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Sub-microsecond temperature measurement in liquid water using laser-induced thermal acoustics

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Abstract: Using laser-induced thermal acoustics, we demonstrate non-intrusive and remote sound speed and temperature measurements over the range 10 – 45 °C in liquid water. Averaged accuracy of sound speed and temperature measurements (10 s) are 0.64 m/s and 0.45 °C respectively. Single-shot precisions based on one standard deviation of 100 or greater samples range from 1 m/s to 16.5 m/s and 0.3 °C to 9.5 °C for sound speed and temperature measurements respectively. The time resolution of each single-shot measurement was 300 nsec.

OCIS codes: 300.2570, 120.6780, 120.4640, 120.0280, 120.5820, 290.5900.

1. Introduction

Fast non-intrusive temperature measurements are needed for numerous applications. This is especially true for the study of shock waves. Bio-medical shock wave applications such as Extracoporeal Shock Wave Lithotripsy (ESWL) have revealed collateral damage to healthy tissue, in addition to the targeted cancerous tissue.[1,2,3] To facilitate the understanding of the damage mechanism and to predict the amount of damage, scientists are studying shock waves in fluids that have acoustic properties similar to human tissue.

For relatively strong shocks with overpressures of 10-100 GPa (100-1000 katm) propagating through liquids, the associated temperatures of ~ 5000 K generate strong thermal radiation in the visible region and can be measured with optical pyrometry.[4] For weaker shocks (~ 1 katm) used in ESWL the situation is different, since the associated temperature jumps of ~ 10 K are not large enough to generate appreciable visible radiative emission. The pressure histories of these weaker shocks waves are easily studied with hydrophones[1] and reveal sharp (~300 ns) pressure increases. But, temperature measurements with similar temporal resolution are not routine, for example, thermocouples are relatively slow (> 100 μs). Previous work[5] has demonstrated ns-temporal resolution in shock wave studies, however only under special conditions (1-dimensional geometry with a line-of-sight diagnostic) and for parameters other than temperature. Thus, we anticipate that future shock-wave research will benefit from the development of additional non-intrusive, spatially resolved, and fast (≤ 1 μs) temperature measurement techniques.

There are several optical temperature measurement techniques that demand consideration for fast, spatially-precise non-intrusive temperature measurements in liquid water. Karl et al.[6]

describe two optical temperature measurement techniques that use Raman spectroscopy. One uses a single laser beam and the other uses a laser sheet. Both techniques extract temperature from spectroscopic information obtained from the intensity ratio at two wavelengths. Both techniques use long integration times necessitated by the use of continuous wave (cw) lasers. By employing a high-powered pulse laser and two intensified charge coupled device (ICCD) cameras, this technique might have potential for single shot, spatially precise water temperature measurements, but likely would still suffer from low signal-to-noise ratio (SNR) and, thus, imprecise temperature/sound speed measurements. If larger laser intensities were used, the water would most likely "break down", forming bubbles.

Other potential temperature measurement techniques include fluorescence and Brillouin scattering. According to Lou et al.,[7] thermochromic shifts (i.e. the peak fluorescence wavelength shifts to higher frequencies with increasing temperature) from various dyes doped into water are useful for measuring large temperature changes. Typical shifts are on the order of 0.1 nm/°C, making temperature measurement resolution, in water, impractical below temperature differences less than about 10°C, according to the authors. Another fluorescence technique described by the same authors uses the ratio of monomer-to-excimer fluorescence to determine temperature. While this method may be precise enough to study shock waves in liquids, single-shot measurements have not yet been demonstrated. Fry et al.[8], measured the sound speed and temperature and salinity of ocean water by measuring the Brillouin width and shift using Brillouin LIDAR. Because of low signal levels, about 10 laser shots are integrated. In the application of measuring rapidly moving shock waves, ranging requirements for spatial resolution would require such short laser pulses that they would be too spectrally broad to observe the Brillouin linewidth. Furthermore, these optical techniques involve time averaging or multiple shots because of low SNR.

The laser-induced thermal acoustics (LITA) method, an optical method that produces a relatively strong coherent signal, has been used extensively for fast, remote sound speed and temperature measurements in gaseous flows.[9-13] In liquids, Nagasaka et al.[14] used a technique very similar to our LITA setup (however, using thermal gratings instead of electrostriction gratings), called forced Rayleigh scattering. They used this method to measure the thermal conductivity of various samples, including water. This technique, using thermal gratings, has the disadvantage of adding significantly more heat to the sample medium than the electrostriction gratings version used in the present paper. Similarly, Maznev et al.[15] used a laser-induced grating technique to make quick non-contact acoustic measurements in water and on transparent biological materials over a frequency range 30 MHz to 1 GHz to determine the dependence of the acoustic attenuation on frequency. Measurements were made over several hundreds of laser shots. Like those of Nagasaka et al., these measurements used absorbing laser wavelengths, resulting in the domination of thermal gratings. However, neither of these two research groups reported using the oscillation frequency to determine the sound speed/temperature of the host liquid.

In this paper, we present the use of LITA with electrostrictive gratings for measuring sound speed in liquid water, from which temperature can be inferred, with a time resolution of better than 1 μs. Water, which makes up approximately 95% of the human body, is an obvious candidate for preliminary investigations, and has an acoustic impedance close to living tissue.[2] The thermodynamic properties of water will limit the useful temperature range of this LITA technique to ~ 5-75 °C.

