac{2}{d}\mu_e \leq \sigma_e [\sigma(\mathbf{r})]. \quad (10)$$

Applying the inequalities of Eq. (10) in the case of a rigid/normal mixture, we immediately obtain Eq. (3). In the case of a percolating

(diluted) network, these inequalities lead to  $T \geq t$ , which is a much weaker lower bound on  $T$  than the following inequality, obtained many years ago<sup>(7)</sup>:

$$T \geq 1 + dv. \quad (11)$$

### 3. ELASTIC AND ELECTRICAL NETWORKS

In order to try and demonstrate the other inequalities, we need to consider carefully the elastic potential energy of a network composed of discrete sites connected by elastic bonds, and compare it with the production rate of Joule heat in a corresponding network of electrically conducting bonds. Since we will seek a correspondence between elastic and electrical properties of percolating networks, we need to implement the elastic behavior in such a way that the rigidity threshold coincides with the conductivity threshold, which will obviously coincide with the usual geometrical percolation threshold  $p_c$ . In order to ensure the former coincidence, it is not enough to endow each bond  $i$  with a spring constant  $k_i$ , and the associated bond stretching force  $k_i \delta b_i$  and stretching energy  $k_i (\delta b_i)^2 / 2$ , where  $\delta b_i$  is the change in length of the bond  $i$ . In addition to that force, an elastic force, and a corresponding potential energy, must also be associated with changes of some angles between bonds.

For example, in the case of a 2D network, a change  $\delta \varphi_{ij}$  in the angle between the nearest neighbor ( $nn$ ) bonds  $i$  and  $j$ , which join together at a network site, should involve an elastic energy  $m_{ij} (\delta \varphi_{ij})^2 / 2 \equiv m_{ij} (\delta \varphi_i - \delta \varphi_j)^2 / 2$ . Here  $m_{ij}$  is the angle-bending force constant and  $\delta \varphi_i$ ,  $\delta \varphi_j$  are the changes in the *absolute orientations* or planar azimuth angles of the two bonds—see Fig. 1(a). The total elastic energy of such a 2D network is thus given by<sup>(1)</sup>

$$2E_L = \sum_i k_i (\delta b_i)^2 + \sum_{(ij)=nn} m_{ij} (\delta \varphi_i - \delta \varphi_j)^2. \quad (12)$$

In a diluted version of such a network, where the force constants are either finite with uniform positive values  $k_i = k > 0$ ,  $m_{ij} = m > 0$ , or else vanish, we also need to specify that  $m_{ij} = m > 0$  only if both of the bonds  $i$  and  $j$  are present with  $k_i = k_j = k > 0$ . Similarly, in a rigid/normal version of such a network, where the force constants are either finite with uniform positive values as above, or else they diverge  $k_i = m_{ij} = \infty$ , we also need to specify that  $m_{ij} = \infty$  only if both of the bonds  $i$  and  $j$  are perfectly rigid against stretching, i.e.,  $k_i = k_j = \infty$ . The first requirement ensures that a diluted elastic network will exhibit solid-state-like macroscopic elastic

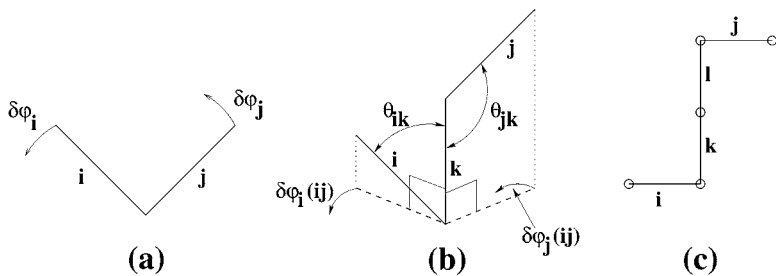


Fig. 1. (a) The angle between the  $nm$  planar bonds  $i$  and  $j$  can be changed by incrementing their absolute azimuth angles by  $\delta\varphi_i$ ,  $\delta\varphi_j$ . (b) The 3D orientations of the  $nmn$  bonds  $i$  and  $j$  are determined by the  $nm$  interbond angles  $\theta_{ik}$ ,  $\theta_{jk}$ , and by the absolute azimuth angles  $\varphi_i(ij)$ ,  $\varphi_j(ij)$ . The latter are defined by projecting the bonds  $i$  and  $j$  onto the plane perpendicular to the intervening bond  $k$ . (c) Quartet of bonds in a diluted simple-cubic network where the third-neighbor pair of bonds  $i$  and  $j$  are separated by the collinear pair of bonds  $k$  and  $l$  in series. Even if elastic energy is associated with torsional distortions of  $nmn$  bond pairs, the third-neighbor bonds  $i$  and  $j$  can be twisted around  $k$  and  $l$  without any cost in energy. (d) Cubic unit cell of a 3D diamond network. Sites are denoted by open circles. The cell contains four of the basic identical tetrads of bonds. All bonds have equal undistorted lengths; all  $nm$  bond pairs subtend the same undistorted angle; there are four basic bond orientations in the undistorted network. In the disordered versions of this network, bonds are either randomly deleted, or else randomly assigned to one of two types—normal or rigid. Additional restrictions, for the case of a *diluted network*, are: (i) There are in fact no bond tetrads—there are at most three bonds attached to any site. (ii) Two such triadic sites must have at least two diadic sites on any path between them. (iii) All the  $nmn$  bonds to a triadic site are of the “fourth type,” i.e., different from the three bonds directly attached to that site. These restrictions impose only short range correlations among the occupation probabilities of different network bonds. Therefore, even though they will alter  $p_c$ , they are not expected to affect the critical exponents. (e) Cluster of elastic bonds (thin solid segments) in a diluted diamond network, along with the corresponding cluster of the covering network (thin dashed line segments represent the bonds with infinite electrical conductance, like  $g_{jp}$ ; thick solid line represents the one bond with finite conductivity  $g_{ij}$ ). The special direction or  $z$  axis lies along the bonds  $k$  and  $s$ . Thus, these are the only bonds in this cluster around which the torsional force constants are finite. That is why  $m_{ij} = g_{ij}$  is finite, but  $m_{kl} = g_{kl} = m_{js} = g_{js} = m_{jp} = g_{jp} = \infty$ . If the  $nm$  angle-bending force constants are also set to  $\infty$ , then the azimuthal angle increments of the bonds  $l$  and  $p$  around the  $z$ -parallel bond  $s$  must be equal  $\delta\varphi_l(lp) = \delta\varphi_p(lp)$ , therefore we add another perfectly conducting bond to the covering network  $g_{lp} = \infty$ , in order to ensure that  $V_l = V_p$  too. Elastic bonds are denoted by thin solid lines, sites of the elastic network are denoted by open circles, while sites of the covering network are denoted by filled circles. Note that the bonds  $j$ ,  $l$ ,  $p$ ,  $s$  can only move by twisting around the bond  $k$  in *unison*, i.e., as a rigid cluster, thereby changing their orientation with respect to the bond  $i$ , as measured by the difference in azimuthal angle increments  $\delta\varphi_i(ij) - \delta\varphi_j(ij)$ . In the covering network cluster we will have  $V_j = V_s = V_p = V_l = V_k$ , thus only  $V_i$  can differ from  $V_j$ . (f) Two adjacent loops in a 3D diluted network. Assuming that loop  $A$  is closed, the bonds  $i$ ,  $j$ ,  $k$ , which are oriented along three independent directions in the undistorted network, can be exploited to ensure that loop  $B$  also closes by an appropriate choice of  $\delta b_i$ ,  $\delta b_j$ , and  $\delta b_k$ . The bonds  $l$ ,  $p$ ,  $s$  can be used, in similar fashion, to ensure closure of a third loop which includes them.



