A study of fractons in polymers

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Inelastic neutron scattering and low-frequency Raman scattering have been employed to study low-energy vibrational excitations in PMMA. The density of vibrational states obtained via neutron scattering in the 2.5–10 meV range follows a power law in energy, with a spectrum corresponding to a fracton dimension $\tilde{d} = 1.8 \pm 0.05$. Over the same spectral range, the low-frequency Raman spectrum provides additional information on fractons. The Raman intensity is also described by a power law in frequency, but the spectral index corresponds to a different fractal dimension than in the case of neutron scattering. The model that we employ for the scattering of light by fractons makes it possible to express the slope of the intensity as a function of frequency on a log-log scale in terms of the spectral dimension and fracton superlocalization index d_Φ. A comparison with experimental data yields $d_\Phi \approx 1.5$. Similar values of $\tilde{d}$ and d_Φ have also been found for deuterated PMMA.

1. INTRODUCTION

Interest in the vibrational properties of disordered fractal systems has lately been on the rise.[1-13] A disordered structure that exhibits self-similarity can be described by a noninteger Hausdorff or fractal dimension D that is less than the dimension d of the Euclidean space in which the fractal object is embedded. For a fractal system, the mass rises as r^D and the density accordingly falls as r^{D-d}. Vibrational excitations of a fractal system—fractons—are localizable, and they have a density of states described by yet another fractal index, the spectral or fracton dimension[1,3] $\tilde{d}$:

$$g(\omega) \propto \omega^{\tilde{d}-1}. \tag{1}$$

In real systems like polymers, aerosols, and the like, fractal structure is manifested only over a certain range of scales $L < r_0$, while on scales $L \gg r_0$ the systems act like an ordinary continuous medium of dimension d. There is accordingly some limiting frequency ω_0 at which the system passes from the fractal regime represented by (1) to the ordinary phonon regime, where

$$g_{ph}(\omega) \propto \omega^{d-1}, \quad \omega \ll \omega_0. \tag{2}$$

Percolating clusters comprise a classic model imbued with fractal properties, for which it has been shown[1] that $\tilde{d} = 4/3$ for all values of $d \geqslant 2$.

Little is presently known about the localization properties of fractons in a fractal. It has been suggested[4,14] that the wave function of a fracton is superlocalized, taking the form

$$|\psi(r)| \propto l_\omega^{-D/2} \exp[-(r/l_\omega)^{d_\Phi}], \tag{3}$$

where l_ω is a frequency-independent localization length for a fracton of frequency ω, and $d_\Phi > 1$ is the so-called superlocalization index. The data relating to d_Φ are inconsistent. It has been claimed,[15] for example, that

$$d_\Phi = D/\tilde{d} \tag{4}$$

for percolating clusters. In two dimensions, it is known[16] that $D = 91/48$ for a percolating cluster, and thus $d_\Phi = 1.42$. Two-dimensional fractal systems have also been modeled numerically. Detailed calculations[17] based on the equations of motion of percolating clusters have yielded a surprisingly high superlocalization index, $d_\Phi \approx 2.3 \pm 0.1$. On the other hand, numerical modeling of a similar system[18,19] suggests that there is no superlocalization at all, $d_\Phi = 1$. Likewise, numerical modeling of the vagaries of an ordered fractal—the Sierpinski gasket[20]—has also failed to confirm superlocalization. Clearly, then, further study is required if we are to ascertain the value of d_Φ.

In recent years, the study of fractons has been actively pursued using low-energy inelastic neutron scattering and low-frequency Raman and Brillouin scattering.[5-13] Inelastic neutron scattering makes it possible to determine the density of vibrational states (1), and thus the value of the spectral dimension $\tilde{d}$. This quantity has been derived for aerosols[8]—experimentally, the best-studied of the fractal systems—epoxy resins,[6,21] and polymers.[10-12] The vibrational-state density spectra that have been obtained for these systems have confirmed that a fracton regime does indeed exist, as given by (1) with $\tilde{d} = 1.3$–2.0, and they have also helped establish both the low-energy boundary at ω_0, where bulk phonon behavior takes over, and the high-energy boundary at ω_1, where the fracton localization length is of the same order as the size of the molecule or cluster composing the fractal.