2. LITA Background

The LITA method involves the crossing of three laser beams. These three beams interact with the medium to produce a fourth called the signal beam. Two input pump beams form a grating in the medium. A third input beam, the probe, scatters from this grating, producing a signal beam that is temporally modulated according to the properties of the medium.

The simplified idea behind LITA is that an intense stationary electromagnetic interference pattern is produced in the medium (pure water in our experiment) during the 10-ns pump pulse. In water, there is very little absorption of the light wave energy at 532 and 488 nm, and the electrostriction process dominates over the thermal grating process. Water molecules, attracted to areas of high electric field, rapidly begin moving from low- to high-field regions. The movement of water molecules necessitates an increase in density in the regions of high electric field and a decrease in the density in the regions of the lower electric field. A pair of induced sound waves counter-propagate away from the interaction region, creating a dynamic density grating, and thus dynamic index of refraction grating that modulates the scattering of the incident probe beam according to the properties of the medium.

3. Measurement Method

In this paper, temperature is inferred from speed of sound, which is measured from the ultrasonic modulation frequency of the LITA signal. We have used an empirical equation, the sum of an exponential and two sine-exponential products with a LabVIEW curve-fitting algorithm, to determine the oscillation frequency of the signal.

To determine the sound speed from the measured frequency we start with the sound speed relationship $c = \Lambda\nu$, where c, Λ, and ν are the sound speed, acoustic wavelength, and acoustic frequency respectively. Eichler et al.[16] express Λ in terms of the laser pump wavelength (λ_p) by the following expression:

$$\Lambda = \frac{\lambda_p}{2\sin(\frac{\theta}{2})} \qquad (1)$$

where θ is the total pump beam crossing angle. The beat frequency for electrostriction gratings is twice the thermalization beat frequency as discussed by Cummings et al.[17] Therefore ν = f/2, where f is the observed/measured electrostriction frequency. If we substitute Equation 1 and ν = f/2 into c = Λν we arrive at the LITA electrostriction frequency relationship with the sound speed:

$$c = \frac{\lambda_p f}{4\sin(\frac{\theta}{2})} \qquad (2)$$

Temperature is determined from a polynomial fit of a transposed expression developed by Chavez, Sosa, and Tsumura[18] shown in Figure 1. Using a commercial spreadsheet, we plot Chávez et al. sound speed versus temperature between the region of 10 – 75 °C with sound speed plotted as the independent variable. We then fit a 6th order polynomial to this data. Using the 6th order polynomial we then were able to directly relate LITA measured sound speed to temperature. In water, the sound speed approaches 1555 m/s near 75 °C and then decreases after that, thus the technique is double valued and very insensitive near this peak. Furthermore, this peak sound speed value presents a problem when noise in the measurement causes the sound speed to be measured above 1555 m/s because there is no temperature correlation above this value. Therefore, as a simple solution, we extrapolate the range of the 6th order polynomial above 75 °C so that sound speeds above 1555 m/s registered as temperatures above 75 °C, although this is not physically possible. As mentioned above, the sound speed increases more rapidly with temperature at lower temperatures and less rapidly at moderate temperatures, reaching a zero rate of increase near 75 °C and decreasing thereafter. This is visually demonstrated in Figure 1, where it is shown that there is an increase in the temperature uncertainty (represented by the distance between the vertical lines) with increasing temperature in the region between 10 - 75 °C, assuming constant uncertainty (distance between the horizontal lines) in speed of sound.

Using this method requires either a very accurate measurement of the beam crossing angle or a calibration point determined at a known temperature. We use the latter method at ambient temperature, where the water is the most uniform in temperature and the least agitated, i.e. the least amount of bubbles and thermal currents.

4. Experimental setup

Figure 2 shows a diagram of the experiment. The setup included two lasers: a 10-ns pulsed, frequency-doubled Nd:YAG laser operating at 532 nm and 10 Hz, and a continuous wave Argon-ion laser operating at 488 nm. After splitting the 532 nm pump beam with a 50/50 beam splitter, the energy of each pump beam used was about 1 mJ/pulse. The power of the 488-nm probe beam was approximately 400 mW. The probe beam was acoustically-optically modulated, with an approximately 10 μs pulse, to prevent scattered light from continuously entering the photodetector.

An electrostriction grating can be formed and read out by pump and probe beams of nearly any wavelength. In contrast, thermal gratings are formed and dominate, only, when the pump laser wavelength matches a strong absorption in the sample. For the grating-forming pump beams, 532 nm was chosen for two reasons. First, there is very little absorption in water at 532 nm, which is paramount when using electrostriction as the dominating grating-forming mechanism. Secondly, 532 nm light was readily available from a 10-ns pulse Nd:YAG laser.

The probe wavelength was chosen as 488 nm because it also has little absorption in water and because its wavelength separation from 532 nm makes it feasible to spectrally reject, with an interference filter, the 532-nm pump scatter and the intermittent stimulated-Raman scatter.

The probe beam, from the Argon-ion laser, intersects and crosses the grating at an angle of approximately 0.5 degrees determined from phase matching considerations. A photo-multiplier tube (Hamamatsu H6780) connected to a digital oscilloscope (Tektronix TDS 584D) was used to monitor and acquire the oscillating signature of the signal.