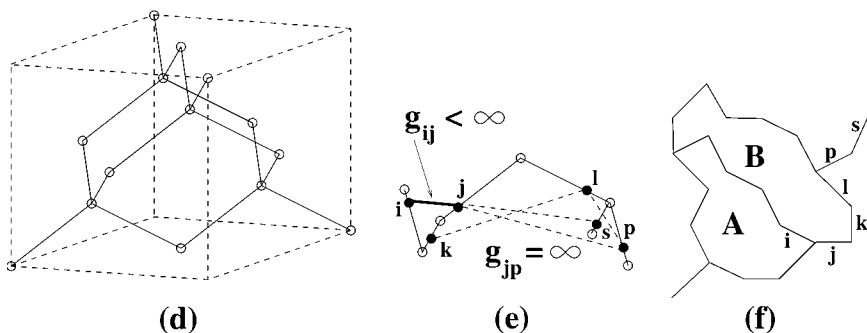


Fig. 1. (Continued).

response, with nonzero values for all the macroscopic elastic stiffness moduli, if and only if it percolates. Similarly, the second requirement ensures that a rigid/normal elastic network is totally rigid (i.e., all its macroscopic elastic stiffness moduli are infinite) if and only if the rigid bonds percolate.

The two types of requirements described above can be summarized concisely, and also generalized, by saying that, in a two-constituent mixture of large  $k_i$ ,  $m_{ij}$  and small  $k_i$ ,  $m_{ij}$ , the large  $m_{ij}$  value applies if and only if both  $k_i$  and  $k_j$  are large. In all the other cases,  $m_{ij}$  will take the small value.

Elastic network models that do not include any angle-bending forces, also known as “spring networks” or “central force models,” not only have a rigidity threshold that is higher than  $p_c$ , but they also apparently have different values for the critical exponents  $T$  and  $\nu$ .<sup>(8,9)</sup> Thus, they belong to a different universality class of percolation. Experiments on real continuum 2D percolating elastic systems, where the basic elastic response is due to the (macroscopic) elastic properties of each constituent, (this excludes polymer networks, where the basic elastic properties have an entropic origin) are consistent with the universality class of 2D networks with angle-bending forces but inconsistent with the universality class of 2D spring networks.<sup>(10–13)</sup>

In 3D networks, angle-bending forces between  $nm$  bonds are not enough to ensure that the rigidity threshold coincides with the percolation threshold. Torsional forces are also required in order to achieve that goal. This means it must cost energy to twist a pair of next-nearest-neighbor ( $nmn$ ) bonds around the intervening bond. This is expressed quantitatively by including two angle-bending terms in the elastic potential energy, one that depends on the change  $\delta\theta_{ij}$  in the angle between *nearest-neighbor*

bonds  $i$  and  $j$ , and another that depends on the change  $\delta\varphi_{ij}$  of the *azimuthal angle* between *next-nearest-neighbor* bonds  $i$  and  $j$ , where that azimuthal angle is defined by projecting both bonds onto the plane perpendicular to the intermediate bond. A crucial property of the latter change is that it can always be expressed as the difference between the increments  $\delta\varphi_i(ij)$ ,  $\delta\varphi_j(ij)$  of the *absolute azimuth angles* of those two bonds  $\delta\varphi_{ij} = \delta\varphi_i(ij) - \delta\varphi_j(ij)$ . The notation  $\delta\varphi_i(ij)$  identifies the type of azimuth angle associated with the bond  $i$  as measuring the orientation of its projection onto a plane perpendicular to the intermediate bond connecting between  $i$  and its *nnn* bond  $j$ —see Fig. 1(b).

The picture presented above is strictly true only if there is no possibility of having straight, collinear chains of bonds in the system. If such straight chains can appear, then it is possible to have an unrestricted torsional distortion—see, e.g., the configuration shown in Fig. 1(c), where the bonds  $i$  and  $j$ , which are third neighbor bonds, can be twisted around the intervening collinear bonds  $k$  and  $l$  without any cost in energy. That is why we do not consider the simplest type of 3D network, namely a simple cubic network, or even a body-centered-cubic or face-centered-cubic network. Instead, we focus our discussion on the case of an elastic network with a diamond lattice microstructure. That makes sense also because it is the natural way to generalize a 2D honeycomb network to a 3D system. In fact, if a diamond network [see Fig. 1(d)] is projected onto a plane perpendicular to any one of its bonds, the result is a honeycomb network. The latter network, because it has the most “open structure” of any 2D network, i.e., the smallest number of nearest neighbor bonds, involves fewer constraints when applying the variational principles, and was therefore the preferred system for establishing the relations of Eq. (1) in the 2D case.<sup>(1)</sup> We then appeal to the universality principle in order to argue that the critical exponents of other types of percolating networks, or even of many types of continuum percolating systems, will have the same values as in the case of the particular type of percolating network considered here.<sup>(14)</sup>

Because there are four different types of bonds in a diamond network, [see Fig. 1(d)] there will be four different types of azimuth angles, corresponding to differently oriented families of planes that are orthogonal to each of the four types of bonds. Each bond will thus have three different types of azimuth angles associated with it. However, the elastic force and energy associated with any pair of *nnn* bonds will depend only on the azimuthal angles that refer to a plane perpendicular to the intermediate bond, which is of course unique.

When the dimensionality of the network increases to values higher than 3, we need to include yet additional types of angle-bending forces in order to ensure that the rigidity threshold coincides with  $p_c$ . This can be

appreciated by considering the augmentation of an existing  $d$ -dimensional cluster by addition of a single bond at any one of its sites. The orientation of that bond is defined by  $d-1$  independent angles. In order for the augmented cluster to exhibit a solid-state-like rigidity, elastic energy must be associated with changes in *any one of those angles*. Percolating elastic networks with  $d > 3$  are briefly discussed in Section 6 later.

In contrast with elastic network models, where a new type of elementary angle-bending force must be introduced with every increase in the dimensionality from  $d$  to  $d+1$ , electrical network models do not need to change the elementary properties of the conducting bonds when  $d$  is increased. The rate of production of Joule heat in a network of any  $d$  can be expressed as

$$W_L = \sum_{(ij)} g_{ij}(V_i - V_j)^2, \quad (13)$$

where  $V_i$  is the potential at site  $i$  of the network,  $g_{ij}$  is the electrical conductance of the bond  $ij$ , which connects the sites  $i$  and  $j$ , and the summation ranges over all those bonds. This expression has the property that it attains its unique minimum value when these site potentials have their correct physical values. In seeking this minimum, some of the  $V_i$ 's should have their values fixed, at sites which can be called "boundary sites." All the other  $V_i$ 's can then be allowed to vary independently over *all possible real values*, both positive and negative. This variational property is a simple consequence of the classical variational principle of Eq. (7).

### 3.1. One-Dimensional Elastic Chains

An argument presented in ref. 7, to show that the bond stretching forces are irrelevant for long 1D chains in a 2D network, is extended here to demonstrate a similar property for the case of 1D chains in a 3D network.

