

Percolation in the effective-medium approximation: Crossover between phonon and fracton excitations

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The d -dimensional bond-percolating network has been examined with the use of the effective-medium approximation (EMA) of Odagaki and Lax and of Webman. We have found that the fracton dimensionality $\bar{d}=1$ for $2 < d < 4$, and have obtained explicit values for \bar{d} between $1 < d < 2$. We have calculated the vibrational density of states, $N(\omega)$, for percolating networks within the EMA for d in these ranges. We find at $2 < d < 4$ that a steep change in $N(\omega)$ takes place between phonon, $N_{\text{ph}}(\omega)$, and fracton, $N_{\text{fr}}(\omega)$, excitation regimes at a critical frequency ω_c which scales as $p - p_c$. The ratio $N_{\text{fr}}(\omega_c)/N_{\text{ph}}(\omega_c)$ is found to scale as $(p - p_c)^{1-d/2}$. These results provide substantial support for the fracton interpretation of the thermal properties of epoxy resin, glasses, and neutron-irradiated quartz as hypothesized by Alexander, Laermans, Orbach, and Rosenberg. At $d=2$, the transition between the two regimes is smoother (logarithmic), but a clearly defined phonon and fracton regime can be ascertained. The velocity of sound in the phonon regime scales as $(p - p_c)^{1/2}$, independent of d for $2 < d < 4$. Finally, we have obtained within the EMA a closed expression for the mean-square diffusion length $\langle R^2(t) \rangle$ for all times of order $(p - p_c)^{-2}$. It is found to be a smooth function of time between the fractal and homogeneous diffusion regimes.

I. INTRODUCTION

A recent assertion¹ claimed that the thermal properties of epoxy resin, glasses, and neutron-irradiated quartz could be understood on the basis of fracton excitations above, and phonon excitations below, a crossover frequency ω_{co} . This treatment assumed a continuous crossover in the dispersion spectra of phonons and fractons (more specifically, that the force constant and mass scaled independently and continuously between the two regimes). This led to the prediction of a sudden drop in the density of vibrational states $N(\omega)$ at ω_{co} by an amount \bar{d}/d , where \bar{d} is the fracton dimensionality² and d the Euclidean dimensionality. A more recent analysis³ claimed an overlap of parts of the phonon and fracton spectra, in particular that the dispersion law itself was discontinuous at a characteristic crossover length (corresponding to ω_{co} in the work of Ref. 2). This led to a sudden increase in $N(\omega)$ at the lowest frequency of phonon-fracton overlap. Agreement with the thermal measurements on epoxy resin by Kelham and Rosenberg⁴ and glasses by Zaitlin and Anderson⁵ was satisfactory with the latter approach, but reasons for the discontinuity in the dispersion law were lacking.

A simulation of a physical system which exhibited both phonon and fracton excitations was necessary to choose between the two theoretical models. The percolating network provides an ideal example of such a system because the thermal excitations at low frequencies are certainly phononlike, whereas those at higher frequencies are known to be fractonlike.^{6,7} The crossover occurs at length scales equal to ξ_p , the percolation correlation length.

This paper contains the results of an effective-medium

approximation (EMA) treatment of the bond-percolation network on a (Euclidean) d -dimensional hypercubic lattice. We use the formulation of Odagaki and Lax⁸ and of Webman.⁹ Specifically, we calculate the Laplace transform of the autocorrelation function $\bar{P}(\vec{s}, u | \vec{s})$ for diffusion in terms of the spectral parameter u and spatial position \vec{s} . The density of states for vibrational excitations is obtained by allowing $u \rightarrow -\omega^2$ and taking the imaginary part of $\bar{P}(\vec{s}, -\omega^2 | \vec{s})$. Surprisingly, though $\bar{P}(\vec{s}, u | \vec{s})$ is a smooth function of u for positive u [said another way, $\langle R^2(t) \rangle$ behaves smoothly as a function of time t (see our explicit evaluation in Sec. IV)], $N(\omega)$ will be shown to exhibit a steep change as a function of ω for the range of Euclidean dimensions we have investigated, $2 < d < 4$. We have verified this behavior both numerically and analytically (see Sec. III below). Briefly, our results are in fact consistent with the general form for $N(\omega)$ proposed by Tua *et al.*,³ exhibiting an increase in $N_{\text{fr}}(\omega)/N_{\text{ph}}(\omega)$ at $\omega = \omega_c$ for $2 < d < 4$, where the subscripts fr and ph designate fracton and phonon, respectively. We identify our ω_c with the crossover frequency ω_{co} of Ref. 3. The critical frequency ω_c at which the crossover takes place will be shown to scale as $p - p_c$, while the ratio $N_{\text{fr}}(\omega)/N_{\text{ph}}(\omega)$ scales as $(p - p_c)^{1-d/2}$ at ω_c . In addition, $N_{\text{fr}}(\omega)$ is constant away from ω_c , implying that $\bar{d}=1$ for $2 < d < 4$. This result is not too far from the result $\bar{d} = \frac{4}{3}$, $2 \leq d$, conjectured by Alexander and Orbach² for percolating networks. We see the EMA may be a reasonable model for the physical behavior of a random system, while not generating precise values for the critical exponents.