The number of observed oscillations of the signal is dependent on the wavelength of the laser, the crossing angle (see Equation 2), the three input beam diameters; the shape of the decay is a function of the size of the interaction region and the acoustic-decay properties of the medium. The effect of the pump-beam diameter is shown in Figure 3. Both signal traces, in Figure 3, are produced using 532-nm laser beams crossed at about 1 deg. In the top signal trace, the pump beams (about 1-cm diameter) are not apertured before the focusing lens, resulting in a calculated pump beam width and height of approximately 50 x 50 um in the interaction region. About 8 oscillations occur in this configuration.

For the bottom trace, we inserted a 4.5 mm internal diameter stainless steel flat washer into the pump beam before the 50/50 beam splitter and then rotated the washer nearly 70 deg about the vertical axis, effectively aperturing the pump beams, mainly in the horizontal dimension. Thus, in the interaction region, the calculated diffraction spreads the horizontal pump-beam width to about 250 μm, but leaves the vertical portion only slightly larger - about 110 μm. This effect increased the number of oscillations to approximately 30. Not only did spreading the beam width at the crossing point create more oscillations, making the frequency determination more accurate, but it also reduced the pump-beam peak intensity which had the effect of reducing stimulated Raman interference and the probability for laser-induced breakdown in the focal region.

Each single-shot temperature measurement consisted of 500 temporal points digitized at 1 GHz. Mean temperatures were determined by averaging the temperatures determined from either 100 or 125 laser shots. We obtained 56 averaged measurements at various temperatures between 10 – 75 °C. A LabVIEW curve-fitting algorithm was used to fit the data to an empirical equation involving the sums of exponentially-damped sine waves. This empirical model was not an accurate physical model of the LITA signal, but gave quick and accurate determinations of the LITA signal modulation frequency. Data that were determined to be errant (outliers) were discarded when the signal amplitude was below threshold (< 20% of average signal amplitude). Data below threshold level consisted of less than 10% of the total data. A fast Fourier transform (FFT) algorithm was also used to determine the signal modulation frequencies, but was found to produce larger root-mean-square (RMS) fluctuations.

5. Results

A comparison of three single shots and a 100-shot average measurement, taken at room temperature, is shown in Figure 4. The traces show the characteristic oscillations associated with dynamic diffraction gratings caused by the counter-propagating acoustic waves. They also show the exponential-like decay caused by the combination of the acoustic dissipation and the acoustic waves passing out of the probe volume. The first three traces are single-shot acquisitions and the fourth trace is an average of 100 single-shot acquisitions. The data has been offset for comparison. The quality and SNR for the single-shot signals compare well with the 100-shot average, making single-shot measurements very practicable. Also, alternating valley heights (particularly evident in the bottom averaged signal) indicate a small contribution from thermal gratings. See Cummings for discussion on thermal and electrostriction contributions.[17] We believe that this is caused by a small amount of absorption of the 532-nm pump beams in water of approximately 0.0004 cm^{-1}.[19] However, impurities in the water could also be absorbing some of the laser energy.

The large SNR of the single-shot measurements is very important for making temperature measurements that are accurate and fast. Less precise determination of the frequency could reasonably be made in much less than 300 ns (e.g., three oscillations over 30 ns) with the data shown in Figure 4, since only a few oscillations are needed for FFT or curve-fit analysis. Thus we could, in principle, have made 30-ns measurements, albeit with larger uncertainties than we are quoting below, instead of 300-ns measurements. Since the frequency of the oscillation is governed by the crossing angle of the pump beams, increasing the pump-beam crossing angle could also, potentially, improve the measurement quality or shorten the measurement time. Caution is required to keep the period of the acoustical oscillations longer than the duration of the pump-laser pulse; otherwise the grating would be washed out. This was a limiting factor in the present experiment. It limited our pump beam total crossing angle to about 2 ° or less.

In Figure 5, single-shot LITA acoustic frequency traces are plotted as a function of time for six different temperatures between 10 – 60 °C. Upon careful inspection, the oscillation frequency is observed to increase with temperature. The SNR degrades as the temperature is varied away from room temperature. There are several reasons for this. The system was aligned and the crossing angle was calibrated at a room temperature of about 17 °C. As the temperature was changed from ambient, temperature gradients were formed in the water, due to minimal insulation around the water oven. This natural convection produced time-dependent beam steering for the three input laser beams and the diffracted signal beam. This beam steering could change the crossing angle and thus the measured frequency. Furthermore, it could cause the overlap of the beams to not be optimal. Beam steering also caused the diffracted signal beam to move with respect to the pinhole that was used as a spatial filter, hence changing the transmission of the signal through the pinhole. The effects of the temperature gradients worsened with increasing temperature deviation from ambient. Despite the problems with beam steering, the LITA sound speed measurement technique produced accurate and precise sound speed measurements over the range 10 - 45 °C.

Figure 6 shows a simultaneous plot of calculated sound speed and 56 LITA mean sound speed measurements versus temperature. Of these 56 measurements, 40 were in the range 10 – 45 °C. In this temperature range, the averaged accuracy of the mean sound speed measurements was 0.64 m/s. Here, the averaged accuracy is defined as the averaged absolute difference between the LITA sound speed measurement and the calculated sound speed. The precision of single-shot measurements (based on 1 σ, or standard deviation of ≥100 samples) ranged from 1 m/s to 16.5 m/s. The error bars plotted are ± 1 σ. The uncertainties in these mean values (not plotted) are about 10 times smaller than these plotted error bars, owing to the averaging of ≥ 100 measurements. As can be seen from the data and selected error bars on the plot, the scatter appears to increase with deviation from room temperature. This suggests that beam steering is a major contributor to the decreasing SNR with variance from room temperature. In situations without the strong thermal gradients and beam steering, as in our current experiment, one could reasonably expect somewhat better results than that shown at the higher temperatures of Figure 6.