In a 1D chain of  $N$  identical bonds  $i = 1 \cdots N$ , randomly constructed upon an underlying diamond network, the elastic potential energy  $E_L$  can be written as

$$2E_L = k \sum_{i=1}^N (\delta b_i)^2 + q \sum_{i=2}^N (\delta \theta_{i,i-1})^2 + m \sum_{i=3}^N (\delta \varphi_{i,i-2})^2, \quad (14)$$

where  $\delta b_i$  is the change in length of the bond  $i$ ,  $\delta \theta_{i,i-1}$  is the change in angle between that bond and its predecessor, and  $\delta \varphi_{i,i-2}$  is the change of azimuth angle of that bond, relative to the change of azimuth angle of the

bond  $i-2$ , both measured in a plane perpendicular to the intervening bond  $i-1$ . All the bond parameters which appear in this equation, namely  $\delta b_i$ ,  $\delta\theta_{i,i-1}$ ,  $\delta\varphi_{i,i-2}$ , are *independent variables*, and they can be used to solve the elastic problem, where a force  $\mathbf{F}$  is applied to the two ends of the chain, by minimizing the quantity  $E_L - \mathbf{F} \cdot \mathbf{R}_N$ , where  $\mathbf{R}_N$  is the vector separation between the two ends of the chain. To leading (first) order in the (mutually independent) bond variables, the change in  $\mathbf{R}_N$  can be written as

$$\delta\mathbf{R}_N = \sum_{i=1}^N \mathbf{b}_i \delta b_i + \sum_{i=2}^N \delta\theta_{i,i-1} \left( \frac{(\mathbf{b}_i \times \mathbf{b}_{i-1})}{|(\mathbf{b}_i \times \mathbf{b}_{i-1})|} \times \mathbf{R}_{Ni} \right) + \sum_{i=2}^N \delta\varphi_{i,i-2} (\mathbf{b}_{i-1} \times \mathbf{R}_{Ni}),$$

where  $\mathbf{b}_i$  is a unit vector along the undistorted bond  $i$ ,  $b_0$  is the undistorted bond length of all the bonds, and  $\mathbf{R}_{Ni} \equiv b_0 \sum_{j=i}^N \mathbf{b}_j$  is the vector separation between the beginning of the bond  $i$  and the end of the bond  $N$ . Clearly,  $\mathbf{R}_{N1} = \mathbf{R}_N$  and  $|\mathbf{R}_{Ni}|$  is usually of the same order as  $|\mathbf{R}_N|$ . We will denote by  $\alpha_0$  the common value of the undistorted angle between any two  $m$  bonds, thus  $|(\mathbf{b}_i \times \mathbf{b}_{i-1})| = \sin \alpha_0$ .

Minimization of  $E_L - \mathbf{F} \cdot \mathbf{R}_N$  yields the following results for the equilibrium values of the bond variables:

$$\delta b_i = \frac{\mathbf{F} \cdot \mathbf{b}_i}{k},$$

$$\delta\theta_{i,i-1} = \frac{1}{q \sin \alpha_0} \mathbf{F} \cdot [(\mathbf{b}_i \times \mathbf{b}_{i-1}) \times \mathbf{R}_{Ni}],$$

$$\delta\varphi_{i,i-2} = \frac{1}{m} \mathbf{F} \cdot (\mathbf{b}_{i-1} \times \mathbf{R}_{Ni}),$$

and consequently the following form for the equilibrium value of  $E_L$ :

$$2E_L = \frac{1}{k} \sum_{i=1}^N (\mathbf{F} \cdot \mathbf{b}_i)^2 + \frac{1}{q(\sin \alpha_0)^2} \sum_{i=2}^N (\mathbf{F} \cdot [\mathbf{R}_{Ni} \times (\mathbf{b}_i \times \mathbf{b}_{i-1})])^2 + \frac{1}{m} \sum_{i=3}^N [\mathbf{F} \cdot (\mathbf{R}_{Ni-1} \times \mathbf{b}_{i-1})]^2.$$

As the number of bonds  $N$  in the randomly meandering chain increases, the first sum will increase as  $N$  itself, while the second and third sums will increase faster, as  $NR^2$ , where  $R$  is of order of the end-to-end distance in the chain  $|\mathbf{R}_N|$ , or more precisely, of its "radius of gyration," which also increases as some positive power of  $N$ . Consequently, when  $N$  is very large, the elastic energy at equilibrium is predominantly due to the angle-bending

terms. The bond stretching force constant  $k$  becomes “irrelevant,” and the macroscopic response of the chain is determined by the other force constants  $q$  and  $m$ . However, it should be noted that, while this is true when  $q$  and  $m$  are comparable to  $kb_0^2$ , it will surely fail if  $k$  is small enough so that  $kR^2 \ll q$  and  $kR^2 \ll m$ . Due to the different dependence on  $N$  and  $R$ , the critical aspects of the macroscopic elastic response will then be different from what they are when  $kb_0^2$  is comparable to either  $q$  or  $m$ . Another fact worth noting is that, if  $q$  and  $m$  are vastly different, i.e., if  $q \ll m$  or  $m \ll q$ , then the larger of the two also becomes irrelevant. Nevertheless, because both of the angle-bending contributions to  $E_L$  have the same dependence on  $N$  and  $R$ , the critical aspects of the 1D chain will be the same, irrespective of whether  $q \ll m$ ,  $m \ll q$ , or  $m \approx q$ .

### 3.2. Three-Dimensional Percolating Elastic Networks

In a random network near  $p_c$ , the percolating backbone is very tenuous and includes many long sections that are 1D chains of bonds. Therefore, even though the critical behavior differs from that of a 1D chain, we can expect that  $k$  will still be irrelevant, and that the relative sizes of  $q$  and  $m$  will also be irrelevant. The first of these expectations has been tested, in the case of 2D networks, by numerical simulations. These simulations showed that the macroscopic elastic moduli of a 2D network at  $p_c$  are insensitive to the precise value of  $k$ , and depend only on the single angle-bending force constant in the plane.<sup>(15, 16)</sup> Similar simulations have not been made for 3D percolating networks. Nevertheless, we will assume that, near  $p_c$ ,  $k$  is irrelevant. More precisely, we will assume that the network response is in the same universality class, and that the critical exponents therefore have the same values, as long as the total bond stretching energy at equilibrium is less than at least one of the total angle-bending energies at equilibrium.

The elastic potential energy of the network can be written in the following form

$$2E_L = \sum_i k_i (\delta b_i)^2 + \sum_{(ij)=nm} q_{ij} (\delta \theta_{ij})^2 + \sum_{(ij)=nmn} m_{ij} [\delta \varphi_i(ij) - \delta \varphi_j(ij)]^2. \quad (15)$$

Here the first term represents the bond stretching energy, with  $\delta b_i$  as the change in length of the bond  $i$ , the second term represents the  $nm$  angle-bending energy, with  $\delta \theta_{ij}$  as the change of angle between the  $nm$  bonds  $i$  and  $j$ , and the third term represents the  $nmn$  torsional energy, with  $\delta \varphi_i(ij)$ ,  $\delta \varphi_j(ij)$  denoting the changes in azimuth angles that the  $nmn$  bonds  $i$ ,  $j$

subtend in a plane perpendicular to the intermediate bond. Although the latter term bears some similarity to the angle-bending term in the 2D expression (12), it must be kept in mind that each bond can now have more than one type of azimuth angle associated with it, depending on the orientations of its various  $mn$  bonds.

