
187th Meeting of the Acoustical Society of America*Virtual Meeting 2024*

18-22 November 2024

***Engineering Acoustics: Paper 3pEA2**

A low-cost ultrasonic absorption spectrometer mainly using off-the-shelf parts**Michelle R. Crouse***Department of Chemistry & Biochemistry, California State University, Dominguez Hills, Carson, CA, 90747; mruth.sci@gmail.com***Małgorzata Musiał and Jason A. Widegren***Materials Measurement Laboratory, National Institute of Standards and Technology, Boulder, CO, 80305; malgorzata.musial@nist.gov; jason.widegren@nist.gov***Jacob Pawlik***Communications Technology Laboratory, National Institute of Standards and Technology, Boulder, CO, 80305; jacob.pawlik@nist.gov***Bryan Bosworth***Materials Measurement Laboratory, National Institute of Standards and Technology, Boulder, CO, 80305; bryan.bosworth@nist.gov***Nathan Orloff, Aaron Hagerstrom, Angela C. Stelson and Robert Lirette***Communications Technology Laboratory, National Institute of Standards and Technology, Boulder, CO, 80305; nathan.orloff@nist.gov; aaron.hagerstrom@nist.gov; angela.stelson@nist.gov; robert.lirette@nist.gov*

Ultrasonic absorption spectroscopy can probe intermolecular interactions that inform research into chemical engineering and pharmaceutical manufacturing processes. The only commercial ultrasonic spectrometer costs over one hundred thousand dollars, putting it out of reach for many institutions. We designed an inexpensive ultrasonic absorption spectrometer comprised of off-the-shelf components and parts from a rapid prototyping service. We employed the through-transmission method for quantifying absorption, using thirty-one pulse measurements at varying distances between the transmitter and receiver. These measurements significantly improved absorption measurement accuracy compared to the fixed path technique and the pulse-echo method, both of which rely on only two measurements. We measured pure water to correct for diffraction effects and used repeated water measurements to propagate the uncertainty in the absorption measurement. We measured aqueous methylcellulose solutions and various salts to validate the spectrometer, comparing the data to the results obtained from a commercial spectrometer. The results were well within the margin of error for both devices. Notably, from these measurements, we observed a relaxation peak at approximately 1 MHz in scandium sulfate, which was reported once before using a resonator method. Our instrument offers an inexpensive alternative to a commercial ultrasonic absorption spectrometer accessible to university researchers and students.

**POMA Student Paper Competition Winner*

INTRODUCTION

Precise ultrasound absorption measurements can reveal information about a liquid's physical, chemical, and structural properties and relaxation phenomena.^{1,2} By studying these relaxation phenomena, we can directly link molecular structure, thermal motion, and side group dynamics to the mechanical properties of the materials.^{3,4} Acquisition of a commercial acoustic spectrometer requires significant capital investment. Universities typically operate within strict budget constraints, where high-tech equipment purchases compete with other priorities, such as lab renovations, general-purpose equipment, and faculty salaries. Universities must also balance research needs with teaching priorities. Acoustic spectrometers are typically research-focused, making it difficult to justify their cost if they do not contribute enough to student instruction. The primary expense lies in the acoustic spectrometer itself, and the cost of a commercially available absorption spectrometer can exceed \$100,000. Previous researchers have used elaborately machined devices, with costs approaching the commercial version due to machine shop fees.⁵ Here, we present a spectrometer made primarily from off-the-shelf parts, costing less by at least a factor of ten. This reduces institutions' expenses and gives them access to ultrasonic absorption spectroscopy measurements.

We used a variable path pulse through-transmission technique to measure the absorption coefficient, α , in various liquids.⁶ Absorption in liquids is the exponential attenuation in acoustic pressure amplitude, P , over a distance, x (Eq. (1)). Our technique used a pulse-transmitting transducer, which moves to various distances along a line extending to a receiving transducer. We measured these distances with a micrometer-adjusted translation stage. From the definition of the absorption coefficient, we calculated α from

$$\ln \frac{P(x)}{P_0} = -\alpha x. \quad (1)$$

We performed a linear fit of the natural logarithm of the frequency-resolved pulse amplitudes versus the distance between the transducers and then calculated the slope of that fit. This slope corresponds to $-\alpha$.