In Figure 7, 55 of the mean LITA temperatures inferred from the 56 LITA sound speed measurements of Figure 6 are plotted against simultaneous thermocouple temperature measurements. The thermocouple was located approximately 6 mm above the LITA temperature measurement location. The 56th LITA temperature was, anomalously, determined to be about 125 °C when the thermocouple read 73 °C, and lies outside the range of Figure 7. The dashed diagonal line represents perfect agreement between the LITA measurement and the thermocouple measurement. The figure shows very good agreement between LITA-measured temperature and the thermocouple in the region 10 – 45 °C. In this region, the averaged accuracy of the LITA single shot temperature measurements was 0.45 °C and the single shot precisions (based on 1 σ of > 100 shots) of the temperature measurements range from 0.3 °C to 9.5 °C. Here averaged accuracy is defined as the averaged absolute difference between the LITA inferred temperature measurement and the thermocouple temperature measurement. Due to the sound speed maximum at about 75°C, the measured LITA temperatures approaching 75 °C become inaccurate.

6. Conclusion

We have made non-intrusive sound speed and temperature measurements in pure liquid water over a range 10 – 75 °C in less than 300 nanoseconds per measurement using LITA. In the range of 10 – 45 °C, the averaged accuracy of mean sound speed measurements was 0.64 m/s and single shot precisions (1 σ) ranged from 1 m/s to 16.5 m/s. The averaged accuracy of the LITA inferred mean temperatures over the same range was 0.45 °C and the single shot precisions (1 σ) ranged from 0.3 °C to 9.5 °C. While LITA temperature measurements have been made previously in various gases, we believe that this is the first time that this technique has been extended to the sound speed and temperature measurement of liquids. It is our belief that this technique will have future potential for making spatially-precise, non-intrusive, accurate and precise temperature measurements in water for a wide spectrum of experiments, especially the study of shock waves in water for bio-medical applications. This technique is preferred over the other techniques mentioned because of the coherent nature of the signal beam, which allows for fast single-shot acquisitions that are accurate and precise.

Our study proved that making fast (300 ns), accurate, spatially resolved (.1 x 0.25 x 30 mm^3),[height x width x length] and non-intrusive temperature measurements in water is possible. Using an improved LITA system, with a shorter pulse duration (a few hundred pico-seconds) laser and crossing the pump beams with a larger angle, faster temperature measurements with much better spatial resolution may be possible. In addition, liquids other than water, that also simulate the acoustic properties of body fluids, may provide a larger usable temperature range.

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References

1. S. Hayakawa, K. Takayama, "Shock wave propagation in model tissue for medical application of shock waves," International Symposium on Shock Waves **21,** Paper 5836, (1997).

2. K. Nagayama, Y. Mori, K. Shimada, M. Nakahara, "Shock hugoniot compression curve for water up to 1 GPa by using a compressed gas gun," J. of Appl. Phys. **91,** 476-482 (2002).

3. Kazuyoshi Takayama, "Applications of shock wave research to medicine," International Symposium on Shock Waves **22,** Paper 2010 (1999).

4. G. A. Lyzenga, Thomas J. Ahrens, W. J. Nellis, A. C. Mitchell, "The temperature of shock-compressed water," J. Chem. Phys. **76,** (1982).

5. N. C. Holmes, R. Chau, "Fast time-resolved spectroscopy in shock compressed matter," J. Chem. Phys. **119,** 3316-3319, (2003).

6. J. Karl and D. Hein, "Measuring water temperature profiles at stratified flow by means of linear raman spectroscopy," Proceedings 2nd Japanese German Symp. on Multi-Phase Flow, Tokyo University, Tokyo, Japan (1997).

7. Jianfeng Lou, Timothy M. Finegan, Paulash Mohsen, T. Alan Hatton, and Paul E. Laibinis "Fluorescence-based thermometry: principles and applications," Reviews In Analytical Chemistry **18,** 235-284 (1999).

8. Edward S. Fry, Jeffrey Katz, Dahe Liu, Thomas Walther, "Temperature dependence of the brillouin linewidth in water" J. of Mod. Opt, **49,** 411-418 (2002).

9. Eric B. Cummings, Hans G. Hornung, Michael S. Brown, and Peter A. DeBarber, "Measurement of gas-phase sound speed and thermal diffusivity over a broad pressure range using laser-induced thermal acoustics," Opt. Lett. **20,** 1577-1579 (1995).

10. Roger C. Hart, R. Jeffrey Balla, and G. C. Herring, "Optical measurement of the speed of sound in air over the temperature range 300-650 K," J. Acoust. Soc. Am. **108,** 1946-1948 (2000).

11. A. Stampanoni-Panariello, B. Hemmerling, and W. Hubschmid, "Temperature measurements in gases using laser induced electrostrictive gratings," Appl. Phys. B **67,** 125-130 (1998).

12. Michael S. Brown and William L. Roberts, "Single-point thermometry in high-pressure, sooting, premixed combustion environments," J. Propulsion Power **15 (No 1)**, 119-127 (1999).

13. Roger C. Hart, R. Jeffrey Balla, and G. C. Herring, "Nonresonant referenced laser-induced thermal acoustics thermometry in air," Appl. Opt. **38,** 577-584 (1999).

