

Neutron inelastic scattering from fumed silica

J. H. Page

Department of Physics, University of Manitoba, Winnipeg, Manitoba, Canada R3T 2N2

W. J. L. Buyers, G. Dolling, and P. Gerlach*

Atomic Energy of Canada Limited, Chalk River Nuclear Laboratories, Chalk River, Ontario, Canada K0J 1J0

J. P. Harrison

Department of Physics, Queen's University, Kingston, Ontario, Canada K7L 3N6

(Received 29 January 1988; revised manuscript received 30 August 1988)

Neutron inelastic scattering measurements have been performed on samples of fumed silica ("Cab-O-Sil") as a function of energy, momentum-transfer vector Q , and temperature T , and compared with analogous results for amorphous quartz. No evidence is seen for a sharp crossover in the vibrational density of states of Cab-O-Sil between phonon and fracton regimes, but the inelastic scattering shows anomalous behavior in several respects. Neither the T dependence nor the Q dependence nor the absolute intensity of the scattering from Cab-O-Sil is in accord with simple phonon models.

I. INTRODUCTION

Due to its technical and scientific importance, the process of aggregation of small particles to form larger clusters has been of increasing interest in recent times. Structural studies (e.g., see Refs. 1–3) have shown that many of these systems exhibit scale invariance and can be described as fractals,⁴ with values of the fractal dimension D that depend on the aggregation kinetics. However, considerably less work has been done experimentally to investigate their dynamical properties, such as the vibrational density of states $g(\omega)$. Alexander and Orbach⁵ were the first to predict that the quantized vibrational states of a fractal, referred to as fractons, are characterized by an additional dimension \bar{d} such that their density of states $g(\omega) \propto \omega^{\bar{d}-1}$. Estimates of \bar{d} range from the Alexander-Orbach conjecture,⁵ $\bar{d} = \frac{4}{3}$ for scalar elasticity, to $\bar{d} = 0.9$ for the bond-bending model in three dimensions,⁶ implying that the frequency dependence of $g(\omega)$ is much weaker for fractons than for the more familiar phonon excitations of a d -dimensional Euclidean system [$g(\omega) \propto \omega^{d-1}$ in the Debye regime]. Another consequence of the small values of \bar{d} is that the fracton modes are expected to be localized.⁷

To look for experimental evidence of fracton excitations, Page and McCulloch⁸ and Courtens *et al.*⁹ have used ultrasonics and Brillouin scattering to study the vibrational modes of sintered powders and silica aerogel. Both systems were shown to be physical realizations of percolating structures above threshold, for which fractal behavior is expected on length scales between the primary particle size and the correlation length ξ . The behavior of the velocity and attenuation damping of the modes as the wavelength λ approaches ξ from above was taken as evidence for a crossover from phonons for $\lambda > \xi$ to fractons for $\lambda < \xi$. However, even though values of \bar{d} could be deduced from the scaling behavior of the data,

there is as yet little direct experimental information on the fracton density of states in such aggregated systems.

In the present paper, we report a study of coherent elastic and inelastic neutron scattering from Cab-O-Sil,¹⁰ a fumed silica which consists of three-dimensional branched aggregates of amorphous SiO_2 spheres. The sphere radius as deduced from Brunauer-Emmet-Teller (BET) gas-adsorption isotherms varies from $a = 35 \text{ \AA}$ to 140 \AA depending on the grade. The aggregates have an open random structure, forming a white fluffy powder whose density is 2% of that of solid SiO_2 . Electron micrographs of Cab-O-Sil aggregates have recently been analyzed³ to determine a fractal dimension $D = 1.71 \pm 0.05$, with fractal behavior extending over nearly 2 orders of magnitude in length scale. This result is in good agreement with an earlier measurement¹¹ $1.55 < D < 1.85$ and with the prediction of the diffusion-limited cluster aggregation model,¹² $D = 1.75$. It also agrees with the value of $D = 1.81 \pm 0.1$ determined from combined light scattering and small-angle neutron scattering (SANS) data.¹³ However there is disagreement with the results of other SANS measurements¹⁴ on compacted samples ($D = 2.6 \pm 0.1$), possibly due to difficulties in accounting for the distribution of aggregate sizes and the effects of cluster entanglement and to the rather limited range of scattering vectors Q over which the data were sensitive to the fractal structure.¹⁵