This paper presents the basic EMA equations in Sec. II. Section III provides a discussion of their significance for

the vibrational excitation problem, and makes explicit the identification of the phonon-fracton regimes. A numerical solution for $N(\omega)$ at $d=3$ and 2 is exhibited, as well as analytical expressions valid for finite ω/ω_c . Section IV contains an explicit evaluation of $\langle R^2(t) \rangle$ within the EMA. It is found that a crossover takes place at a time t precisely equal to ω_c^{-2} between two different time regimes (homogeneous and fractal), but that the transition is smooth, in contrast to the density of vibrational states. Section V contains a discussion of the results of this paper, and presents some concluding remarks.

II. EMA FOR PERCOLATING NETWORKS

The details of the EMA for percolating networks are outlined in great detail in Refs. 8 and 9. We reproduce their results in this section in order to establish notation, and to give a sense of the character of their derivation. The reader is referred to these two excellent references for further details. We assume a diffusing particle is at the position \vec{s}_0 at time $t=0$, and that the probability density for finding it at \vec{s} at time t is given by the master equation:

$$\frac{\partial P(\vec{s}, t | \vec{s}_0)}{\partial t} = -\Gamma_{\vec{s}} P(\vec{s}, t | \vec{s}_0) + \sum_{\vec{s}' (\neq \vec{s})} W_{\vec{s}, \vec{s}'} P(\vec{s}', t | \vec{s}_0), \quad (1)$$

$$Q = (1/d\bar{W}) \left[1 - (1/2\pi)^d \int_0^{2\pi} \cdots \int_0^{2\pi} \frac{u dq_1 \cdots dq_d}{u + 2\bar{W} \left[\sum_{i=1}^d (1 - \cos q_i) \right]} \right]. \quad (5)$$

Our task is therefore to calculate \bar{W} as a function of p and u , to obtain $\bar{P}(\vec{s}, u | \vec{s})$. We then obtain $N(\omega)$ by setting¹⁰

$$N(\omega) = -(2/\pi)\omega \operatorname{Im}[\bar{P}(\vec{s}, -\omega^2 | \vec{s})]. \quad (6)$$

This completes the formal aspect of the EMA treatment. It is important, however, to recognize some specific points. First, at $p=p_c$, one is entirely within the fracton regime because ξ_p is infinite, and all length scales are therefore less than ξ_p . We shall show in the next section that $\bar{P}(\vec{s}, u | \vec{s}) \sim 1/u^{1/2}$ for $2 < d < 4$ at $p=p_c$, leading to $N(\omega) = \text{const}$ from Eq. (6). This is interpreted by us in the same spirit as Ref. 2, which notes that $N(\omega) \sim \omega^{\bar{d}-1}$ in the fracton regime. Thus, $\bar{d}_{\text{EMA}} = 1$, $2 < d < 4$. The value conjectured by Alexander and co-workers^{2,11} is $\bar{d} = \frac{4}{3}$, $2 \leq d$. Second, for $p > p_c$, one encounters both phonon and fracton excitations, depending on the excitation length scale. Hence, at very small ω , the excitations must be phononlike, while at larger (but still perhaps small) ω , the excitations will be fractons (note that within the EMA there is no distinction between the finite and infinite clusters). The advantage of using percolation theory is that one can control the magnitude of the frequency ω_c , which separates the phonon and fracton regimes, by mere-

where $\Gamma_{\vec{s}} = \sum_{\vec{s}'} W_{\vec{s}, \vec{s}'}$. We work in the symmetric case (e.g., zero-electric-field limit) where $W_{\vec{s}, \vec{s}'} = W_{\vec{s}', \vec{s}}$. For bond percolation, the $W_{\vec{s}, \vec{s}'}$ obey the probability density

$$P(W_{\vec{s}, \vec{s}'}) = p\delta(W_{\vec{s}, \vec{s}'} - W_0) + (1-p)\delta(W_{\vec{s}, \vec{s}'}). \quad (2)$$

