NOVEL TECHNIQUES IN ULTRASONIC CORRELATION SPECTROSCOPY:

Characterizing the Dynamics of Strongly Scattering Materials

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1. INTRODUCTION

In strongly scattering materials, especially those containing a large concentration of high-contrast scattering objects (e.g. particles or inclusions), traditional ultrasonic imaging techniques break down, motivating the search for other approaches to characterize the properties of such materials. Even though acoustic wave scattering cannot readily be used to image the position of each scatterer unambiguously under these circumstances, much can be learned about the dynamic properties of the medium when the scatterers are moving. For particles suspended in a liquid, much has already been achieved through the development of sophisticated Doppler ultrasound imaging techniques, which have been extensively used to measure fluid flow when the scattering particles move together collectively throughout the scattering volume probed by the ultrasonic beam. However, there are many important cases where the motion is more complex, requiring new experimental approaches and methods of analysis to extract meaningful information from the scattered ultrasonic signals. To address this need, we have developed two ultrasonic correlation spectroscopies, Dynamic Sound Scattering (DSS) and Diffusing Acoustic Wave Spectroscopy (DAWS)[1], which exploit the dynamic information present in ultrasonic speckles that are ubiquitous to wave scattering experiments. These techniques use correlation spectroscopy to measure the motion of the scatterers even in situations where the variance exceeds the mean velocity of the

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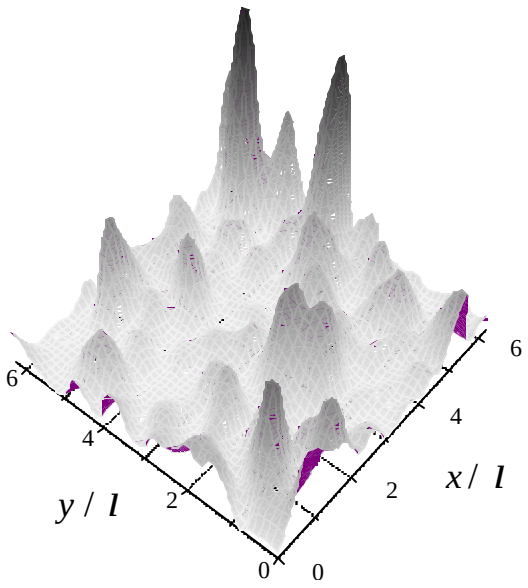

Figure 1. Ultrasonic near-field speckle pattern.

scatterers, thus greatly extending the type of dynamic information that can be obtained from ultrasonic scattering experiments. In this chapter, we will describe how DSS and DAWS can be used to obtain new information about the dynamics of strongly scattering materials, illustrating the potential of these techniques with experimental results on fluidized suspensions of particles.

2. BASIC PRINCIPLES OF ULTRASONIC CORRELATION SPECTROSCOPY

Whenever there is more than one scatterer in the volume probed by the scattered ultrasonic waves, a speckle pattern is formed due to the interference of waves that have traveled different paths through the sample. Figure 1 shows a typical example of the ultrasonic speckle pattern measured at intermediate frequencies (wavelength λ ~ particle size) in a strongly scattering medium. This picture shows the variation of the scattered ultrasonic intensity across the face of the sample when illuminated uniformly from the opposite side; the intensity pattern is seen to vary randomly in position on the scale of the ultrasonic wavelength, and can be thought of most simply as the diffraction pattern of all the randomly positioned scatterers in the sample. While speckles are a nuisance in static imaging applications, the fluctuations in the speckle pattern that occur when the scatterers are moving contain a wealth of information about the scatterers' dynamics.