As in the case of 2D networks, we need to specify that, in a two-constituent mixture of small and large values of the force constants, the  $mn$  angle-bending force constant  $q_{ij}$  takes the large value only if both of the stretching force constants  $k_i$  and  $k_j$  have large values. In addition to that, we also need to specify that the  $nnn$  azimuthal angle-bending constant  $m_{ij}$  takes the large value only if *all three associated bonds* have large stretching force constants. Thus, not only must  $k_i$  and  $k_j$  be large, but also the stretching constant  $k_l$  of the bond  $l$  that connects between the  $nnn$  bond pair  $ij$ . These requirements ensure that the macroscopic elastic stiffness moduli of the network will be large if and only if the bonds with the large stretching force constants percolate.

### 3.3. Electrical Networks Related to Elastic Networks

It is evident that the expression (13) for  $W_L$  bears some formal similarity to the last sum in Eq. (15). Nevertheless there are also some important differences, which hinder the establishment of a close correspondence between the variables  $V_i$  and  $\delta\phi_i(ij)$ : (a) The potentials  $V_i$  refer to sites whereas the azimuth angle increments  $\delta\phi_i(ij)$  refer to bonds. (b) Whereas a conducting site has only one potential  $V_i$  associated with it, an elastic bond in a 3D network usually has more than one type of azimuth angle associated with it.

The difficulty posed by item (a) is resolved by considering the electrical problem on a "covering network" of the original elastic network. This covering network is constructed as follows: Every bond of the original network is replaced by a site of the covering network; any two sites of the covering network are connected by a bond if and only if the corresponding bonds of the original network were *next-nearest-neighbors*, i.e., if they were separated by one intermediate bond. This is a variation on the original concept of "covering network," where two sites of that network were connected by a bond only if the corresponding bonds of the original network were *nearest-neighbors*.<sup>(17, 18)</sup> Covering networks of the original type were invoked in the recent discussion of exact relations between elastic and electrical responses of 2D percolating networks.<sup>(1)</sup>

Although the covering network differs from the original network, the two will have identical percolation thresholds: That is so because the existence/nonexistence of a percolating cluster in the system is a common

property of the original network and the covering network, irrespective of whether the microstructure is ordered or disordered.<sup>(18)</sup> Furthermore, we can expect that the covering network belongs to the same universality class of percolation as the original network, since no long range correlations are introduced by the transformation from one to the other.

The difficulty posed by item (b) is dealt with by a more devious trick: Recalling that there are four different types of azimuth angles  $\delta\varphi_i(ij)$ , corresponding to the four possible orientations of the intermediate bond connecting the  $nmn$  bonds  $i$  and  $j$ , we change the torsional force constants  $m_{ij}$  for three of those types of torsional distortions either to 0 or to  $\infty$ , if they were originally set to be finite. The orientation of the fourth bond type will be chosen as the  $z$  axis. The result of this alteration of the force constants is that there will be no elastic energy associated with torsional distortions of  $nmn$  bond pairs around the other three directions. Therefore the torsional energy term in Eq. (15) will include only terms with one type of azimuthal angle increments, which will be denoted simply by  $\delta\varphi_i$

$$\sum_{(ij)=nmn} m_{ij}(\delta\varphi_i - \delta\varphi_j)^2.$$

Obviously, these azimuthal angle increments are measured in the  $x, y$  plane. That is why the above expression has almost the same form as the angle-bending energy in the case of a 2D network—cf. Eq. (12). The conductances  $g_{ij}$  of the covering network are now set equal to the corresponding values of  $m_{ij}$  in the original network, while the finite  $m_{ij}$  that were altered to become 0 or  $\infty$  are translated into similarly altered values of  $g_{ij}$ .

The choice between setting the other  $m_{ij}$  and  $g_{ij}$  to 0 or to  $\infty$  depends on the type of percolation problem we wish to consider: In the case of a diluted network slightly above the percolation threshold  $p_c$ , any pair of sites on a connected cluster, separated by a distance of order  $\xi$ , have a large number of “singly connected bonds (SCB)” associated with them, in fact, their number is of order  $1/\Delta p$ .<sup>(19)</sup> Those are bonds, connected in series, such that if any one of them is deleted, the pair of sites become disconnected. In this case, replacing a certain fraction of all the (normal) bonds by totally rigid or perfectly conducting bonds will usually not make the connection between the two sites totally rigid or superconducting. Therefore, in this case we choose  $\infty$  for the non-finite value of  $m_{ij}$  or  $g_{ij}$ .

By contrast, in the case of a rigid/normal (superelastic) or normal/perfectly conducting (superconducting) random network, slightly below  $p_c$  of the totally rigid or perfectly conducting constituent, we need to choose 0 for the non-finite value of  $m_{ij}$  or  $g_{ij}$ : Any pair of unconnected sites,

(i.e., sites not on the same rigid or perfectly conducting cluster) at a distance  $\xi$  from each other, have a similarly large number (of order  $1/|\Delta p|$ ) of “singly disconnecting bonds (SDB)” associated with them.<sup>(20)</sup> These are normal bonds, situated in parallel, such that if any of them is made rigid or superconducting, it will place the two sites on the same rigid or superconducting cluster. In this case, deleting a certain fraction of all the normal bonds will usually not sever the *normal connection* between the two sites—they will still have a large number of (parallel) SDB’s associated with them. Therefore, in this case we choose 0 for the non-finite value of  $m_{ij}$  of *nnn* normal bond pairs where the intermediate bond lies in one of the three “wrong directions,” and also  $g_{ij} = 0$  for the conductance of the corresponding covering network bond.

Note that force constants  $m_{ij}$  that were either 0 or  $\infty$  to begin with are not changed in the above described procedure. In particular, the “absent bonds” in a diluted network have zero values for all of the associated force constants, and those are not changed. Similarly, a rigid bond  $i$  in a superelastic network has  $k_i = \infty$ , as well as possible infinite values for some of the associated angle-bending force constants  $m_{ij}$ ,  $q_{ij}$ , and those are not changed either. Only finite values of  $m_{ij}$  can be changed in that procedure. These caveats are necessary so as not to change the overall properties of such networks: If we changed a large fraction of the absent *nnn* bond pairs in a diluted network near  $p_c$  to *nnn* bond pairs with  $m_{ij} = \infty$ , then the network would become totally rigid. Similarly, if we changed a large fraction of the totally rigid *nnn* bond pairs of a nearly percolating superelastic network to pairs with  $m_{ij} = 0$ , then that network would cease to be nearly rigid.

As explained above, in order to establish a close correspondence between the elastic network and the covering electrical network, we set  $g_{ij} = m_{ij}$  for all the bonds of the latter network. If the finite values of  $m_{ij}$  are all identical  $m_{ij} = m$ , then the covering network will be a *three constituent mixture*, where bonds can have either a fixed finite value of the conductivity  $g_{ij} = m$ , or  $g_{ij} = 0$ , or  $g_{ij} = \infty$ .

