Ultrasound Propagation in Sintered Metal Powder: Evidence for a Crossover from Phonons to Fractons

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Ultrasonic experiments performed on a three-dimensional percolating system made from sintered copper powder show a rapid increase in the attenuation, corresponding to a sudden decrease in the localization length for vibrations, at wavelengths approaching the percolation correlation length. The data support recent theoretical predictions of a crossover from extended phonon excitations to localized fracton excitations at a length scale below which the structure has the self-similarity of a fractal system.

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Recently there has been considerable interest in the elastic and vibrational properties of percolating systems above the percolation threshold p_c. Such percolating networks are known to have a homogeneous (Euclidean) structure at long length scales, but have a self-similar (fractal) geometry at short length scales.[1] The transition between these two regions occurs at the percolation correlation length ξ, which corresponds above p_c to the radius of the largest holes in the structure, and which diverges as p_c is approached. Orbach *et al.*[2-5] have used scaling arguments and effective-medium-approximation calculations to predict a crossover in the vibrational spectrum of percolating networks from extended (phonon) excitations for wavelengths $\lambda > \xi$ to localized excitations, known as fractons, for $\lambda < \xi$. Furthermore they have postulated[3,4,6] that the vibrational modes of amorphous materials such as epoxy resins and glasses may exhibit analogous behavior, and that this is responsible for their anomalous thermal properties at low temperatures. To date, however, there have been few experiments that can directly test the fundamental ingredients of these theories. In this Letter we address this question by presenting the results of new ultrasonic attenuation experiments that give evidence for phonon localization, consistent with the phonon-fracton model, in a three-dimensional percolating structure comprised of sintered metal powder.

In previous experimental work,[7,8] Maliepaard *et al.* used conventional ultrasonic techniques to study the vibrational modes that are excited thermally at millikelvin temperatures in the sintered metal powder used as low-temperature heat exchangers. At long wavelengths, they found that the ultrasonic attenuation is low and that the vibrational modes are correctly described by phonons with "soft" phonon velocities compared with bulk metal. The softening of the longitudinal and transverse sound velocities with occupied volume fraction f is simply related to the behavior of the static elasticity, and was found to be in good agreement with a recent theoretical model for the elasticity of a percolating system in which bond bending as well as central forces between particles was included.[9] These data, in conjunction with the elasticity and conductivity measurements of Deptuck, Harrison, and Zawadski[10] close to the percolation threshold, have established that sintered metal powder is well described as a percolating structure above threshold and therefore can be taken as a model system for the testing of the theoretical ideas outlined above. Indeed, Maliepaard *et al.*[7] reported a sudden decrease in the ultrasonic signal transmitted through the sinter specimens as the wavelength was lowered, and interpreted their results as evidence of a band edge between low-frequency propagating phonons and high-frequency localized vibrational modes. However, they were unable to make accurate measurements of the change in the ultrasonic attenuation because of possible phase interferences effects in the conventional piezoelectric transducers used in their experiments. In the present work, we have used a new phase-insensitive transducer to make the first reliable measurements of the variation with frequency of the ultrasonic attenuation in porous copper-powder sinters, thus enabling the results to be compared quantitatively with recent theoretical predictions for the excitation spectrum of a percolating system.

The ultrasonic attenuation experiments were performed in the 1–20-MHz frequency range on sintered copper-powder samples with powder diameters $d = 0.5$ and 10 μm, and occupied-volume fractions from 0.3 to 0.6. Ultrasonic pulses, generated by a lead zirconium titanate transducer, were transmitted through thin disk-shaped samples and detected by use of a CdS phase-insensitive transducer.[11] This transducer makes use of the acoustoelectric effect to measure the ultrasonic energy flux,[12] and is therefore insensitive to wave-front distortion which plagues measurements in inhomogeneous materials made with conventional piezoelectric transducers. The frequency dependence