METHODS

EXPERIMENTAL METHODS

Our spectrometer was made from primarily off-the-shelf parts (Fig. 1a). The instrument covers a bandwidth of approximately 0.6 MHz to 6 MHz by employing interchangeable transducer pairs. The center frequency for Transducer A is approximately 1 MHz, Transducer B is approximately 2.25 MHz, and Transducer C is approximately 5 MHz. We designed custom aluminum mounts for the transducers and had them machined using an online rapid prototyping service for less than \$100. An off-the-shelf optics bench micrometer-adjusted translation stage moved the transmitting transducer (Fig. 1a). We purchased from a vendor a flexible sample holder that was molded from silicone. The sample holder has two holes on each side, which fit onto the two transducers, forming a seal (Fig. 1a), and it holds approximately 20 mL of liquid. This sample holder can also be made inexpensively using an online rapid prototyping service. The most expensive part of our instrument is the pulser-receiver, which transmits high-voltage pulses to one of the transducers (transmitting transducer) and receives low-voltage pulses from the other transducer (receiving transducer). The receiver section of the pulser-receiver amplifies and filters the received pulse. A 70 MHz bandwidth digital oscilloscope collected the data from the pulse receiver (Fig. 1b).

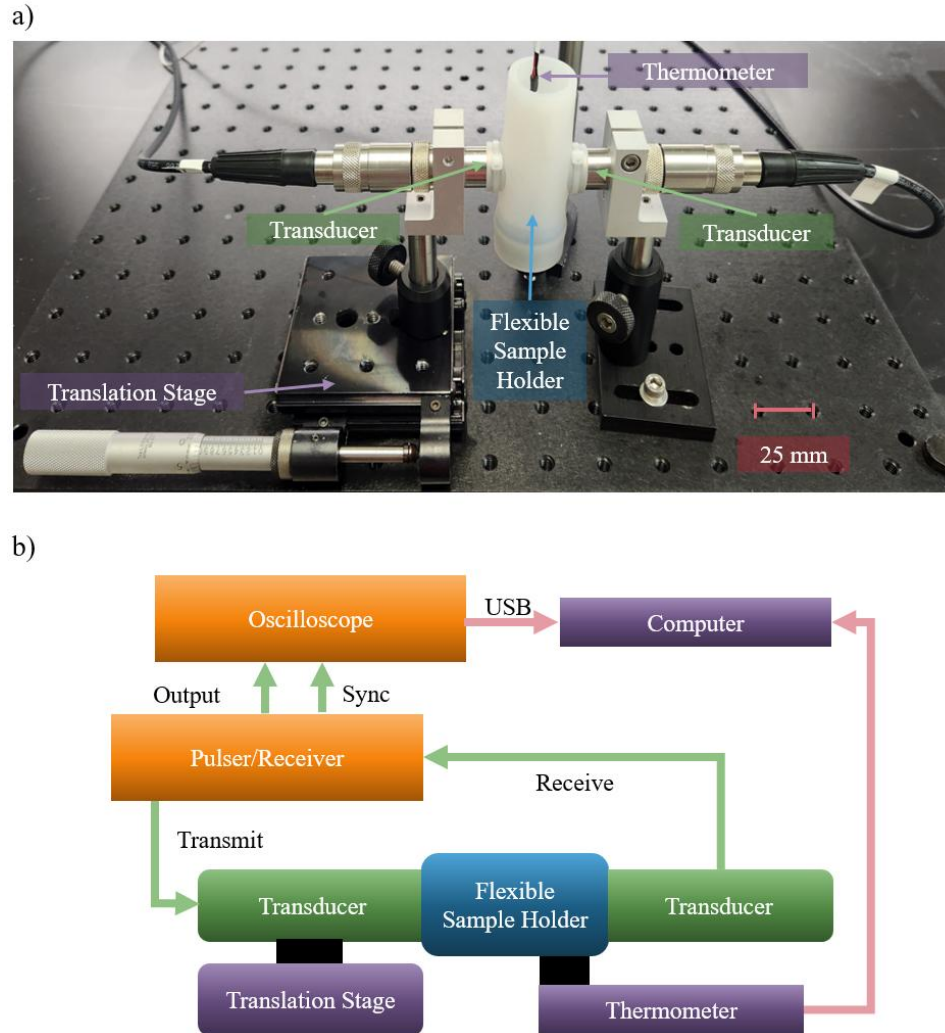


Figure 1: a) Image of our measurement setup. The translation stage moves the transmitting transducer to various distances towards the receiving transducer with the sample holder in between the transducers. The sample holder holds approximately 20 mL of sample volume. b) Diagram of our experiment. We used an oscilloscope to capture data from a pulser-receiver, which transmits and receives pulses to the transducers and samples. We also monitored the temperature of our samples via a thermocouple inside the sample holder, which was positioned out of the way of the acoustic path. The green arrows represent analog signal flow and the pink arrows represent digital signal flow.

We collected thirty-one pulse measurements at different distances, moving the transducers from approximately 25.400 mm to 6.350 mm apart in steps of approximately 0.635 mm. We performed these measurements at room temperature, approximately 21 °C to 23 °C. We performed an identical measurement run with the sample holder filled with deionized water for each sample measurement. This water data was used as a reference to correct for diffraction effects with our sample data, as discussed below.