From the above considerations it is clear that the substitution of 0 or  $\infty$  for all the finite  $m_{ij}$  or  $g_{ij}$  that lie along the three “wrong” bond directions in the system will not change the rigidity or conductivity threshold away from the geometrical percolation threshold  $p_c$ . However, these changes may make the macroscopic response somewhat anisotropic. Nevertheless, we will assume that these substitutions do not change the universality class of the system, and that the various components of  $\hat{C}_e$  are still characterized by the same values of the critical exponents  $T$  and  $S$ , and that  $\hat{\sigma}_e$  is similarly still characterized by the same values of  $t$  and  $s$ . In Section 7 later we will argue that it would be sufficient to verify this



universality property for conducting networks with such substitutions—the analogous property for elastic networks would then follow from the results obtained in this article.

## 4. APPLICATION OF THE NETWORK VARIATIONAL PROPERTIES

### 4.1. Elastic Solution as Trial Values for Electrical Problem

Because the variational properties of Eq. (13) allow independent and unrestricted variations of the site potentials  $V_i$ , it becomes a straightforward matter to use the exact solution of the elastic network problem in order to provide a set of trial values for the electrical problem on the covering network—we simply set  $g_{ij} = m_{ij}$  and  $V_i = \delta\varphi_i$  everywhere. In this way we find

$$W_L \leq \sum_{(ij)=nm} m_{ij} (\delta\varphi_i - \delta\varphi_j)^2 \leq 2E_L. \quad (16)$$

In Section 5 below, this inequality is applied to specific macroscopic configurations of random networks in order to derive inequalities for the critical exponents of elastic and electrical percolating networks.

### 4.2. Electrical Solution as Trial Values for Elastic Problem

In contrast with the brief discussion in the previous subsection, the use of the exact solution of an electrical problem in order to provide trial values for the elastic bond variables must overcome certain obstacles: (a) Although  $E_L$  achieves its unique minimum value, equal to its physical value at equilibrium, when  $\delta b_i$ ,  $\delta\theta_{ij}$ ,  $\delta\varphi_i$  are given their correct physical values, in seeking that minimum these variables cannot be varied independently. Thus, any trial values must obey certain constraints. Some of these constraints result from the existence of closed loops in the network. These involve long range correlations, since there are loops of all sizes up to  $\xi$ . Therefore a straightforward imposition of constraints on the  $V_i$ 's of the covering network, similar to those which are satisfied by the  $\delta\varphi_i$ 's of the original network, might possibly change the universality class of the electrical percolation problem. That option should therefore be avoided. Instead of that, we will attempt to satisfy the loop constraints by a judicious choice of trial values for the  $\delta b_i$ 's, which are not determined by the solution of the electrical problem. Other short range constraints also exist,

among values of  $\delta\varphi_i$  and  $\delta\theta_{ij}$  for neighboring interbond angles—those will be discussed below. These constraints are not automatically satisfied if we simply try to assign  $\delta\varphi_i = V_i$ , where  $V_i$  is an exact solution of an electrical problem on the covering network. (b) Even if we somehow managed to satisfy all the constraints, we would still only have an estimate for the torsional part of the elastic energy, whereas we need an upper bound for the entire elastic energy.

Since we will only need to apply these trial values in the case of a diluted network slightly above  $p_c$ , we now show how to overcome the above obstacles in that case.

Item (b) is dealt with by first changing to  $\infty$  all the originally finite (i.e., nonzero)  $nm$  angle-bending force constants  $q_{ij}$ . This will result in a vanishing contribution to  $E_L$  from all the  $nm$  angle-bending energies, since all the  $q_{ij}$  are now either 0 or  $\infty$ . As shown in Section 3.1, this does not change the macroscopic critical behavior of 1D chains. Due to the large number of SCB's in a randomly diluted network that is slightly above  $p_c$ , such chains will be abundant in the system. Consequently, the rigidity threshold will not be changed. We will assume further that the universality class also remains unaltered when this change has been implemented. As a result of this, together with the fact that many of the nonzero  $m_{ij}$  have also been set to  $\infty$ , there will be many bonds whose azimuth angles  $\delta\varphi_i$  can only change in unison: That is true for any pair or triplet of  $nm$  bonds none of which lie along the special direction  $z$ . This constitutes another constraint on the values allowed for the  $\delta\varphi_i$ 's. In order to ensure that the corresponding site potentials in the covering network also obey these additional restrictions, we add superconducting bonds  $g_{ij} = \infty$  between every such pair of covering network sites—see Fig. 1(e), where the covering network bond (dashed line) between the covering network sites (filled circles)  $l$  and  $p$  is such a conductor with  $g_{lp} = \infty$ .

Assuming we have managed to find values for  $\delta b_i$  that will satisfy all of the loop constraints, after making the assignments  $\delta\varphi_i = V_i$ , we will have the following upper bound for  $E_L$ :

$$2E_L \leq \sum_i k_i (\delta b_i)^2 + \sum_{(ij)=nm} m_{ij} (\delta\varphi_i - \delta\varphi_j)^2, \quad (17)$$

$$\sum_{(ij)=nm} m_{ij} (\delta\varphi_i - \delta\varphi_j)^2 = W_L. \quad (18)$$

Since the  $\delta b_i$  in this upper bound were chosen so as to satisfy the loop constraints, without any regard for the cost in energy, this can result in

a very large value for the sum  $\sum_i k_i (\delta b_i)^2$ . We therefore now *choose a value*  $k$  for the bond stretching force constants that will make that sum not greater than  $W_L$ . Thus we will have  $2E_L \leq 2W_L$ . This may require a very small value of  $k_i \equiv k$ , and we need to worry about whether that may take the system to a different universality class, as explained in Section 3.1. We therefore allow both  $\delta b_i$  and  $\delta \varphi_i$  to vary further, subject to all the relevant constraints, so as to minimize the sum of two sums on the r.h.s. of Eq. (17). That minimum will be the correct equilibrium value of  $2E_L$ . In reaching that minimum, the second sum on the r.h.s. of that equation will in fact have *increased* to a value greater than  $W_L$ , since the latter value was already the *unconstrained minimum* of that sum. Therefore, the first sum on the r.h.s. of that equation will have decreased, by an even greater amount, to a value that is less than  $W_L$ . We will then have

$$2E_L = k \sum_i (\delta b_i)^2 + \sum_{(ij)=nnn} m_{ij} (\delta \varphi_i - \delta \varphi_j)^2 \leq 2W_L, \quad (19)$$

$$k \sum_i (\delta b_i)^2 \leq W_L \leq \sum_{(ij)=nnn} m_{ij} (\delta \varphi_i - \delta \varphi_j)^2. \quad (20)$$

The first line is the sought after relation between  $E_L$  and  $W_L$ . We note that it is *not inconsistent* with the previously obtained inequality (16). The second line is important because it allows us to expect that  $k$  is not so small as to place the system in a different universality class. In fact, we need to choose the value of  $k$  for a system of size  $\xi \propto \Delta p^{-\nu}$  for some definite value of  $\Delta p$ . Then,  $W_L \propto \xi^{d-2-1/\nu}$  increases with increasing  $\Delta p$  for any dimensionality  $d$ , (in particular, for  $d=3$ , where we have  $\nu \cong 0.89^{(22)}$  and  $t \cong 2.0$ ,<sup>(23)</sup> we get  $W_L \propto \xi^{-1.2} \propto \Delta p^{1.1}$ ) but the value of  $k \sum_i (\delta b_i)^2$  dictated by the constraints can only decrease, since the total number of bonds in closed loops decreases when  $\xi$  decreases from a large value (recall that  $\xi$  is also the total size of the system). Therefore, a choice of  $k$  that makes  $k \sum_i (\delta b_i)^2 \leq W_L$  for some value of  $\Delta p$  should also suffice for any larger value of  $\Delta p$ .