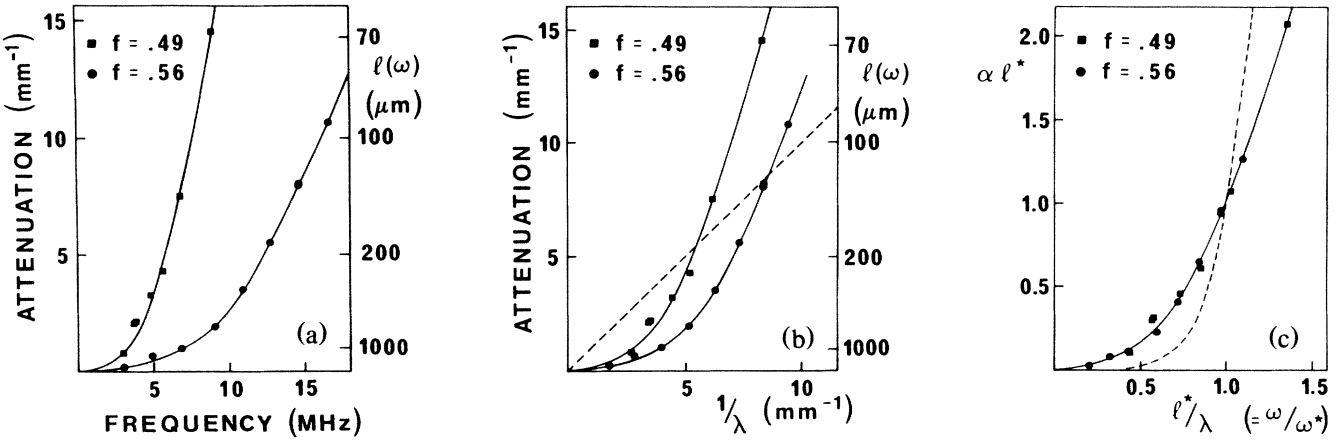

FIG. 1. Attenuation of longitudinal ultrasonic waves in 10-μm sinters. (a) Attenuation vs frequency. The localization length l is indicated by the scale on the right. (b) Attenuation vs inverse wavelength, obtained from (a) by use of the measured sound velocities. The intersection with the dashed line $\alpha = \lambda^{-1}$ determines the inverse of the critical localization length l^*. (c) Attenuation vs inverse wavelength, scaled by l^*. The dashed curve is the EMA prediction of Ref. 4.

of the ultrasonic attenuation in the 10- and 0.5-μm powder sinters is shown in Figs. 1(a) and 2(a). For both sets of data, there is a dramatic increase in the ultrasonic attenuation with frequency, with the increase occurring at lower frequencies in samples with lower occupied-volume fractions. The maximum attenuations observed are very much larger than the attenuation in bulk copper, implying that the attenuation is not caused by the usual sound-absorption mechanisms in metals[13] but is presumably associated with scattering from the geometric disorder in sinter. While the frequency dependence of the attenuation approximately follows a power law over the rather limited range of the data, the measured exponents are in the range 1.6 to 2.4, thus ruling out ordinary Rayleigh scattering which varies as the fourth power of the frequency. Such a contribution would be expected only at lower frequencies where the ultrasonic wavelengths are much larger than the inhomogeneities in the structure. Instead, we believe that the very large attenuations observed in the present experiments are associated with a transition to localized vibrational modes in sinter, as predicted for a percolating structure at length scales near the percolation correlation length.[2-5] For an ultrasonic wave propagating in, say, the x direction, the intensity $I(x)$ falls off[14] as $I_0 e^{-\alpha(\omega)x}$, so that when absorption of ultrasonic energy by the sinter medium is negligible, the attenuation $\alpha(\omega)$ can be interpreted as the inverse of a frequency-dependent localization length for vibrations $l(\omega)$, i.e., $I(x) = I_0 e^{-x/l(\omega)}$. At low frequencies the localization length is large ($l \gg \lambda$), implying extended vibrational states ap-

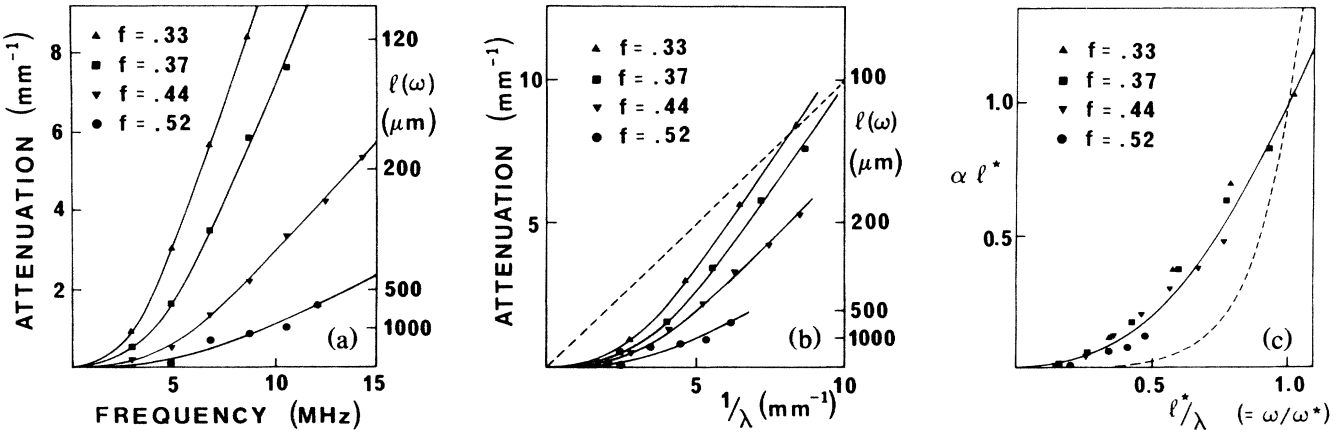

FIG. 2. Attenuation of longitudinal ultrasonic waves in 0.5-μm sinters. Parts as in Fig. 1.