Low-frequency Raman scattering of light has been utilized to study fractons in these same materials—particularly in preliminary investigations, since these experiments are much easier to carry out.[5,6,9-12] On the whole, the Raman spectra for scattering from fractons resemble the neutron scattering spectra. A fairly general formula derived by Shuker and Gammon[22] indicates that the Raman scattering intensity in a disordered material is proportional to $g(\omega)$, and that the coupling constant $C(\omega)$ varies smoothly with frequency:

$$\frac{I}{n+1} = C(\omega) g(\omega)/\omega. \tag{5}$$

Here $n + 1$ is the Bose factor. The factor $C(\omega)$ in (5) obviously implies that data derived from the inelastic scattering of light will be less directly related to structural properties than neutron scattering data, and they may well depend on all of the fractal exponents mentioned above. The theory of low-frequency Raman scattering from fractons plays an important role here, as it should enable one to interpret the

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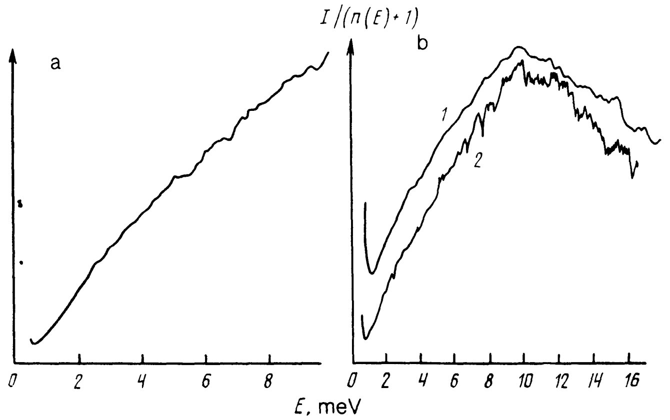

FIG. 1. Low-energy spectra of PMMA at $T = 300$ K. a—Neutron scattering data, thin-film sample; b—Raman scattering spectrum for thin-film sample (1) and bulk sample (2).

experimental Raman scattering data in terms of the fractal parameters of the medium.

The first such model was proposed by Duval *et al.*,[5,6] with a Raman spectrum $I/(n+1) \propto \omega^{\mu}$, where μ is given in terms of D, $\tilde{d}$, and d_{Φ}. The expression obtained by Boukenter et al.,[6] however, which provides a reasonably good description of their experimental Raman scattering data in epoxy resins, leads to an unacceptable value $d_{\Phi} < 1$ for the experimental data that we consider here (for other polymers). In Sec. 3 of the present paper, we present a model of Raman scattering from fractons that yields a good fit to all of the experimental data of which we are aware.

Note also that Alexander[2] has proposed a new fractal exponent σ, which reflects the scaling properties of the deformation tensor, to describe the Raman scattering spectrum. That model will be discussed in more detail in Sec. 4.

Our current objective has been to carry out a close investigation of fractons in a typical kind of fractal system, a polymer—specifically, polymethylmethacrylate (PMMA). As we have shown previously,[10–12] PMMA exhibits fractal behavior on scales $L \leqslant 30$–50 Å. We have made a comparative study of both ordinary and deuterated PMMA using low-energy inelastic neutron scattering and low-frequency Raman scattering of light. The model that we propose and the spectral dimension obtained from the neutron scattering experiment imply a superlocalization index $d_{\Phi} = 1.5$.

2. THE EXPERIMENT

We chose to study the polymer polymethylmethacrylate (PMMA), which, like all disordered polymers, exhibits fractal structure. Thin-film samples of PMMA 30–50 μm thick were prepared by the method described in detail in Ref. 23. Commercially prepared thin-film samples of deuterated PMMA were obtained from the Izoton company.

Time-of-flight measurements of the neutron scattering spectrum were made at scattering angles 30, 45, 60, 75, and 90°. The resulting data were processed in the usual way, with all necessary corrections.[24] In the incoherent approximation, making no assumptions about the model involved, one can obtain directly from the neutron scattering data the so-called generalized vibrational spectrum $\theta(E)$, which is related to $g(E)$ by

$$\theta(E) = \sum_i \sigma_i c_i M_i^{-1} |\mathbf{e}_i(E)|^2 \exp(-2W_i) g(E), \qquad (6)$$

where σ_i, M_i, c_i, and $\mathbf{e}_i$ are the scattering cross section, mass, number density, and vibration vector of atoms of type i, and W is the Debye-Waller factor. Energies $E < 5.5$ meV involve averaging over momentum transfers in the range 1–3 Å^{-1}. In Fig. 1 we show the low-energy $\theta(E)$ and Raman spectra for ordinary PMMA, and in Fig. 2, for deuterated PMMA. There is a large contribution to the Raman spectrum of the thin-film samples at $E < 3$ meV from quasielastic scattering, and at $E \gtrsim 12$ meV from photoluminescence. These additional contributions complicate the processing of the Raman spectrum and the identification of scattering from vibrational excitations. For comparison, we have also plotted the Raman spectrum of a bulk sample of PMMA at $T = 100$ K in Fig. 1b; there the quasielastic scattering level is much lower.