Then, within the EMA, Refs. 8 and 9 show that the Laplace transform of the autocorrelation function for diffusion on the percolating network is given by

$$\bar{P}(\vec{s}, u | \vec{s}) = (1/2\pi)^d \times \int_0^{2\pi} \cdots \int_0^{2\pi} \frac{dq_1 \cdots dq_d}{u + 2\bar{W} \left[\sum_{i=1}^d (1 - \cos q_i) \right]} \quad (3)$$

for a hypercubic lattice in Euclidean dimension d . Here \bar{W} is the effective conductivity at frequency $\Omega = iu$, and \bar{W} and u are measured in units of W_0 .⁹ Therefore, $\sigma(\Omega) = d\bar{W}(iu)$, where \bar{W} depends on u and is given by the solution of

$$\bar{W}^2 - \bar{W} - Q\bar{W} + p = 0. \quad (4)$$

The function Q is itself a function of u and \bar{W} :

ly adjusting $p - p_c$. That is, $\xi_p \sim (p - p_c)^{-\nu}$, so that ξ_p can be made arbitrarily large as $p \rightarrow p_c$. Hence, ω_c can be made as small as one wishes by allowing $p \rightarrow p_c$ [Ref. 2 obtains the relationship $\omega_c \sim \xi_p^{-(1+\theta/2)}$ where θ represents the range dependence of the effective diffusion constant, $D(r) \sim r^{-\theta}$, in the fractal regime¹²]. We shall be able to exploit this property in the next section when we expand \bar{P} and \bar{W} in powers of ω in the vicinity of ω_c .

Combining these ideas, we have some useful checks on our results in the next section. (1) $N(\omega)_{p > p_c}$ must tend to a constant, $N(\omega)_{p=p_c}$, for ω greater than ω_c . (2) $N(\omega)_{p > p_c}$ must be proportional to the usual phonon density of states, ω^{d-1} , for ω less than ω_c . That there may be a steep change in $N(\omega)$ at ω_c will be shown for $d \geq 2$, but at this stage it is by no means obvious.

III. DISCUSSION OF THE EMA RESULTS

Equations (4) and (5) are coupled, and very difficult to solve in general. We present first an approximate numerical solution for $d=3$ and 2 in order to exhibit the general form for $N(\omega)$ explicitly. We then use insights from the numerical work to develop a number of exact analytic expansions for the entire range of values of u such that

$u/(p-p_c)^2$ is finite. In the EMA, $p_c = 1/d$.

First, in three dimensions, the integrals in Eqs. (3) and (5) are very complicated for any u . We follow an approximation used by Odagaki and Lax⁸ for the denominator of Eq. (5) [see Eq. (4.7) of their paper]:

$$\bar{P}_{\text{approx}}(\vec{s}, u | \vec{s}) = 2\{u + 2d\bar{W} + [u(u + 4d\bar{W})]^{1/2}\}^{-1}. \quad (7)$$

Using Eq. (6), we are now able to obtain numerical solutions for $N(\omega)$. We display our results in Fig. 1 for $d=3$ and different $p-p_c$. The form is surprising.

One finds, as expected, a phonon regime for $\omega < \omega_c$, and a constant $N(\omega)$ in the fracton regime ($\omega > \omega_c$). However, instead of a "smooth" crossover between the two regions, a steep change appears in $N(\omega)$ (defining ω_c). We shall show in Sec. IV that the related quantity $\langle R^2(t) \rangle$, the mean-square distance that a particle diffuses in a time t , is a very smooth function of time, and does indeed possess a smooth crossover between fractal and homogeneous behavior at a time $t = 1/\omega_c^2$. The difference in behavior for the two quantities will be shown to rest with the effects of the substitution of $-\omega^2$ for u . One sees the two distinct features discussed earlier in Fig. 1. First, $N(\omega) \sim \omega^2$ as $\omega \rightarrow 0$. This is just the phonon regime since $d=3$ and $\omega < \omega_c$. At $\omega > \omega_c$, the density of states $N(\omega)$ tends to a constant of order unity. This is the fracton regime, implying the fracton dimensionality $\bar{d}=1$ [since $N(\omega) \approx \omega^{\bar{d}-1}$ in the fracton regime, according to Ref. 2]. The transition between the two regimes is abrupt. The change in $N(\omega)$ is in the direction favored by Tua *et al.*,³ and therefore consistent with a fracton interpretation of the vibrational properties of amorphous materials.^{4,5} The

steep change in $N(\omega)$ allows us to define a critical frequency ω_c quite precisely. Numerically (from Fig. 1), we find

$$\omega_c = 0.58[(p-p_c)/p_c] = 1.8(p-p_c).$$

Note that the magnitude of the change between the phonon density of states extrapolated to ω_c from the phonon side, $N_{\text{ph}}(\omega_c)$, and the fracton density of states extrapolated to ω_c from the fracton side, $N_{\text{fr}}(\omega_c)$, diminishes with increasing $p-p_c$. We shall show that the ratio $N_{\text{fr}}(\omega_c)/N_{\text{ph}}(\omega_c)$ scales with $(p-p_c)^{1-d/2}$ when we treat $N(\omega)$ analytically. This behavior is clearly followed in Fig. 1.