At this stage we can relax the infinite values assumed for  $q_{ij}$ , as well as the infinite values assumed for the  $m_{ij}$  that were in the “wrong directions,” without spoiling the correspondence established between the elastic and the electrical networks: If we now let the  $q_{ij}$ ’s and all the “wrong direction”  $m_{ij}$ ’s have finite values, say comparable to the “right direction”  $m_{ij}$ ’s, but leave the infinite values of  $g_{ij}$  unchanged in the covering network, then this can only decrease the equilibrium value of  $E_L$ , thereby strengthening the inequality (19). The significance of this step is that it allows us to consider an elastic network that will exhibit a macroscopic response which is isotropic.

We still need to deal with item (a), namely, we need to show that the assignments  $\delta\varphi_i = V_i$  involve no inconsistencies, and that values of  $\delta b_i$  exist that will satisfy all of the loop constraints.

We have already limited all the azimuthal angle increments to rotations in the  $x, y$  plane, around bonds that are perpendicular to that plane. Moreover, different bonds attached to such a bond at one of its ends must all undergo the same azimuth angle increment  $\delta\varphi_i$  [see Fig. 1(e)]. This requirement is reflected in a similar requirement that is satisfied by the corresponding site potentials  $V_i$ , as discussed earlier in connection with item (b) above. Therefore, the assignments  $\delta\varphi_i = V_i$  can proceed without encountering any inconsistency. The only remaining problem is that a closed loop will usually transform into an un-closed loop, and we need three independent and unused degrees of freedom in order to close it back again. These can be provided by an appropriate choice of three  $\delta b_i$ 's in the loop, corresponding to bonds along *three different directions*. In order to ensure the existence of such bonds, without having to impose any long range correlations, we introduce the following additional requirements: (i) Any site has at most three bonds attached to it. Naturally, these bonds lie along three different directions. Such a site will be called a "triadic site." (ii) Two triadic sites are separated by at least two non-triadic (i.e., diadic) sites in series. (iii) The  $nmn$  bonds of a triadic site all lie in the "fourth direction," i.e., the one that differs from those of the three bonds attached to the triadic site itself.

These restrictions ensure that any consecutive sequence of three bonds, where one of the two internal sites is a triadic site, must have *different undistorted orientations* that span the entire 3D space, thus these bonds can be exploited to close a distorted loop—see Fig. 1(f). The final step of this argument is to note that the percolating cluster can be constructed by starting with one of its loops, built around a triadic site (loop  $A$  in that figure), which must include at least one more triadic site, and then adding a chain at that second triadic site. If that chain evolves into a new loop (loop  $B$  in the same figure), which must have at least three bonds, as well as two triadic sites and two diadic sites, in common with the original loop, then a sequence of three bonds ( $i, j, k$  in that figure), which includes one of the common triadic sites as an internal site, can be used to ensure closure of the new loop.

We note that the additional restrictions (i)–(iii) introduce short range correlations into the random configurations of occupied bonds in the diluted network. This will complicate any attempts to simulate such a network, and will most likely change the value of  $p_c$ . However, such correlations are not expected to change the universality class of percolation. Therefore they should not affect the values of any of the critical exponents.

## 5. MACROSCOPIC SAMPLES OF PERCOLATING NETWORKS

In order to obtain results for the critical behavior of the macroscopic moduli, we need to apply the inequalities (16) and (19), obtained in Section 4, to appropriate boundary value problems on some macroscopic network samples.

### 5.1. Diluted Networks

Taking a cue from the case of diluted 2D networks, where we considered a planar ring shaped sample,<sup>(1)</sup> we consider for the 3D case a long circular cylinder, with its axis along the special direction  $z$  and its length  $L_z$ , and with a ring shaped cross section—see Fig. 2(a). The boundary at the outer radius of the sample is held fixed, i.e., all the bonds at that boundary have their lengths and orientations unchanged. The boundary at the inner radius of the sample is rigidly rotated around the cylinder axis by an angle  $\delta\varphi_0$ , i.e., all the relevant bonds (those that are not parallel to  $z$ ) which intersect that boundary have  $\delta\varphi_i = \delta\varphi_0$ . This boundary value problem is solvable in continuum elasticity, when the medium is homogeneous and isotropic in the  $x, y$  plane, which is the case if the inner radius, denoted by  $L$ , is much greater than the percolation correlation length  $\xi$ . If the outer

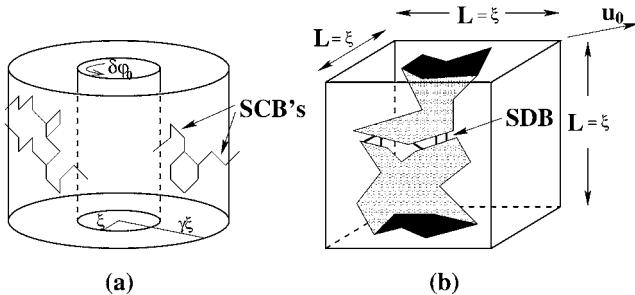


Fig. 2. (a) Macroscopic sample of a diluted elastic diamond network, shaped as a cylinder with a circular-ring-shaped cross section, and its axis along the elementary bond direction  $z$ , around which the elementary torsional or azimuthal force constants  $m_{ij}$  remain finite. Only some of the network bonds are shown, including two “spanning clusters.” Of the azimuthal force constants, only those that refer to azimuth angles in the  $(x, y)$  ring plane have finite values. All the other  $m_{ij}$  of non-absent bonds are infinite. (b) Macroscopic sample of a rigid/normal elastic diamond network, shaped as a  $\xi \times \xi \times \xi$  cube. Shown are two large rigid clusters (cross-hatched), attached to opposite edges of the sample, (the flat attachment areas are shown in black) and separated from each other by, among other things, a large number of SDB's in parallel. Those SDB's are denoted by short thick solid segments.

radius, denoted by  $\gamma L$ , is also much greater than the inner radius, i.e.,  $\gamma \gg 1$ , then the total elastic energy per unit length of the cylinder is given by

$$\frac{E_L}{L_z} = 2\pi\mu_e L^2 (\delta\varphi_0)^2, \quad (21)$$

where  $\mu_e \equiv C_{xyxy}^{(e)}$  is the macroscopic or bulk effective shear modulus in the  $x, y$  plane. Presumably, all the macroscopic stiffness moduli will exhibit the same critical behavior.

For the covering network of the above described elastic network, we apply the boundary condition  $V_i = 0$  at all sites on the outer radius of the ring-shaped cylinder and  $V_i = \delta\varphi_0$  at all sites on the inner radius that correspond to non- $z$ -parallel bonds of the elastic network. This problem is also easily solvable in continuum conductivity, when the medium is homogeneous and isotropic in the  $x, y$  plane. The total production rate of Joule heat per unit length of the cylinder is given by

$$\frac{W_L}{L_z} = 2\pi\sigma_e (\delta\varphi_0)^2 \ln \gamma, \quad (22)$$

where  $\sigma_e$  is the macroscopic or bulk effective conductivity in the  $x, y$  plane. Again, we presume that all the macroscopic conductivities will exhibit the same critical behavior.

