A statistical approach to direct density of states measurements in disordered systems

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A statistical method for measuring the modal density of elastic waves through direct mode counting in strongly scattering disordered systems is presented. To illustrate this approach, the results of ultrasonic experiments in a highly porous sintered glass bead network are reported. This method is shown to yield a reliable and robust measurement of the density of states, enabling mode-counting techniques to be applied to increasingly complex systems, where modal overlap and sensitivity to experimental conditions have previously hampered definitive results.
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I. INTRODUCTION

During the last 2 decades, there has been a growing interest in wave transport in strongly scattering random media.[1–5] This interest had been motivated both by curiosity in the fundamentals of the novel wave phenomena that can occur in complex media and by practical applications in nondestructive evaluation of such materials. Ultrasonics has played an important role in studying these strongly scattering disordered systems.[6–20] Initial investigations focused on the applicability of the diffusion approximation to describe the transport of energy by multiply scattered waves in pulsed experiments, where a time-dependent "coda" is observed after the initial arrivals due to ballistic transport.[6,7] Even though the diffusion approximation neglects all phase information, it gives a remarkably accurate description of wave transport under a wide variety of experimental conditions, including transmission and reflection (coherent backscattering) experiments.[7–10] Dispersion due to scattering resonances has also been investigated and modeled, providing new insights into the group velocity that describes coherent pulse propagation.[11] More recently, evidence for the breakdown of normal diffusive wave transport due to Anderson localization of ultrasound in the presence of very strong disorder has been demonstrated for both two and three dimensional systems,[15,16] further extending our knowledge of the profound effects that scattering can have on wave propagation. The knowledge gained from the fundamental studies has also led to useful applications. Even though imaging inside multiply scattering media is still a challenge, multiply scattered waves have been demonstrated to be very sensitive to motion of the scatterers, leading to the new techniques of diffusing acoustic wave spectroscopy and diffusing reverberant acoustic wave spectroscopy for characterizing scatterer dynamics.[17–19] Despite the wealth of information that has been gleaned on wave transport from ultrasonic experiments, relatively little attention has been paid to another fundamental quantity—the density of states—and a reliable method of measuring this quantity remains essential to obtaining a complete picture of wave transport.

Density of states measurements have been performed in several systems using a direct mode-counting technique.[20–23] In such experiments, samples are excited using a short, broadband impulse (either from a piezoelectric transducer or by a physical impact), and the resulting vibration is measured using a transducer in contact with the sample. Since the number of modes increases with system size, the samples must be sufficiently small to enable the individual modes of vibration to be resolved and counted in the Fourier transform of the received signal, yielding the density of states. In these measurements, one must ensure that *all* the modes are indeed being accounted for. Degeneracies (or near degeneracies), finite peak width, and nodes at the excitation or measurement location may cause some modes to be missed, and electronic noise may make the unambiguous identification of some of the modes difficult. All of these factors may contribute to an incorrect enumeration of the modes.

Methods for addressing these difficulties have been developed in the field of structural acoustics, where sophisticated techniques of experimental modal analysis have been used to identify resonances under difficult conditions.[24] However, these techniques rely largely on multiple input/output schemes, which may be impossible due to physical size limitations and/or modal shifting, caused by interaction with the source and detector in the experiment. Furthermore, strong coupling between modes, along with noise in the data, may prohibit mode identification through phase rotations.

In this paper, we propose a new approach that aims to overcome these limitations. In our experiments, the vibrational modes of a highly porous, disordered sintered glass bead network are investigated using ultrasonic techniques in the strong-scattering frequency regime, where the range of wavelengths is comparable to the pore sizes. We present a robust statistical method of measuring the density of states using a mode-counting technique designed to account for the possibility of "missing" modes.

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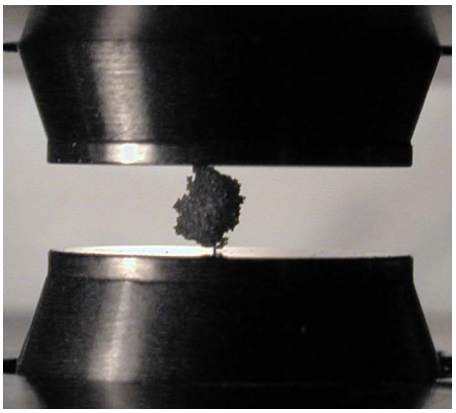

FIG. 1. (Color online) One of the sintered glass bead samples, in contact at two points with two 1/4 in. diameter piezoelectric transducers. The samples range in volume from 0.6 to 5.8 mm^3, and have a glass volume fraction of 33.2%.

