

Phonon and fracton vibrational modes in disordered harmonic structures: A self-consistent theory

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The density of vibrational modes $N(\omega)$ of a harmonic lattice with bond or site disorder is calculated. We predict the existence in three dimensions of a crossover frequency $\omega_c \sim |c - c^*|^{3/2}$, where c^* is the critical concentration for percolation. For $c > c^*$, $N(\omega) \sim \omega^2$ if $\omega \ll \omega_c$ (phonon regime), and $N(\omega) \sim \omega^{1/3}$ if $\omega \gg \omega_c$ (fracton regime). This is the first analytical theory to predict a value of $\frac{1}{3}$ for the spectral dimension of a percolation cluster in three dimensions, in agreement with the conjecture of Alexander and Orbach and with recent simulations.

It has been suggested that the thermal properties of amorphous materials reflect the existence of a crossover between fractal excitations (fractons) at high frequencies and ordinary acoustic modes (phonons) at low frequencies.¹⁻³ A model system that is known to display fractal behavior over a range of length (frequency) scales is a prepercolation cluster.⁴⁻⁹ Below the critical percolation concentration, only finite clusters are formed. Such structures are self-similar for distance scales that are large compared to the lattice spacing, but small compared to the cluster size. At the critical point, an infinite cluster is formed, which is known to be self-similar for length scales large compared to the lattice spacing. Above the critical concentration, an infinite cluster exists, which is self-similar for a range of length scales that is large compared to the lattice spacing, but which becomes homogeneous over sufficiently large lengths. Let us consider a site or bond percolation system above the critical concentration in which the bonds are harmonic. At very small frequencies, the density of vibrational modes has the Debye form $N(\omega) \sim \omega^{d-1}$, at very large frequencies it reflects the details of the lattice structure, and at intermediate frequencies fractal dynamics obtain, and $N(\omega) \sim \omega^{d_s-1}$, where d_s is the spectral dimensionality.^{6,7}

We consider a lattice, characterized by either site or bond disorder, that is composed of N particles of equal mass that are connected by harmonic bonds. In the case of site disorder, a given lattice site is occupied by a particle with probability c and is unoccupied with probability $1 - c$. Two particles occupying nearest-neighbor sites are connected by a bond. In the bond-disorder problem, all lattice sites are occupied, and two particles occupying nearest-neighbor sites are connected by a bond with probability c and are not connected with probability $1 - c$. The harmonic bonds have identical force constants. The equation of motion of a particular realization of the random system is¹⁰

$$\frac{d^2 \mathbf{p}}{dt^2} = \mathbf{W} \cdot \mathbf{p}, \tag{1a}$$

$$W_{nm} = w_{nm} - \delta_{nm} \sum_j w_{jn}, \tag{1b}$$

where \mathbf{p} is an N -component vector whose n th element is

the displacement of the particle labeled n . w_{nm} is the ratio of the force constant of the bond connecting particles labeled m and n to the particle mass. For either type of disorder, w_{nm} is zero if particles m and n do not occupy nearest-neighbor lattice sites. In the site problem, $w_{nm} = w$, if particles m and n are nearest neighbors on the lattice, and in the bond problem, $w_{nm} = w$ with probability c , and $w_{nm} = 0$ with probability $1 - c$. The configuration-averaged density of vibrational modes $N(\omega)$ is given by¹⁰

$$N(\omega) = (-2\omega/\pi) \text{Im}[P_0(-\omega^2)], \tag{2a}$$

$$P_0(\varepsilon) = \int_0^\infty dt \exp(-\varepsilon t) \langle \text{[exp}(\mathbf{W}t)\text{]}_{11} \rangle. \tag{2b}$$

The angular brackets in Eq. (2b) denote a configuration average.

The relationship between the present problem and the determination of transport properties of a localized excitation in a disordered medium has been used by Alexander, *et al.* in their calculations of the density of vibrational modes of disordered, harmonic structures.¹⁰ Replacement of $d^2 \mathbf{p}/dt^2$ in Eq. (1a) by $d \mathbf{p}/dt$ yields the Pauli master equation for the motion of a localized excitation. In the transport problem, the n th element of \mathbf{p} is the probability that the excitation resides on the n th lattice site, and w_{nm} is the excitation transfer rate between sites m and n . $P_0(\varepsilon)$ is the Laplace transform of the configuration-averaged probability that an excitation which initially occupies a given site can be found at that site at a later time. We have recently developed a self-consistent mode-coupling procedure (SCMC) for calculating transport properties of excitations in disordered media.¹¹⁻¹³ This technique has been applied to the quantum percolation model,^{12(a)} to the Anderson model,^{12(b)} and to a model of incoherent motion that is based on the Pauli master equation.¹³ In this work, we extend the SCMC to the determination of $N(\omega)$. The SCMC is based on the following pair of coupled equations for $P_0(\varepsilon)$ and the diffusion kernel $D(\mathbf{k}, \varepsilon)$:

$$P_0(\varepsilon) = \Omega^{-1} \int d\mathbf{k} [\varepsilon + k^2 D(\mathbf{k}, \varepsilon)]^{-1}, \tag{3a}$$

$$D(\mathbf{k}, \varepsilon) \equiv \tilde{D}[\mathbf{k}, P_0(\varepsilon)] = \sum_{m=1}^\infty c^m \tilde{D}_m[\mathbf{k}, P_0(\varepsilon)]. \tag{3b}$$

The integration in Eq. (3a) is carried out over the first Brillouin zone. [$\Omega = (2\pi)^d/\Omega_0$, where Ω_0 is the volume of the unit cell.] Equation (3a) is an exact relation that follows from the definitions of P_0 and D . The first equality in Eq. (3b) is an ansatz that is made in order to provide a second independent relation between these two quantities. According to this ansatz, D is expressed as a functional of P_0 , which depends implicitly on ε through the dependence of P_0 on ε . \bar{D} depends explicitly on c as well as depending implicitly on c through the dependence of P_0 on c . The second equality in Eq. (3b) is an expansion of \bar{D} with respect to its *explicit* c dependence. Each term in the expansion depends on c to infinite order. Equation (3b) should be viewed as a partially resummed density expansion of D . In Appendix A of Ref. 12(a), we present a procedure by which \bar{D}_n can be uniquely determined from the coefficients of c^m for $m \leq n$ in the unresummed density expansion of $D(k, \varepsilon)$ and P_0 . Such coefficients can be calculated using the standard Mayer cluster expansion, as discussed in Appendix B of Ref. 12(a). We shall make use of an additional piece of information in obtaining an approximation to $\bar{D}(k, P_0)$. At $c=1$ (ordered lattice limit), $D(k, \varepsilon)$ can be determined exactly: $k^2 D(k, \varepsilon) = 2w[d - \sum_j \cos(k \cdot a_j)]$. a_j is the lattice vector in the direction j . Following the procedure described in Ref. 12(a), we construct a Padé approximant to $\bar{D}_1(k, P_0)$ in the variable $c/(1-c)$, which approaches $c\bar{D}_1(k, P_0)$ for $c \ll 1$, and which yields the exact result at $c=1$. (It should be noted that \bar{D}_1 has the same form for site and bond disorder. This is not the case for \bar{D}_n with $n > 1$.) With this approximation to \bar{D} , the SCMC equation for the d -dimensional analog of a simple cubic lattice with nearest-neighbor interactions is¹³

$$P_0(\varepsilon) = [2w_{\text{eff}}(\varepsilon)]^{-1} I_d[\varepsilon/2w_{\text{eff}}(\varepsilon)], \quad (4a)$$

$$w_{\text{eff}}(\varepsilon) = cw/[1 + 2(1-c)wP_0(\varepsilon)], \quad (4b)$$

$$I_d(x) = \pi^{-d} \int_0^\pi dq_1 \dots \int_0^\pi dq_d \left[x + d - \sum_{j=1}^d \cos(q_j) \right]^{-1}. \quad (4c)$$

Equations (4a) and (4b) are coupled equations for P_0 and $w_{\text{eff}}(\varepsilon)$, which is the long-wavelength ($k \rightarrow 0$) limit of the diffusion kernel $D(k, \varepsilon)$, divided by the square of the lattice spacing. $w_{\text{eff}}(\varepsilon)$ can be interpreted as an effective, frequency dependent force constant in the vibrational problem and as an effective transfer rate in the transport problem. Equation (4) is applicable to either site or bond percolation, if c is given the appropriate interpretation. Equation (4) is exactly correct to first order in c and at $c=1$ (the ordered lattice limit). I_d , which is defined in Eq. (4c), is the diagonal element of the lattice Green's function. The density of vibrational modes $N(\omega)$ for a disordered d -dimensional lattice is obtained by solving Eqs. (4a) and (4b) for $P_0(\varepsilon)$, setting $\varepsilon \rightarrow -\omega^2$, and substituting the result into Eq. (2a).