In such aggregates, we expect a crossover in the vibrational spectrum at a frequency ω_c determined by the size of the primary particles. For $\omega > \omega_c$, modes characteristic of bulk silica will be excited, but for lower frequencies corresponding to wavelengths greater than the particle size, these modes will be replaced by fracton excitations. A simple mode-counting argument, equating the number of fracton modes to the number of Debye phonon modes which are cut off from the bulk silica spectrum below ω_c , gives $\omega_c = 3(\pi/6)^{1/3}v_D/a$, where v_D is the Debye velocity

for bulk silica. For $a = 35 \text{ \AA}$ and $v_D = 4.1 \times 10^3 \text{ m/s}$, this gives $\omega_c/2\pi = 0.45 \text{ THz}$, a frequency which is easily accessible by neutron scattering techniques. A simple model¹⁶ developed for amorphous materials can be used to predict a qualitative change in the inelastic neutron scattering from Cab-O-Sil at ω_c , reflecting the difference in the density of states $g(\omega)$ for the phonon and fracton regimes. We find, however, that our experimental data, presented in Sec. II, do not show any evidence of a sharp crossover in the frequency range 0.3–0.7 THz; this result suggests that the crossover from phonon to fracton modes occurs at a lower frequency, consistent with the recent SANS data of Hurd *et al.*¹³ who find that the primary particles are larger than expected from BET measurements because of the effects of surface roughness. Our inelastic scattering data, which exhibit several novel features that distinguish the results for Cab-O-Sil from those for bulk silica, are discussed in Sec. III and attributed to the importance of anharmonic interactions in this system.

II. EXPERIMENT

The experiments were all performed on grade EH-5 Cab-O-Sil which has a surface area of $400 \text{ m}^2/\text{g}$. For smooth spherical particles, this surface area corresponds to a primary particle radius of 35 \AA . However, the SANS data of Hurd *et al.*¹³ show that this interpretation of the surface-area measurements is not valid since the particles have fractally rough surfaces. From the value of Q at which their data crossover from mass fractal to surface fractal behavior, the primary particle radius is found to be about 100 \AA . While the chemical purity of Cab-O-Sil is very high,¹⁷ it is known that the silica surfaces are normally coated with hydrogen atoms in the form of OH ions and H_2O molecules.¹⁸ Since hydrogen is an intense incoherent scatterer of neutrons, this surface layer can dominate the neutron scattering from Cab-O-Sil, providing a method for studying the dynamics of the surface atoms to which the hydrogen atoms are tightly bound.¹⁹ In the present experiments, however, we are interested in studying the dynamics of the bulk Cab-O-Sil particles, since these constitute the mass fractal structure described above. We therefore removed as much of the surface hydrogen as possible by heating the samples to temperatures as high as 1100 K before collecting any scattering data.

Three Cab-O-Sil samples were studied. Sample 1 was baked overnight under vacuum at 500 K and then heated to 1050 K while monitoring the removal of hydrogen by measuring the neutron transmission through the sample. After this heat treatment, we estimate that less than 1% of the total scattering was due to hydrogen. A second sample was prepared in a similar way except that it was only heated to 700 K . Sample 3 was compacted by soaking the sample in water, resulting in a threefold increase in density. The gel produced in this way was dried under a vacuum at 900 K for several hours, thereby removing all free water molecules and leaving only a small quantity of OH radicals bound to the surface.

All scattered neutron spectra were measured with the

N5 triple-axis crystal spectrometer at the NRU reactor, Chalk River. To measure the inelastic scattering from Cab-O-Sil, the instrument was operated in the energy-loss mode, with scattering vector $\mathbf{Q} = \mathbf{k}_i - \mathbf{k}_f$ held constant and with fixed scattered neutron energy $E_f = \hbar^2 k_f^2 / 2m$. Most of the measurements were made with $E_f/h = 3.52 \text{ THz}$, for which the experimental energy resolution was 0.13 THz . Both monochromator and analyzer crystals were Si(111). Use of a Si(111) monochromator crystal virtually eliminated all second-order neutrons from the incident beam. A pyrolytic graphite filter in front of the analyzer collimator removed both second- and third-order contamination from the scattered beam. Some additional scans were made with $E_f/h = 8.20 \text{ THz}$. All inelastic spectra were corrected for fast neutron background and for the scattering from the tantalum sample containers and the radiation shields of the furnace. The integrated background showed no Q or ω dependence and never exceeded 40% of the total scattering. Elastic scattering was also investigated as a function of Q up to 4 \AA^{-1} with an analyzer set to the same values of E_f . To facilitate the identification of the scattering from Cab-O-Sil due to its aggregated structure, we carried out exactly analogous scans on a thin-walled amorphous silica tube, as a reference bulk sample having comparable scattering intensity.

