Vibrations of fractals and scattering of light from aerogels

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The inelastic polarized and depolarized scattering of light from the vibrations in aerogels recently studied experimentally by Courtens, Vacher, and collaborators are analyzed with use of fracton-model scaling and crossover considerations. We discuss the implications of the short-range fractal structure and the way the strongly localized vibrations on the short-range fractal structure of the gel (fractons) show up in the scattering. For this purpose we extend existing discussions of the implications of assuming a connected self-similar fractal structure for the vibrations, and also discuss the relationship of fractons to elastic-scattering theory and to Anderson localization. The disordered fractal structure of the underlying mass density and the three-dimensional character of the vibrations are included by treating the inelastic light scattering as Mie scattering from the vibrating fractal blobs. We find a maximum in the scattering cross section when the wavelength of the scattered light is comparable to the size of the fracton and smaller than the coherence length of the structure, and discuss the different frequency dependence of the polarized and depolarized scattering. The results contradict extrapolations of the Brillouin-scattering expressions from the low-frequency Rayleigh regime. We also discuss the structure of the vibrational eigenmodes and show that the indices measured are new phenomenological indices which are not related to standard fractal dimensions or to superlocalization.

I. INTRODUCTION

The direct motivation for this paper arises from the detailed experimental investigations of light scattering from silica aerogels by Courtens, Vacher, and their collaborators.[1-4] Together with the elastic[5] and inelastic[6] neutron diffraction results,[5] and other experiments[7] of these authors reviewed in Ref. 8, these experiments constitute a very detailed test of the scaling considerations of the fracton model[9-15] for the eigenmode spectrum of fractals and of materials with a short-range fractal structure. They also require a reconsideration and extension of the qualitative results in these references. Neither the specific aspects of light scattering from the vibrations nor the vector character of the vibrations and the way it shows up in the *observable* strains were properly considered in these references or elsewhere in the literature. The purpose of this paper is to try to obtain as much theoretical information on this problem as is possible without a specific physical model.

In Secs. II and III we derive the scaling predictions of the fracton model for the vibrations. In Sec. II we discuss the vibrations of a fractal (fractons) and the associated strains. We emphasize the distinction between an intrinsic strain (when such a quantity can be defined) and the externally observable strains of the mass distribution in the embedding space which are actually measured. In Sec. III we discuss crossover effects for a real material, such as aerogels, which are macroscopically homogeneous and are only fractal below some crossover length (ξ_{co}). We also discuss the relation of these results to scattering theory and the way fractal correlations in the disorder overcome the difficulties encountered in straightforward perturbational scattering calculations and can generate a low-frequency, strong-scattering regime. We believe these considerations are relevant to numerous experimental situations with tenuous connectivity. They may also have some relevance to glasses and to the design and interpretation of experiments on optical localization.

In Sec. IV we derive our results for polarized and depolarized inelastic light scattering from fracton vibrations of disordered fractals including both the large- and the small-scattering-wave-vector limits and the crossover to the Brillouin scattering from longitudinal phonons using an approach related to Mie scattering—as in Ref. 3. In Sec. V we discuss the problems encountered if one attempts to do this calculation using an explicit ansatz for the fracton eigenfunctions. In Sec. VI we discuss the results and conclusions and the interpretation of experimental results.

Experiments have shown[5] that silica aerogels, when carefully prepared, show a fractal self-similar structure for a considerable range of length scales, between the grain size (a) and a density- (ρ-) dependent crossover length (ξ_{co}):

$$\rho \propto (\xi_{co}/a)^{D-3} , \tag{1.1}$$

where D is the fractal dimension of the aerogel. Depending on the preparation conditions,[5] the experimental value of D is either ~ 2.4 or ~ 1.8. Experiments also determine a scaling form for the elastic constants,[7]

$$\mathcal{K} \propto \rho^{a} \tag{1.2}$$

and for the velocity of sound,[1,2] $v^2 \propto \mathcal{K}/\rho$. Polarized Brillouin-scattering experiments[1,2] also show that

phonons—for which $\omega \simeq vq$—only describe the long-wavelength ($\lambda \ll \xi_{co}$) vibrational excitations below a crossover frequency (ω_{co}):

$$\omega_{co} \propto v/\xi_{co} , \tag{1.3}$$

and are also consistent with a scaling ansatz for the density of states per unit mass at higher frequencies:[6]

$$N(\omega) = C\omega^{\bar{\bar{d}}-1} . \tag{1.4}$$

The Brillouin-scattering experiments[1,2] also measure the elastic scattering Rayleigh width of the phonons (Γ):

$$\Gamma(\omega) = \omega(\omega/\omega_{IR}^{el})^3 . \tag{1.5}$$

They show that, for these materials, the Ioffe-Regel frequency[16] for *elastic* scattering [ω_{IR}^{el}—defined by Eq. (1.5)], at which strong elastic scattering sets in, can be identified with ω_{co} (or, at least, scales with ω_{co}).[8]

The results in Eqs. (1.1)–(1.4) agree with the detailed scaling predictions of the fracton model[9–15] for the vibrations of fractals and for crossover effects. Equation (1.5) confirms the identification of the fracton regime with the strong- (elastic) scattering, strong-localization regime.[11,12,14,15] Since the three-dimensional Anderson-localization edge occurs[17] at a frequency (ω_{An}) which is comparable to the elastic Ioffe-Regel frequency (ω_{IR}^{el}) but somewhat lower ($\omega_{An} < \omega_{IR}^{el}$), it also shows that the crossover occurs at a frequency higher than the localization edge for the phonons. While this is not a proof, it certainly supports the assumption of Refs. 11 and 12 and of 14 and 15 that the vibrational eigenmodes in the strong-scattering regime—fractons—are controlled by a *single* length scale which simultaneously plays the role of a wavelength, a scattering length, and a localization length. We note that this is a much stronger claim than that made by Rammal and Toulouse[18] when they applied the Abrahams-Anderson-Licciardello-Ramakrishnan weak-localization arguments[19] to fractals showing that the fracton (spectral) dimension determines localization.

This suggests that one try to check other predictions of this model and, in particular, work out the scaling predictions for inelastic light scattering from the high-frequency (fracton) vibrations,[1–4] which, except for a brief discussion of Raman scattering in Ref. 3, are not at present available. This is the main object of this paper.

We note that there exists fairly extensive literature, both experimental and theoretical, on somewhat related problems—e.g. on "superlocalization"—which is actually irrelevant to the problems we want to discuss and, in our opinion, often misleading in the present context. We neither use nor discuss this work, except implicitly in Sec. V.

In the context of theoretical treatments of amorphous materials, the fracton model constitutes an attempt to see how far one can go with a harmonic model, having only elastic scattering by a disordered medium, in explaining universal features observed in amorphous tenuous structures, in glasses and in other such materials. For this to be meaningful one needs a large strong-scattering frequency range with strongly localized vibrational eigenmodes. It is therefore crucial to have an Ioffe-Regel frequency for elastic scattering [ω_{IR}^{el}—Eq. (1.5)] that is low compared to the Debye frequency.

There are essentially two reasons for rejecting such an approach to the spectrum of amorphous materials. One is the obvious, and dramatic, success of the Anderson-Halperin-Varma-Phillips[20] two-level-system theory in explaining and predicting many of the universal low-temperature features. This makes it reasonable to assume that inelastic scattering, from two-level systems, also explains features such as the plateau in the thermal conductivity and anomalies in the density of states.[20–22] The second reason is that it is difficult to construct a strong-elastic-scattering model for lattice vibrations (or EM waves) because of the way in which the disorder enters the wave equation. For lattice vibrations the implication is that most treatments of the disorder will not show a transition to strong scattering at low frequencies or even a localization edge much lower than the Debye frequency (in three dimensions). The latter problem has been discussed extensively recently (e.g., Refs. 17 and 23) in the context of recent interest in phonon and photon localization. The fracton model is directly relevant to this problem and shows under what conditions one can actually have the elastic scattering play an important role at relatively low frequencies.[15] We discuss this below, in Sec. III, and in somewhat more detail than required for the aerogels.

The experiments on aerogels and, in particular, the observation of a large frequency range of phonon Rayleigh scattering [Eq. (1.5)], by the Brillouin scattering of light, demonstrates that strong *elastic* scattering (of phonons) must occur and that it plays an important role in these materials. The relevance to other materials depends crucially on the question of the relative importance of elastic and inelastic scattering.