In this work, we shall focus on the physically relevant case of $d=3$. Before doing so, we shall briefly review the predictions of the SCMC for $d=1$ and $d=2$. Let us consider the solution of Eqs. (4) in the limit of small ε . The

behavior of this equation in d dimensions depends on the properties of $I_d(x)$, which is defined in Eq. (4c). The argument of I_d in Eq. (4c) is $\varepsilon/2w_{\text{eff}}$. For $c > c^*$, the critical concentration, we expect that w_{eff} approaches a finite limit as ε approaches zero, and hence that the argument of I_d becomes arbitrarily small for sufficiently small ε . For $c < c^*$, we expect w_{eff} to have the form $f(c)\varepsilon$ as ε approaches zero, where $f(c)$ is a function of c that diverges as c approaches c^* from below. Thus, in evaluating Eq. (4a) in the $\varepsilon \rightarrow 0$ limit for $c > c^*$ or c very close to but less than c^* , we need only consider the behavior of $I_d(x)$ for small values of x .^{14,15} In this limit, $I_1(x) = (2x)^{-1/2}$, $I_2(x) = (2\pi)^{-1} \ln(1/x)$, and $I_3(x) = I_3(0) - \pi^{-1}(x/2)^{1/2}$, where $I_3(0) \cong 0.5055$. Substitution of these expressions into Eq. (4a) yields a simplified pair of coupled equations for P and w_{eff} , that are valid for small frequencies and in the concentration regime described above. For $d=1$, this pair of equations yields the following solution for w_{eff} . For $c < 1$, $w_{\text{eff}} = [c/(1-c)]^2 \varepsilon$, and at $c=1$, $w_{\text{eff}} = w$. For a disordered linear chain, Eqs. (4) predict that the effective force constant vanishes in the zero-frequency limit for $c < 1$, and is finite at $c=1$. The percolation threshold is correctly predicted to occur at $c=1$ in one dimension. Equations (4) yield the following behavior for w_{eff} in the small-frequency limit in the vicinity of the percolation transition for $d=2$. For $c < 1$, $w_{\text{eff}} = (\varepsilon/2) \exp[2\pi c/(1-c)]$, and at $c=1$, $w_{\text{eff}} = w$. The percolation threshold is predicted to occur at $c=1$ in two dimensions. For a square lattice, the critical concentrations for site and bond percolation have been determined by computer simulation⁴ to occur, respectively, at $c^* \cong 0.593$ and $c^* \cong 0.500$.

Substitution of the limiting form of I_3 for small values of its argument into Eq. (4a), and of Eq. (4a) into Eq. (4b) yields a cubic self-consistent equation for $w_{\text{eff}}(\varepsilon)$ in $d=3$ that is valid for small frequencies in the vicinity of the critical point [$\varepsilon/w_{\text{eff}}(\varepsilon) \ll 1$].

$$w_{\text{eff}}[w_{\text{eff}}/w - (c - c^*)/(1 - c^*)]^2 = [(1 - c)/2\pi]^2 \varepsilon, \quad (5a)$$

$$c^* = I_3(0)/[1 + I_3(0)] \cong 0.3358. \quad (5b)$$

Inspection of the physically reasonable root of Eq. (5a) shows that c^* is a critical point. For $c < c^*$, $w_{\text{eff}}(\varepsilon) \sim (c^* - c)^{-2} \varepsilon$ in the limit of small ε , for $c > c^*$, $w_{\text{eff}}(\varepsilon) \sim w(c - c^*)/(1 - c^*)$ in this limit, and at $c = c^*$, $w_{\text{eff}}(\varepsilon) \sim \varepsilon^{1/3}$. The critical concentrations for site and bond percolation on a simple cubic lattice have been determined from simulations⁴ to be $c^* \cong 0.311$ and $c^* \cong 0.249$, respectively. $N(\omega)$ is determined from Eq. (5a) as follows. The physically reasonable solution of Eq. (5a) is obtained and the analytic continuation $\varepsilon \rightarrow -\omega^2$ is performed. Equation (4b) provides a simple relation between w_{eff} and P_0 . $P_0(-\omega^2)$ is determined by substituting $w_{\text{eff}}(-\omega^2)$ from Eq. (5a) into the left side of Eq. (4b). $N(\omega)$ is related to $P_0(-\omega^2)$ in Eq. (2a). We shall focus on the crossover from phonon to fracton behavior and shall therefore restrict our attention to $c \geq c^*$. For $c > c^*$, delocalized vibrations (acoustic modes) will exist on the infinite percolation cluster in the limit of small frequencies. For $c < c^*$, there is no infinite cluster, and low-frequency