14. Y. Nagasaka, T. Hatakeyama, M. Okuda, and A. Nagashima, "Measurement of the thermal diffusivity of liquids by the forced Rayleigh scattering method: Theory and experiment," Rev. Sci. Instrum. **59,** 1156-1168 (1988).

15. Alexei A. Maznev, Daniel J. McAuliffe, Apostolos G. Doukas and Keith A. Nelson, "Wide-band acoustic spectroscopy of biological material based on a laser-induced grating technique," Ultrasound in Med. & Biol. **25,** 601-607 (1999).

16. H. J. Eichler, P. Günter, D. W. Pohl, "Production and Detection of Dynamic Gratings," in *Laser-Induced Dynamic Gratings*, (Springer-Verlag, Berlin, Heidelberg, 1986), pp. 13-37.

17. E. B. Cummings, I. A. Leyva, and H. G. Hornung, "Laser-induced thermal acoustics (LITA) signals from finite beams," Appl. Opt. **34,** 3290-3302 (1995).

18. Martín Chávez, Victor Sosa, and Ricardo Tsumura, "Speed of sound in saturated pure water," J. Acoust. Soc. Am. **77,** 420-423 (1985).

19. George M. Hale and Marvin R. Querry, "Optical constants of water in the 200-nm to 200-μm wavelength region," Appl. Opt. **12,** 555-563 (1973).

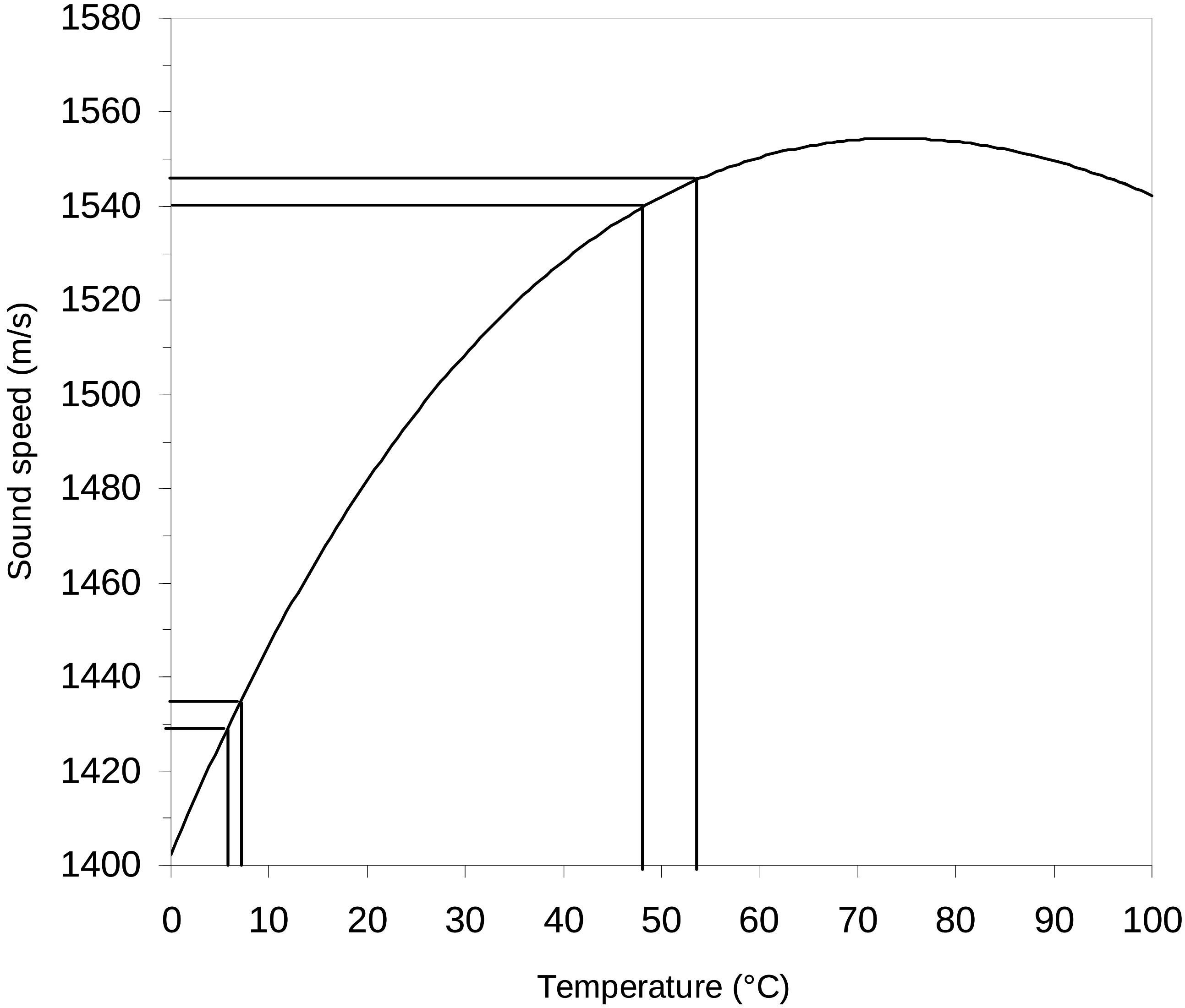

Figure 1. Visual demonstration of how temperature is inferred from LITA sound speed measurements, and how, for a constant sound speed uncertainty (constant distance between horizontal lines), temperature uncertainty increases with temperature (increasing distance between vertical lines with increase with temperature).