In Fig. 3, we show the density of states and Raman scattering spectra in log-log coordinates. For the ordinary PMMA samples in the energy range $E \geqslant 2.5$ meV we observe power-law behavior, with spectral indices $\mu = 0.8 \pm 0.05$ for $\theta(E)$ and $\mu = 1.7 \pm 0.15$ for Raman scattering. The

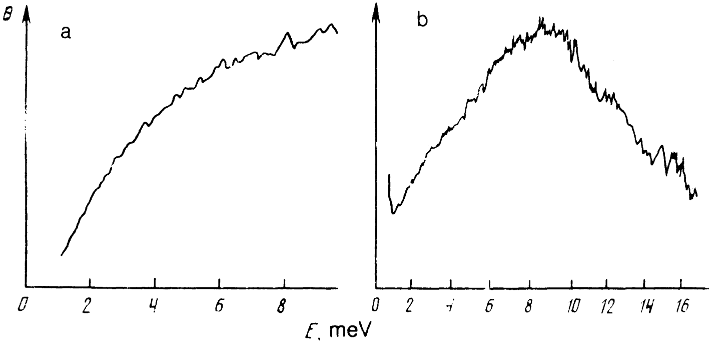

FIG. 2. Low-energy spectrum of deuterated PMMA films. a—Neutron scattering data; b—Raman spectrum.

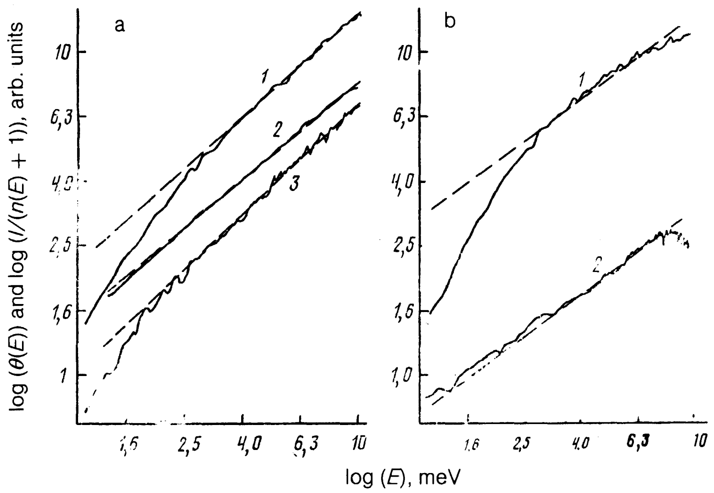

FIG. 3. Log-log plots of experimental data for different samples. a— PMMA: density of states for thin-film sample, $\mu = 0.8$ (1); normalized Raman scattering intensity for thin-film sample, $\mu = 0.7$ (2); normalized Raman scattering intensity for bulk sample, $\mu = 0.83$ (3); b—deuterated PMMA: density of states, $\mu = 0.65$ (1); normalized Raman scattering intensity, $\mu = 0.66$ (2).

large error in the latter is related to the aforementioned contributions of quasielastic scattering of light and photoluminescence. For a bulk sample of PMMA, where those effects are negligible, the spectral index can be determined quite accurately: $\mu = 1.83 \pm 0.05$ (Fig. 3). In deuterated PMMA, the power-law behavior is less clear-cut, and the corresponding spectral index is smaller: $\mu = 0.65 \pm 0.05$ for $\theta(E)$ and $\mu = 1.65 \pm 0.15$ for Raman scattering.