It is standard to relate inelastic light scattering to the modulation of the local index of refraction by the lattice vibrations. This means that we want to calculate the correlation function for the modulation of the local (optical) index of refraction—$n(\mathbf{r},t)$—by the vibrations. In general this involves a (local, fourth-rank) material tensor relating the local strains ($\underset{\sim}{e}$) to the tensorial index of refraction ($\underset{\sim}{n}$). In bulk materials the dominant contribution is usually that due to the modulation of the local density [$\delta n \propto \delta\rho = \mathrm{Tr}(\underset{\sim}{e}) = \mathrm{div}\mathbf{u}$]. For physical reasons it is reasonable to assume this also for the aerogels (contrary to the situation for small molecules). We therefore assume in the following that

$$n(\mathbf{r},t) \sim p\rho(\mathbf{r},t) , \tag{1.6}$$

where p is a material constant and ρ the (average) local density. Any depolarized scattering is then due to the shape anisotropies of the vibrating region. While one could use a more general tensorial notation, there is, obviously, no way one could really say something meaningful about locally anisotropic couplings even if they were of significance. We also believe that Eq. (1.6) must be a good approximation. To emphasize the fact that the scattering is due to the strains, we shall frequently talk about the strains ($\underset{\sim}{e}$) due to the vibrations below without repeating the above argument and stating explicitly that

we assume that only the change in the density (i.e., the mass distribution) is important in the light-scattering experiments. We can write

$$\rho(\mathbf{r},t)=\sum_i \delta(\mathbf{R}_i+\mathbf{u}_i(t)-\mathbf{r})=\rho_0(\mathbf{r})+\delta\rho(\mathbf{r},t)\ , \quad (1.7)$$

$$\rho_0(\mathbf{r})=\sum_i \delta(\mathbf{r}-\mathbf{R}_i)\ , \quad (1.8)$$

where $\mathbf{R}_i$ is the equilibrium position of the grain i, $\rho_0(\mathbf{r})$ the underlying static density, and $\delta\rho(\mathbf{r},t)$ the time-dependent vibrating density responsible for the inelastic scattering.

Formally one can write, as usual,

$$\delta\rho(\mathbf{r},t)=\rho_0(\mathbf{r})\mathrm{Tr}[\underset{\sim}{e}(\mathbf{r},t)]\ , \quad (1.9)$$

where $\underset{\sim}{e}(\mathbf{r},t)$ [$\propto\nabla\cdot\mathbf{u}(\mathbf{r},t)$] is the local strain. One has to remember, however, that derivatives of the vibrational amplitudes $\mathbf{u}(\mathbf{r},t)$ are not properly defined (see Sec. V below), and that the fractal structure factor $\rho_0(\mathbf{r})$ is essential. In studying the vibrations, it is important to remember that the displacement $\mathbf{u}$ is a vector in the embedding space, and that one may be interested in features of the vibrations, such as the way they modulate the density, which cannot even be defined as intrinsic properties of the fractal. One expects $\rho_0(\mathbf{r})$, $\mathbf{u}(\mathbf{r},t)$, and the strains $\underset{\sim}{e}(\mathbf{r},t)$ [$\propto\nabla\cdot\mathbf{u}(\mathbf{r},t)$] to fluctuate rapidly on short length scales ($a<\delta r<\xi_{\mathrm{co}}$), for the situations we shall be interested in, so that the strains are only defined as scale-dependent local averages. The relevant correlation function is

$$\langle\delta\rho(\mathbf{r}_0,t_0)\delta\rho(\mathbf{r}_0+\mathbf{r},t_0+t)\rangle$$
$$=\langle\rho_0(\mathbf{r}_0)\rho_0(\mathbf{r}_0+\mathbf{r})\underset{\sim}{e}(\mathbf{r}_0,t_0)\underset{\sim}{e}(\mathbf{r}_0+\mathbf{r},t_0+t)\rangle\ , \quad (1.10)$$

and the scattering intensity is given, as usual, by the Fourier transform:

$$I(q,\omega)=A\langle|\delta\rho(q,\omega)|^2\rangle\ , \quad (1.11)$$

which is a convolution of the Fourier transforms of the fractal static density-density correlation function of $\rho_0(\mathbf{r})$ [$g(r)$], which plays the role of a structure factor, and of the strain—$\underset{\sim}{e}_\alpha(\mathbf{r},t)$—associated with vibrations at frequency $\omega_\alpha=\omega$. In this q is the scattering wave vector of the scattered light.

An obvious approach to the calculation of $I(q,\omega)$ is to use an ansatz for the (average) form of the high-frequency ($\omega_\alpha>\omega_{\mathrm{co}}$) fracton vibrational eigenmodes:

$$\mathbf{u}_\alpha(\mathbf{r},t)=u_\alpha\cdot\boldsymbol{\psi}_\alpha(\mathbf{r})\exp(i\omega_\alpha t)\ . \quad (1.12)$$

If one knew $\rho_0(\mathbf{r})$ and $\boldsymbol{\psi}_\alpha(\mathbf{r})$ for a specific realization and eigenmode, one could, of course, calculate the correlation function in Eqs. (1.10) and (1.11) directly. These are, however, complicated random functions with fractal correlations. Describing them by an "average" eigenmode can be very misleading. At best, it is unreliable and hard to justify. We shall discuss this approach in some detail in Sec. V of this paper with rather discouraging conclusions as to its viability.

To calculate the scattering, in Sec. IV, we shall therefore follow Refs. 2 and 3 and use a different approach related to the way one describes Mie scattering from random scatterers. We shall see that the correlation function [Eqs. (1.10) and (1.11)] is only sensitive to some very general scaling features of the vibration spectrum. Any additional details implied by an ansatz are redundant and cannot be checked by the experiments. For the regime most relevant to the depolarized (called "Raman" in Ref. 3) light-scattering experiments, but also to part of the polarized ("Brillouin") results in Refs. 1 and 2,

$$ql(\omega)\ll 1,\quad \omega_{\mathrm{co}}\ll\omega\ , \quad (1.13)$$

we largely reproduce the results of the analysis in Ref. 3 but not those of Ref. 2. We find that the crossover (at ω_{co}) to scattering from phonons is complex and expected to be broad. In Eq. (1.13), $l(\omega)$ is the length scale relevant for the spatial coherence of the fractal vibrations. We extend these results to the large-[$l(\omega)$] (small-q) regime.

II. VIBRATIONS OF FRACTALS: THE FRACTON MODEL

Our purpose here is to use scaling arguments to obtain information about the vibrations of a connected, rigid, self-similar fractal cluster. Examples to which these results are directly relevant would be gels, frozen polymer chains, aggregation clusters, or physical realizations of percolation clusters. While scaling arguments similar to those of this section are contained in Refs. 9–15 and explained in fair detail in Ref. 12, the derivation here is different and, we believe, clearer. The focus of the discussion there was on problems which could be completely described *on* the fractal—such as diffusion on the fractal or the solution of the Schrödinger equation in a fractal geometry. The problem of vibrations is more complex because the $\mathbf{u}_i$ are always vectors in the embedding (three-dimensional) space and cannot be mapped into some intrinsic internal geometry of the connected fractal.

One result of this difference is the complex tensorial character of the force constants[24–28]—the importance of angular (bending) and twist force constants—which tend to dominate the large-scale mechanical properties. In general, this makes a tenuous fractal, or, in fact, any lower-dimensional solid object embedded in a higher-dimensional space, much softer than it would be if no normal components of $\mathbf{u}$ were allowed. For wet gels the result is the dominance of stress-induced scalar elasticity[24,25] near criticality (just as surface tension, when present, dominates the capillary waves of a free film). It seems unlikely that a similar mechanism plays any role in mechanically rigid, self-supporting aerogels (as can be seen from the detailed analysis of the requirements in Refs. 24 and 25). Since the aerogels are found to be fairly rigid, one concludes that they must have a much more closely interconnected structure than treelike fractals like percolation and aggregation clusters. A model with the appropriate properties would be very instructive, but is not available at present.

A second effect of the three-dimensional vector character of $\mathbf{u}$ is more technical, but extremely important in interpreting experiments. Experimental probes, like light

waves, propagate in the embedding (three-dimensional) space and are not sensitive to the way the vibrations are related to the complicated internal geometry of the fractal. One therefore has to worry about the way the vibrations of a complicated fractal object show up in the density (or, more generally, the effective observable strain) in the embedding space. This complication is, of course, related to the type of experiment one does. It is less severe and easier to treat in scalar problems, like diffusion and quantum percolation, which can be defined completely in the *internal* geometry of the fractals.