In two dimensions, we have carried out an analogous self-consistent evaluation of the two equations (4) and (5). The results are plotted for $N(\omega)$ in Fig. 2. The results are different from those at $d=3$. The phonon and fracton regimes in $d=2$ are seen to merge smoothly into one another. The ratio $N_{\text{fr}}(\omega_c)/N_{\text{ph}}(\omega_c)$ now appears to be independent of $p-p_c$, consistent with the scaling law mentioned above.

It is now worthwhile to probe the analytic character of the structure of these solutions. As a consequence, we shall obtain better insight into the physical content of the problem.

Glancing at Fig. 1, we see that that we can make ω_c as small as we wish by allowing $p \rightarrow p_c$ as close as we please, and that ω_c is linear in $p-p_c$. We are now going to describe the region of values for u such that $u/(p-p_c)^2$ remains finite. This will lead us to the (consistent) observation that $\bar{W}/(p-p_c)$ will remain finite as $p \rightarrow p_c$. If this be true, then in the vicinity of p_c , $u/\bar{W} \ll 1$. This

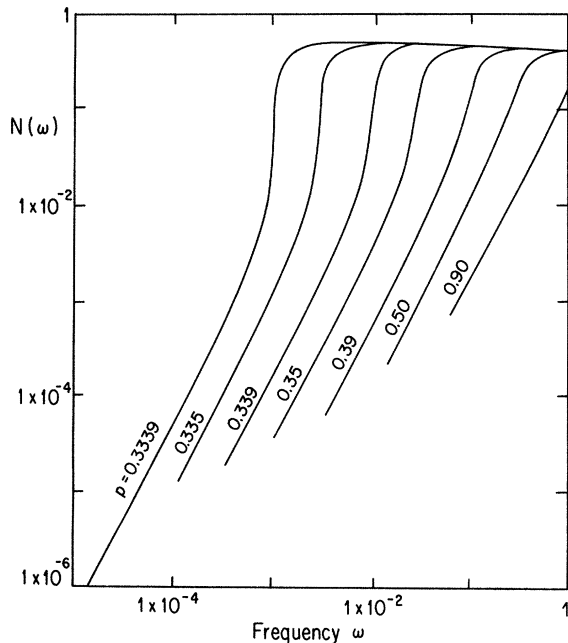


FIG. 1. Vibrational density of states for the simple-cubic lattice percolating network, $d=3$, as a function of frequency ω , calculated within the EMA for differing bond densities p . Here $p_c = \frac{1}{3} = 0.33333 \dots$

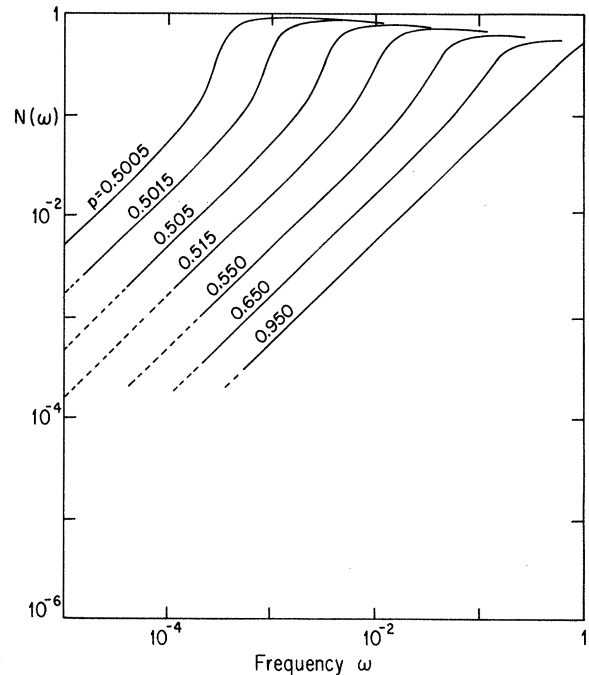


FIG. 2. Vibrational density of states for the simple-square lattice, percolating network, $d=2$, as a function of frequency ω , calculated within the EMA, for differing bond densities p . Here $p_c = \frac{1}{2} = 0.5$.

provides us with an expansion parameter. Expanding Q in powers of u/\bar{W} , for $2 < d < 4$, one finds

$$Q = (1/\bar{W}d)[1 - (u/2\bar{W})\psi_d + (u/\bar{W})^{d/2}\chi_d + \dots], \quad (8)$$

where ψ_d and χ_d are real numbers given by

$$\psi_d = (1/2\pi)^d \int_0^{2\pi} \dots \int_0^{2\pi} \frac{dq_1 \dots dq_d}{\sum_{i=1}^d (1 - \cos q_i)}, \quad (9a)$$

$$\chi_d = S_d (1/2\pi)^d \int_0^\infty x^{d-1} dx / [x^2(x^2 + 1)], \quad (9b)$$

where S_d is the surface area of the unit sphere in d dimensions [$S_2 = 2\pi$, $S_3 = 4\pi$, and in general $S_d = 2(\pi)^{d/2}/\Gamma(d/2)$]. For $d=3$, the quantities in Eq. (9) have been evaluated⁸ to give $\psi_3 = 1.51638/3$, $\chi_3 = 1/4\pi$.