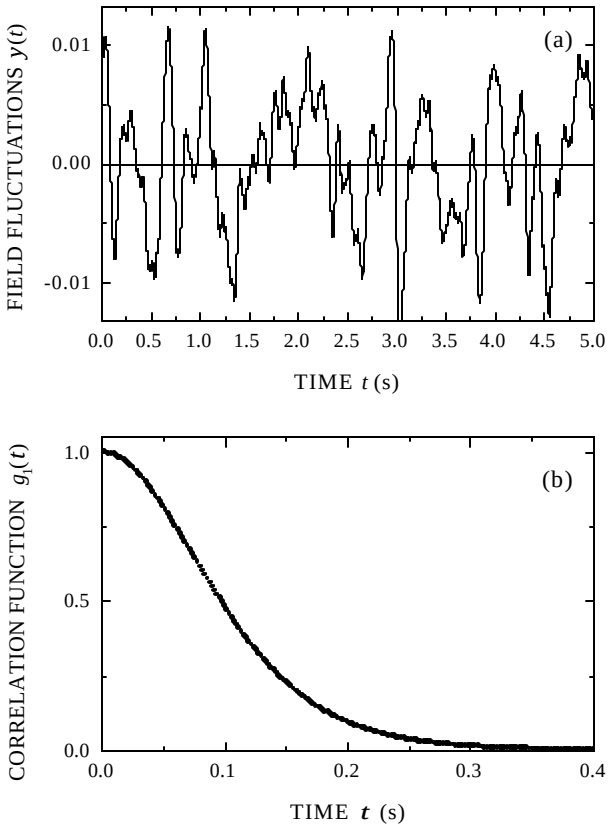

Figure 2. (a) Field fluctuations in a single speckle due to the motion of the scattering objects (b) Field autocorrelation function of these field fluctuations.

Figure 2(a) shows an example of the temporal field fluctuations $y(t)$ in a single speckle spot in a sample of glass beads suspended at a volume fraction of 40% by flowing the surrounding fluid upwards to counterbalance sedimentation. In ultrasonic correlation spectroscopy, the motion of the particles causing these field fluctuations is determined from the temporal field auto-correlation function,

$$g_1(\tau) = \int y^*(t)\, y(t+\tau)\, dt \Big/ \int |y(t)|^2 dt \qquad (1)$$

which we calculate from digitized records of the field fluctuations using a Fast Fourier Transform method. Figure 2(b) shows the auto-correlation function $g_1(\tau)$ for the data illustrated in Fig. 2(a). The decay of the autocorrelation function is caused by the increase with time in the variance of total change in the phase of scattering paths through the sample, $\langle \Delta \phi^2 \rangle$, which can be directly related to the motion of the particles.

3. DYNAMIC SOUND SCATTERING (DSS)

In Ultrasonic Correlation Spectroscopy, there are two regimes in which quantitative information on the dynamics of the scatterers can be obtained directly from $g_1(\tau)$: single scattering and multiple scattering. In Dynamic Sound Scattering, singly scattered ultrasonic waves are measured, and in this limit the field autocorrelation function has the relatively simple form

$$g_1(\tau) = \exp\left[-\frac{1}{2}q^2\langle\Delta r_{\vec{q}}^2(\tau)\rangle\right] \tag{2}$$

Here $q = 2k\sin(\theta/2)$ is the scattering wave vector (the difference between the incident and scattered wave vectors), $k = 2\pi/\lambda$ is the magnitude of the ultrasonic wave vector in the medium, θ is the scattering angle, and $\langle\Delta r_{\vec{q}}^2(\tau)\rangle$ is the mean square displacement of the scattering particles in the direction of $\vec{q}$. Thus by measuring the field fluctuations at a particular scattering angle, the temporal evolution of the mean square displacement of the particles can be measured.

We have developed two different implementations of DSS, a near-field and a far-field technique. Both of these methods use pulsed ultrasonic techniques, where a train of ultrasonic pulses is incident on the sample and the scattered field is measured at the same time in each transmitted pulse, allowing us to measure field fluctuations due to the motion of the scatterers at a rate determined by the pulse repetition frequency (e.g. see Fig. 2(a)). The near-field technique uses a small detector, having dimensions less than the wavelength, placed close to the sample on the side directly opposite the incident beam. Since, for an incident plane wave, it takes longer for ultrasound to reach the detector when it is scattered through a larger scattering angle, the average scattering angle can be determined from the time in the pulse at which the scattered field is measured. Because of the axial symmetry of the scattering geometry, this technique essentially measures a combination of the two Cartesian components of $\langle\Delta r^2(\tau)\rangle$ in the plane perpendicular to the incident beam direction. By contrast, the far-field technique requires the use of long path lengths in order to collimate the incident and scattered beams. This is done in a large water tank, with both the generating and detecting transducers placed at least 50 cm away from the sample, giving direct control over the scattering angle of the measured ultrasound with good angular resolution. In addition, by using the appropriate scattering geometry, the direction of $\vec{q}$ can be selected, and thus all three components of the mean square displacement can be measured separately. By combining these individual components, the total 3-dimensional rms displacement of the particles can also be determined. Thus while the near field apparatus is more compact and convenient to set up, the far-field technique provides more information about the dynamics of the particles.