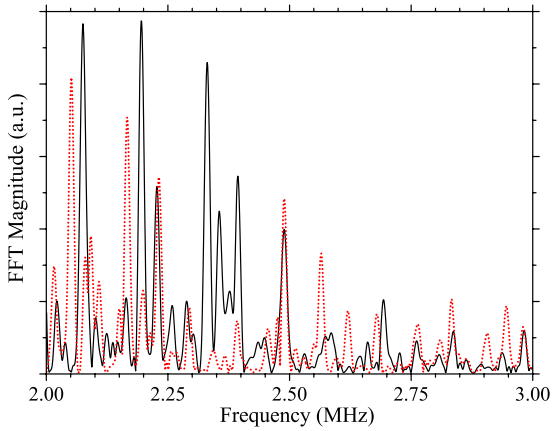

FIG. 3. (Color online) The spectra from one sample with two different sets of contact points. Though the spectra look quite similar at some frequencies, the location and number of modes are not the same.

II. MODE-COUNTING EXPERIMENTS

Our experiments were performed on a highly porous sintered network of glass beads. This network was constructed by sintering a random, 1:1 mixture of glass and iron beads (diameter ≈ 120 μm), and subsequently removing the iron through etching. The resulting material had a glass volume fraction of $\phi=0.33$, and was cut into small samples with volumes ranging from 0.6 to 5.8 mm^3.

The samples were carefully placed between two broadband transducers, which were each in contact with only one or at most a few points on the sample surface (see Fig. 1), in order to minimize the effects of coupling with the transducers on the modal structure of the sample. The contact pressure between the sample and the transducers was very light, so that damping of the vibrations by the transducer surface would not significantly broaden the modes. A short pulse (≈ 0.2 μs) from one transducer was used to excite the modes of the sample, and the multiply scattered, transmitted signal was detected by the other transducer. An example of one of the acquired signals is shown in Fig. 2. The experiments were performed in a weak vacuum (≈ 100 mTorr) to eliminate any signals traveling between the transducers through air, and to reduce losses to the air, which cause the modes to broaden. The pulses were repeated and averaged 10 000 to 20 000 times at a rate of 100 Hz, in order to improve the signal-to-noise ratios. Over the required acquisition time, despite the very long path lengths involved, no change in the waveforms due to temperature fluctuations or drifts[25] was observed. The fast Fourier transform (FFT) of the signals was taken using a −74 dB four-term Blackman–Harris window,[26] which was found to yield consistent peak shapes and minimize spectral leakage, at the expense of only a very slight increase in peak width overall—this made the peaks significantly easier to identify.

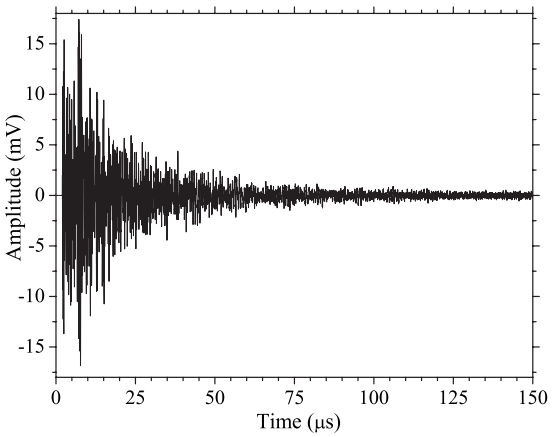

FIG. 2. A typical signal transmitted through one of the samples. The nominal frequency of the transducers is 5 MHz, and the waveform has been averaged about 15 000 times.