3. A MODEL FOR THE INELASTIC SCATTERING OF LIGHT FROM FRACTONS

We now consider the inelastic scattering of light from vibrational excitations in a medium with fractal dimension D. Small oscillations of atoms lead to time-dependent fluctuations in the dielectric constant; light scatters from these inhomogeneities with an attendant change in frequency. The fluctuations in the dielectric constant are proportional to the gradient of the atomic displacements resulting from vibrational excitations, the elastooptic coefficients being the proportionality constants. Omitting constants unrelated to the frequency dependence, we may write the matrix element M_i for the interaction of light with a fracton located at the point r_i in the form

$$M_i \propto \int d^D r \exp(i\mathbf{k}\mathbf{r}) \nabla\psi(\mathbf{r}-\mathbf{r}_i) \exp(-i\mathbf{k}'\mathbf{r}). \tag{7}$$

Here $\mathbf{k}$ and $\mathbf{k}'$ are the momenta of the incident and scattered photons, and $\psi(r)$ is the fracton wave function (we have omitted any reference to polarization, which has no bearing on the present development). As noted above, the fracton wave function falls off faster than exponentially, and is given by

$$|\psi(r)| \propto l_\omega^{-D/2} \exp[-(r/l_\omega)^{d_\Phi}], \tag{8}$$

where l_ω is the localization scale length of a fracton of frequency ω. We assume that this is the only parameter with dimensions of length relevant to the fracton regime.

Scaling considerations enable us to determine the frequency dependence:[1,2]

$$l_\omega \propto \omega^{-\tilde{d}/D}. \tag{9}$$

The superlocalization index d_Φ in (8) allows for the fact that the wave function along a fractal will fall off somewhat more rapidly in the Euclidean embedding space than usual (hence the term "superlocalization index"). In other words, the localized fracton wave function has a simple exponential dependence on the length l along the fractal:

$$|\psi(l)| \propto \exp(-\alpha l). \tag{10}$$

The gradient of the fracton wave function along a fractal, which appears in (7), is

$$\nabla_l |\psi_\omega| \propto l_\omega^{-d_\Phi} \psi_\omega. \tag{11}$$

We also note that to order of magnitude, the scattering wave vector $\mathbf{q} = \mathbf{k}' - \mathbf{k}$ is approximately $2\pi/\lambda$, where λ is the wavelength of the scattered light. Since $\lambda \sim 10^{-4}$ cm is much greater than the localization length $l_\omega \lesssim 10^{-6}$–$10^{-7}$ cm, we can replace $\exp(i\mathbf{q}\mathbf{r})$ in (7) by unity. As a result, the scattering matrix element becomes

$$|M_i| \propto l_\omega^{-d_\Phi} \int d^D r \, |\psi_\omega| \propto l_\omega^{D/2-d_\Phi}. \tag{12}$$

It must be borne in mind that fractons localized at different points r_i may differ in phase, so that the matrix element will contain an additional factor $\exp(i\alpha_i)$, where α_i is the overall phase:

$$M_i \propto l_\omega^{D/2-d_\Phi} \exp(i\alpha_i). \tag{13}$$

The total matrix element for scattering from all fractons is obtained by summing over the matrix elements given by (13). Hence, the square of the matrix element, which determines the scattering intensity from the whole sample, is

$$|M|^2 = \Big|\sum_i M_i\Big|^2 \propto l_\omega^{D-2d_\Phi} \sum_{i,j} \exp[i(\alpha_i - \alpha_j)]. \tag{14}$$

This quantity depends on the correlation properties of the phase factor. By analogy with Ref. 6, we consider the following two situations.

1. Incoherent oscillations. The phases α_i are random and uncorrelated. This means that

$$\langle \exp[i(\alpha_i - \alpha_j)] \rangle = \delta_{ij}, \tag{15}$$

where the averaging takes place over distinct pairs of fractons. Then

$$|M|^2_{\text{incoh}} \propto l_\omega^{D-2d_\Phi}. \tag{16}$$

2. Coherence length of vibrations of the order of fracton localization length l_ω. The sum in (14) is now proportional to l_ω^D, and for $|M|^2_{\rm coh}$ we obtain

$$|M|^2_{\rm coh} \propto l_\omega^{2D-2d_\Phi}. \tag{17}$$

Fermi's Golden Rule then says that the scattered intensity, normalized by the Bose factor, is

$$\frac{\omega I}{n+1} \propto |M|^2 g(\omega). \tag{18}$$

Using the expression (1) for the density of states and the dispersion relation (9) for fractons, we obtain

$$\frac{\omega I}{n+1} \propto E^\mu, \tag{19}$$

where in the incoherent case

$$\mu=\mu_{\rm incoh}=\frac{2\tilde{d}d_\Phi}{D}-1 \tag{20}$$

and in the coherent case

$$\mu=\mu_{\rm coh}=\frac{2\tilde{d}d_\Phi}{D}-1-\tilde{d}. \tag{21}$$

These are the expressions for μ that we compare below with the experimental data on the inelastic scattering of light and neutrons in polymers.