It is noteworthy that  $W_L$  is independent of the inner radius of the system  $L$ , while  $E_L \propto L^2$ . It follows that the inequality (16) will be satisfied in a trivial fashion if  $L$  is large enough. In order to get a useful result from that inequality, we should therefore make  $L$  as small as possible. The smallest value of  $L$  for which the homogeneous continuum expressions for  $E_L$  and  $W_L$  are still valid is  $\xi \propto \Delta p^{-\nu}$ , the percolation correlation length. That is so because on length scales much greater than  $\xi$ , the heterogeneity is averaged out and the system response to uniform external perturbations is like that of a homogeneous material characterized by the bulk effective values  $\sigma_e, C_e$  of macroscopic conductivity and macroscopic elastic stiffness. By contrast, when the system size is smaller than  $\xi$ , the response is not averaged out, and it fluctuates strongly from sample to sample, depending on whether a particular sample happens to percolate and on other microstructural details. This property of  $\xi$  lies at the basis of the finite size scaling phenomenon, originally found in a discussion of the 2D Ising model,<sup>(24)</sup> and later formulated as a hypothesis for generic second order phase transitions.<sup>(25)</sup> It has been widely used in the interpretation of simulations of percolating networks<sup>(14-16, 22, 23)</sup> and other statistical models.<sup>(26, 27)</sup> From the definition of  $\xi$  implied by the description given above, it is obvious that  $\xi$  is

similar to the average cluster size in systems below the percolation threshold. Above that threshold  $\xi$  will be similar to the average distance between adjacent nodes, where a node is a site from which more than 2 independent paths exist that lead to the edges of the system. It is generally assumed that the critical exponent  $\nu$  of Eq. (2) has the same value above and below the threshold  $p_c$ ,<sup>(14)</sup> as found exactly in 2D, where  $\nu = 4/3$ ,<sup>(14, 21)</sup> and numerically in 3D, where  $\nu = 0.89 \pm 0.01$ .<sup>(22)</sup> In fact, we only need to use the value of  $\nu$  above  $p_c$ , therefore this question is moot.

When we put  $L = \xi$  in the expressions for  $E_L/L_z$  and  $W_L/L_z$ , the results depend only upon  $\Delta p$ , and the inequality (16) leads to

$$\sigma_e \ln \gamma < 2\mu_e \xi^2 \Rightarrow \Delta p^t < \Delta p^{T-2\nu} \Rightarrow T \leq t + 2\nu. \quad (23)$$

Applying the inequality (19) to the same network leads to the opposite inequality between the critical exponents

$$T \geq t + 2\nu. \quad (24)$$

Taken together, the two inequalities lead to the exact equality

$$T = t + 2\nu$$

for 3D networks.

## 5.2. Rigid/Normal Networks

In the case of a rigid/normal network, a more convenient boundary value problem is obtained by considering a macroscopic sample in the shape of a cube with edge length  $L$ —see Fig. 2(b). At the boundaries of the elastic network, in the limit of homogeneous continuum elasticity, we apply a linearly varying displacement field  $\mathbf{u}_0(\mathbf{r}) = \hat{\epsilon}_0 \cdot \mathbf{r}$ . This ensures that the volume averaged strain tensor is equal to  $\hat{\epsilon}_0$ , and that the total elastic energy is given by

$$E_L = \frac{1}{2} L^3 \hat{\epsilon}_0 \cdot \hat{C}_e \cdot \hat{\epsilon}_0. \quad (25)$$

In order to find a relation between the boundary conditions of the continuum problem and the network problem, we recall that, slightly below the percolation threshold of such a network, there are a large number of “singly disconnecting bonds (SDB)” for any two distant sites of the system: If two network sites are at a distance  $\xi$ , then the total number of associated SDB’s is of order  $1/|\Delta p| \propto \xi^{1/\nu} \gg 1$ .<sup>(20)</sup> From this observation there emerges a simple picture of what a rigid/normal network looks like when it

is just below the percolation threshold of the rigid constituent and its size is  $\xi \times \xi \times \xi$ : There are a few very large rigid clusters, which do not form a system-spanning rigid cluster due to a large number of SDB's *arranged in parallel*—see Fig. 2(b). These SDB's will carry essentially the entire boundary displacement  $\mathbf{u}_0(\mathbf{r})$ . Thus their lengths will change by an amount of order  $|\mathbf{u}_0| \propto \varepsilon_0 \xi$ , where  $\varepsilon_0$  represents the magnitude of the average strain tensor  $\hat{\varepsilon}_0$ . Similarly, their orientations, including azimuth angles, will change by an amount of order  $\varepsilon_0 \xi / b_0 \equiv \delta\varphi_0$ , where  $b_0$  is the length of an undistorted bond. Many bonds at the boundary will also have their azimuthal orientations changed by an amount of order  $\delta\varphi_0$ . The total elastic energy of such a network is given by

$$E_L \propto \xi^3 C_e (\varepsilon_0)^2 \propto \xi C_e b_0^2 (\delta\varphi_0)^2, \quad (26)$$

where the quantity  $C_e$  represents a generic macroscopic elastic stiffness modulus or component of  $\hat{C}_e$ .

The corresponding (covering) electrical network of this rigid/normal elastic network is an electrical superconducting/normal network, just below the percolation threshold of the superconducting constituent. In order to correspond to the elastic network problem, we apply a potential  $V_i$  at each boundary site that is equal to the azimuthal angle increment  $\delta\varphi_i$  of the corresponding boundary bond. As explained above, many of those will be of order  $\delta\varphi_0$ , hence the total rate of production of Joule heat will be given by

$$W_L \propto \xi \sigma_e (\delta\varphi_0)^2, \quad (27)$$

where  $\sigma_e$  is one of the macroscopic conductivities, all of which should exhibit similar critical behavior.

Applying the inequality (16) to the expressions (26) and (27) we get

$$\sigma_e < C_e b_0^2 \Rightarrow |\Delta p|^{-s} < |\Delta p|^{-S} \Rightarrow S \geq s.$$

Taken together with the previous result  $S \leq s$ , which was demonstrated in Section 2, this leads to the exact equality

$$S = s \quad (28)$$

for 3D networks.

## 6. EXTENSION TO NETWORKS OF ARBITRARY DIMENSIONALITY $d$

In order to extend the proofs presented above to percolating elastic networks of higher dimensionalities  $d$ , we probably need to use a basic



network structure that is a hyper-diamond lattice. That would be an ordered network constructed from identical sites, with  $d+1$  bonds attached to each site in a manner such that the undistorted angle between any pair of those bonds is the same, and any  $(d-1)$ -member subset of those bonds spans a different  $(d-1)$ -dimensional subspace. In order to ensure that the rigidity threshold coincides with  $p_c$ , we need to have angle-bending forces between further than nearest-neighbor bond pairs, out to furthest neighbor bond pairs that are separated by  $d-2$  intervening bonds in series. None of those intermediate bonds should be parallel to any of the end bonds, and they should span a unique  $(d-2)$ -dimensional subspace. This will uniquely define a family of 2D parallel planes orthogonal to that subspace, which will not be perpendicular to either of the two end bonds. These two bonds will therefore have non-vanishing projections onto that family of planes, and we will be able to define absolute azimuth angles  $\delta\varphi_i(ij)$ ,  $\delta\varphi_j(ij)$  for those projections. The elastic potential energy will be a sum of  $d$  positive terms: A bond stretching term, plus  $d-1$  angle-bending terms, involving angles between bond pairs separated by a successively increasing number of intermediate bonds in series, from 0 up to  $d-2$ . The term involving the “furthest neighbor bond (*FNB*) pairs” will have a form similar to the third term of Eq. (15), namely

$$\sum_{(ij)=FNB} m_{ij}[\delta\varphi_i(ij) - \delta\varphi_j(ij)]^2. \quad (29)$$

The only differences are that  $i$  and  $j$  are now an *FNB* pair instead of an *nnn* pair, and that there are many more different types of azimuth angles  $\delta\varphi_i(ij)$ , depending on an entire chain of bonds between the *FNB*'s  $i$  and  $j$ .