Each sample investigated was studied under several orientations relative to the axis defined by the normal to the transducer surfaces, so that each orientation caused different points on the sample surface to be in contact with the transducers. The spectra acquired from one sample for two different sets of contact points are shown in Fig. 3. While some features are common to both spectra, several peaks seem to be either shifted or missing in one spectrum compared to the other. Through a careful analysis of the very small samples (wherein the modes were very sparse, and therefore easier to identify), we were able to determine that the modes shifted in frequency unpredictably as a result of changed contact or ambient (vacuum) pressure, and that some modes present in one orientation were indeed missing in another, when different points were in contact with the transducer faces.[27] Furthermore, the peaks due to the weakest modes (where the displacement at the detecting transducer is very small) are difficult to unambiguously distinguish from the electronic noise. Because of these random factors that influence mode detectability, a statistical approach to mode-counting was required to obtain a reliable measurement of the density of states.

III. ACCOUNTING FOR "MISSING" MODES

It is reasonable to assume that for any given mode of vibration in the strong-scattering frequency regime, nodes will be present on the surface of the sample—this is akin to the presence of dark spots in a speckle pattern. If the contact

points with one of the transducers (nearly) coincide with these nodes, the mode will simply not be observed.

If *all* the modes present in a sample were observed for *each* set of contact points, one would, of course, count the same number of modes in each orientation. If, however, the modes are being either found or missed in a probabilistic fashion, a distribution of mode counts will be found when a single sample is studied under different orientations. The width of this distribution can be used to estimate the proportion of modes that are being missed, and therefore can be used to calculate the true number of modes in the sample.

Assume, first of all, that for the spectrum acquired for one set of contact points for a particular sample, there is a probability p of detecting any one of the n total modes present in the frequency range observed. For each mode, there are only two possible outcomes: either the mode is detected or it is not. It is reasonable to assume that mode detectability is a random process, based on the observed character of modal responses to changes in the transducer contact points and the measured speckle statistics in similar samples, which show that the wave fields are Gaussian random variables.[20] Therefore, the binomial distribution should be used as a parent distribution for the number of modes observed.[28] The detection probability and total number of modes can then be written in terms of the measured mean $\mu=np$ and standard deviation $\sigma=\sqrt{np(1-p)}$:

$$p=\frac{\mu-\sigma^2}{\mu}, \tag{1}$$

$$n=\frac{\mu^2}{\mu-\sigma^2}. \tag{2}$$

If there are N trials (for a particular sample), it is straightforward to write down the uncertainty in the mean and standard deviation, and standard propagation of uncertainties then yields the following for the uncertainties in p and n:

$$\Delta_p=\frac{\sigma^2}{\mu^2}\sqrt{\frac{\sigma^2+2\mu^2}{N}}, \tag{3}$$

$$\Delta_n=\frac{\mu\sigma}{(\sigma^2-\mu^2)^2}\sqrt{[(\mu-2\sigma^2)^2+2\mu^2\sigma^2]/N}. \tag{4}$$

This gives an estimate (with a measurable uncertainty) of the true number of modes in a given frequency range for a *particular* sample. However, the microscopic nature of the samples is inherently different from one to the next, and the exact number of modes in a given frequency range for each sample will fluctuate accordingly. That is, even if the density of states for each sample could be measured perfectly, there would still be statistical fluctuations from one sample to the next. To account for this, an additional contribution to the uncertainty in n should be included to account for the expected variance in the actual density of states from one sample to another. This contribution can be estimated from random matrix theory,[21,29] and is added in quadrature to Eq. (4), in order to better represent the uncertainty in the density of states of the ensemble average, i.e.,

$$\Delta_n=\sqrt{\frac{2\ln n}{\pi^2}+0.44+\frac{\mu^2\sigma^2[(\mu-2\sigma^2)^2+2\mu^2\sigma^2]}{N(\sigma^2-\mu)^4}}. \tag{5}$$

Provided the number of measurements and the number of modes is high enough to obtain meaningful statistics, this method can be used independently for each frequency bin used, in order to measure the dependence of the density of states on frequency.

The density of states is also expected to scale with sample volume. While a surface term[21] should generally be included in any expression for the density of states of a porous system, the total surface area (including internal surfaces) of any sample will also be proportional to sample volume, provided that the size of the sample is significantly larger than the average size ξ of the largest pores. In our samples, ξ was previously measured to be 240 μm,[27] yielding a pore volume of about 0.01 mm^3, which is typically 100 times smaller than the volume of any of the samples. Furthermore, when the mean free path is significantly less than the wavelength and sample size (as in our samples, where the sizes of the pores and beads are also less than a wavelength), the distinction between "surface" and "bulk" modes breaks down, as strong multiple scattering ensures that the modes are diffuse and therefore extend throughout the sample. For these reasons, the density of states is indeed expected to scale with volume. Measurements from several samples can then be normalized by their respective volumes, and by taking a weighted average, the ensemble-averaged density of states of the medium can be determined.