III. RESULTS AND DISCUSSION

The Q dependence of the elastic scattering from Cab-O-Sil is compared with that from bulk amorphous silica (quartz tube) in Fig. 1(a). Both scans were taken at room temperature. The figure clearly shows that the elastic scattering between $Q = 1.0$ and 4.0 \AA^{-1} is nearly identical for the two materials, indicating that the microscopic structure inside the primary particles of Cab-O-Sil is essentially the same as in bulk amorphous silica. The similarity of the two scattering curves also confirms that there is no significant scattering from surface hydrogen in our Cab-O-Sil samples, all of which gave identical elastic scattering data when corrected for the different sample densities. At low Q the scattering from Cab-O-Sil is enhanced by small-angle scattering^{13,14} due to the aggregated structure formed from the small particles.

Figure 1(b) shows the elastic scattering from Cab-O-Sil at the two temperatures 290 and 670 K . The scattering intensity is reduced at the higher temperature, with the difference $\Delta I(Q) = I(Q, 290 \text{ K}) - I(Q, 670 \text{ K})$ varying linearly with Q . In the usual phonon picture, the temperature dependence of the elastic scattering is described by the Debye-Waller factor which predicts that this difference should be proportional to Q^2 . Thus the observed linear Q dependence of $\Delta I(Q)$ in Cab-O-Sil is anomalous, implying that the conventional phonon description of vibrations is inappropriate for this material.

To further study the vibrational properties at low energies, we performed inelastic neutron scattering from both Cab-O-Sil and bulk amorphous silica at energy transfers $E_i - E_f$ up to 0.7 THz . Typical spectra at temperatures of 290 and 670 K are shown in Fig. 2 for $Q = 2.95 \text{ \AA}^{-1}$,

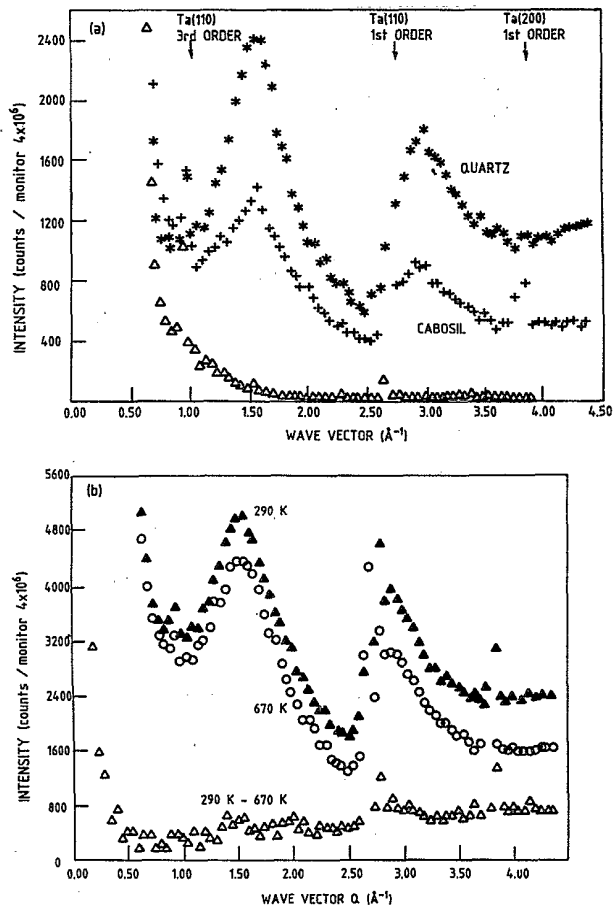


FIG. 1. (a) Elastic scattering at $E_f = 3.52$ THz from bulk amorphous silica (*) and Cab-O-Sil sample 1(+). $T = 290$ K. The positions of the main diffraction peaks from the tantalum can surrounding the sample are also indicated. The background with the specimen removed from the can is shown by the open triangles. (b) Comparison of the elastic scattering from Cab-O-Sil sample 3 at $T = 290$ K (\blacktriangle) and 670 K (\circ). The Q dependence of the difference is shown by the open triangles. (The intensity is plotted for a fixed number of neutrons incident on the specimen, as measured by a monitor detector, in this figure, 4×10^6 .)