Also, in a two-constituent mixture of large and small values of the elementary force constants, the large value of any angle-bending constant will apply if and only if all the relevant bonds have large stretching constants  $k_i$ . I.e., for a bond pair  $ij$ , both  $k_i$  and  $k_j$  must be large, as well as all the  $k_i$  for at least one continuous chain of  $d-2$  intermediate bonds.

The electrical problem now needs to be considered on a covering network where two sites are connected by a bond only if the corresponding bonds of the original network were *FNB*'s.

In order to establish a close correspondence, between the expression for  $W_L$  of the covering network and the *FNB* term in the expression for  $2E_L$  of the original elastic network, we will again have to set many of the  $m_{ij}$  force constants, along with the corresponding conductances  $g_{ij}$  of the covering network, either to  $\infty$  (in the case of a diluted network near  $p_c$ ) or to 0 (in the case of a superelastic network near  $p_c$ ), leaving finite values only for those  $m_{ij}$  which refer to azimuth angles in one family of parallel

2D planes. In order to obtain the analogues of Eqs. (19) and (20), we will have to set all the other angle-bending force constants to  $\infty$  in the case of a diluted network, and we will have to introduce additional covering network bonds with infinite conductance between nearby covering network sites whose potentials need to be equal in order to satisfy certain elastic network constraints. Some short range correlations will have to be imposed on the occupation of bonds in the randomly diluted elastic network, in order to ensure the consistency of the assignments  $\delta\varphi_i = V_i$  and the possibility of closing all the distorted loops by an appropriate choice of the  $\delta b_i$ 's. I have not worked out the precise nature of these short range correlations, since that requires taking account of the detailed microstructure of the hyper-diamond networks upon which the discussion is presently based.

From the above discussion, it will follow that the equalities (1) should hold for percolating networks of any dimensionality  $d$ . This conclusion is based on assumptions regarding universality classes of percolation which are widely believed but unproven, as in the case  $d = 3$ .

## 7. SUMMARY AND DISCUSSION

Relations between the macroscopic critical behaviors of percolating elastic networks with angle-bending forces and percolating electrical networks were discussed in the case where the dimensionality is 3 or greater. Diluted networks as well as superelastic networks were considered. Using exact variational principles, arguments were presented to show that  $T = t + 2v$  and  $S = s$ . Those arguments involved consideration of special types of random networks, with specific "open" microstructures and short range correlations, as well as reassignments of extreme values (0 or  $\infty$ ) to some of the originally finite force constants or conductances of elementary network bonds. Therefore the validity of those arguments hinges upon certain universality properties of percolating systems. Though widely believed, those properties have not been examined in a critical fashion for the percolation process, and therefore they remain unproven. In particular, replacement of a finite fraction of finite force constants  $m_{ij}$  or finite conductances  $g_{ij}$  by  $\infty$  or 0, in diluted networks or in superelastic networks respectively, needs to be studied in order to verify the assumption that such replacements do not change the universality class. Because of this, the proofs presented here are conditional upon verification of these assumptions. Nevertheless, I am confident that the conclusions reached are valid. If they fail because any of the universality assumptions that were made turns out to be wrong, then that breakdown of universality would become an interesting discovery in its own right. I hope that the results presented

here will motivate other researchers to try and prove or disprove some of those assumptions.

I note that Eq. (1) was obtained independently of whether those universality assumptions are valid. Those assumptions are crucial only as regards the actual values attained by the critical exponents  $t$ ,  $s$ ,  $T$ ,  $S$ . I.e., only if those universality assumptions are valid, will it follow that the relations of Eq. (1) also hold for the properties of the unaltered elastic and electrical networks, which can serve as models for real, two-constituent, continuum percolating composites. It would be sufficient to check those universality assumptions using appropriately altered three constituent conducting networks with normal, perfectly insulating, and perfectly conducting bonds, without having to also check those assumptions on elastic networks. Obviously, such a study would be much easier to perform on conducting networks than on the analogous elastic networks. The only assumptions that need to be tested specifically by simulations of elastic networks are the ones having to do with the irrelevance of  $k$  and of the ratio  $q/m$  when that ratio tends to  $\infty$  or to 0. I also hope that researchers with a better understanding of the microstructural details of hyperdiamond networks will try to fill in the gaps in the outlined arguments for  $d > 3$ .

It should also be noted that the arguments and proofs presented here do not rely on the fact that the fractal percolating cluster is produced by a random assignment of the network bonds to two different classes. Therefore they should also be applicable to other fractal systems, such as the one discussed in ref. 28, which is based on the fractals known as Koch curves. They might also be applicable to percolating systems that lie in different universality classes. A relevant example is provided by the so-called "Swiss cheese" models, discussed in ref. 29. Although these models were introduced in the context of continuum percolation, they can always be mimicked using networks with a specially contrived distribution of bond conductances or elastic force constants.

Simulations have not been performed for any elastic system with  $d > 2$ . This is due not only to the difficulty of carrying out such simulations, but also to the fact that some upper<sup>(5)</sup> and lower<sup>(7)</sup> bounds for  $T$ , known for many years, appeared to be so close to each other, e.g.,

$$1 + dv \cong 3.67 \leq T \leq t + 2v \cong 3.78 \quad \text{for } d = 3. \quad (30)$$

Such simulations would clearly be valuable in verifying the assumptions made regarding the irrelevance of  $k$  and of  $q$  (or  $m$ ) when  $kb_0^2 \sim q \ll m$  (or  $kb_0^2 \sim m \ll q$ ) in 3D diluted elastic networks.

I am aware of only two experiments performed on 3D continuum percolating elastic systems:

(a) Using a set of solid beams, made by sintering a powder of submicron grains of metallic Silver, both the electrical conductivity and Young's modulus were measured over a range of volume fractions approaching the rigidity threshold *from above*.<sup>(30)</sup> The results were  $T = 3.8 \pm 0.5$ ,  $t = 2.15 \pm 0.25$ . Using  $\nu = 0.89 \pm 0.01$ ,<sup>(22)</sup> these results are of course consistent with the equality  $T = t + 2\nu$ . It is also clear that, due to the error bars, the results are considerably less tight than the theoretical bounds of Eq. (30).

(b) Using samples of a Silicon-based gel mixed with small particles of metallic Aluminum, both the dielectric constant and Young's modulus were measured over a range of volume fractions approaching the rigidity threshold *from below*.<sup>(31)</sup> By plotting Young's modulus vs. the dielectric constant, the need to measure volume fractions precisely was avoided. As a result of this, the ratio  $S/s$  could be obtained directly, without having to measure  $S$  and  $s$  separately. The result found was  $S/s = 0.80 \pm 0.05$ , which differs from the conclusion of the present study that this ratio should be 1. At present I can offer no explanation for this discrepancy. I hope that other researchers will now be motivated to perform similar experiments, and thus shed more light on this discrepancy.

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