4. DISCUSSION

It is clear from Fig. 3 that in the energy range $E_{\rm min} \approx 2$ meV to $E_{\rm max} \simeq 10$ meV, both the density of vibrational states and the Raman scattering intensity in thin-film PMMA exhibit a power-law energy dependence. Moreover, the power-law behavior of the deuterated PMMA spectrum is less well-defined. According to the fractal model of polymer structure, this spectral range corresponds to the fracton regime.

The high value of the depolarization, $\rho = 0.7$, suggests that transverse vibrational modes contribute more to the low-frequency spectrum than longitudinal modes.

At $E<2$ meV, ordinary three-dimensional phonons dominate. The slope of the curves increases, and at low enough energies it should tend to $\tilde{d}=3$ for the density of states and $\mu = 4$ for Raman spectra. The point of inflection of the spectrum (located at E_0) yields an estimate of the characteristic size of a fractal cluster in the polymer: $L \approx v_t/E_0$, where v_t is the speed of sound for transverse vibrational modes, which make the largest contribution to the low-frequency spectrum. For $v_t = 1.22\cdot 10^5$ cm/sec (Ref. 25), we have $L \approx 30$ Å in PMMA.

For $E \gtrsim 10$ meV, the break in the power-law behavior of the spectra is due to the boundary of the vibrational spectrum (the analog of the Debye limit in the ordinary phonon spectrum). An estimate indicates that the frequency range of interest corresponds to vibrations with $\lambda \approx 5$–8 Å, which is just about the size of a link in the polymer chain. Thus, the lower limit on the size of a fractal structure is that of a molecular link.

In the fracton region of the spectrum, the slope of the curve for the density of states yields a spectral dimension $\tilde{d} = 1.8$ for a thin-film sample of PMMA, and $\tilde{d} = 1.65$ for deuterated PMMA.

Let us turn now to the Raman scattering data. The exponent μ in (19) depends in a more complicated way on the actual parameters of the fractal system than it did for neutron scattering, since the interaction of molecular vibrations with light is governed by the gradient of the wave function. The fractal dimension for an ideal linear disordered polymer is $D=2$. A number of experimental techniques[26–28] have made it possible to determine D in real polymers, with the result that $D = 2.0$–2.2. Then, knowing μ, D, and $\tilde{d}$, and making use of the model described in Sec. 3, we can determine the superlocalization index:

$$d_{\Phi,\rm incoh}=(\mu+1)D/2\tilde{d}, \tag{22}$$

$$d_{\Phi,\rm coh}=(\mu+1+\tilde{d})D/2\tilde{d}. \tag{23}$$

Substituting the previously derived spectral dimension $\tilde{d} = 1.8$ and $D = 2$, we obtain $d_{\Phi\ \rm incoh} \approx 1.5$ and $d_{\Phi\ \rm coh} \approx 2.5$ for ordinary samples of PMMA, and $d_{\Phi\ \rm incoh} = 1.6$ and $d_{\Phi\ \rm coh} = 2.6$ for deuterated samples. As might be expected, the value of $d_{\Phi\ \rm coh}$ obtained in this way is unrealistic. The point is that in a chaotic polymer chain with segment length l_0, the distance along the chain between some origin O and a point N segments away is $L = Nl_0$; on the other hand, motion along the chain can be viewed as a random walk in the embedding space. After N steps, the distance to the original point in a Euclidean space will be $r \approx (l_0^2N)^{1/2} \approx (Ll_0)^{1/2}$, so that $L \sim r^2$. We therefore expect that for real polymers, $d_\Phi \lesssim 2$ always. Comparing this with the two results for d_Φ, it would appear that the one for $d_{\Phi\ \rm incoh}$ is the correct one, implying that fractons are incoherent.

We next consider a number of other models for inelastic scattering of neutrons and light from fractons, and we apply Eq. (22) for d_Φ to a variety of experimental data obtained in different materials.

Instead of the superlocalization index d_Φ, Alexander[2] has introduced another more phenomenological parameter σ, which determines the polarization tensor e_{ij}, with

$$e_{ij}=\frac{\partial u_i}{\partial r_j}+\frac{\partial u_j}{\partial r_i}. \tag{24}$$

If u_i and u_j are the displacements of the two elements comprising the fractal, which are separated by a distance r from one another in the external embedding space, the parameter σ may be introduced via the phenomenological relation

$$e^2 \propto \frac{(u_i-u_j)^2}{l^2}, \tag{25}$$

where $l \sim r^\sigma$. One argument for using σ is that the deformation tensor is more directly related to the scattering intensity. In this model, the expression for the exponent [Eq. (20)] is similar to the one we obtained, but with σ instead of d_Φ:

$$\mu=\frac{2\tilde{d}\sigma}{D}-1. \tag{26}$$

Using the experimental data obtained in our present investigation, the model proposed by Alexander[2] yields $\sigma = 1.5$. This is the same as the value previously obtained for aerosols,[13] and agrees with the value of d_Φ obtained for PMMA in the present work.