modes are localized. The result for $c \geq c^*$ is

$$N(\omega) = \omega^{-1/2} \{3^{1/2}/[2(4\pi)^{1/3}]\} \\ \times [c/(1-c)^{5/3}] (\omega/\omega_c)^{1/3} F(\omega/\omega_c), \quad (6a)$$

$$F(y) = y^{2/3} \frac{\sinh Q \cosh Q}{(\frac{3}{4} \cosh^2 Q - \frac{1}{4} \sinh^2 Q)^2 + \frac{3}{4} \sinh^2 Q \cosh^2 Q}, \quad (6b)$$

$$Q = \frac{1}{3} \tanh^{-1} \{ [y^2/(y^2+1)]^{1/2} \}, \quad (6c)$$

$$\omega_c = \omega^{1/2} (4\pi/3^{3/2}) (1-c^*)^{-3/2} (1-c)^{-1} |c-c^*|^{3/2}. \quad (6d)$$

The scaling function $F(y)$, which is given in Eqs. (6b) and (6c) is proportional to $y^{5/3}$ for $y \ll 1$, and is independent of y for $y \gg 1$. In the limits $\omega \ll \omega_c$ and $\omega \gg \omega_c$, Eq. (6a) reduces to

$$N(\omega) = \begin{cases} \omega^{-2/3} [c/(1-c)^{5/3}] (16/\pi)^{1/3} (2/3^{5/2}) \omega_c^{-5/3} \omega^2, & \omega \ll \omega_c, \\ \omega^{-2/3} [c/(1-c)^{5/3}] (2\pi)^{-1/3} 3^{1/2} \omega^{1/3}, & \omega \gg \omega_c. \end{cases} \quad (7)$$

Inspection of Eq. (7) shows that ω_c is the crossover frequency between fractal and homogeneous dynamics. For $\omega \ll \omega_c$, $N(\omega) \sim \omega^2$, in agreement with the Debye density of modes for acoustic phonons in $d=3$. For $\omega \gg \omega_c$, $N(\omega) \sim \omega^{1/3}$, which corresponds to $d_s = \frac{4}{3}$. The crossover frequency ω_c vanishes as $|c-c^*|^{3/2}$. At $c=c^*$, $\omega_c=0$, and $N(\omega) \sim \omega^{1/3}$ for all values of ω . Alexander and Orbach⁷ have used a scaling argument to conjecture that $d_s = \frac{4}{3}$ for $d \geq 2$. Recent simulations of random walks on percolation clusters in $d=3$ by Ben-Avraham and Havlin,⁸ and by Argyrakis and Kopelman⁹ have yielded $d_s = 1.26 \pm 0.01$ and $d_s = 1.32 \pm 0.06$, respectively. This value is also consistent with experimental data. Orbach has pointed out that the neutron scattering measurements of the vibrational density of states of vitreous silica, carried out by Buchenau, Nucker, and Dianoux¹⁶ are consistent with $d_s \cong 1.4$. Thus, the value of d_s predicted by the SCMC in $d=3$ is in perfect agreement with the currently accepted value. The SCMC also yields a prediction of d_f , the fractal or Hausdorff dimension of a percolation cluster.¹⁷ The mean-squared displacement, $\langle r^2(t) \rangle$, for a random walk on a fractal⁵⁻⁷ grows at long times as t^{d_s/d_f} . Within the SCMC,¹¹⁻¹³ $\langle r^2(t) \rangle$ is proportional to the inverse Laplace transform of $\varepsilon^{-2} w_{\text{eff}}(\varepsilon)$. In the fractal regime $w_{\text{eff}}(\varepsilon) \sim \varepsilon^{1/3}$, which implies that $\langle r^2(t) \rangle \sim t^{2/3}$, and hence that $d_f=2$. This result is in good agreement with the current best estimate from scaling arguments of $d_f \cong 2.5$ in $d=3$. Alexander and Orbach⁷ have derived a scaling relation between d_f , d_s , ν (the correlation length exponent), and the exponent that characterizes the vanishing of ω_c as c approaches c^* , which is obeyed by the SCMC: $\omega_c \sim |c-c^*|^{3\nu/d_s}$. The exponent ν describes the divergence of ξ , the correlation length, which is related to a