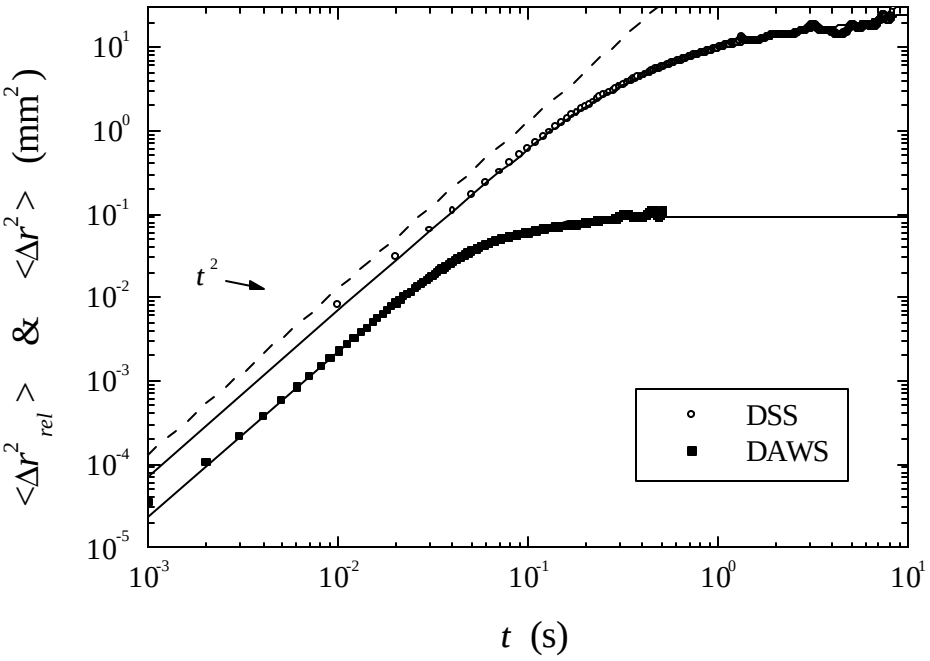

Figure 3. Mean square displacement of glass beads in a fluidized suspension measured in DSS, and the relative mean square displacement measured in DAWS.

Figure 3 shows an example of $\langle \Delta r^2(\boldsymbol{t}) \rangle$ measured by DSS. As is evident in this figure, $\langle \Delta r^2(\boldsymbol{t}) \rangle \propto \boldsymbol{t}^2$ at early times, indicating that the particles move ballistically at early times, with a root mean square velocity $V_{\rm rms}$ given by $\langle \Delta r^2(\boldsymbol{t}) \rangle^{1/2} / \boldsymbol{t}$. Typical results for $V_{\rm rms}$ as a function of the volume fraction of particles are shown in Fig. 4, where the values are normalized by the fluid flow velocity V_f. At later times, $\langle \Delta r^2(\boldsymbol{t}) \rangle$ no longer follows a quadratic time dependence but increases at a slower rate, as the particle trajectories deviate from straight-line paths. Modeling this behaviour using the phenomenological expression

$$\langle \Delta r^2(\boldsymbol{t}) \rangle = \langle \Delta V^2 \rangle \boldsymbol{t}^2 \Big/ \left[1 + \left(\boldsymbol{t}/\boldsymbol{t}_c \right)^{2-m} \right] \tag{3}$$

gives a good description of the data over the entire range of measurement times, as shown by the solid line in Fig. 3. Here $\boldsymbol{t}_c$ is the velocity crossover time and m is an exponent that characterizes the long time behaviour of $\langle \Delta r^2(\boldsymbol{t}) \rangle$. By fitting this expression (Eq. (3)) to our data, the crossover time $\boldsymbol{t}_c$ can be measured, allowing the dynamic velocity correlation length $d_{\rm c} = V_{\rm rms}\, \boldsymbol{t}_c$ to also be determined. Thus DSS gives important information on the temporal and spatial extent of correlations in the particles' motion, as well as measuring the magnitude of the rms particle velocity.