propriate to the phonon regime. At higher frequencies, there is a rapid decrease in localization length with frequency, with $l(\omega)$ becoming smaller than the ultrasonic wavelength at the maximum attenuations observable in the present experiments. Such small values of the localization length indicate that plane waves do not propagate at high frequencies, implying a crossover to localized vibrational modes. Note that since the localization length $l(\omega)$ can be identified with the phonon elastic mean free path l_{el}, this conclusion is consistent with the familiar Ioffe-Regel criterion[15] for localization, $l_{el} \lesssim \lambda$. It is interesting that over the range of the data, the approach to the localized regime appears to be continuous; we note, however, that the transition occurs over a very narrow range of length scales as can by seen by a plot of attenuation versus wavelength (cf. Fig. 12 of Ref. 7).

To investigate whether this evidence for phonon localization in sinter can be interpreted in terms of a crossover from phonons to fractons, we consider the scaling of the ultrasonic attenuation with the percolation correlation length $\xi(f)$ that is believed to mark the boundary between the two regimes in a percolation system. Single-parameter scaling at frequencies below crossover[4,5] predicts that the localization lengths in sinters with different occupied-volume fractions f_1 and f_2 should scale in the following way: For $\lambda_1/\xi(f_1) = \lambda_2/\xi(f_2)$,

$$\frac{l_1(\omega_1 = 2\pi v_1/\lambda_1)}{\xi(f_1)} = \frac{l_2(\omega_2 = 2\pi v_2/\lambda_2)}{\xi(f_2)} \quad (1)$$

where λ_1, λ_2 and v_1, v_2 are the wavelengths and sound velocities, respectively. Since the magnitude of $\xi(f)$ is not known *a priori,* we have tested this prediction by scaling the data using a critical value of the localization length, l_f^*, which is expected to be proportional to ξ and which is defined as the value of $l(\omega)$ at which the ultrasonic intensity decreases by $1/e$ in propagating a single wavelength, i.e., $l_f^* = l(\omega^*)$ at the frequency ω^* such that $l(\omega^*) = \lambda = 2\pi v/\omega^*$. For the 10-$\mu$m sinters, l_f^* is determined from the plot of attenuation versus $1/\lambda$ in Fig. 1(b), giving $l_{0.56}^* = 116$ μm and $l_{0.49}^* = 141$ μm,[16] so that $l_{0.56}^*/l_{0.49}^* = 0.82$ in good agreement with the expected ratio of the correlation lengths $\xi(0.56)/\xi(0.49) = 0.83$ obtained from the relation $\xi \propto (f - f_c)^{-\nu}$ with $\nu = 0.88$.[17] Here we have taken $f_c = 0.2$[18] as determined from the scaling of the longitudinal and shear velocities for sinter, $v \propto f^{-1/2}(f - f_c)^{\tau/2}$ with $\tau = 3.6$.[7] Figure 1(c) shows that when the attenuation and inverse wavelength are both scaled by l_f^* the data for the two sinters fall on the same curve, thus establishing the scaling relation (1). For the 0.5-μm sinters, the data only extend to high enough attenuations and frequencies to determine l_f^* for the lowest value of $f = 0.33$ [Fig. 2(b)]. The data were therefore scaled by our assuming that $l_f^* \propto \xi(f)$ with $f_c = 0.06$[10] and $l_{0.33}^* = 120$ μm.[19] The result, Fig. 2(c), shows reasonable agreement with the scaling predictions, confirming that the onset of localization for vibrations in sinter occurs at a length scale determined by the correlation length ξ, i.e., by the boundary between the Euclidean and fractal regions.