Somewhat of a different expression was obtained for μ in Ref. 6,

$$\mu=\frac{2\tilde{d}d_{\Phi,\rm incoh}}{D}+\tilde{d}-1, \tag{27}$$

TABLE I.

Material	$3-\tilde{d}$	$\tilde{d}$	μ	$d_{\Phi,\mathrm{incoh}}$	$d_{\Phi,\mathrm{coh}}$
DGEBA EDA 1X*	1,56	1,44	1,63	1,83	0,83
DGEBA EDA 2X*	1,56	1,44	1,67	1,85	0,85
Amorphous DGEBA*	1,48	1,52	1,68	1,68	0,68
PMMA (thin-film sample)	1,20	1,8±0,05	1,70	1,50	0,50
Bulk PMMA	–	–	1,83	1,54	0,54
Deuterated PMMA	1,35	1,65±0,05	1,65±0,15	1,61	0,61

* Data from Ref. 6.

which was thereupon used to describe the scattering of light in DGEBA epoxy resins. For the polymers that we have investigated here, Eq. (27) yields an unacceptable value for d_Φ, $d_\Phi \approx 0.5$, which is inconsistent with the theoretical requirement[4,14] that $d_\Phi > 1$. Other definitions of d_Φ were also invoked in Refs. 6 and 21, where use was made of the suggestion put forward in Ref. 29 that the superlocalization index d_Φ should be identified with the power-law exponent ζ that enters into the expression for the resistance of the fractal. The resistance R between two points on the fractal that are separated by a distance L ($L \ll \xi$, where ξ is the correlation length in the percolation theory) is given by $R \propto L^{\zeta}$. It has been shown[3] that

$$\zeta = \frac{(2-\tilde{d})D}{\tilde{d}}. \tag{28}$$

For this particular case, the expression relating μ and $\tilde{d}$ is[6,21]

$$\mu = 3-\tilde{d}. \tag{29}$$

which has been used to analyze the Raman scattering spectra of a number of gels and polymers and thereby obtain values of $\tilde{d}$ that are quite close to the experimental values.[6,21] This last result, however, was probably accidental, being related to the small value $\tilde{d} = 1.44$. In our case, $\tilde{d} = 1.8$, and (20) yields $\mu = 1.2$, which is significantly different from the experimental value $\mu = 1.7 \pm 0.15$. The fact that (29) is in error is also apparent from a more careful analysis of previous results on DGEBA polymers[21] (Table I): μ increases with increasing $\tilde{d}$, rather than decreasing as predicted by (29).

In Table I, we have listed the experimental values of μ, $\tilde{d}$, and $3-\tilde{d}$ for a number of materials; we also give the value of d_Φ predicted by various models—in particular, using (22) and (27). Clearly, (27) yields $d_\Phi < 1$ even for bulk PMMA, the Raman spectrum of which has been measured to high accuracy. The values of d_Φ obtained from (22) are in fact consistent with the theoretical bounds (for an ideal linear disordered polymer, $1 < d_\Phi < 2$). Furthermore, d_Φ is observed to grow as the spectral dimension $\tilde{d}$ decreases. The latter is a fairly obvious result, as lowering of the spectral dimension corresponds to increasing one-dimensionality of vibrational excitations and an increase in the geometrical factor d_Φ.

CONCLUSION

In the work described here, we have carried out a comprehensive investigation of fractal vibrational excitations in polymers, using inelastic scattering of both light and neutrons. The scale invariance that characterizes the structure of a fractal cluster results in a power-law scattering spectrum in the fracton regime. However, the spectral indices for inelastic neutron scattering and Raman scattering of light depend quite differently on the fractal dimension. In the context of the model proposed in Sec. 2, this makes it possible to combine two types of experimental data to determine the spectral dimension and superlocalization index in polymers. An analysis of several formulas previously used to express the Raman scattering intensity in terms of fractal dimension has shown that our experimental data do not conform to them for any reasonable values of those dimensions. On the other hand, the value $\sigma \approx 1.5$, as determined in Ref. 2, turns out to be the same in polymers as in aerosols.[13]

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