For both materials the frequency dependence of the inelastic scattering is quite similar, the spectra being approximately independent of frequency in the range 0.3–0.7 THz. Below 0.3 THz the inelastic scattering becomes masked by the tail of the much more intense elastic scattering peak. There is, however, a marked difference in the temperature dependence of the spectra for the two materials. For amorphous silica [Fig. 2(a)], the inelastic scattering is significantly more intense at higher temperatures, approximately as expected for a system of normal phonons (see below). On the other hand, there is almost no change in the scattering with temperature for Cab-O-Sil [Fig. 2(b)]. There are also differences in the Q dependence of the inelastic scattering. Figure 3 shows the integrated intensity detected in an energy interval between 0.3 and 0.7 THz plotted as a function of selected scattering vectors Q . The empty can background

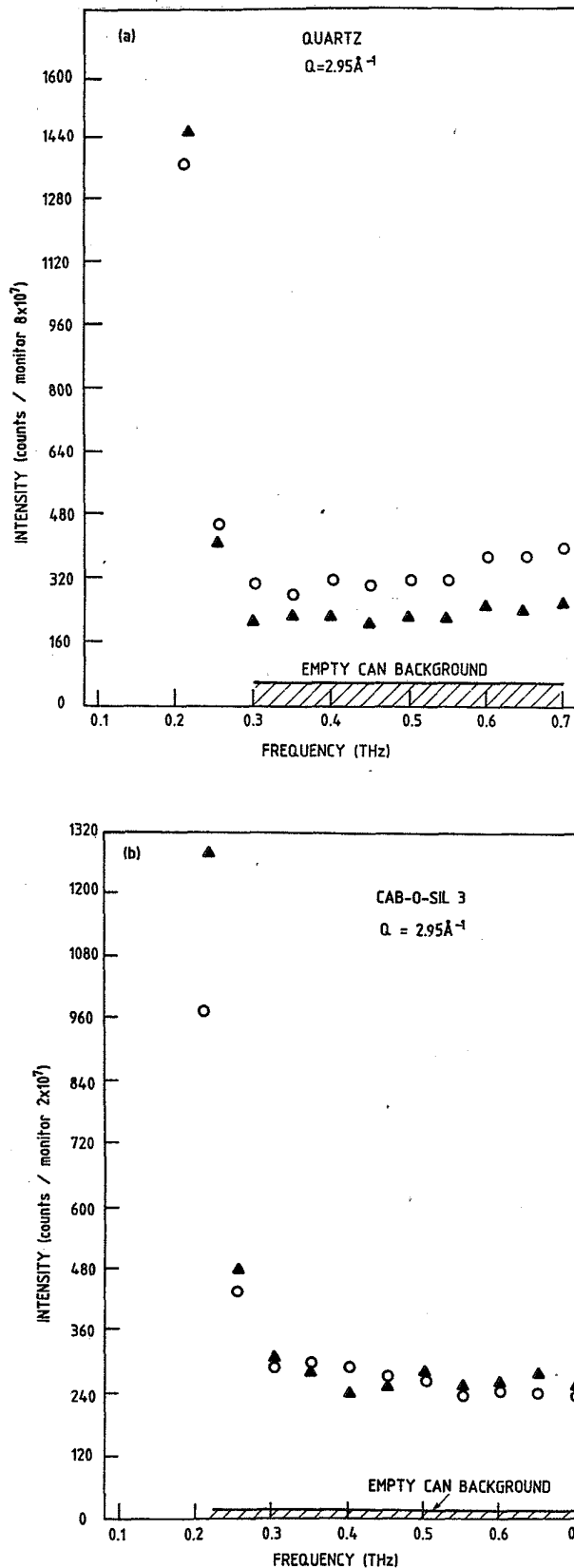


FIG. 2. Inelastic scattering at $Q = 2.95 \text{\AA}^{-1}$ for $T = 290$ K (\blacktriangle) and 670 K (\circ) from (a) amorphous silica tube and (b) Cab-O-Sil sample 3. The shaded regions indicate the empty can background.