For $1 < d < 2$, one finds

$$Q = (1/d\bar{W})[1 - (u/\bar{W})^{d/2}\tilde{\chi}_d], \quad (10)$$

where

$$\tilde{\chi}_d = (1/2\pi)^3 S_d \int_0^\infty x^{d-1} dx / (x^2 + 1). \quad (11)$$

Now that we have obtained Q , we need to evaluate \bar{W} from Eq. (4). We restrict ourselves to $2 < d < 4$ for the moment, and expand \bar{W} in powers of $p - p_c$ to order $(p - p_c)^{d/2}$. We find

$$\bar{W}/d - \bar{W} - 1/d + (\psi_d/2\bar{W}d)u - (\chi_d/d)(u/\bar{W})^{d/2} + p = 0. \quad (12)$$

We obtain $\bar{W} = W_0(p, u) + W_1(p, u) + \dots$, where $W_0 \sim p - p_c$ and $W_1 \sim (p - p_c)^{d/2}$. Explicitly,

$$W_0 = [d/2(d-1)]\{(p - p_c) + [(p - p_c)^2 + 2u(d-1)\psi_d/d^2]^{1/2}\} \quad (13a)$$

and

$$W_1 = - \frac{\frac{\chi_d}{d} \left[\frac{u}{W_0} \right]^{d/2}}{\left[1 - \frac{1}{d} \right] + \frac{\psi_d u}{2dW_0^2} - \frac{\chi_d}{2} \frac{u^{d/2}}{W_0^{d/2+1}}}. \quad (13b)$$

Note that we shall be setting $u = -\omega^2$. Hence, from the form of Eq. (13) it is clear that structure may occur in the vicinity of

$$u = -d^2(p - p_c)^2/2(d-1)\psi_d. \quad (14)$$

Glancing at Eq. (13), we see that because $\bar{W} \sim p - p_c$, and our assumption that $u/(p - p_c)^2$ is finite, $W_1 \sim (p - p_c)^{d/2}$ consistently. Hence, $\bar{W} \sim p - p_c$, as hypothesized. Thus, returning to Eq. (3), we can evaluate $\bar{P}(\vec{s}, u | \vec{s})$. Because $u/\bar{W} \ll 1$, we have

$$\bar{P}(\vec{s}, u | \vec{s}) = \psi_d/2\bar{W} - u^{d/2-1}\chi_d/\bar{W}^{d/2} + \dots$$

Using the expansion (13) we find

$$\bar{P}(\vec{s}, u | \vec{s}) = (\psi_d/2W_0)\{1 + (\chi_d/d)(u/W_0)^{d/2}[(1 - 1/d)W_0 + \psi_d u/2dW_0]^{-1}\} - u^{d/2-1}\chi_d/W_0^{d/2} + \dots \quad (15)$$

It is clear that ω_c defined by

$$\omega_c = [d^2/2(d-1)\psi_d]^{1/2}(p - p_c) \quad (16)$$

is a zero of the square root found in the expressions for W_0 [Eq. (13a)] and $\bar{P}(\vec{s}, u | \vec{s})$ [Eq. (15)]. This should be compared with the numerically determined value presented for $d=3$: $\omega_c \cong 1.8(p - p_c)$. At $d=3$, Eq. (16) reduces to $\omega_c = 2.1(p - p_c)$. The difference between these results is due to the approximate expression for $\bar{P}(\vec{s}, u | \vec{s})$ used in the numerical calculation. Nevertheless, the proportionality of ω_c to $p - p_c$ is the same for both approaches.

It is clear that for $d > 2$, ω_c separates two distinct regions. Consider first the fracton regime ($\omega > \omega_c$). Then the imaginary part of \bar{W} is linear in $p - p_c$. Because the imaginary part of $\bar{P}(\vec{s}, -\omega^2 | \vec{s})$ is proportional to the density of states $N(\omega)$, we find that

$$\bar{P}(\vec{s}, -\omega^2 | \vec{s}) = [\psi_d(d-1)/d(p - p_c)]\{1 - i[(\omega/\omega_c)^2 - 1]^{1/2}\}/(\omega/\omega_c)^2. \quad (17)$$

Using Eqs. (16) and (6), we obtain from Eq. (17),

$$N_{fr}(\omega) = \begin{cases} (2/\pi)[\psi_d(d-1)/2]^{1/2}[(\omega^2 - \omega_c^2)]^{1/2}/\omega, & \omega > \omega_c \\ (2/\pi)[\psi_d(d-1)/2]^{1/2}[1 - \frac{1}{2}(\omega_c/\omega)^2], & \omega \gg \omega_c \end{cases} \quad (18a)$$

Consider now the second or phonon regime, $\omega < \omega_c$. The leading order [of order $(p - p_c)^{-1}$] does not contribute, and one must go to the next higher-order term. We find

$$N_{ph}(\omega) = (2/\pi)\sin[(d/2 - 1)\pi][(d-1)/2\psi_d]^{d/4}\chi_d[\omega^{d-1}/(\omega_c)^{d/2}], \quad \omega \ll \omega_c. \quad (19)$$

Equations (18b) and (19) are our principal results away from ω_c .