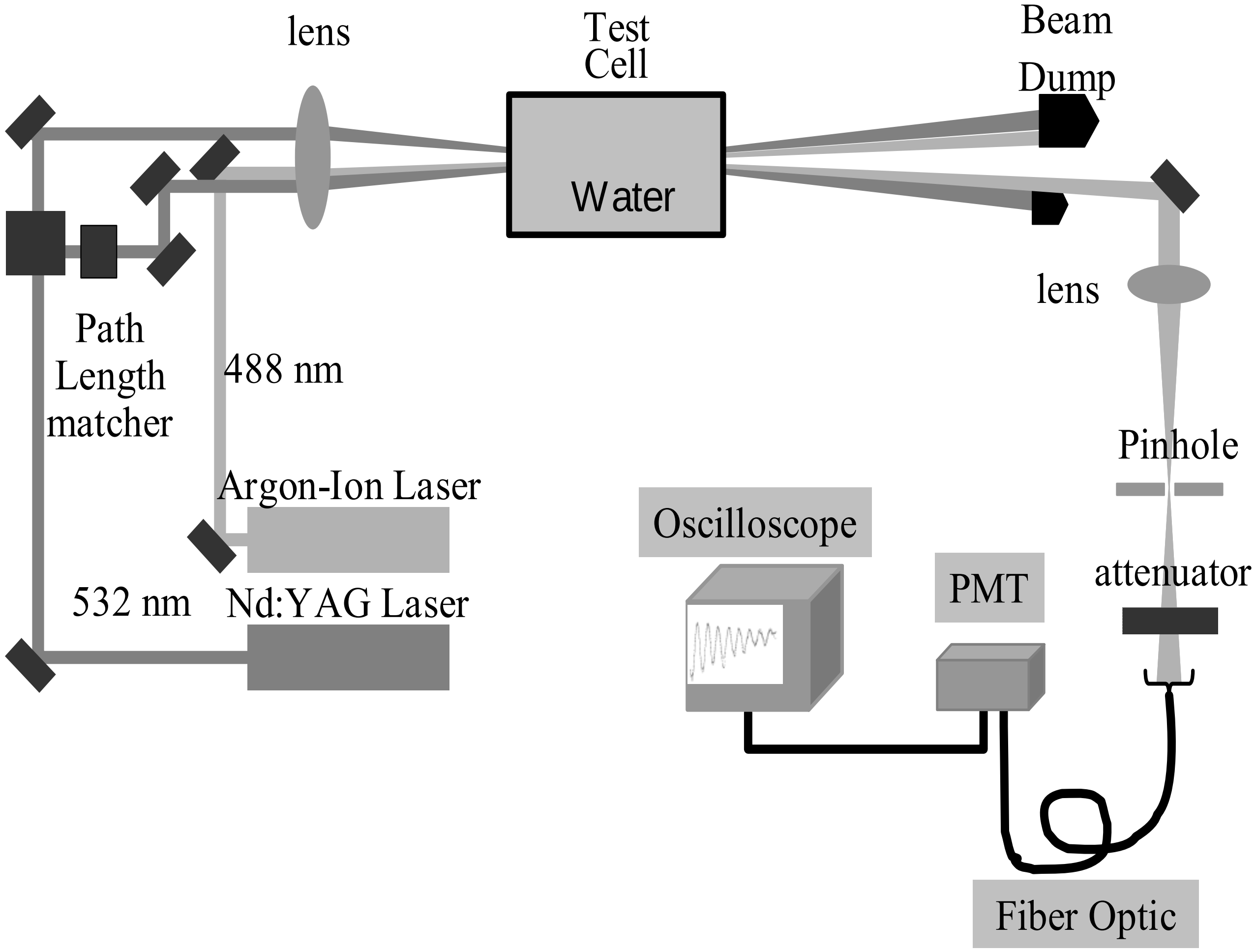

Figure 2. Diagram of the LITA experiment. Grating and probe generating optics are on the left side of the test cell and collection/reading optics are shown on the right side of the test cell.