Recently Entin-Wohlman *et al.*,[4] have used the effective-medium approximation (EMA) to solve for the vibrational spectrum of a simple bond-percolation network. In their model, uniform-mass particles are placed on a simple cubic lattice and are connected by isotropic force constants which are assigned randomly with probability p. Even though this scalar elasticity model, for which the elasticity belongs to the same universality class as the conductivity, is too simple to give a good description of the elastic properties of sinter,[7,10] it is still of interest to compare their calculations with our data. By calcuation of the ultrasonic displacements from the EMA lattice Green's function, it is straightforward to show that, in the absence of dissipation, the ultrasonic attenuation length is just the localization length calculated by Entin-Wohlman *et al.* Figure 2 of their paper shows that $l(\omega)$ decreases precipitously at the crossover frequency ω_c corresponding to $q_c \sim \xi^{-1}$, in good qualitative agreement with our experimental results.[20] It is interesting to note that their model predicts that $l(\omega)$ continues to decrease with decreasing length scale right through crossover, consistent with the continuous increase in the attenuation observed experimentally. Probably the most meaningful way of making a quantitative comparison of theory and experiment is to scale the values of localization length determined numerically from Eqs. (4), (9), and (19) of Ref. 4 in the same way that the data are scaled in Figs. 1 and 2, with the calculated values of v and l^* appropriate to the model.[21] These scaled theoretical predictions are shown by the dashed curves in Figs. 1(c) and 2(c), indicating that the calculated attenuation rises more steeply near crossover than is observed experimentally. It seems unlikely that the difference between theory and experiment can be attributed to rounding of the data due to a variation of the occupied-volume fraction or particle size within the sinter samples.[22] The discrepancy may simply reflect the limitations of applying a lattice-based percolation model to a "truly random" medium such as sinter, or it may result from use of the scalar Born elasticity model and the EMA as a basis for describing the vibrational modes. It would be very interesting to see if further calculations, based on the bond-bending model[9] which is known to describe the elasticity of sinter,[7,10] can give better agreement with our experimental results.

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[14]This exponential decay law has been checked experimentally by measurement of the attenuation for different sample thicknesses. In this context, it is worth pointing out the distinction between ballistic propagation, which is expected to be exponentially attenuated, and the transmission of the scattered radiation, which need not obey this relationship [see, e.g., P. W. Anderson, Philos. Mag. B **52**, 505 (1985)]. In the present experiments, the dominant signals are ballistic, as is evident from careful analysis of time-resolved transmission data such as those shown in Fig. 2 of Ref. 8. (At shorter wavelengths than those used here, this figure also shows a large diffuse signal. The fact that the diffusely scattered radiation does not show up more significantly in the range of the present experiments results from our experimental geometry; much of the diffuse radiation arrives at the sample-transducer interface at oblique angles of incidence for which it undergoes total internal reflection and is therefore not detected.)

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[16]Preliminary analysis of scanning electron micrographs of one of the 10-μm sinters indicates that l^* is probably within a factor of 3 of the percolation correlation length. This was obtained by measurement of the area-area autocorrelation function $\langle A(0)A(R)\rangle$ of a polished two-dimensional cross section of an epoxy-impregnated sinter sample. Since $\langle A(0)A(R)\rangle \propto R^{\bar{d}-3}$ for lengths $R<\xi$, where $\bar{d}$ is the fractal dimensionality of sinter, and $\langle A(0)A(R)\rangle \propto R^0$ for $R>\xi$, ξ can be obtained from the crossover in power-law dependence of the autocorrelation function. This analysis, supplemented by measurements of perimeter-area scaling of the two-dimensional clusters, also gives a preliminary estimate of the fractal dimensionality, $\bar{d}\sim 2.7\pm 0.2$. (A similar analysis of the submicron sinters has not yet been successful because of the greater difficulty in obtaining accurate two-dimensional cross sections.)

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[18]This value of f_c for the 10-μm sinters (0.2) is larger than the value measured for submicron sinters (Ref. 10) (0.06), reflecting a difference in the way the particles pack together in the two sets of sinters. For the submicron sinters, the powder first clumps together through presintering, forming isolated clusters which are then joined together during the sintering process. As a result, the threshold value of the occupied-volume fraction f_c that is appropriate to the packing of these low-density clusters is correspondingly smaller than for the simple packing of bulk-density particles. For the 10-μm sinters, on the other hand, there is no evidence of cluster formation prior to sintering, consistent with the larger value of f_c found experimentally. Note that the value of f_c for the 10-μm sinters is closer to theoretical predictions for continuum percolation ($f_c=0.15$) and for site percolation of spheres on a simple cubic lattice ($f_c=0.16$) as discussed in Ref. 7.

[19]Note that l^*/d is about an order of magnitude larger for the 0.5-μm sinters than for 10-μm sinters, presumably reflecting the cluster-of-clusters structure seen in electron microscope pictures of submicron sinter.

[20]Fig. 2 of Ref. 4 also shows dispersion in the velocity near crossover; however, this effect becomes relatively small well away from threshold where the present experiments, which were insensitive to small changes in velocity, were conducted. Further experiments nearer p_c are planned to investigate this effect.

[21]This avoids making a three-parameter fit of $l(\omega)$ to the data which is complicated by the fact that the predicted variation of velocity and hence $l(\omega)$ with $p-p_c$ is quite different from the measured dependences on $f-f_c$.

[22]For example, for a sample with an average occupied-volume fraction $f=0.44$, an unrealistically large variation in f, characterized by a Gaussian distribution with full width at half maximum of 0.24, is required to round the theory sufficiently to coincide with the solid curve in Fig. 2(c).