DATA ANALYSIS

To process the data, we gated the waveform to remove the echoes between the transducers. We then measured the pulse's group sound speed, c , in our samples and water reference. To do this, we calculated the Hilbert envelope (the energy in the pulse) of each pulse we collected. Then,

by finding the maximum of this envelope, we determined the time delay between when each pulse arrived at the receiving transducer relative to when it arrived with the transducers in their nearest configuration, Δt . We then performed a linear fit of the distance between the transducers, x , versus time, Δt , to determine the group sound speed, c , using $x = c\Delta t$, where c is the slope. This assumes the fluid is not dispersive. We took relative distance measurements with our micrometer. Thus, the intercept of this fit represents the distance between the transducers in their nearest position.

Next, we took the Fast Fourier transform (FFT) to convert our data to the frequency domain. We then converted the data into the wavelength, λ , domain using the sound speed we measured as described above and in $\lambda = c/f$. This wavelength-resolved sample and water data were used to correct diffraction effects in our sample data. The transducers have a finite aperture size relative to the wavelength, which leads to diffractive spreading.⁷ We corrected this diffraction by using water as a reference and with the assumption that water has a known absorption coefficient as a function of the wavelength. We used the value from a table given by Pinkerton in Ref. 6 and extrapolated it to our measured temperature.⁸ The diffraction correction factor, $D(x, \lambda)$, was constructed from our wavelength-resolved water data, P_{water} , by dividing out its assumed attenuation due to absorption,

$$D(\lambda, x) = \frac{P_{water}(x, \lambda)}{e^{-\alpha x}}. \quad (2)$$

We then corrected our wavelength-resolved sample data, $P_S(x, \lambda)$, by dividing it by the diffraction correction,

$$P_c(x, \lambda) = \frac{P_S(x, \lambda)}{D(x, \lambda)}. \quad (3)$$

Using this method, we assume that any changes in amplitude are due only to absorption and diffraction. We previously attempted to use a theoretical expression for the Lommel-diffraction correction.⁹ However, this led to high uncertainties in our data, especially below 1 MHz.