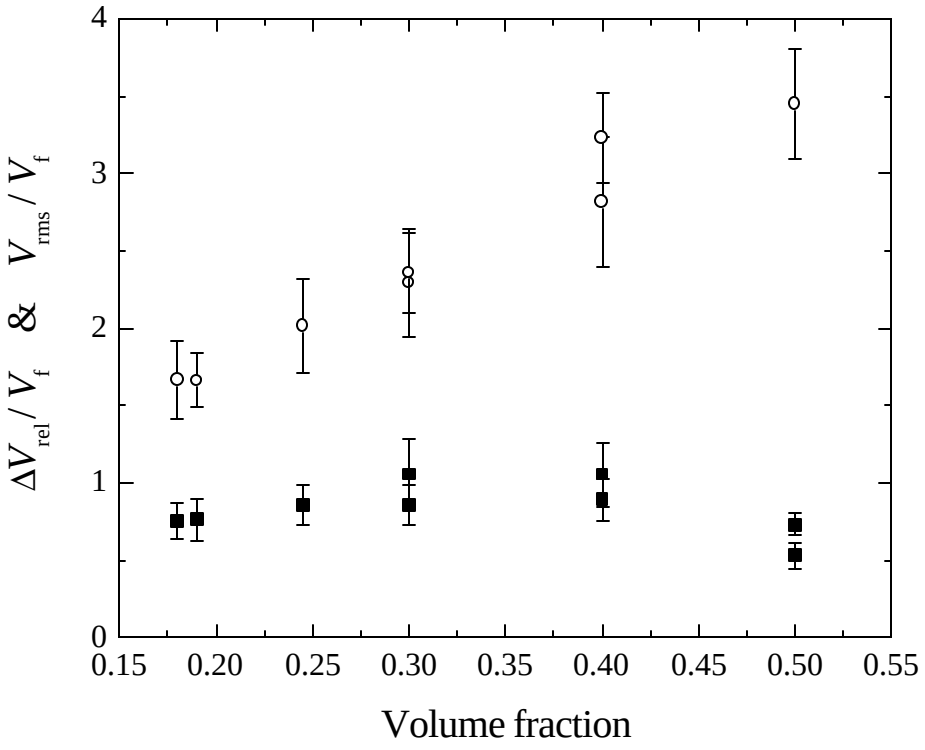

Figure 4. V_{rms} (open symbols) from DSS compared with ΔV_{rel} (solid symbols) from DAWS.

4. DIFFUSING ACOUSTIC WAVE SPECTROSCOPY (DAWS)

Additional information on the dynamics can be obtained using Diffusing Acoustic Wave Spectroscopy, which can be performed at higher frequencies where the ultrasonic waves scatter many times before leaving the sample. This technique, which is based on the analogous optical technique Diffusing Wave Spectroscopy [2-4], is made feasible by recent progress in understanding the diffusion of multiply scattered ultrasonic waves. We have shown the diffusion approximation to be an extremely powerful and remarkably accurate model for describing acoustic wave transport under these conditions [5-6]. By modeling the ultrasound propagation using the diffusion approximation, this novel technique can be used to determine the *relative* motion of particles that are separated, on average, by a distance equal to the ultrasonic mean free path l^*. Within the diffusion approximation, the field autocorrelation function for ultrasound that has undergone n scattering events can be written as

$$g_1(\boldsymbol{t}) \approx \exp\left[-\frac{nk^2}{6}\left\langle \Delta r_{rel}^2\left(\boldsymbol{t}, l^*\right)\right\rangle\right] \tag{4}$$

Thus by measuring the field autocorrelation function of scattered sound that has traveled a particular path length, the relative mean square displacement of neighbors in the scattering paths can be measured.