Special consideration must be given to the case where no modes are measured for *any* trial for a particular sample in a given frequency bin. In this special case, $\mu=\sigma=0$, leaving n, p, Δ_n, and Δ_p indeterminate. One might consider setting $n=\Delta_n=0$ for this case, but this would essentially give the zero-value infinite weight in the weighted ensemble average, and still leave p and Δ_p undefined. This means that a density of states of zero would be "measured" at a particular frequency, even though there is a statistically significant probability of finding a mode at that frequency in other samples. This possibility should be taken into account, so that the "zero-measurement" case is properly included in the weighted average.

For any one trial, with no *a priori* knowledge of the uncertainty, it is reasonable to assume Poisson statistics, since one is simply counting modes. In order to estimate the expected mean and uncertainty in a zero-measurement (for several trials), the probability of *observing* $\mu=0$ (in one single measurement) when the *actual* value is $\nu\neq 0$ (Ref. 30) must first be found. This is given by the Poisson distribution

$$P_p(\mu;\nu)\equiv\frac{\nu^\mu}{\mu!}e^{-\nu} \tag{6}$$

evaluated at $\mu=0$. The probability of observing zero *every* time, if N measurements are made, is then given by

$$(P_p(0;\nu))^N=e^{-\nu N}. \tag{7}$$

Summing over all possible ν, the probability of consistently observing zero for N measurements with an unknown $\nu\neq 0$ is given by

$$P(0;N,\nu\neq 0)\equiv\sum_{\nu=1}^{\infty}e^{-\nu N}=\frac{1}{e^N-1}. \tag{8}$$

The probability that the actual value is zero ($\nu=0$) is then simply given by subtracting the above quantity from unity:

$$P(0;N,\nu=0)=1-\frac{1}{e^N-1}. \tag{9}$$

The best estimate for n is then given by the expected value of ν (the "actual" value) when zero is consistently measured:

$$n(0)=\langle\nu\rangle=\sum_{\nu=0}^{\infty}\nu(P-p(0;\nu))^N=\frac{e^N}{(e^N-1)^2}. \tag{10}$$

The best estimate for Δ_n is then obtained in the usual way, i.e.,

$$\Delta_n{}^2(0)=\langle\nu^2\rangle-\langle\nu\rangle^2=\frac{e^N(e^N+1)}{(e^N-1)^3}-\frac{e^{2N}}{(e^N-1)^4}. \tag{11}$$

Therefore, when no modes are observed in a frequency bin for a particular sample, a reasonable value and uncertainty can be assigned to n based on the number of trials performed. For large N, Eqs. (10) and (11) both tend to e^{-N}, which seems to be a value of a sensible order of magnitude to be used as a weighting factor in the ensemble average, for the case of a zero measurement.

The most probable value for p could also be calculated based on the above assumption of Poisson statistics. However, that assumption was only intended to give a rough idea of how to assign an appropriate value to n in the case of a zero measurement, so that it might be sensibly included in the average. There is no obvious reason to infer a particular value for p, given that one simply does not know if there are *actually* no modes in the given frequency range, or if they have simply all been missed. For this reason, in the case of a zero measurement, a uniform probability distribution for p is simply assumed, yielding $p(0)=\Delta_p(0)=0.5$.

IV. RESULTS AND DISCUSSION

One of the most challenging aspects of the analysis of these experiments was to accurately and objectively identify the peaks in the spectra among the background noise. The statistical argument of Sec. III only accounts for missed modes in the spectra, and will give erroneous results if peaks due to electronic noise are additionally counted. The modes were discriminated from the noise by comparing their height (relative to adjacent minima) to a threshold value expressed as a percentage of the average peak height. To ensure that the modes are being identified correctly, this threshold was varied, and the calculated "true" number of modes (n) and the mean number of mode counts (μ) were plotted as a function of threshold value, as shown in Fig. 4. A region is sought where n is independent of μ, that is, where the measured density of states is unaffected by missing modes. The threshold value was then chosen at the onset of this region to avoid missing modes unnecessarily. This threshold was then used for all trials performed on the sample in question, in order to keep the analysis as consistent and objective as possible.