UNCERTAINTY ANALYSIS

Our most significant sources of overall measurement uncertainty are in our measurements of pressure amplitude and the distance between the transducers. The uncertainty in the pressure amplitude is mainly due to the uncertainties in the transduced voltage. The uncertainty in our distance measurements is due to alignment and the uncertainty of our micrometer translation stage. We measured pressure amplitudes versus distance twenty-four times with our device to quantify these uncertainties. Then, we calculated the standard deviation of the amplitude versus distance and frequency for each transducer pair. We used water for this measurement because it has low dispersion and absorption, which yielded a relatively large amplitude measurement and allowed us to resolve the uncertainty. Figure 2 shows Transducer A's standard deviation of the normalized pressure amplitudes, plotted over its 6 dB bandwidth. Generally, the standard deviation is a minimum near the fundamental frequency of each transducer, where the transducer is operating optimally.

For the uncertainty in distance, we first measured the sound speed in this water data using the method described above. We determined the error in our sound speed measurement by comparing our measurement to a 5th-order polynomial fit based on temperature.¹⁰ We assume the error in time to be due to the jitter in the clock of our digital sampling oscilloscope, approximately 1 ns per the

manufacturer's specification with the time base settings we used. This error is small compared to our measured times (approximately 1.2 ns), by 4 orders of magnitude, and is neglected. Thus, we conclude that the relative uncertainty in our distance measurement is approximately the same as the relative uncertainty in our sound speed measurement. We used this value in our uncertainty analysis. We propagated these uncertainties through the linear regression algorithm used in our absorption measurements.¹¹

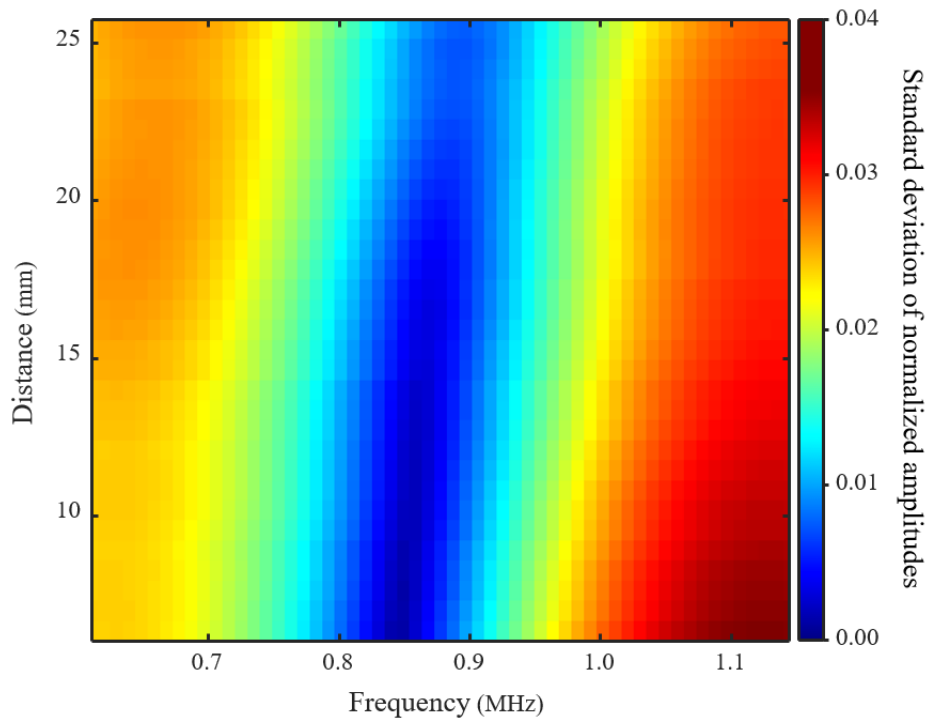


Figure 2: *Standard deviation (unitless) of the normalized pressure amplitudes measured with the Transducer A pair versus frequency and the distance between the pair. The minimum standard deviation for this pair was near its fundamental frequency, approximately 0.87 MHz. The error appears to shift as the transducers are pulled apart, indicating a slight misalignment of the transducers. We normalized the standard deviation by the amplitudes and then multiplied it by our measurement amplitudes to obtain the standard uncertainty in the pressure amplitude measurement.*