To avoid these difficulties, we therefore present the scaling argument in as general a form as possible, and in a form which is directly relevant to the vibrations. We derive some new results and shall also try to stress the assumptions made. In essence, we are trying to see how much one can say about the vibration spectrum using only the self-similar dilation symmetry. The fractal "looks" the same at different resolutions and, therefore, presumably also "acts" the same way.

The static density-density correlation function around a point on a self-similar fractal[29] has the form

$$g(r) \propto (r/a)^{-(d-D)} , \tag{2.1}$$

where a is the size of the elementary grains constituting the fractal, d the dimension of the external embedding space, and D the fractal dimension. This is equivalent to saying that a "blob"[30] of the fractal of size r will, on the average, have a mass

$$M(r) = m_a (r/a)^D . \tag{2.2}$$

Since the density of such a blob is much higher than the average density of a large fractal—of size $r' \gg r$—it follows that the fractal is mostly empty, and that this also holds when we look at the distribution of fractal blobs in a large fractal.[29,30] This also means that one can heuristically think of a fracton vibration as the "free" vibration of a fractal blob of appropriate size (l) relatively weakly bound to its, mainly empty, surroundings. This follows from the fractal structure and is relatively insensitive to the detailed internal connectivity model. We note that because of the low ramification dimension[29] of most connected fractals known, one expects "surface modes" to be even less important than in finite solids.

It seems natural to assume, and this is confirmed in models for which one can do the calculation,[9,13] that the scaling form carries over to the density of states, at least for sufficiently low frequencies:

$$N(\omega) \propto \omega^{\bar{\bar{d}}-1} , \tag{2.3}$$

as in Eq. (1.4). This defines the spectral (fracton) dimension $\bar{\bar{d}}$, which is, in essence, an independent fractal dimension. $\bar{\bar{d}}$ depends on the internal geometry of the fractal—on the way it is internally connected—but for the vibrational problem also on the way it is embedded in the external space and on the nature of the terms in the harmonic expansion that determine its rigidity.[24–28] Because $\bar{\bar{d}}$, in this context, is a property of the vibration spectrum and not an intrinsic geometric property of the connected fractal (as it is for scalar problems), we follow Rammal and Toulouse[18] and shall call it a (vibrational) spectral dimension.

We can now associate a length scale $l = l(\omega)$ with the vibrations. It is easiest to do this in terms of the effect of boundary conditions. We consider the vibrations of an *isolated* fractal blob of size L. High-frequency modes (of the infinite fractal) for which $l(\omega) \ll L$ will not be affected by the change in the boundary conditions, at scale L, involved in disconnecting the blob from the rest of the fractal. On the other hand, the low-frequency modes for which $l(\omega) \gg L$ will disappear from the spectrum. The crossover occurs at some frequency ω_L such that

$$L \sim l(\omega_L) . \tag{2.4}$$

The integrated spectral weight of the *missing* low-frequency modes is lumped together in the center-of-mass (and rotational) degrees of freedom of the disconnected blob. This is a number that cannot depend on L and ω (Ref. 12). Therefore, using Eqs. (2.2) and (2.3),

$$[l(\omega)]^D \int_0^{\omega_l} d\omega\, \omega^{\bar{\bar{d}}-1} \propto [l(\omega)]^D \omega_l^{\bar{\bar{d}}} = \text{const} , \tag{2.5}$$

from these very general dimensional considerations. (This assumes a normalization of the density of states to unit mass rather than unit volume.) The dispersion relation between l and ω is therefore[10–12]

$$l(\omega) \propto \omega^{-\bar{\bar{d}}/D} , \tag{2.6}$$

independent of any model-dependent considerations for the calculation of D and $\bar{\bar{d}}$ or of the geometrical interpretation of these dimensions.[31]

We want to clarify the meaning of this definition. If one would apply this definition of the relevant frequency-dependent length scale $[l(\omega)]$ to free waves in a homogeneous medium ($D = \bar{\bar{d}}$), one would, of course, get the *wavelength* $[l(\omega) \sim \lambda(\omega) = 2\pi v/\omega]$. The lowest vibrational frequency of a grain of size l is, approximately, $\omega_l \sim 2\pi v/l$. This also holds in the weak-localization regime for localized phonons near the Anderson-localization edge. The density of states is not changed (for weak localization) and the length scale $l(\omega)$, as defined above, then, is not related to the localization length or to the scattering length, but only to the underlying wavelength of the scattered wave. On random strongly disordered fractals, one can assume strong localization,[11,12,15] so that $l(\omega)$ is *also* the localization length.[32] The vibrational eigenmodes are then called fractons[11,15,32] and are characterized by a single unique length scale related to ω, D, and $\bar{\bar{d}}$ by Eq. (2.6).

Since the fractal is denser at small length scales, it is intuitively plausible that its elastic constants will also be scale dependent. For this reason, and because of the complicated random mass distribution, it will deform in a complicated (usually nonaffine) way under external stresses.

Assume the equations of motion of the fractal are

$$-m_1 \omega^2 \mathbf{u}_i = T_1 \sum_j \tau_{ij} \cdot (\mathbf{u}_j - \mathbf{u}_i) , \tag{2.7}$$

where m_1 is a mass at the initial scale (1), the $\underset{\sim}{\tau}_{ij}$ are coupling tensors, and T_1 is a characteristic strength of the force constants. We note that one can write the equations in this form even when one has both mass and force-constant disorder. We assume a normalization so that the upper cutoff "Debye frequency" for these equations is

$$\Omega_a^2 \sim \Omega_{D1}^2 = T_1/m_1 \ . \tag{2.8}$$

We now coarse-grain the fractal by collapsing all blobs of size l to points and construct a new net on this scale. This will eliminate all modes with frequencies $\omega > \omega_l$ and replace m_1 by $m_l = m_1 l^D$. Since the fractal is self-similar, we assume that the structure of the $\underset{\sim}{\tau}_{ij}$ remains unchanged (in a statistical sense) and adjust T so that the coarse-grained model has the same low-frequency spectrum as the original one. This requires $\Omega_{Dl} \sim \omega_l$ and therefore, using Eq. (2.6),

$$T_l = m_l \omega_l^2 \propto l^D \omega_l^2 \propto l^{(D/\bar{\bar{d}})(\bar{\bar{d}}-2)} \propto \omega_l^{(2-\bar{\bar{d}})} \ . \tag{2.9}$$

As defined here, T_l is a *force constant* relevant to the length scale l—that is, the coefficient for the restoring force associated with the relative displacements of points a distance l apart. We shall, however, need the changes in density associated with the vibrations and, therefore, the *strains* $\underset{\sim}{e}_l$.

There is some ambiguity in the definition of the strain for a fractal object. One approach would relate the strain to the *internal* geometry of the fractal defined by the way the fractal is connected internally. This obviously has some intuitive heuristic attractions, but is difficult to implement when one does not have a detailed microscopic model. The second approach, which we adopt here, defines the strain directly in terms of the effect of the deformations [i.e., the $\mathbf{u}(\mathbf{r})$] on the (suitably averaged) density distribution in the embedding space. This is, of course, the definition *directly relevant* to the experimentally measured quantities. Depending on the situation, the *internal* strain (discussed briefly later in this section and defined explicitly in Sec. V) may or may not coincide with the *externally observable* strain we shall use (or at least may be simply related to it). We are not aware of any basis on which one could make general statements about this relationship.

We *assume* a phenomenological scaling form for the strains:

$$\underset{\sim}{e}_l^2 = \langle (\mathbf{u}_i - \mathbf{u}_j)^2 \rangle_l / s_l^2, \quad s_l \propto l^\sigma \tag{2.10}$$

where $\langle (\mathbf{u}_i - \mathbf{u}_j)^2 \rangle_l$ is the average relative motion of two grains a distance l apart. The trace of $\underset{\sim}{e}_l$, defined in this phenomenological way, is the change in density used in Eq. (1.9). This is an explicit and separate scaling assumption. It is the closest one can get to the definition of an externally observable strain for a fractal. It should be regarded as a purely *phenomenological* scaling ansatz relating the strains one observes in the embedding space to the amplitude of the vibrations. $\underset{\sim}{e}_l$ measures the relative changes in the average density (and the strains of the averaged mass distribution) associated with the vibrations. In general, the scaling index σ is expected to depend on the actual mechanical properties, on the internal geometry, and on the way the fractal is embedded in space. While $\sigma = 1$ cannot be ruled out, it is almost impossible to imagine a situation in which the fractal vibrations will arrange themselves nicely to give an ordinary homogeneous strain of the mass distribution. It is, for example, easy to convince oneself that even in the heuristically simplest cases, such as a fractal polymer chain or a crumpled sheet with purely internal vibrations, one finds $\sigma \neq 1$. We claim that, in general, σ should be regarded as an *independent* scaling index which cannot be determined from the fractal and spectral dimensions (D, $\bar{\bar{d}}$) or, as has repeatedly been implied in the literature, from the "superlocalization" index.