For $\omega < \omega_c$ we find

$$N_{ph}(\omega) = (2/\pi)\omega^{d-1}(\chi_d/W_0^{d/2})\sin[\pi(d/2 - 1)]\{1 - (\omega^2/2dW_0)[(1 - 1/d)W_0 - \omega^2\psi_d/2dW_0]^{-1}\}. \quad (20)$$

One sees that, because W_0 is singular at ω_c , Eqs. (18a) and (20) are singular at ω_c . However, as one can see from Fig. 1, the transition between the phonon and fracton $N(\omega)$ regimes is steep but smooth. This occurs because of the following argument. The expressions contained in Eqs. (18b) and (20) do not mean that, for $p - p_c$ finite, $N(\omega)$ has a singularity at $\omega = \omega_c$. These expressions are only valid in the limit $p \rightarrow p_c$, $\omega^2/(p - p_c)$ finite. Indeed, one sees clearly from Fig. 1 that as $p \rightarrow p_c$ the crossover between the two regimes becomes steeper and steeper.

One should note that the expansion $\bar{W} = W_0 + W_1 + \dots$ for the solution of Eq. (4) is valid only for $\omega \neq \omega_c$. At $\omega = \omega_c$, Eq. (4) has a double solution to leading order in $p - p_c$. Therefore, one must go to higher order to describe the density of states analytically in the immediate vicinity of ω_c .

These are our primary analytic results for $d > 2$. One sees clearly that $N_{fr}(\omega)$, as ω moves away from ω_c , $\omega > \omega_c$, approaches a constant,

$$N_{fr}(\omega \gg \omega_c) = (2/\pi)[\psi_d(d-1)/2]^{1/2},$$

remembering that ω must remain small (this is all possible as long as one is not too far away from $p - p_c$). If we make the Alexander-Orbach interpretation of the fracton density of states,² $N_{fr}(\omega) \sim \omega^{\bar{d}-1}$, we can interpret this limit by setting

$$\bar{d}_{EMA} = 1, \quad 2 < d < 4.$$

This result strays from the conjecture² that $\bar{d} = \frac{4}{3}$, $2 \leq d$ for percolating networks, but is consistent in the sense that it holds for all $2 < d < 4$. It is quite common for the EMA to give the correct physical properties of disordered systems, but to yield incorrect critical exponents. We suggest that this is occurring for percolation too.

The phonon regime is certainly correct for the frequency dependence of $N_{ph}(\omega)$. It is also interesting to note that since in the homogeneous limit $N_{ph}(\omega) \sim \omega^{d-1}/v_s^d$, one can extract the $p - p_c$ dependence of the velocity of sound v_s . This variation was taken into account in the work of Tua *et al.*,³ but no explicit exponent was given. From Eq. (19)

$$v_s \sim \omega_c^{1/2} \sim (p - p_c)^{1/2}. \quad (21)$$

This factor is, of course, responsible for the shift in position of the plot of $N(\omega)$ versus ω in Fig. 1 in the phonon ($\omega < \omega_c$) region with increasing $p - p_c$.

Finally, it is interesting to determine the size of the increase in $N(\omega)$ at ω_c . We take $N_{ph}(\omega)$ from Eq. (19) and extrapolate the power dependence ω^{d-1} to ω_c^{d-1} , $\omega < \omega_c$. In an analogous manner, we take the "high"-frequency limit of $N_{fr}(\omega)$, and extrapolate the constant value to ω_c , $\omega > \omega_c$. The ratio of these two limits gives us ($2 < d < 4$)

$$N_{fr}(\omega)/N_{ph}(\omega) |_{\omega \rightarrow \omega_c} \sim \omega_c^{1-d/2} \sim (p - p_c)^{1-d/2}. \quad (22)$$

At $d = 3$, the extrapolated ratio given by Eq. (22) is proportional to $(p - p_c)^{-1/2}$. This can be clearly seen in the numerical results exhibited in Fig. 1.