RESULTS

To validate our spectrometer, we measured a variety of liquids with a range of expected absorption coefficients. We compared our results of the same liquids to measurements made with a commercially available acoustic absorption spectrometer. The result for the highest absorbing liquid we measured, zinc chloride (1 M), is shown in Fig. 3a. Zinc chloride has a pronounced chemical relaxation at approximately 11 MHz, which can be seen in Fig. 3a. The plot line shown in dark blue with error bars represents data obtained from the commercial spectrometer, and the error bars are the manufacturer's stated uncertainty for the device, 0.1 dB/(cm·MHz). Data obtained with Transducer A are in light blue, Transducer B are in orange, and Transducer C are in green. The shaded regions in each frequency range represent the standard uncertainty discussed above. We used this coloring scheme for all results presented below. The bandwidth of the commercial system ranged from approximately 3 MHz to 100 MHz. This overlaps with the bandwidth of Transducer C, which is approximately 3 MHz to 6 MHz, allowing us to compare our

measurements with measurements taken by the commercial device. The total frequency range of our system was from approximately 0.6 MHz to 6 MHz.

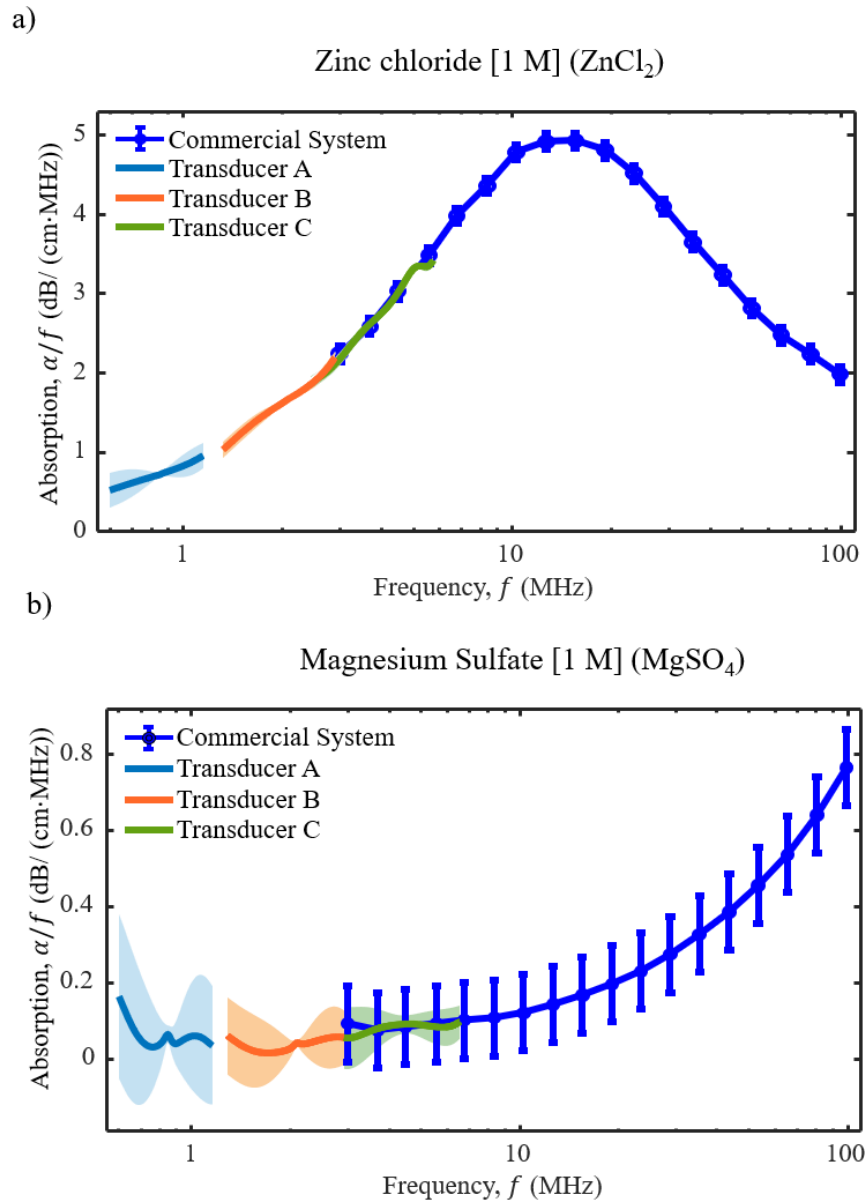


Figure 3: Validation with high- and low-absorbing liquids. Absorption coefficient versus frequency for a) Zinc chloride (approximately 1 M) has a much lower absorption coefficient overall.