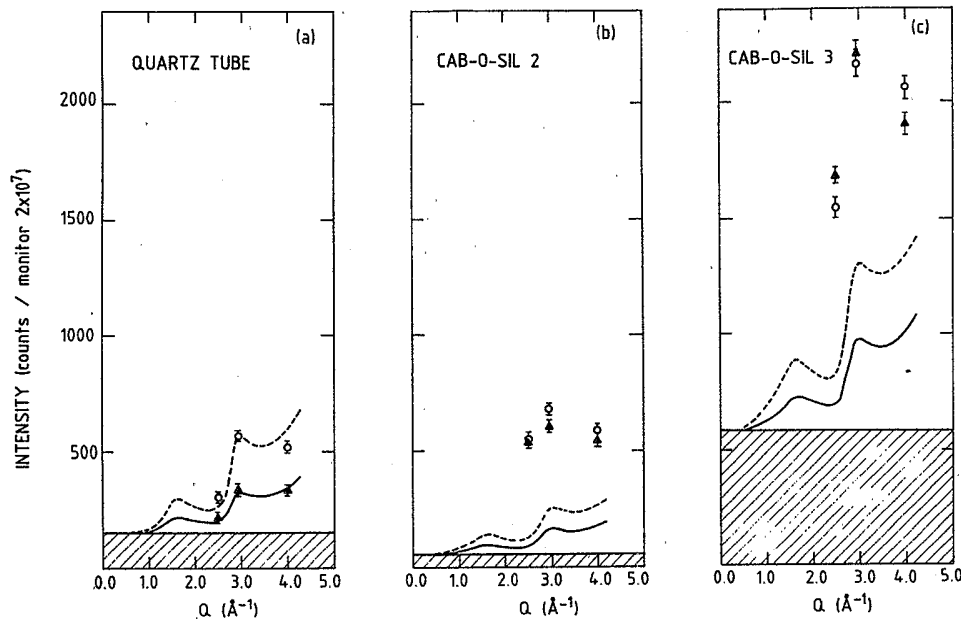


FIG. 3. Integrated inelastic intensity between 0.3 and 0.7 THz vs scattering vector Q at 290 K (\blacktriangle) and 670 K (\circ) for (a) amorphous silica, (b) Cab-O-Sil sample 2, and (c) Cab-O-Sil sample 3. The solid and dashed curves represent the one-phonon contribution to the scattering at 290 and 670 K, respectively (see text). The shaded regions correspond to the estimated multiple-scattering contributions which should scale approximately as the square of the sample mass.

has been subtracted from the data in this figure. It can be seen that there is a tendency for the intensity to increase at larger Q , but that the Q dependence is less pronounced in Cab-O-Sil than it is in amorphous silica.

Before discussing these results in detail, it is of interest to compare the elastic and inelastic scattering intensities for the amorphous silica and Cab-O-Sil samples. Table I summarizes the relevant data. We find that the elastic scattering from any of the three forms of SiO_2 scales as the mass of the specimen in the neutron beam (\propto number of SiO_2 molecules). However, the inelastic scattering from Cab-O-Sil is much greater than expected, some three to four times that of quartz for the same mass. Nonetheless the inelastic intensity from the differently treated Cab-O-Sil specimens does scale as their relative masses. Thus much more of the density response of the fractal materials is in their inelastic spectrum, implying

that their structure is less rigid.

We next compare our data with the predictions by Carpenter and Pelizzari¹⁶ and more recently by Buchenau *et al.*²⁰ for the inelastic scattering from long-wavelength acoustic phonons in amorphous materials. At the low vibrational frequencies investigated in the present experiments, the wave vectors of any phonon excitations are much smaller than the scattering vectors Q , and in this case their calculations show that the inelastic scattering intensity is proportional to

$$\frac{d^2\sigma}{d\Omega d\omega} \propto \frac{g(\omega)}{\omega} n(\omega) Q^2 I(Q). \quad (1)$$

Here $I(Q)$ is the elastic scattering intensity, and $n(\omega)$ is the Bose factor, proportional to T/ω for $k_B T \gg \hbar\omega$ as is appropriate for our data. For long-wavelength acoustic phonons in three dimensions, the density of states

TABLE I. Elastic and inelastic scattering intensities for the three Cab-O-Sil samples relative to bulk amorphous silica. For comparison, the relative densities and masses of the samples are also included.