The *elastic constants* ($\mathcal{K}$) are defined as the coefficients of the strain $\underset{\sim}{e}$ in the elastic deformation-energy density[33] ($E_{\rm el}$). Using the definition of the strains in Eq. (2.10), we can also define the equivalent of an elastic constant for the fractal ($\mathcal{K}_l^f$):

$$\mathcal{K}_l^f \propto T_l s_l^2 / l^d \propto l^{(D/\bar{\bar{d}})(\bar{\bar{d}}-2)+2\sigma-d} \ , \tag{2.11a}$$

$$E_{\rm el} = \int T_l u_l^2 = \int \mathcal{K}_l^f \underset{\sim}{e}_l^2 \ . \tag{2.11b}$$

It is heuristically attractive, and meaningful at least for simple situations, to look for an intrinsic definition of the strain related to the internal geometry of the fractal, and try to identify this *internal* strain with the *external* strain defined phenomenologically in Eq. (2.10). One interprets s as an *internal* length scale defined on the fractal.[34–37] This obviously implies $\sigma \geq 1$. One can then try to relate T_l directly to the intrinsic lengths and to an internal dimension of the fractal (δ) in the usual way:

$$T_l \propto s_l^{(\delta-2)} \propto l^{\sigma(\delta-2)} \propto \omega_l^{\bar{\sigma}(2-\delta)} \ , \tag{2.12}$$

where we have defined $\bar{\sigma} = \bar{\bar{d}}\sigma/D$. The meaning of Eq. (2.12) is that we assume that the force constant between the two ends of a "string" of springs, of strength T_1, connected in series is T_1/s_l when the ends of the string are a distance l apart and that $s_l^{(\delta-1)}$ such strings appear in parallel for the effective force constant of a "cube" of size l^3. Assuming σ (and s) defined by the strains [Eq. (2.10)], this may be taken as the definition of δ. Even this way one notes that the procedure cannot be simply extended to explicit bending forces, which show up as three-body terms in the harmonic expansion or to four-body twist terms[24,25] [because Eq. (2.12) does not hold]. If such forces are important, δ no longer has a simple geometric interpretation and can only be regarded as a formal, alternative parametrization of σ. Comparing Eq. (2.12) with Eq. (2.9) gives

$$\begin{aligned} \sigma &= (D/\bar{\bar{d}})[(\bar{\bar{d}}-2)/(\delta-2)] \ , \\ \bar{\sigma} &= (\bar{\bar{d}}/D)\sigma = (\bar{\bar{d}}-2)/(\delta-2) \ , \end{aligned} \tag{2.13}$$

which may be a somewhat better, or at least more intuitive, parametrization of the scaling ansatz (2.10). This sort of interpretation is implicit in discussions of the "chemical distance" and, appears in somewhat more gen-

eral form, in Refs. 35–37. All these discussions assume a constant ramification[29] ($\delta=1$), and this was also used in Ref. 3. We note, however, that a simple geometric interpretation of δ as a "ramification" dimension is meaningless when bending forces, whose existence depends on the embedding space, are dominant,[28] so that T no longer scales with s like a resistance.[37] Thus the geometric interpretation of δ, σ, and s should not be taken too literally.

III. CROSSOVER AND SCATTERING

A solid material, such as an aerogel, has a finite density and is of course never a fractal on a macroscopic scale. At most, it can be fractal locally up to a crossover length (ξ_{co}). Qualitatively, one can then think of such a material as a dense packing of connected fractal blobs of size ξ_{co}—analogous to the description of a semidilute polymer solution[30] and to that of the infinite cluster in the percolation problem. On large scales the material is homogeneous with matching crossover relations at ξ_{co} determining the macroscopic properties. Using Eq. (2.1) the macroscopic density is then given by Eq. (1.1).

Using standard crossover-scaling considerations for the dispersion relating frequency and length scales, and combining Eqs. (1.3) and (2.6), one finds for the velocity of sound

$$v \propto \omega_{co}^{1-(\bar{\bar{d}}/D)} \propto \xi_{co}^{-[(D/\bar{\bar{d}})-1]} . \tag{3.1}$$

One now notes that one cannot simultaneously assume smooth crossover-scaling relations for the vibrational frequencies (as implicit in the above argument) and for the elastic constants.

The macroscopic bulk elastic constants [$\mathcal{K}_b(\rho)$] must be related to the density and to the velocity of sound in the usual way:

$$\mathcal{K}_b(\rho)=\rho v^2 \propto (\xi_{co})^{\chi}, \quad \chi=(D/\bar{\bar{d}})(\bar{\bar{d}}-2)+2-d , \tag{3.2}$$

where we have used Eqs. (1.1) and (3.1). This is clearly consistent with the fractal result for $\mathcal{K}_l^f$ at $l=\xi_{co}$—Eq. (2.11)—only for $\sigma=1$. The origin of this discrepancy is in the definition of the strains in Eq. (2.10) ($\underline{e}_l \propto u/l^{\sigma}$), which reflected the fact that the vibrations of the fractal are not directly related to changes in the density (or to other deformations of the vibrating "blob") and are therefore less effective in inducing such deformations (for $\sigma>1$). A fracton with $l=\xi_{co}$ is less effective in modulating the density than the "phonon" vibration, at the same frequency, of a homogeneous medium with the same average density (ρ) and the same stiffness force constants (T). The implication is that one predicts a *qualitative* change in the structure of the eigenfunctions $\psi_\alpha(\mathbf{r})$ in the crossover region that is not fully reflected in the dispersion relation between l and ω.

Just as a comment, we remark that this is not simply a question of mixing longitudinal and transverse modes. Because of the nonaffine nature of the deformations associated with the vibrations of a fractal, the averaging simply eliminates part of the local effects of the $\mathbf{u}$. The substance of the effect is somewhat analogous to situations where optical modes are mixed into the low-frequency phonons in crystals. Our argument above shows that these effects cannot be incorporated into a universal crossover function for $\mathcal{K}$.

The low-frequency vibrations are propagating phonons. They are scattered elastically from fluctuations in the local velocity of sound—$v^2(\mathbf{r})=\mathcal{K}(\mathbf{r})/\rho(\mathbf{r})$—and this shows up in the Rayleigh width Γ [Eq. (1.5)]. Scattering is enhanced by the fractal correlations at short distances, and the characteristic length scale for disorder becomes ξ_{co} rather than the grain size a.[15] This is one of the most striking effects of the short-range fractal correlations. We want to discuss it briefly in the context of (Rayleigh-) scattering theory, extending the brief discussion in Ref. 15. Because of the importance of the problem in other contexts, we do this in a somewhat more general form than is actually relevant to the aerogels.

Quite generally, one can relate Rayleigh scattering to the fluctuations in the *local* velocity of sound [$v^2(\mathbf{r})$] in a disordered medium, averaged over the wavelength (λ). Macroscopically,

$$v^2=\langle v^2(\mathbf{r})\rangle, \quad \omega=vq=2\pi/\lambda , \tag{3.3}$$

and, from standard statistical considerations, the variance would be expected to be

$$\mathcal{V}(\lambda)=[\langle v^4(\mathbf{r})\rangle_\lambda/\langle v^2(\mathbf{r})\rangle_\lambda^2]-1 \sim \mathcal{V}(b)/(\lambda/b)^d, \quad \lambda/b \gg 1 \tag{3.4}$$

where b is the length scale at which the elementary fluctuations occur.

Scattering at $\omega(\lambda)$ is proportional to the variance $\mathcal{V}(\lambda)$, and one obtains the Rayleigh result—$\Gamma(\omega)\propto\omega^{(d+1)}$—as long as Eq. (3.4) holds and $\mathcal{V}(\lambda)$ is not too large, say

$$\mathcal{V}(\omega)\sim(\omega/\omega_{IR}^{el})^{-d} \leq \mathcal{V}_{IR}=O(1) . \tag{3.5}$$

Except for numerical coefficients, this qualitative argument is equivalent to the treatment of the fluctuations in v^2 [$v^2(\mathbf{r})-\langle v^2\rangle$] as a perturbation in the wave equation and is therefore implicit in the Born-approximation (Rayleigh) result for $\Gamma(\omega)$. This approach can, however, never be extended to situations where $\mathcal{V}(b)$ becomes large.