To test the limits of our measurement capabilities, we also measured a liquid that we expected to have an extremely low absorption coefficient in our frequency range, magnesium sulfate (1 M). Figure 3b shows the results for magnesium sulfate, a well-studied salt solution.⁷ Due to magnesium sulfate's much lower absorption than zinc chloride, it has a higher relative error. The results obtained with Transducer C overlap well within the error bars of the commercial spectrometer. Measurements with Transducers A and B demonstrate a consistent trend versus frequency, validating our measurement results even with weakly absorbing solutions.

To further validate our results, we measured the absorption coefficient of scandium sulfate to compare our results with published data (see Fig. 4). Our results were within the uncertainty

bounds of results obtained with the commercial spectrometer, similar to our previous results. As shown in Fig. 4, our results show a relaxation peak at approximately 1 MHz, which the commercial system does not capture due to bandwidth limitations. This same relaxation peak was previously reported in 1978 using a fixed path-length resonator method.¹² The black dashed curve in Fig. 4 was calculated from the three Debye relaxation coefficients reported by Bensen et al. in Ref. 12. There is less than a 0.1 dB/(cm·MHz) difference between our data and this previously published result despite the vastly different measurement methods. This difference in magnitude of the absorption coefficient may be due to slight differences in concentration and sample temperatures compared to data published by Bensen et al. in Ref. 12.

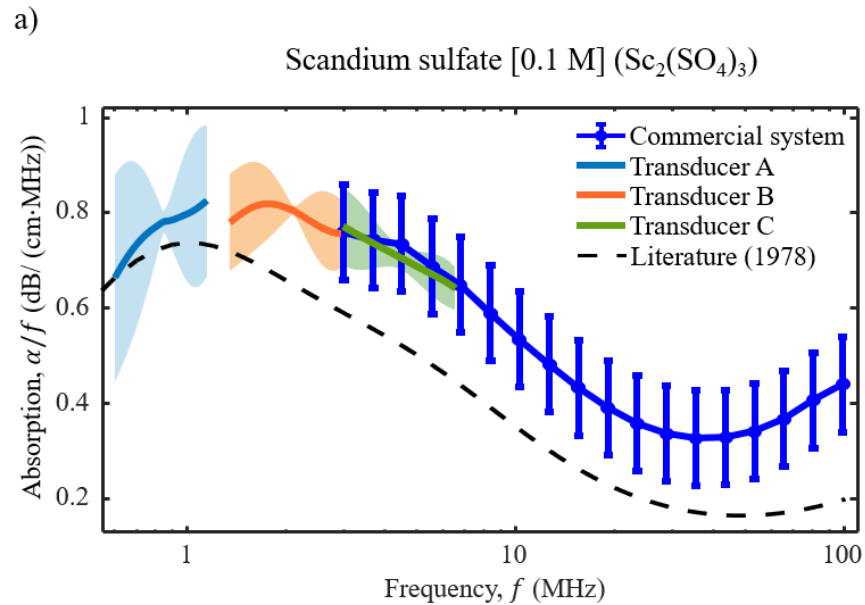


Figure 4: Absorption coefficient versus frequency of approximately 0.1 M concentration by mass of scandium sulfate was compared to data collected with the commercial system and published data⁴. There is a relaxation peak at approximately 1 MHz, which can also be seen in the previously published result.