	Bulk amorphous silica	Cab-O-Sil 1 and Cab-O-Sil 2	Cab-O-Sil 3
Density, ρ	$\rho_0 = 2.2 \text{ g/cm}^3$	0.03 g/cm ³	0.12 g/cm ³
Sample mass, M	$M_0 = 6.6 \text{ g}$	3.3 g	13 g
Elastic scattering intensity $I(Q)$ at 290 K	1	0.53	2.2
Inelastic scattering intensity $I(Q, 0.5)$ at 290 K	1	2.0	6.7

$g(\omega) \propto \omega^2$, so that the scattering law simplifies to

$$\frac{d^2\sigma}{d\Omega d\omega} \propto TQ^2I(Q). \quad (2)$$

This expression is no longer ω dependent, consistent with the apparent lack of any significant variation with frequency in our inelastic scattering data over the range 0.3 to 0.7 THz (Fig. 2). In Fig. 3 we show the temperature and Q -vector dependence of the inelastic scattering calculated using Eq. (2) with $I(Q)$ taken from the measured elastic scattering data for each sample (e.g., see Fig. 1). For bulk amorphous silica [Fig. 3(a)], this model and the data are in approximate agreement once a Q -independent background which can be attributed to multiple scattering²⁰ is subtracted from the data. However for Cab-O-Sil [Figs. 3(b) and 3(c)], neither the temperature dependence, nor the Q dependence, nor the absolute intensity of the inelastic scattering is in agreement with these predictions for one-phonon scattering, implying that the vibrational properties of Cab-O-Sil are anomalous over the entire frequency range studied in our experiments.²¹

The disagreement with the $Q^2S(Q)$ dependence expected for an amorphous solid, and the even stronger departure from a linear dependence on temperature suggests that the vibrational properties are radically different from those of a harmonic solid. Note that even if the modes in the primary particles of Cab-O-Sil are localized, one would still expect the inelastic intensity to depend on Q^2 and on T provided the modes are harmonic. One might expect that the dynamics at room temperature and above would be highly anharmonic. This is because an almost constant density of low-frequency vibrational states allows a phonon to decay into many more lower frequency vibrations than are possible in the limited phase space of a solid with well-defined dispersion relations. Thus higher frequency modes, e.g., the rotational modes, could develop a low-frequency wing as a result of anharmonic damping. However the intensity in the wing should increase at least as fast as T^2 unless there is a large Debye-Waller factor that would attenuate the vibrational scattering at high temperatures as well as at large wave vectors Q . Another consequence of the greater phase space (i.e., lack of momentum conservation combined with a near constant density of states) is that two-phonon and higher phonon processes become more likely at a particular Q and T than in a crystal. However, such multiphonon scattering normally gives a faster variation with Q and T than one-phonon scattering, in contrast to what is observed.

These difficulties suggest that, at low frequencies, an expansion of the scattering cross section in terms of phonons, localized or not, breaks down in a fractal like Cab-O-Sil. Indeed the concept of vibrational excitations taking place about an equilibrium structure may not be valid. Note that many theories of fractal dynamics assume essentially harmonic forces.^{5,6,22} Consider the mean-square displacement in a material which has a density of states $g(\omega) \propto \omega^{\bar{d}-1}$ between a lower cutoff frequency ω_0 and the crossover frequency ω_c to bulk Debye-type behavior. For $kT < \hbar\omega_c$, the thermally activated contribu-

tion to the mean-square displacement arising from the fracton modes is

$$\begin{aligned} \langle x^2 \rangle_{\text{fr}} &\propto kT \int_{\omega_0}^{\omega_c} d\omega g(\omega)/\omega^2 \\ &\propto kT(\omega_0^{\bar{d}-2} - \omega_c^{\bar{d}-2})/(2-\bar{d}). \end{aligned}$$

Thus the mean-square displacement for $\bar{d} < 2$ becomes extremely large for the small-cutoff frequency ω_0 below which the sound waves are hydrodynamic. There is then no distinction at low frequencies between statics and dynamics, in analogy to liquids. At high frequencies, on the other hand, this softness of the structure will produce little difference between the vibrational response of a fractal-like Cab-O-Sil and an amorphous solid.