An expansion similar to that which led to Eq. (17), but for $1 < d < 2$, can be made using the relationship Eq. (10). One finds now that at p_c , and for the higher frequencies

for $p > p_c$,

$$N_{fr}(\omega) \sim \omega^{(d-2)/(d+2)}, \quad 1 < d < 2 \quad (23)$$

or

$$\bar{d}_{EMA} = 2d/(d+2), \quad 1 < d < 2. \quad (24)$$

The low-frequency behavior has the same form as Eq. (19), viz.,

$$N_{ph}(\omega) \sim \omega^{d-1}/(p - p_c)^{d/2}.$$

At $d = 2$, logarithmic terms enter so that the explicit expressions for $N_{ph}(\omega)$ and $N_{fr}(\omega)$ become much more complex than Eqs. (18) and (19) for $d > 2$. Nevertheless, a glance at Fig. 2 does seem to show that the inflection point in $N(\omega)$ occurs at the same $N(\omega)$ value, independent of $p - p_c$. Such a prediction is in accord with Eq. (22).

The steep change in $N(\omega)$ at ω_c is important because it is also the feature that is required for the analysis of the specific-heat data of epoxy resin by Kelham and Rosenberg⁴. A glance at their Fig. 4 and a comparison with Fig. 1 of this paper show the compatibility of the experimental and EMA results. This, of course, had been anticipated by Tua *et al.*³ in their phenomenological analysis of the epoxy-resin data of Kelham and Rosenberg. The interest in the EMA result is that it finally adds a relative firm foundation to the supposition of Alexander *et al.* that the higher-frequency vibrational properties of epoxy resin can be usefully described by fracton excitations. The implications are quite exciting, for Alexander *et al.*¹ also analyzed the thermal properties of glass and neutron-irradiated quartz. The results of this paper suggest that the similar thermal behavior of nearly all amorphous systems¹³ may be describable by the density of states of Tua *et al.*,³ and that the supposition of Alexander *et al.*,¹ [above a "crossover frequency" ω_{co} , fracton excitations enter into the vibrational spectra of epoxy resin and glasses] may apply to nearly all amorphous materials. In terms of a characteristic length scale ξ , one can make an equivalent distinction: for length scales shorter than ξ , the vibrational excitations are of fracton character. For percolation, $\xi = \xi_p$. Further evidence for the features in the vibrational dispersion law posited by Tua *et al.*³ can be found in a recent EMA analysis by Entin-Wohlman *et al.*¹⁴ for the "dispersion law" for a percolating network.

IV. EVALUATION OF $\langle R^2(t) \rangle$

The "ant" problem of de Gennes¹⁵ can also be solved within the EMA. The Laplace transform of the mean-square distance that a particle diffuses on a d -dimensional percolating network in a time t has been exhibited by Odagaki and Lax. They show $\langle R^2(u) \rangle = 2d\bar{W}(u)/u^2$. To leading order, we can use Eq. (13a) for $\bar{W}(u) = W_0$ to obtain

$$\begin{aligned} \langle R^2(u) \rangle &= [d^2/u^2(d-1)] \\ &\times \{ (p - p_c) + [(p - p_c)^2 + 2u(d-1)\psi_d/d^2]^{1/2} \}. \end{aligned} \quad (25)$$

This expression holds for all $p - p_c$ (positive or negative). Looking at the argument of the square root, we are naturally led to a characteristic time t_0 given by

$$t_0 = 2(d-1)\psi_d/d^2(p-p_c)^2. \quad (26)$$

We shall see that indeed this corresponds to the crossover time for diffusion: for $t < t_0$, diffusion is in a fractal space, while for $t > t_0$, diffusion is in a homogeneous space. The mean-square distance traversed by the ant in the time t_0 will therefore be designated as the square of the crossover length (the percolation-correlation length ξ_p ,

$$\langle R^2(t) \rangle = [d^2(p-p_c)/(d-1)] \left[t \pm (2t_0/\sqrt{\pi}) \int_0^{(t/t_0)^{1/2}} e^{-u^2}(t/t_0 - u^2 + 1) du \right], \quad (27)$$

where \pm refers to $p \lesseqgtr p_c$. The expression for Eq. (27) is valid only for $2 < d < 4$ because of the use of Eq. (13a).

The expression for $\langle R^2(t) \rangle$ can be written quite naturally as a scaling function. Explicitly,

$$\langle R^2(t) \rangle = [d^2(p-p_c)/(d-1)] t_0 f(t/t_0), \quad (28)$$

where

$$f(x) = x \pm (2/\sqrt{\pi}) \int_0^{\sqrt{x}} (x+1-u^2)e^{-u^2} du. \quad (29)$$

For convenience, we have plotted $|f(x)|$ against x in Fig. 3. The short- and long-time limits (small and large x) for $\langle R^2(t) \rangle$ [for $f(x)$] are easily found: For $t \ll t_0$,

$$\langle R^2(t) \rangle = [2d^2/(d-1)\sqrt{\pi}] [2(d-1)\psi_d/d^2]^{1/2} \sqrt{t}, \quad (30a)$$

and for $t \gg t_0$,

$$\langle R^2(t) \rangle = \begin{cases} [2d^2/(d-1)](p-p_c)t, & p > p_c \\ \psi_d/(p_c-p), & p < p_c \end{cases} \quad (30b)$$

$$(30c)$$

Apart from the value of the critical exponents, the results of Eq. (30) are entirely consistent with the scaling solution of the ant problem of Gefen *et al.*¹² At short times (of

referred to at the end of Sec. III).