We measured aqueous methylcellulose solution at a 1% by mass concentration (Fig. 5). This system may have relaxations due to the local motion of chain segments through glucosidic linkages, known as beta-relaxations, previously seen in dielectric spectra for methylcellulose.^{13,14} However, we cannot resolve them within the uncertainties of our measurements.

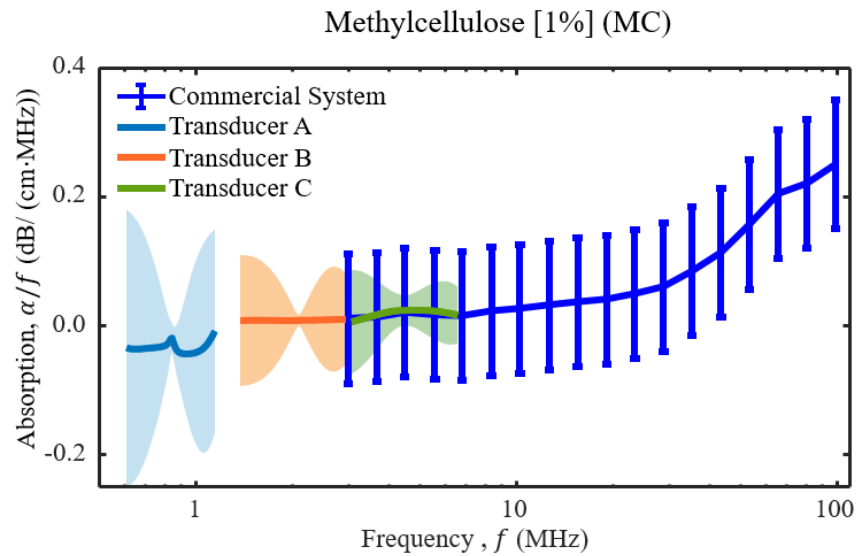


Figure 5: Absorption coefficient versus frequency for aqueous methylcellulose solution at 1% by mass concentration. This fluid has a very low overall absorption coefficient.

CONCLUSION

Barriers faced by universities to the acquisition of an acoustic spectrometer include the prohibitive cost of the instrument acquisition, justification of the use of the instrument, and ongoing maintenance requirements. To overcome these barriers, one typically must secure dedicated funding and provide clear justification for the instrument's long-term research and educational value. The ultrasonic absorption spectrometer we described here offers an alternative to commercial systems, costing around one-tenth the amount and enabling university researchers and students to explore acoustic absorption metrology in liquids. This enables investing in an inexpensive alternative to a commercial or highly customized system. We have demonstrated the accuracy of this spectrometer by comparing data taken on our spectrometer to data obtained using a commercial system.

We observed low-frequency relaxation peaks in liquid ultrasonic absorption data taken with our spectrometer. Our spectrometer's low-frequency bandwidth captured the approximately 1 MHz relaxation peak in scandium sulfate that was not measurable on the commercial system. The data agrees with data taken with the commercial spectrometer within the uncertainty bounds, and with published data.¹²

A key innovation of our spectrometer was a measurement-based correction for diffraction effects. We devised a method to directly measure diffraction effects instead of relying on a theoretical estimate. We measured the attenuation of a water sample and divided out an assumed known value for the absorption in water. We then normalized this data by wavelength to create our custom diffraction correction unique to our instrument.

Despite our demonstrated accuracy and precision, our instrument has a few limitations. With the current design, each transducer pair must be the same diameter to fit in the sample holder. The transducer size affects its possible frequency range. The transducers we used were approximately 13 mm in diameter. This limited our accessible frequency range from approximately 0.6 MHz to 6 MHz using the same size and series transducers from the manufacturer. Using another set of mounts that hold different sizes of transducers could further extend the instrument's frequency range. Another limitation of our method is manual manipulation of the transducer's position, which is a significant source of uncertainty in our measurements. This could be significantly improved

with motorized automation. Lastly, with this setup, we could not control the temperature of our samples, so we obtained all our measurements at ambient (room) temperature. Nevertheless, our methods demonstrated accuracy and precision comparable to the commercially available spectrometer within the uncertainties we assessed. To further illustrate the utility of our instrument, we plan to test our device with other liquids, such as hydroxyethyl cellulose and carboxymethyl cellulose. We also plan to test more transducers to extend the measurable frequency range. Lastly, we plan to automate the setup with a motorized, digitally controlled translation stage instead of the manual micrometer stage, which could lead to lower measurement uncertainties.