One might naively argue that when $\mathcal{V}(b)$ is large at the elementary scale (b), the limit $\mathcal{V}(\omega)=\mathcal{V}_{IR}$ is reached at some low frequency:

$$\omega=\omega_{IR}^{el} \ll \Omega_b \propto 2\pi v/b , \tag{3.6}$$

where ω_{IR}^{el} is defined as in Eq. (1.5). As noted in the Introduction, this is clearly the experimental situation in the aerogels. The disorder occurs at the scale of the grain size (a), but $\xi_{co}/a \gg 1$ $\omega_{IR}^{el} \ll \Omega_a$.

When one tries to do an actual scattering calculation (e.g., Refs. 17 and 23), one finds, however, that it is difficult to construct a model which will exhibit this type of behavior [Eq. (3.6)]. Even when one starts with $\mathcal{V}(b)\gg 1$, one finds $\omega_{IR}^{el}\sim\Omega_b$ for the low-frequency Rayleigh scattering [Eq. (1.5)]. The reason for this is that the simple averaging process implied by Eq. (3.4) cannot describe the wavelength dependence of the scattering when $\mathcal{V}$ is large. The variance experienced at some larger scale

is not given by the simple average of $v^4(\mathbf{r})$. Most detailed descriptions of the strong-scattering regime give a much faster decay of $\mathcal{V}(\lambda)$ than Eq. (3.4), in the large-variance regime, and are essentially inconsistent with the inequality (3.6).

It is of some interest to see where the difficulty originates and why the problem does not arise in the aerogels and in other situations with strong short-range correlations in the disorder.

Consider first the purely formal meaning of having a large variance in $v^2(\mathbf{r})$ at some length scale l — $\mathcal{V}(l) \gg 1$. Since $v^2(\mathbf{r})$ is always positive, the only way this can occur is by having a *small* number of "hard" regions for which v^2 is much larger than the average ($v^2 \sim v_h^2 \gg \langle v^2 \rangle$), which dominate the variance, and a much larger number of "soft" regions for which v^2 is smaller than the average. The probability of finding a region with velocity v^2, $P_l(v^2)$, must be double-humped or have a long, high-v^2, tail. Thus, in essence, $\mathcal{V} \gg 1$ implies a *dilute* scattering situation with a small concentration of "hard" ($v^2 \gg \langle v^2 \rangle$) scatterers embedded in a soft, low-v^2, medium (and possibly a much larger concentration of "soft" scatterers).

Consider the effect of the coarse-graining process. We are interested in the (average) local velocity of sound, $v_p^2(\mathbf{r})$, of a box of size pl ($p \gg 1$) (to be definite, we could, for example, define v_p as the velocity of sound of a medium obtained by applying periodic boundary conditions to the box, or as in Ref. 15). One notes immediately that one cannot simply use the average of v^2 in calculating the effect of the hard scatterers [as assumed in Eq. (3.4)] because of the mismatch between them and the soft background. The proper averaging procedure is very sensitive to the correlations in the distribution of the dilute, high-v^2 regions. Fluctuations in the elastic constants and in the average density should be treated separately.

If the hard scatterers (of size l) are isolated, the only propagating modes are those of the soft background medium. The scatterers act as rigid inclusions in the medium. Roughly, the hard scatterers simply increase the effective elastic constant by a factor proportional to the (small) volume fraction they occupy:

$$\mathcal{K}_s \rightarrow \mathcal{K}_s(1+f_h) , \tag{3.7}$$

where $\mathcal{K}_s$ is an elastic constant of the soft medium and f_h ($\ll 1$) is the volume fraction occupied by the hard scatterers. This is a small correction essentially independent of the magnitude of the (large) elastic constant $\mathcal{K}_h$ of the hard scatterers. The mass is, of course, the total mass, and therefore

$$\rho_s \rightarrow \rho_s - (\rho_s - \rho_h) f_h , \tag{3.8}$$

where ρ_s and ρ_h are the two densities. The fluctuations in ρ can become large only when the hard scatterers are also heavy and dominate the mass ($\rho_h f_h \geq \rho_s$). Thus the rescaled velocity of sound is

$$v_p^2 \sim \mathcal{K}_s(1+f_h)/[\rho_s - (\rho_s - \rho_h) f_h] . \tag{3.9}$$

Assume $\rho_s > \rho_h > 1$.[38] Fluctuations in v_p^2 are then small—of order f_h—even when v_h^2 ($\sim \mathcal{K}_h/\rho_h$) is much larger than v_s^2 ($\sim \mathcal{K}_s/\rho_s$). A more careful averaging procedure does not change the qualitative features of this result. The variance in v^2 at the scale pl is controlled by fluctuations in f_h^2 ($\langle f_h^2 \rangle / \langle f_h \rangle^2$) and loses all memory of the large $\mathcal{V}_l$. The implication is, of course, that the variance becomes much smaller than one would expect from Eq. (3.4).

The argument above neglects the "local modes" centered around the hard scatterers. For a single scatterer they would be localized and have a high frequency—analogous to localized defect modes in a solid. When they are dilute and uncorrelated, so that they are coupled weakly, they can only form a high-frequency "optical" impurity band. On the other hand, when they are correlated and coupled sufficiently strongly, they can dominate the elastic properties and the *low-frequency* velocity of sound. In other words, even a low density of hard regions can dominate the rigidity if they form a sufficiently rigid connected net. In a way this is the inverse of the "slow-sound" situation for fluids in porous media (e.g., fourth sound for He).[39]

For aerogels we have the extreme limit of this last situation. Fluctuations occur at the scale of the grain size (a) and $\mathcal{V}_a \gg 1$. The "hard" grains form a connected fractal net and dominate both the rigidity and the density. The result is that the velocity of sound [$\langle v^2(l) \rangle$] is scale dependent [Eq. (3.1)], but the variance $\mathcal{V}$ is scale invariant—up to the scale $\xi_{\rm co}$—because of the self-similarity.[15] This was expressed in the scaling relations for the fractal regime that we discussed in Sec. II and in Eq. (3.1) for the velocity of sound.

In the fractal regime there are two types of fluctuations. One relates the "velocity of sound" of the "blobs" ($v_l^2 \sim T_l l^2/m_l$) to those of the empty background ($v_B^2 \sim 0$).[40] The second one, which is relevant for the Rayleigh scattering, is the fluctuation in v_l^2 among different blobs. In the spirit of the definition of a Haussdorf-Besicovitz dimension,[29] one could, for example, cover a fractal with spheres of radius l and look at the distribution—$P_l(v^2)$—of v_l^2 among these blobs. One expects a scaling form

$$P_l(v^2) = (1/\langle v_l^2 \rangle) f(v^2/\langle v_l^2 \rangle) , \tag{3.10}$$

where $f(x)$ is a scale-invariant function of its argument. This implies a scale-invariant variance—$\mathcal{V}_l = \mathcal{V}_g$. The implication is that the elastic Ioffe-Regel frequency $\omega_{\rm IR}^{\rm el}$ for different gels should scale like the crossover frequency $\omega_{\rm co}$. The fact that the two frequencies are comparable shows that $f(x)$ is broad, so that $\mathcal{V}_g \sim 1$ for the actual fractal structures of these gels.

We note that these large fluctuations do not follow from the fractal structure, but are model dependent and depend on the detailed nature of the disorder in the fractal geometry. Models where such fluctuations are large (for other properties) are, however, frequently encountered for complicated fractals (see, e.g., the extensive recent literature on multifractals[41]). We are not aware of any attempt to study these fluctuations for the velocity of sound or other properties relevant to the vibration spectrum. As already noted, the experimental results for the

Rayleigh scattering in aerogels demonstrate that $\mathcal{V}_g$ is large for the fractals generated by the specific preparation process used. It also seems large for percolation clusters.

IV. SCATTERING FROM FRACTONS

We want to calculate the scattering of light from fractons using Eqs. (1.6)–(1.11). For frequency shift ω and scattering wave vector q, we need the Fourier transform for a single fracton mode (α):

$$\delta\rho_\alpha(q,\omega)=\delta(\omega_\alpha-\omega)\int d\mathbf{r}\,\rho_0(\mathbf{r})\cdot\underset{\sim}{\mathbf{e}}_\alpha(\mathbf{r})\exp[i(\mathbf{q}\cdot\mathbf{r})]\ , \quad (4.1)$$

where we have introduced the strain $\underset{\sim}{\mathbf{e}}$ explicitly.[42] More accurately, one should say that the source of the scattered radiation is a vibrating polarization induced by the incoming field, $\mathbf{P}_\alpha(\mathbf{r},t)$, in the vibrating region of size $l(\omega)$.