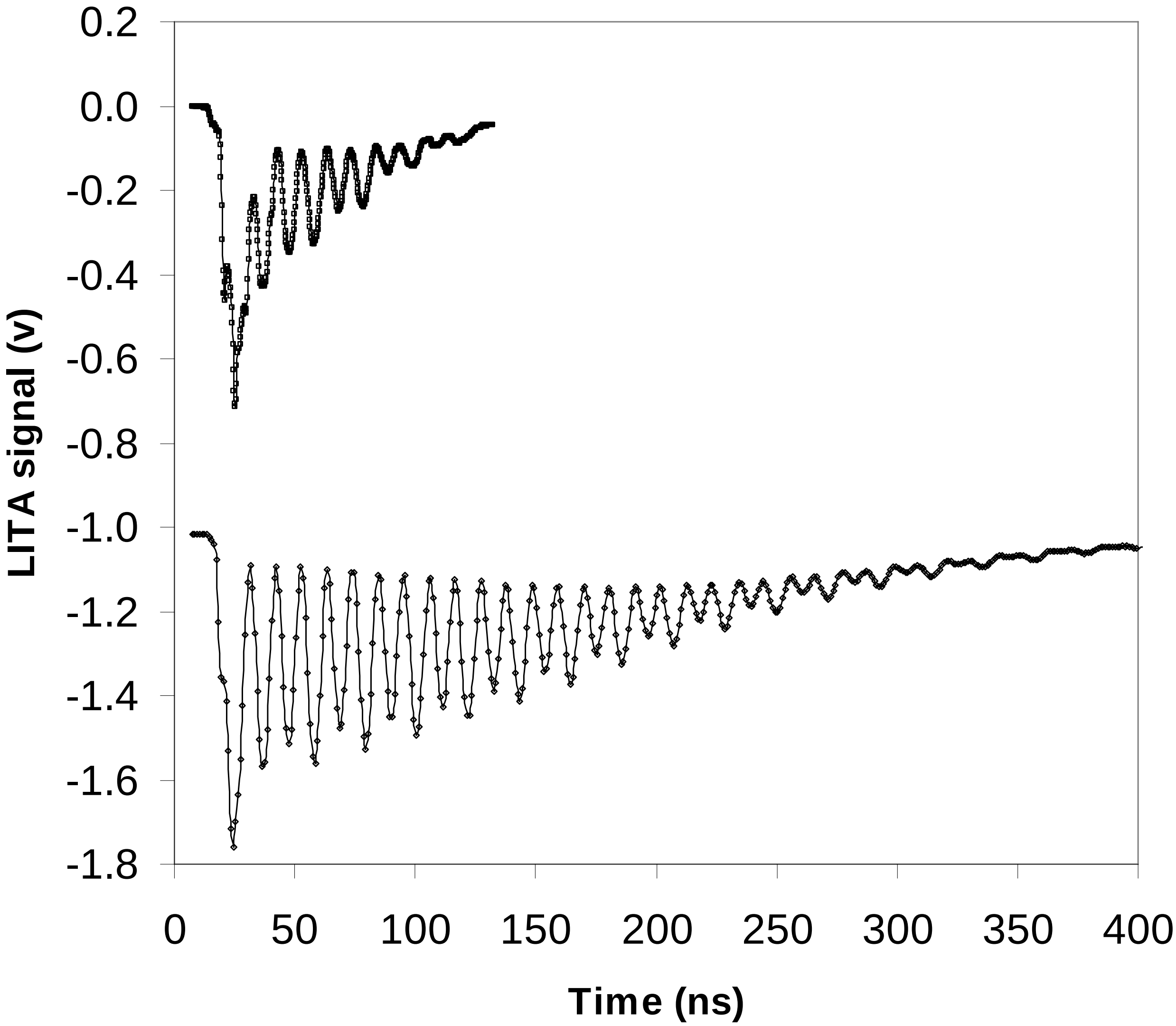

Figure 3. Comparison of the oscillation of the “signal” diffracted off a smaller grating (from a narrow pump beam ~30 μm) and a larger grating (from a wide pump beam ~340 μm), respectively top and bottom.