ACKNOWLEDGMENTS

This work was partially supported by a National Institute of Standards and Technology (NIST) Summer Undergraduate Research Fellowship (SURF). NIST does not specify commercial manufacturers or endorse products.

REFERENCES

- ¹F. Eggers, and Th. Funck, “Ultrasonic Measurements with Milliliter Liquid Samples in the 0.5–100 MHz Range,” *Rev. Sci. Instrum.* **44**, 969–977 (1973). doi:10.1063/1.1686339
- ²H. E. Bass, H.-J. Bauer, and L. B. Evans, “Atmospheric Absorption of Sound: Analytical Expressions,” *J. Acoust. Soc. Am.* **52**, 821 (1972). doi:10.1121/1.1913183
- ³U. Kaatze, R. Behrends, and K. Lautscham, “Acoustic relaxation spectrometers for liquids,” *Ultrasonics* **39**, 393–406 (2001). doi:10.1016/S0041-624X(01)00078-6
- ⁴U. Kaatze, T. O. Hushcha, and F. Eggers, “Ultrasonic Broadband Spectrometry of Liquids: A Research Tool in Pure and Applied Chemistry and Chemical Physics,” *J. Sol. Chem.* **29**, 4 (2000).
- ⁵F. Eggers, and U. Kaatze, “Broad-band ultrasonic measurement techniques for liquids,” *Meas. Sci. Technol.* **7**, 1–19 (1996).
- ⁶L. W. Labaw, and A. O. Williams Jr., “Absorption of Supersonic Waves in Water near One Megacycle,” *J. Acoust. Soc. Am.* **19**, 30–34 (1947). doi:10.1121/1.1916399
- ⁷L. E. Kinsler, A. R. Frey, A. B. Coppens, and J. V. Sanders, *Fundamentals of Acoustics*, 4th Edition, (Wiley, New York, 2000).
- ⁸J. M. M. Pinkerton, “A Pulse Method for the Measurement of Ultrasonic Absorption in Liquids: Results for Water,” *Nature*, **160**, 128–129 (1947).
- ⁹P. H. Rogers, and A. L. Van Buren, “An exact expression for the Lommel-diffraction correction integral,” *J. Acoust. Soc. Am.* **55**, 725 (1974). doi:10.1121/1.1914589
- ¹⁰W. Marczak, “Water as a standard in the measurements of the speed of sound in liquids,” *J. Acoust. Soc. Am.* **102**, 2776–2779 (1997). doi:10.1121/1.420332
- ¹¹D. York, N. M. Evensen, M. L. Martínez, and J. De Basabe Delgado, “Unified equations for the slope, intercept, and standard errors of the best straight line,” *Am. J. Phys.* **72**, 367–375 (2004). doi:10.1119/1.1632486
- ¹²A. Bonsen, W. Knoche, W. Berger, K. Giese, and S. Petrucci, “Ultrasonic relaxation studies in aqueous solutions of aluminium sulphate and scandium sulphate,” *Phys. Chem.* **82**, 678–683 (1978).
- ¹³P. Pissis, and D. Daoukaki-Daimanti, “Dielectric relaxation of water in the water-methylcellulose system,” *Chem. Phys.* **123**, 165–173 (1988). doi:10.1016/0301-0104(88)87042-3
- ¹⁴A. Rachocki, E. Marchewicz, and J. Tritt-Goc, “Dielectric Relaxation in Cellulose and its Derivatives,” *Acta. Phys. Pol. A.* **108**, 137–145 (2005).