Consider first the case $ql(\omega)\ll 1$. The electric field of the incoming wave is then essentially constant over the region, of size $\sim l$, for which $\underset{\sim}{\mathbf{e}}_\alpha$ has an appreciable amplitude. The geometric considerations are the same as for the Mie scattering from a small particle. The source of the scattered wave is the induced dipole ($\delta\boldsymbol{\mu}$) of the vibrating region. Its magnitude is proportional to the mass of the strained region and it will have a transverse, depolarized, component if this region is anisotropic. The dominant fractron scattering is coherent from the vibrationally strained region (assuming strong, single-length-scale localization).[43] Also, one expects this region to be anisotropic and therefore to have an anisotropic polarizability as in scattering from small particles with shape anisotropy. One therefore predicts comparable amplitudes for the polarized $[I_\alpha^{\parallel}(q,\omega)]$ and depolarized $[I_\alpha^{\perp}(q,\omega)]$ components of the scattered light:

$$I_\alpha^{\parallel}(q,\omega)\propto I_\alpha^{\perp}(q,\omega)\propto\delta\mu^2\propto l^{2D}\langle\underset{\sim}{\mathbf{e}}_l^2\rangle\ . \quad (4.2)$$

Since scattering from different fractons is incoherent, the total scattered intensity is then[3]

$$I(q,\omega)=\sum_\alpha\delta(\omega-\omega_\alpha)I_\alpha(q,\omega)=N(\omega)\langle I_\alpha(q,\omega)\rangle\ , \quad (4.3)$$

where $N(\omega)$ is the density of states.

We have to estimate the strain, the relative change in density. From Eq. (2.9) we can estimate the mean-square amplitude of the vibrations:

$$\langle[(\mathbf{u}_i-\mathbf{u}_j)_l]^2\rangle\propto kT/T_l\propto\omega^{-(2-\bar{\bar{d}})}\ , \quad (4.4)$$

and, therefore, using Eq. (2.10) for $\underset{\sim}{\mathbf{e}}$,

$$\langle\underset{\sim}{\mathbf{e}}_l^2\rangle\propto\omega^{-(2-2\bar{\sigma}-\bar{\bar{d}})}\ . \quad (4.5)$$

Using this result with $l(\omega)\propto\omega^{-\bar{\bar{d}}/D}$ [Eq. (2.6)] and Eq. (1.4) for $N(\omega)$, in substituting Eq. (4.2) into Eq. (4.3), one gets

$$S(q,\omega)=(\omega/kT)I(q,\omega)\propto(1/\omega^2)(1/s_l)^2\ , \quad (4.6)$$

so that

$$S(q,\omega)\propto\omega^{2(\bar{\sigma}-1)}=\omega^{2[(\delta-\bar{\bar{d}})/(2-\delta)]},\quad ql(\omega)\ll 1 \quad (4.7)$$

where $\bar{\sigma}$ is defined in Eq. (2.12), where, as usual in this context, we factor out the Bose function ($b\sim kT/\omega$). Equation (4.7) is our final result for the scattering when $ql\ll 1$. An equivalent result was obtained in Ref. 3 using the superlocalization index d_ϕ for σ and subsequently following Ref. 36 in assuming $\delta=1$. This resulted in an expression for σ in terms of $\bar{\bar{d}}$ (and D). There is no justification for the latter assumption or for identifying σ with the superlocalization index. We therefore conclude that the measurement of this frequency dependence in Ref. 3 should be interpreted as a measurement of $\bar{\sigma}$ (or, equivalently, of δ) rather than as a check on the value of $\bar{\bar{d}}$.

We now consider the high-q (or low-frequency) limit. There are two modifications.

First, because of the random structure factor $\rho_0(\mathbf{r})$, the correlations between scattering from different blobs of size $\lambda=2\pi/q$ are small [Eq. (2.1)]. This reduces the coherent scattering from the whole fracton by a factor $\sim(ql)^D$ and makes it comparable to the *incoherent* scattering from blobs of size λ ($q=2\pi/\lambda$).

In addition, the deformation of these blobs can also be different. One cannot assume *a priori* that the strain of the blob of size λ ($\ll l$), as a result of the vibration at scale l, is the same as the average strain at the scale l.

We try a naive estimate. Assume that the force acting on such a blob is related to the force at the scale l by

$$(s_\lambda/s_l)^{(\delta-1)}T_\lambda u_\lambda=T_l u_l\ , \quad (4.8)$$

as would follow from a naive interpretation of the argument at the end of Sec. III. This assumes that the force on the face of a "cube" of size l is carried in parallel by $(s_l/s_\lambda)^{(\delta-1)}$ strands of thickness λ. While this is a geometrically plausible picture, we emphasize that we certainly cannot claim that it is necessarily correct in all cases. This leads to

$$(\underset{\sim}{\mathbf{e}}_\lambda)^2=(T_l^2/T_\lambda)^2(s_\lambda/s_l)^{2(2-\delta)}(e_l)^2=(e_l)^2\ , \quad (4.9)$$

so that the strain is uniform—at least in this model—in spite of the hardening of small blobs. This is, of course, the usual result for strains in a homogeneous material. Multiplying Eq. (4.7) by $(ql)^{-D}$, one ends up with

$$S(q,\omega)\propto\omega^{2(\bar{\sigma}-1)}(\omega/\omega_\lambda)^{\bar{\bar{d}}},\quad ql(\omega)\gg 1\ . \quad (4.10)$$

We have introduced a numerical factor in Eq. (4.10) to ensure that the two results (4.7) and (4.10) match at ω_λ [$l(\omega_\lambda)=\lambda$]. At least for the experimentally observed values[1–8] of the indices ($\bar{\sigma}\sim 0.8$, $\bar{\bar{d}}\sim 1.3$) the scattering $[S(q,\omega)]$ at fixed q ($=2\pi/\lambda$) is predicted to have a maximum for $[l(\omega)/\lambda]\sim 1$, as one would expect intuitively and contrary to the predictions of Ref. 2. For sufficiently light samples one should be able to observe this. In such a measurement, Eq. (4.7) would describe the high-frequency (small-l) and Eq. (4.10) the low-frequency (large-l) behavior.

While the argument leading to Eq. (4.7) is, we believe, unique and follows from the general scaling assumptions, we are less certain of the validity of the derivation of Eq. (4.10). One could imagine fractal models in which the

elastic energy, and therefore the strain, is not uniformly distributed among all the small blobs (of size λ). It has, for example, been suggested that only the backbone is stressed in the vibrations, while those parts of the mass which are located on "dead end" side branches are carried along rigidly without being strained. Behavior of this type is possibly even indicated by simulation results on percolation clusters.[44] Such behavior is certainly expected in the application of a *static* stress to a fractal. For the vibrations, it is, however, hard to imagine a mechanism which would accelerate a blob situated on a "dead end" without straining it, irrespective of its position with respect to the backbone. We would therefore expect equipartition of energy to hold.

A more delicate question is the distribution of strains on much smaller scales. The resistivity of very tenuous structures is known to be dominated by a small concentration of separating bonds (called "red" by Coniglio and Stanley[45]). This must also be true for the vibrations of, say, percolation clusters, and certainly has important effects on the detailed structure of the eigenmodes.[44] It is less obvious how important it would be in the averaged strains that one measures in the scattering experiments. Aerogels are also relatively rigid and air is certainly a very poor "solvent" for the silica grains. One would therefore expect a much more tightly interconnected fractal structure with $\delta > 1$ and relatively few dead end side branches, quite different from the tenuous structures one expects in wet gels. All this supports the assumptions we made in deriving Eq. (4.10). This should, however, be subject to reevaluation if experiments and a reasonable mechanical model for this regime become available.

As we noted, one expects both the polarized and depolarized light scattering to be described by the same expression $[I(q,\omega)]$, Eqs. (4.7) and (4.10), when ω is in the fractal range $\omega \gg \omega_{co}$. This reflected the large fluctuations and anisotropy in the vibrating density on the small relevant length scales.

On the scales relevant for phonons, the material is essentially homogeneous and one expects only polarized scattering (from longitudinal phonons). One therefore expects very different behavior for the polarized (Brillouin) and for the depolarized (Raman) scattering in the crossover region $\omega \sim \omega_{co}$ and in the phonon frequency range. Roughly, one would predict that the depolarized scattering from phonons is dominated by fluctuations in the local index of refraction and is therefore coherent only for regions of size ξ_{co}. The depolarized scattering will always decrease as ω decreases (and λ increases) in the phonon frequency range. One therefore expects a maximum in the depolarized scattering—$S_\perp(q,\omega)$—somewhere in the fracton frequency range even for small q ($q\xi_{co} \ll 1$). In particular, one expects no resonant maximum in the depolarized scattering when $vq = \omega \ll \omega_{co}$. Since for small q the position of this maximum is determined by crossover effects, one predicts a density dependence of its position, but the details cannot be derived from our arguments.