We believe that the anomalous behavior of the excitations in Cab-O-Sil in our frequency range may be associated with the range of length scales that characterize the surface roughness of the Cab-O-Sil particles. Although our measurement frequencies are above the crossover frequency of the largest dimension of the particles, they may lie below the crossover frequency of the modes corresponding to the shortest dimension of the surface roughness. However at this stage we cannot rule out the alternative possibilities, that the excess scattering is caused by anharmonic tails of the higher frequency modes as mentioned above, or by relaxation involving tunnelling modes as invoked by Buchenau *et al.*²³ to explain the low-frequency excitations of *bulk* vitreous silica. Their model involves librations of coupled rotations of SiO_4 tetrahedra, and these local motions could be significantly modified by finite-size effects associated with the large fraction of surface atoms in the small primary particles of Cab-O-Sil. Further work is planned to investigate this possibility more fully.

We conclude that the difficulty in understanding quantitatively the unusual temperature, Q vector, and intensity dependence of the inelastic neutron scattering from Cab-O-Sil seriously complicates the measurement of the vibrational density of states near the expected crossover frequency ω_c . Note that Eq. (1) predicts that the frequency dependence of the inelastic spectra should change from ω^0 above ω_c to $\omega^{\bar{d}-3}$ below ω_c , potentially a large effect if $\bar{d} \approx 1$. The fact that no deviation from ω^0 behavior was observed between 0.3 and 0.7 THz implies that the crossover frequency $\omega_c/2\pi$ is less than 0.3 THz, consistent with the SANS measurements of Hurd *et al.*¹³ that indicate a primary particle radius of $a \approx 100$ Å corresponding to a crossover frequency of about 0.15 THz. (As discussed above, this crossover will nonetheless be broadened by the surface roughness.) Since these measurements were completed, Kjems *et al.*²⁴ have reported preliminary data that extend to lower frequencies the work of Richter and Passell¹⁹ on the inelastic scattering from hydroxyl ions bound to the surface of Cab-O-Sil. They find that below ≈ 0.35 THz there is a gradual increase (slope = -0.7) in the inelastic scattering as the frequency is decreased, possibly suggesting that \bar{d} has the very large value of 2.3. In more recent inelastic neutron scattering experiments on hydrogenated Cab-O-Sil using a time-of-flight spectrometer, Freltoft, Kjems, and

Richter²⁵ also find large values of \bar{d} , 1.8 ± 0.1 at 136 K and 2.1 ± 0.1 at 265 K. While these new data are largely complementary to our results, in that their data extend to lower frequencies and temperatures and their analysis concentrates on the scattering from hydrogenated *surfaces* of the SiO₂ particles, there appear to be some disconcerting discrepancies where the three sets of data overlap. In the time-of-flight data,²⁵ the SiO₂ "background" (which corresponds to our signal) decreases with frequency over the entire frequency range studied (≈ 0.03 – 2 THz). By contrast, our data on hydrogen-free Cab-O-Sil shows no frequency dependence between 0.3 and 0.7 THz [Fig. 2(b)]. The time-of-flight results for the hydroxyl scattering also appear to contradict earlier data by Richter and Passell (replotted in Fig. 4 of Ref. 24) which show the surface hydroxyl ion scattering to be independent of frequency between 0.4 and 1.2 THz. While the origin of this discrepancy is unclear at present, we

note that our data and the data of Refs. 19 and 24 were taken using a triple-axis spectrometer in which the inelastic spectra are measured at constant Q . By contrast, in the time-of-flight technique, both Q and ω simultaneously vary; this could complicate the analysis of the data in Ref. 25 because, as we have shown, the scattering from hydrogen-free Cab-O-Sil depends on Q and this Q dependence should have been allowed for before the background was subtracted to obtain the surface hydroxyl ion scattering. We also note, as pointed out in Sec. II, that the data on hydrogenated Cab-O-Sil directly probe the motion of the surface atoms which, especially for fractally rough surfaces, may be quite different from the bulk vibrational modes of the small particles. In short, despite the interesting results reported so far, it is clear that further experimental and theoretical work is required to fully understand the low-frequency vibrational modes of fractal aggregates such as Cab-O-Sil.