Because there can be only one length scale in the problem (apart from the interatomic spacing), Eq. (26) must also bear some relation to ω_c . Remarkably, as stated in the Introduction, t_0 is just the inverse square root of the crossover frequency ω_c [see the explicit expression for ω_c given by Eq. (16)]. This is because $t \sim 1/u \sim -1/\omega^2$ upon analytic continuation.

After considerable effort, we were able to invert Eq. (25) into the time domain (i.e., take the inverse Laplace transform). The result is the following:

course, the EMA does not take explicitly into account the finite clusters), diffusion is in a fractal space. The precise exponent for t depends upon whether one averages over finite clusters or not (see Ref. 12). The EMA result exhibits a square-root time dependence, and hence an infinite slope in $\langle R^2(t) \rangle$ versus t at $t=0$. All scaling results exhibit this behavior. The crossover at t_0 sets the time scale (and hence length scale) for the problem. We see that for $p < p_c$, crossover from the fractal to homogeneous regime results in a saturation of $\langle R^2(t) \rangle$. It is remarkable that even within the EMA one can obtain results which indicate a "maximum" cluster size [i.e., a "saturation" in $\langle R^2(t) \rangle$ for t large ($t \gg t_0$) for $p < p_c$]. We note in conclusion that Sahimi *et al.*¹⁶ have also evaluated $\langle R^2(t) \rangle$ in the limit $t \rightarrow \infty$ at p_c for $p > p_c$ in the EMA. Our results restricted to these cases agree with theirs.

V. DISCUSSION AND CONCLUSION

We have used the EMA to obtain the vibrational density of states of a percolating network of identical masses and springs. Quite remarkably, for a solution which in the diffusion regime exhibits no sharp structure in, for example $\langle R^2(t) \rangle$, one finds, upon analytically continuing as appropriate for the vibrational problem, a steep change in the vibrational density of states $N(\omega)$ at a critical frequency ω_c (i.e., compare Fig. 3 with Fig. 1) for $2 < d < 4$. We interpret this to be an expression of a rapid transition between pure phononlike excitations for $\omega < \omega_c$, to fractonlike excitations for $\omega > \omega_c$. Remarkably, at or near the percolation threshold, for $2 < d < 4$, $N(\omega)$ is independent of d , and can be associated with a fracton dimensionality² $\bar{d}=1$, not far from that conjectured by Alexander and Orbach² for $d \geq 2$, $\bar{d} = \frac{4}{3}$ for percolating networks.

It would be very interesting to know if the steep change in the density of states $N(\omega)$ is just an effect of the EMA, or if it remains beyond the approximation. The most obvious approach would be, of course, to calculate directly the density of states for vibrations on a percolating network. This seems very difficult to do except by Monte Carlo methods, and then only with very small samples. Another way of solving the problem would be to use a generalized EMA (Ref. 16), or to study the case of

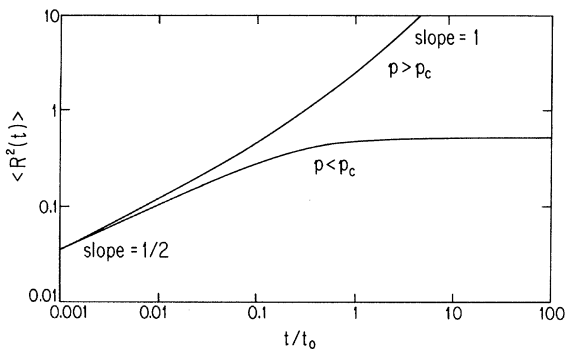


FIG. 3. Mean-square distance $\langle R^2(t) \rangle$ diffused by a particle on a hypercubic lattice, percolating network, normalized by $[d^2(p-p_c)/(d-1)]t_0$, as a function of time t , normalized to the crossover time (defined in the text) t_0 for bond-percolation concentrations above and below the critical percolation threshold p_c .

hierarchical lattices^{17,18} where numerical calculations should be much easier. If, for hierarchical lattices, one continues to find the same steep change in the density of states, it will serve as a strong indication that this feature remains for regular lattices.

The importance of the vibrational density-of-states problem lies, in addition to its intrinsic interest, in the interpretation given by Rosenberg *et al.*¹ to the thermal properties of amorphous systems. The steep change in the density of states found in this EMA treatment is quite consistent with what is required to fit the specific-heat density of states in amorphous systems, as pointed out by Tua *et al.*³ The present work seems to strengthen the case for the fracton interpretation of amorphous-state vibrational excitations.

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