On the other hand, the polarized scattering can have a maximum in the phonon range, for sufficiently small q, and will then decrease rapidly with increasing ω in the crossover region.

The standard expression for the polarized scattering in the phonon regime is[1,2]

$$I(q,\omega) \propto (v^2q^2/\omega^2)\Gamma[(\omega^2+\Gamma^2-v^2q^2)^2+4\Gamma^2v^2q^2]^{-1}\ , \tag{4.11}$$

which is the phonon equivalent of a Lorentzian. This should cross over to Eq. (4.7) or (4.10), depending on the value of $q\xi_{co}$.

A theoretical estimate of the crossover effects is complicated by the direct crossover effects on $\underset{\sim}{e}$ that we discussed in Sec. III. As shown there, one expects the strain to increase by a factor s_l/l because of a change in the form of the fracton eigenfunctions as $l \to \xi_{co}$. One would guess that this will also decrease the depolarized scattering. Since this effect is not understood in detail, and may be model dependent, we cannot describe the details of the broad crossover region observed for the depolarized (Raman) scattering.[3]

In this context it is interesting to compare our results with the expression used in Ref. 2 to fit the Brillouin-(polarized-) scattering results. In Ref. 2 an attempt was made to extrapolate the standard expression for phonons [Eq. (4.11)] into the fracton regime by introducing a frequency-dependent velocity of sound $v = v(\omega)$ and $\Gamma(\omega)$ arranged so that they would have their proper values in the phonon range and cross over to

$$v(\omega)/l(\omega) \sim \Gamma(\omega) \sim \omega, \quad \omega \gg \omega_{co} \tag{4.12}$$

in the fracton frequency range. This gave a very impressive fit to the experimental results up to quite high frequencies. There are, however, serious problems with this approach quite aside from the fact that the derivation of Eq. (4.11) assumes coherent scattering (from phonons) over regions larger than the wavelength and, like all Lorentzian-broadening descriptions of scattering. In essence $\Gamma/\omega \ll 1$.

Using Eq. (4.12) in Eq. (4.11), one finds

$$S(q,\omega) \propto \omega^{-2(1+\bar{\tilde{d}}/D)}, \quad ql \ll 1, \quad \omega \gg \omega_{co} \tag{4.13a}$$

and

$$S(q,\omega) \propto \omega^{-2(1-\bar{\tilde{d}}/D)}, \quad ql \gg 1, \quad \omega \gg \omega_{co}\ . \tag{4.13b}$$

Thus the scattering, at constant q, is monotonically decreasing throughout the fracton regime with a weak crossover in the form of the decay—from (4.13b) to (4.13a) when $ql \sim 1$. When there is no maximum in the phonon regime, the interpolated function has a maximum in the crossover region that is completely controlled by the interpolation formulas for $v(\omega)$ and $\Gamma(\omega)$. Obviously, Eq. (4.13a) predicts a much steeper decay (with ω) than Eqs. (4.7), and (4.13b) even has a slope with the wrong sign when compared to (4.10). One concludes that the extrapolation of Eq. (4.11) into the fracton frequency range can, at most, be meaningful only in the crossover region and for the polarized scattering.

V. THE FRACTON WAVE FUNCTION

As noted in the Introduction, the most straightforward approach to the scattering problem is to write an ansatz for an eigenmode [Eq. (1.12)],

$$\mathbf{u}_\alpha(\mathbf{r},t)=\sum_i \mathbf{u}_\alpha(\mathbf{R}_i,t)\delta(\mathbf{r}-\mathbf{R}_i)$$

$$=u_\alpha\cdot\boldsymbol{\psi}_\alpha(\mathbf{r})\rho_0(\mathbf{r})e^{i\omega_\alpha t}\ , \tag{5.1}$$

and proceed from there. Our purpose in this section is to point out the difficulties and pitfalls of this approach and also to discuss the eigenfunctions.

The vector "eigenfunctions" $\boldsymbol{\psi}_\alpha$ are orthonormal on the fractal:

$$\int d\mathbf{r}\,\rho_0(\mathbf{r})[\boldsymbol{\psi}_\alpha(\mathbf{r})\cdot\boldsymbol{\psi}_\beta(\mathbf{r})]=\sum_i \boldsymbol{\psi}_\alpha^i\cdot\boldsymbol{\psi}_\beta^i=\delta(\alpha,\beta)\ . \tag{5.2}$$

This is, of course, just formal. $\boldsymbol{\psi}(\mathbf{r})$ is only defined *on* the fractal—i.e., only for $\mathbf{r}=\mathbf{R}_i$. In general, one does not expect that the vector field $\boldsymbol{\psi}_\alpha(\mathbf{r})$ can be interpolated with any degree of smoothness in the embedding space, at least for a random fractal. One can, nevertheless, make some general statements.

If the function $\boldsymbol{\psi}$ is localized in a region of size l_α, one must have, from normalization,

$$\langle\psi_\alpha^2\rangle\sim l_\alpha^{-D} \tag{5.3}$$

in this region and small elsewhere.

Because of the factor ρ_0 in Eq. (5.2), there are no exact orthogonality relations for the Fourier components of the $\boldsymbol{\psi}$ in spite of the fact that the low-frequency vibrations are plane-wave-like phonons with a well-defined (longitudinal or transverse) average polarization. Nevertheless, for any vibration one must have

$$\int d\mathbf{r}\,\rho_0(\mathbf{r})\boldsymbol{\psi}_\alpha(\mathbf{r})=\sum_i \boldsymbol{\psi}_\alpha^i\equiv 0 \tag{5.4}$$

from the translational invariance of the equations of motion, and this has to be approximately true in the region of size l in which ψ^2 is large. The sign must change. This should certainly be remembered in thinking about the ψ. By continuity, it also has some (not very serious) implications for the small-q Fourier transforms.

As already noted in Sec. III, there is some ambiguity in the definition of the strains for a fractal geometry. The stresses must, of course, follow the fractal geometry. A local *internal* strain should presumably be defined by the limit

$$(\underset{\sim}{e}_i)_{\text{int}}=\lim_{j\to i}\sum_j(\boldsymbol{\psi}_i-\boldsymbol{\psi}_j)/|\mathbf{R}_i-\mathbf{R}_j|\ . \tag{5.5}$$

It is reasonable to assume that $\boldsymbol{\psi}$ is smooth along the connected paths of the fractal and the strains should be definable if the limit $(j\to i)$ in Eq. (5.5) is taken along these paths and not along general directions in the embedding space. What one really wants are, however, the changes in density in the embedding space, which are not necessarily simply related to these internal strains. To calculate the scattering, one needs the true external changes in shape and density as seen by the scattered wave. Thus one wants, for scattering wave vector $\mathbf{q}$,

$$\underset{\sim}{e}_\alpha(\mathbf{q})\propto i\mathbf{q}^*\cdot\boldsymbol{\psi}_\alpha(\mathbf{q})\ , \tag{5.6}$$

where the Fourier transform $\boldsymbol{\psi}_\alpha(\mathbf{q})$ is

$$\boldsymbol{\psi}_\alpha(\mathbf{q})=\int d\mathbf{r}\,\rho_0(\mathbf{r})\boldsymbol{\psi}_\alpha(\mathbf{r})e^{i\mathbf{q}\cdot\mathbf{r}}\propto\sum_i e^{i\mathbf{q}\cdot\mathbf{R}_i}\boldsymbol{\psi}_\alpha^i\ , \tag{5.7a}$$

$$\boldsymbol{\psi}_\alpha(\mathbf{r})=\int d\mathbf{q}\,\boldsymbol{\psi}_\alpha(\mathbf{q})e^{-i\mathbf{q}\cdot\mathbf{r}}=\sum_i \boldsymbol{\psi}_\alpha^i\delta(\mathbf{r}-\mathbf{R}_i)\ . \tag{5.7b}$$

This is exact and one can, of course, calculate the scattering directly from Eqs. (5.7). Written in this way, the difficulties in handling the dependence of the vector field $\boldsymbol{\psi}(\mathbf{r})$ on $\mathbf{r}$ are then hidden in the dependence of its Fourier transform $\boldsymbol{\psi}(\mathbf{q})$ on its argument $\mathbf{q}$. Both are clearly very messy functions of their arguments. No one has, we believe, come up with a plausible direct ansatz for $\boldsymbol{\psi}(\mathbf{r})$ or for its Fourier transform that one could really use.