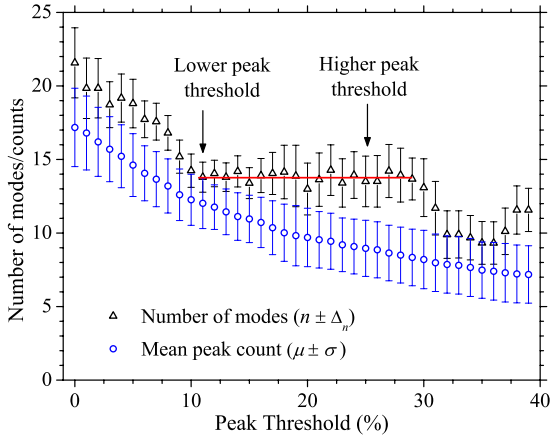

FIG. 4. (Color online) The mean number of peaks counted (μ) and the calculated number of modes (n) are shown as a function of peak threshold (as a percentage of average peak height). A plateau in (n) (while μ is decreasing) is observed, indicating the correct range for the peak threshold (solid line shown as a guide to the eye). While the threshold chosen is indicated by the arrow on the left, the higher peak threshold (arrow on right) yields similar results. The frequency range for the data shown is 2–4 MHz.

The plateau region in n, while μ is still decreasing as the threshold is increased, is seen in Fig. 4. Any threshold value chosen within this region will yield virtually the same results for the density of states. Because this region is not always as easily identified as in the figure shown here (especially when the signal-to-noise ratio is smaller), the spectra were viewed with the peaks above threshold indicated, to verify that the results appear reasonable. That is, threshold values that are too large or too small can be ruled out simply by visual inspection, and the plateau region within this range is used to identify the correct threshold value. One of the spectra, with the above-threshold peaks shown for two values within the plateau region, is shown in Fig. 5.

In order to verify that the density of states is scaling with sample volume as expected, the number of modes per unit frequency was plotted as a function of sample volume, as shown in Fig. 6. This figure shows a limitation of the method presented: when the sample is too large, the spacing of the modes becomes smaller than the peak width, and the modes

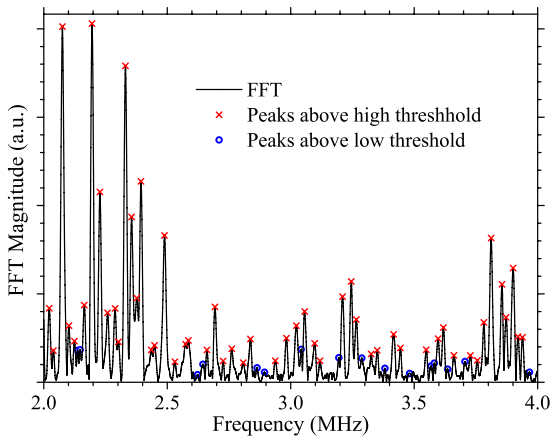

FIG. 5. (Color online) The FFT of one of the signals, with the peaks found above higher and lower threshold values, as indicated in Fig. 4. While some modes are missed when the higher threshold value is used, the statistical analysis yields a similar value for the density of states in both cases.

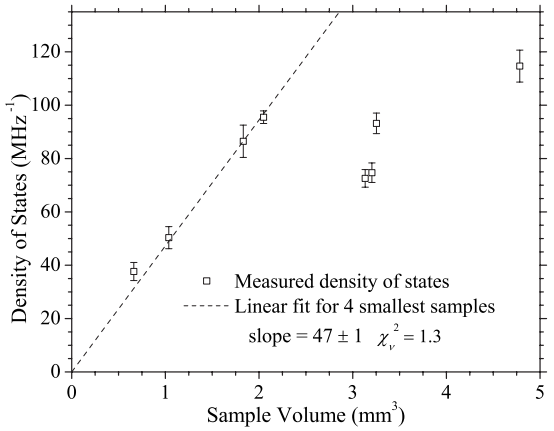

FIG. 6. The density of states as a function of sample volume (frequency range: 2–4 MHz). When the samples become too large, the individual modes can no longer be resolved. This causes the number of modes counted to significantly underestimate the density of states, as shown here by the saturation in the data for large samples.