*Present address: Wacker Chemietronic Abteilung, FOU Postfach 1140, D-8263 Burghausen, West Germany.

¹D. A. Weitz and M. Oliveira, Phys. Rev. Lett. **52**, 1433 (1984).

²D. W. Schaefer, J. E. Martin, P. Wiltzins, and D. S. Cannell, Phys. Rev. Lett. **52**, 2371 (1984).

³J. Forsman, J. P. Harrison, and A. Rutenberg, Can. J. Phys. **65**, 767 (1987).

⁴B. Mandelbrot, *The Fractal Geometry of Nature* (Freeman, New York, 1983).

⁵S. Alexander and R. Orbach, J. Phys. (Paris) Lett. **43**, L625 (1982).

⁶I. Webman and G. S. Grest, Phys. Rev. B **31**, 1689 (1985).

⁷R. Rammal and G. Toulouse, J. Phys. (Paris) Lett. **44**, L13 (1983).

⁸J. H. Page and R. M. McCulloch, Phys. Rev. Lett. **57**, 1324 (1986).

⁹E. Courtens, J. Pelous, J. Phalippou, R. Vacher, and T. Woignier, Phys. Rev. Lett. **58**, 128 (1987).

¹⁰Cabot Corp., P.O. Box 188, Tuscola, IL 61953.

¹¹S. R. Forrest and T. A. Witten, Jr., J. Phys. A **12**, L109 (1979).

¹²P. Meakin, Phys. Rev. Lett. **53**, 2028 (1984).

¹³A. J. Hurd, D. W. Schaefer, and J. E. Martin, Phys. Rev. A **35**, 2361 (1987).

¹⁴T. Freltoft, J. K. Kjems, and S. K. Sinha, Phys. Rev. B **33**, 269 (1986).

¹⁵D. W. Schaefer, A. J. Hurd, and J. E. Martin, Phys. Rev. Lett. **59**, 515 (1987).

¹⁶J. M. Carpenter and C. A. Pelizzari, Phys. Rev. B **12**, 2391 (1975); **12**, 2397 (1975).

¹⁷See *Cab-O-Sil Properties and Functions*, a booklet available on request from Cabot Corp. (Ref. 10).

¹⁸J. B. Peri, J. Phys. Chem. **70**, 2937 (1966).

¹⁹D. Richter and L. Passell, Phys. Rev. Lett. **44**, 1593 (1980); Phys. Rev. B **26**, 4078 (1982).

²⁰U. Buchenau, N. Nücker, and A. J. Dianoux, Phys. Rev. Lett. **53**, 2316 (1984).

²¹Note that even in amorphous silica, Buchenau *et al.* (Ref. 20) showed that there are additional low-frequency vibrational modes, which have been interpreted by Orbach [R. Orbach, in *Scaling Phenomena in Disordered Systems*, Vol. 133 of *NATO Advanced Study Institute, Series B: Physics*, edited by R. Pynn and A. Skjeltorp (Plenum, New York, 1985), p. 335] as evidence for fracton excitations above ≈ 0.6 THz, i.e., at frequencies mainly above those studied here. This would also imply that many of the vibrational modes on length scales shorter than the primary particle size in Cab-O-Sil have fracton characteristics, related presumably to the atomic disorder in amorphous silica.

²²B. W. Southern and A. R. Douchant, Phys. Rev. Lett. **55**, 966 (1985).

²³U. Buchenau, M. Prager, N. Nücker, A. J. Dianoux, N. Ahmad, and W. A. Phillips, Phys. Rev. B **34**, 5665 (1986); U. Buchenau, H. M. Zhou, N. Nücker, K. S. Gilroy, and W. A. Phillips, Phys. Rev. Lett. **60**, 1318 (1988).

²⁴J. K. Kjems, T. Freltoft, D. Richter, and S. K. Sinha, Physica **136B**, 285 (1986).

²⁵T. Freltoft, J. Kjems, and D. Richter, Phys. Rev. Lett. **59**, 1212 (1987).