Since the mean free path can be varied by varying the ultrasonic frequency, DAWS can be used to probe the local relative motion of the particles over a range of length scales, down to the nearest-neighbor separation of the particles. In Figs. 3 and 4, the relative mean square displacements $\langle \Delta r^2_{rel}(\boldsymbol{t})\rangle$ and the local relative velocity ΔV_{rel}, measured using DAWS at the interparticle separation, are compared with the corresponding values on an absolute scale determined by DSS. The local relative velocity of the particles is found to be smaller than the rms velocity, which implies that the motion of neighboring particles is at least partially correlated.

To explore the extent of the velocity correlations, we take advantage of the fact that

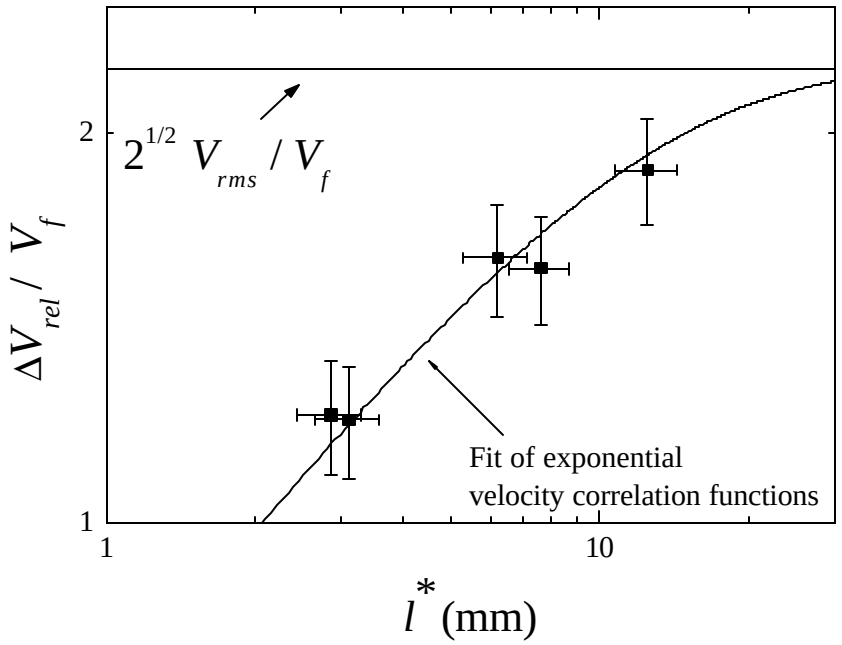

Figure 5. The length scale dependence of V_{rms} in a sample with a volume fraction of 0.15. The points are measurements using DAWS and the line is a fit assuming an exponentially decaying velocity correlation function .

the local relative velocity of particles separated by l^* can be related to the spatial velocity correlation function of the particles[1,7]:

$$\Delta V_{rel}^2\left(l^*\right) = 2\Delta V_{rms}^2\left[1-\frac{\left\langle V(\vec{x})V\left(\vec{x}+\vec{l}^*\right)\right\rangle}{\left\langle V(\vec{x})V(\vec{x})\right\rangle}\right] \tag{5}$$

Thus by measuring ΔV_{rel} as a function of l^* using DAWS and the rms velocity using DSS, the spatial velocity correlation function of the particles can be probed. This is illustrated in Fig. 5, where V_{rms} is plotted as a function of l^*. As the length scale l^* at which ΔV_{rel} is measured is increased, ΔV_{rel} also increases. The data can be fit using Eq. (5), assuming an exponentially decaying spatial velocity correlation function. The exponentially decaying velocity correlation function reproduces the data quite well, with the decay constant giving the instantaneous velocity correlation length $\boldsymbol{x}$. Thus the correlation length $\boldsymbol{x}$, which determines the size of the complex correlated flow patterns (swirls) that are characteristic of fluidized suspensions, can also be measured by combining DSS and DAWS.

5. CONCLUSIONS

We have described two new techniques in ultrasonic correlation spectroscopy, Dynamic Sound Scattering and Diffusing Acoustic Wave Spectroscopy. These results show the power of these two spectroscopic techniques to investigate the dynamics of complex systems, suggesting that DSS and DAWS may have important uses both for fundamental studies of dynamical behavior and for practical applications in the non-destructive evaluation of materials.

6. REFERENCES

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