can no longer be resolved. The spectra obtained from these larger samples indeed show qualitative differences, and spectra from one large and one small sample are shown in Fig. 7 for comparison. The larger sample is characterized by a lack of "dead zones" in the Fourier transform—neighboring modes are immediately adjacent to one another. In this case, modes are being missed due to limited resolvability rather than nodes on the sample surface, and the presented statistical method breaks down. When these samples are ignored, the density of states does indeed scale with volume, as shown by the four smallest samples in Fig. 6, which are consistent with a straight line. This linear dependence of the density of states on sample volume was found for *all* samples investigated at lower frequencies, where the modes are narrower, providing additional justification that this interpretation of the data presented in Fig. 6 is correct.[27]

As discussed, the density of states can be calculated according to this statistical method over many narrow frequency intervals (bins) independently, so that the frequency dependence of the density of states can be measured. (Note that the threshold value is chosen for, and applied to, the

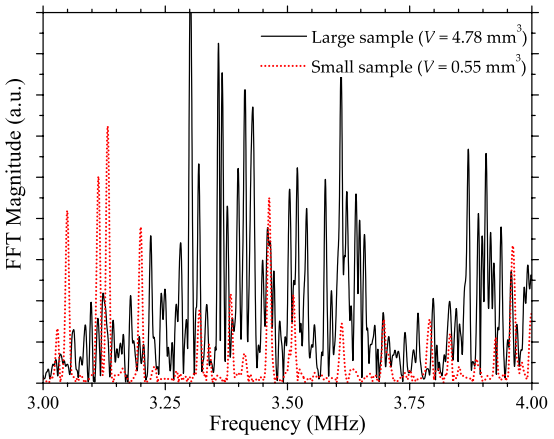

FIG. 7. (Color online) Spectra from a large and small sample. The lack of "dead" frequencies in the larger sample suggests that the modes may be insufficiently spaced to be properly resolved.

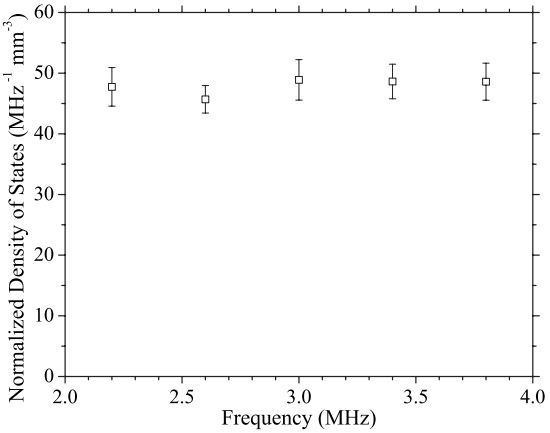

FIG. 8. The normalized density of states is nearly constant in this frequency range (2–4 MHz).

entire available range of frequencies for one set of experiments). The resulting measured density of states is shown in Fig. 8, revealing an unusual nearly constant frequency dependence. A similar result has been reported in aluminum foams,[23] and postulated for sintered metal powders.[31] In view of the unusual character of the density of states in such systems, the implementation of a robust method of measuring the density of states using ultrasonic experiments is especially important. Note that the uncertainties shown in Fig. 8 are not estimated, but are purely derived from the statistics of the measurements. The average probability of observing any mode present in the sample was found to be $p = 0.7 \pm 0.2$, that is, about 70% of the total number of modes was observed in any one single trial.

V. CONCLUSION

We have presented a robust statistical method for directly measuring the density of states of elastic waves in a strongly scattering system. Previous density of states measurements (both in our experiments[20] and elsewhere[23]) attempted to uniquely identify the modes from one spectrum to the next (from the same sample), and thereby estimate the total number of modes based on this correspondence. Because the shape, height, and frequency of a mode can change somewhat unpredictably based on experimental conditions (as exemplified by Fig. 3), this unique identification becomes increasingly difficult as the density of states increases, especially when studying "softer," more sensitive materials.[32] Our statistical method enables direct measurements of the density of states in more types of systems and with larger samples than possible previously. The experiments and analysis described in this paper provide a good foundation for further measurements of this fundamental quantity in similar systems.

ACKNOWLEDGMENTS

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