One procedure that has been used fairly widely in this (or related) context is to make an ansatz for the spherical average of $\boldsymbol{\psi}$:

$$\bar{\boldsymbol{\psi}}_\alpha(r)=\int d\Omega\,\boldsymbol{\psi}_\alpha(\mathbf{r})\sim\boldsymbol{\varepsilon}_\alpha(r)\exp[-(r/l_\alpha)^{d_\phi}]\ , \tag{5.8}$$

where l_α is the localization length and $\boldsymbol{\varepsilon}_\alpha(r)$ a unit vector whose orientation must depend on r. d_ϕ is then called the superlocalization index and it is tempting to identify it with σ. One can then proceed to calculate the radial derivatives (i.e., strains) from this ansatz.

As a radial average for a scalar function, this sort of expression does, we believe, make sense for the asymptotic behavior, and it is therefore useful when one is really interested in the asymptotic, large-r tails of ψ. For scalar fields it is also evident that one expects the angular average $\bar{\psi}(r)$ to be much smoother—in r—than the bare function $\psi(\mathbf{r})$ (in $\mathbf{r}$). The meaning of the (in practice, inevitable) neglect of the radial dependence of $\boldsymbol{\varepsilon}(r)$ is less clear.

The superlocalization index d_ϕ for the asymptotic decay can be identified with the index σ for the strain [Eq. (2.10)] when there are geometrical reasons for doing this, e.g., for a linear chain or for a crumpled sheet. Even when this is justified, one is not necessarily justified in using this expression for the strains for the elastic problem. More precisely, the index d_ϕ in Eq. (5.8) *never* shows up explicitly in the calculation of the strain from this ansatz. In the region where ψ^2 is significant, one must have

$$(\partial\psi/\partial r)^2\sim\psi^2/l^2\sim l^{-(2+D)} \tag{5.9}$$

for any reasonably well-behaved localized function $\psi(r)$ that is localized with localization length l and does not oscillate wildly on some much smaller length scale. We have used Eq. (5.3) for ψ^2, which follows from normalization, and the estimate of the derivatives follows from elementary geometrical considerations. The asymptotic dependence on d_ϕ does not enter at all. Thus the detailed form of the function $\boldsymbol{\psi}$, and, in particular, the superlocalization index d_ϕ in Eq. (5.8), cannot show up in taking the derivatives of such an ansatz. One obtains an identical result if one looks directly at the Fourier transforms (of ψ

or of $\partial\psi/\partial r$) and does the calculation correctly.

When one can define an internal length $s \propto r^{\sigma}$ and the function decays like $e^{-\alpha s}$, it is, of course, trivially true that $d_{\phi}=\sigma$. The internal strain ($\partial u/\partial s$) is then given by

$$(\underset{\sim}{e}_{\rm int})^2 \sim (d\psi/ds)^2 \sim (d\psi/dr)^2 (ds/dr)^{-2} \sim l^{-D-2\sigma} , \tag{5.10}$$

which is consistent with Eq. (2.10). The anomalous power of l (2σ instead of 2) comes from the derivative of s and not from the functional form of ψ. It is far from obvious that the assumed relationship between the asymptotic decay and the local strain does, in fact, hold for a fractal geometry. Moreover, from a purely empirical point of view the strains are accessible, as in the scattering experiments, but presumably also in properly analyzed simulations.[44]

Other problems with such a simple ansatz are that it does not exhibit the transition from coherent to incoherent scattering because the structure factor ρ_0 is missing and also that it cannot exhibit the depolarized scattering. Neither the scaling behavior of the density nor that of the rigidity are included. We therefore believe that a meaningful calculation requires an explicit ansatz for the correlation functions—which can, of course, be averaged. It may be possible to do this better and in a more detailed way than we have done here.

VI. DISCUSSION AND COMPARISON WITH EXPERIMENT

Our purpose in this paper was to discuss the implications of a self-similar scaling structure for the vibration spectrum. This is known to be relevant for the silica aerogels, which have such a structure at short distances. We have tried to do this in as general a form as possible, in part because the relevant results are not available in the literature, so that it seemed necessary to discuss the underlying scaling assumptions, the definition of a strain for a fractal structure, and the role of the static fractal structure factor in Sec. II and the way the crossover comes in (in Sec. III) before proceeding to the Mie-type calculation of the scattering in Sec. IV. Much of the literature on fractons is also concerned with scalar problems, which differ from the discussion of the vibrations in important ways. We have also tried to avoid assumptions based on experience with percolation clusters and other very tenuous fractals which, we believe, cannot be relevant to aerogels. Thus, most of this paper is devoted to the detailed development of the scaling, fracton model for the vibrations of a fractal object in as general a way as seemed feasible. We have also tried in Sec. III to show the connection to scattering theory—thus extending the brief remarks on this subject in Ref. 15. We believe that this discussion clarifies the discrepancies and shows under what conditions fractonlike excitations can appear in dense amorphous materials. Whether these conditions are actually relevant to real materials is, unfortunately, a different question, one which will have to be settled by experiments and more detailed modeling.

We would also like to comment that our discussion casts very serious doubts on the meaning of any effective-medium calculation for this type of problem. Any such calculation treats the disorder in a very rough averaged way and we cannot imagine how the subtle effects of the disorder on the polarization on coherence and on the observable strains could conceivably be incorporated. We therefore feel that fits of experiments to such calculations should be treated with extreme caution and are probably meaningless.

As stated in the Introduction, the motivation for this work was the experiments on inelastic light scattering from aerogels.[1-8] Our relevant results are Eqs. (4.7) and (4.10). Equation (4.7) is similar to that used in Ref. 3, except for the different interpretation of the measured indices, and a scaling regime of suitable form was indeed found in the depolarized-scattering experiments. The analysis in that reference implicitly assumed $\delta=1$. If one gives up this assumption, which is hard to justify, one concludes that the experiment measures the scaling index $\bar{\sigma}$, or equivalently δ, which is a new index. It cannot be considered a crosscheck on other determinations of D and $\bar{\bar{d}}$. The analysis in Ref. 8 gives $\delta=1.15$. The Raman experiments of Tsujimi *et al.* also show a density-dependent maximum in the depolarized scattering in the fracton frequency range that is consistent with our conclusions in Sec. IV. It would be very desirable to have polarized-scattering results for the same frequency range to check our conclusion that the two components of the scattering must scale in the same way in the fracton regime. All the available data for which one can make such a comparison end at frequencies still well inside the broad crossover region for the Raman scattering from the same aerogels.

There are also no experiments for which the crossover from Eq. (4.7) to Eq. (4.10) would be relevant for either polarized or depolarized scattering. We note that the observation of such a crossover would constitute a direct measurement of the frequency-dependent length scale $l(\omega)$, which could be compared with scaling predictions of indices that have been measured. They could also be compared directly with the determination of the dispersion relation from the comparison of the crossover frequency ($\omega_{\rm co}$) and crossover length ($\xi_{\rm co}$) in Ref. 8. Our analysis would predict that the "universal" position of the Brillouin-scattering maximum—when $\omega > \omega_{\rm co}$—which is observed and agrees with the predictions of the extrapolation of Eq. (4.11) is also a crossover effect. The maximum should become frequency dependent at higher frequencies or for lighter samples.

Finally we note that the crossover length $\xi_{\rm co}$ is determined independently from neutron scattering[5] and from the optical scattering experiments.[1,2,8] As predicted, the two values scale the same way, but the dynamically determined correlation length seems considerably larger (by about a factor of 5).[8] The authors of Ref. 8 suggest that this might be a question of definition, which is, of course, quite possible. We would, however, like to add that the two length scales do not have to be identical. As we have shown in Sec. III the elastic Ioffe-Regel frequency, which is the quantity actually measured in the scattering experiments, is determined by the (scale-invariant) variance $\mathcal{V}_g$.

It follows from the scaling ansatz [Eq. (3.10)] that it has to scale like the crossover frequency, but the numerical factors between different determinations of the crossover certainly depend on the actual form of the function $f(x)$ defined in Eq. (3.10) and therefore on the physical structure of the fractal. A scaling argument is simply not sufficient to say anything about this ratio, as emphasized there.

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[32]For convenience, we shall assume this in the discussion below. Actually, localization plays no real role in the scattering from a random fractal. An extended mode on such a fractal would still have essentially incoherent scattering from "one-wavelength blobs" because of the structure factor $\rho_0(\mathbf{r})$.

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