characteristic cluster size: $\xi \sim |c-c^*|^{-\nu}$. The value of ν within the SCMC for $d=3$ can be obtained from the solution of Eq. (5a). For $c < c^*$, the Laplace transform of $\langle r^2(t) \rangle$ is proportional to $\varepsilon \xi^2$. The solution of Eq. (5a) for $c < c^*$ has the form $w_{\text{eff}}(\varepsilon) \sim (c^*-c)^{-2} \varepsilon$ in the limit of small ε , so $\nu=1$. Substitution of the SCMC values for ν , d_f , and d_s into the scaling relation leads to an exponent of $\frac{3}{2}$ for the crossover frequency, in agreement with Eq. (6d). The frequency dependence of $N(\omega)$, calculated from Eqs. (6), is shown in Fig. 1 for three values of c that are greater than $c^* \cong 0.3358$. The crossover from homogeneous (ω^2) to fractal ($\omega^{1/3}$) dynamics is explicitly shown, as is the vanishing of the crossover frequency as c approaches c^* from above.

We next turn to the predictions of the SCMC for $d > 3$. In order to analyze the solution of Eqs. (4a) and (4b) in the vicinity of c^* we need the behavior of I_d for small values of its argument. Analysis of the definition of I_d in Eq. (4c) shows that $I_d(x) \sim A_d + B_d x \ln(x)$, and that $I_d(x) \sim A_d + B_d x$ for $d > 4$, in the limit $x \ll 1$, where A_d and B_d are numbers. Substitution of the latter result into Eqs. (4a) and (4b) leads to a quadratic equation for w_{eff} for $d > 4$, in contrast to the cubic equation given in Eq. (5a) for $d=3$. Repetition of the above analysis shows that the SCMC predicts $d_f=2$ and $d_s=1$ for $d > 4$. For $d=4$, there is a logarithmic correction to the power-law behavior.

A crossover from homogeneous to fractal dynamics for a disordered harmonic network is also predicted by the effective-medium approximation (EMA),¹⁸⁻²⁰ which was applied to this problem by Derrida, Orbach, and Yu.² Like the SCMC, the EMA predicts the existence of a crossover frequency that vanishes at $c=c^*$. However, in $d=3$, the EMA predicts that $\omega_c \sim |c-c^*|$ and that $d_s=1$. The EMA and the SCMC agree in the prediction $d_f=2$. The EMA yields $\nu = \frac{1}{2}$, so that the scaling relation

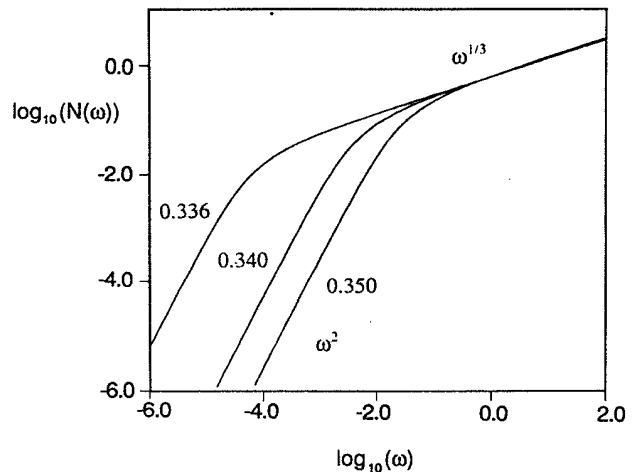


FIG. 1. $N(\omega)$, the density of vibrational modes, is calculated from Eqs. (6) for a disordered simple cubic lattice with $c=0.336, 0.340$, and 0.350 ; $\omega=1$; $c^* \cong 0.3358$. The crossover from homogeneous dynamics (ω^2) to fractal dynamics ($\omega^{1/3}$) is clearly illustrated.

of Orbach and Alexander is satisfied. Comparison of Fig. 1 of the present work to Fig. 1 of Ref. 2, shows that the SCMC and the EMA differ qualitatively in the behavior of $N(\omega)$ in the vicinity of ω_c . The transition from homogeneous to fractal behavior is very abrupt in the EMA, and $N(\omega)$ has an inflection point in the vicinity of ω_c . The SCMC does not predict a drastic change in $N(\omega)$ in the crossover region. The SCMC is the first theory of dynamics in disordered systems to yield the correct value

of d_s in $d=3$, and to allow the calculation of $N(\omega)$ for all frequencies and concentrations.

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