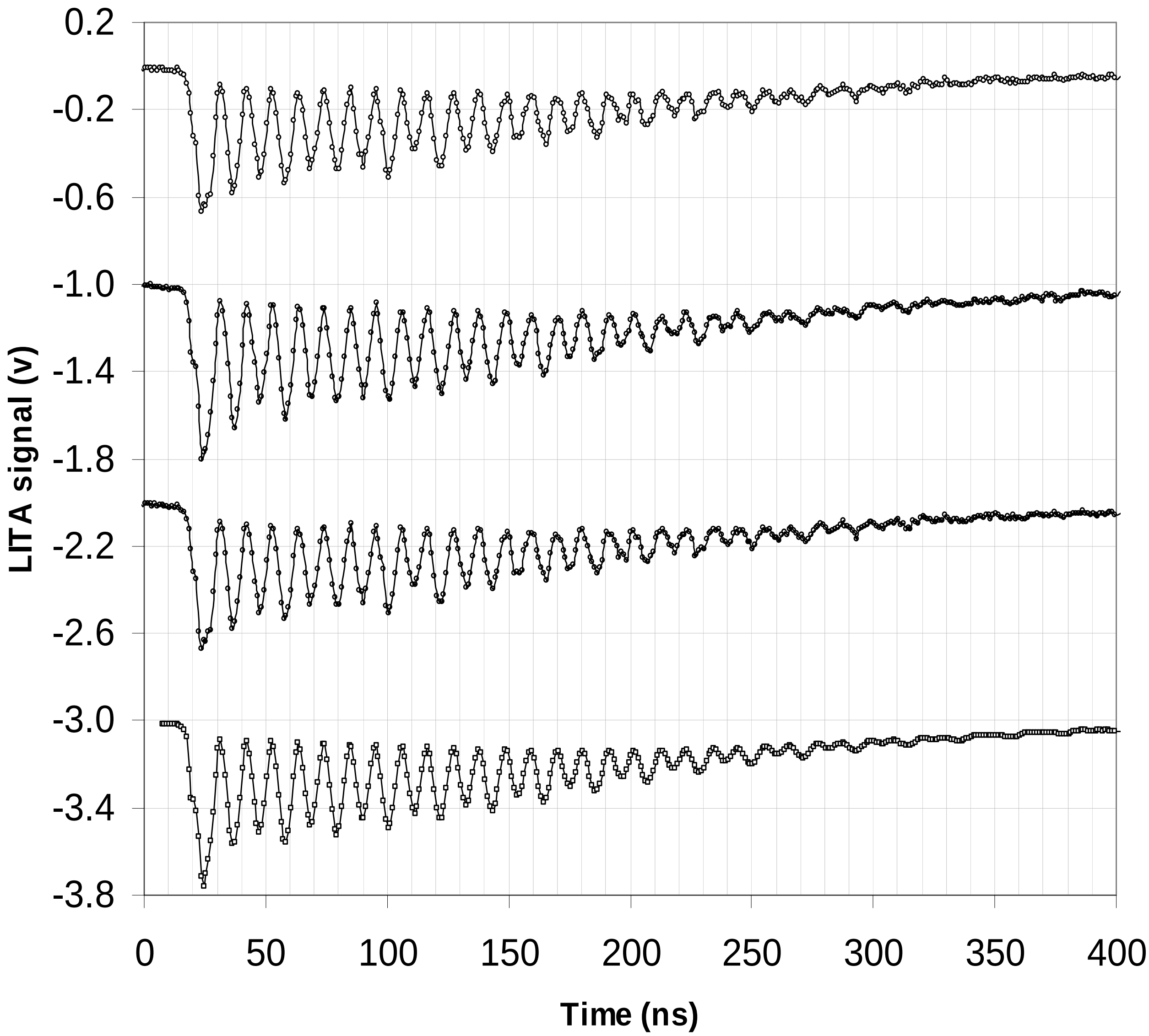

Figure 4. Offset for clarity, this figure compares 4 LITA sound speed signals. The top three are single shots and the bottom is a 100-shot average (10 sec), all at room temperature.

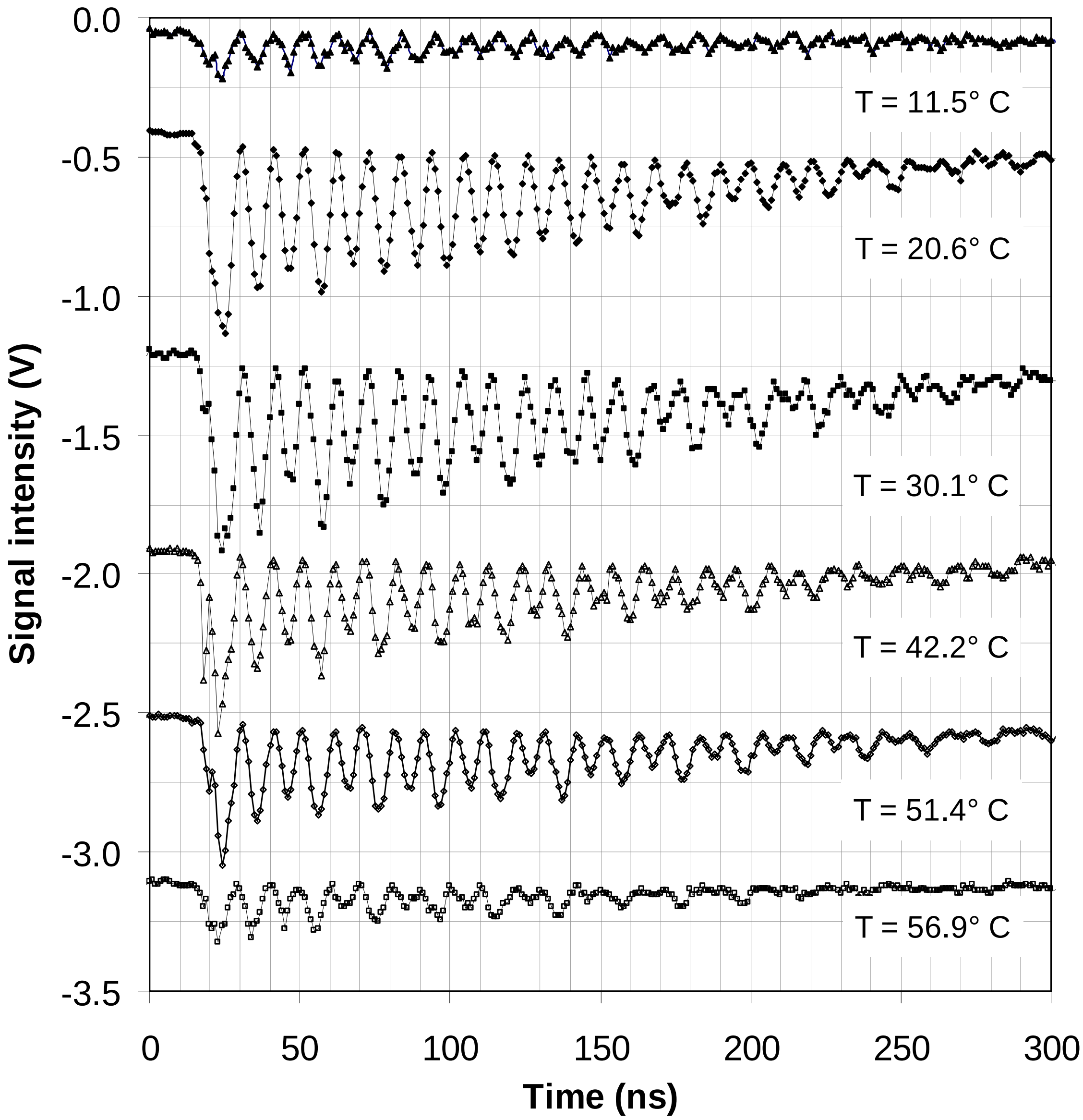

Figure 5. Comparison of the "signal" beam vs time over an approximate temperature span of about 10 – 60 °C. Data is offset for clarity.

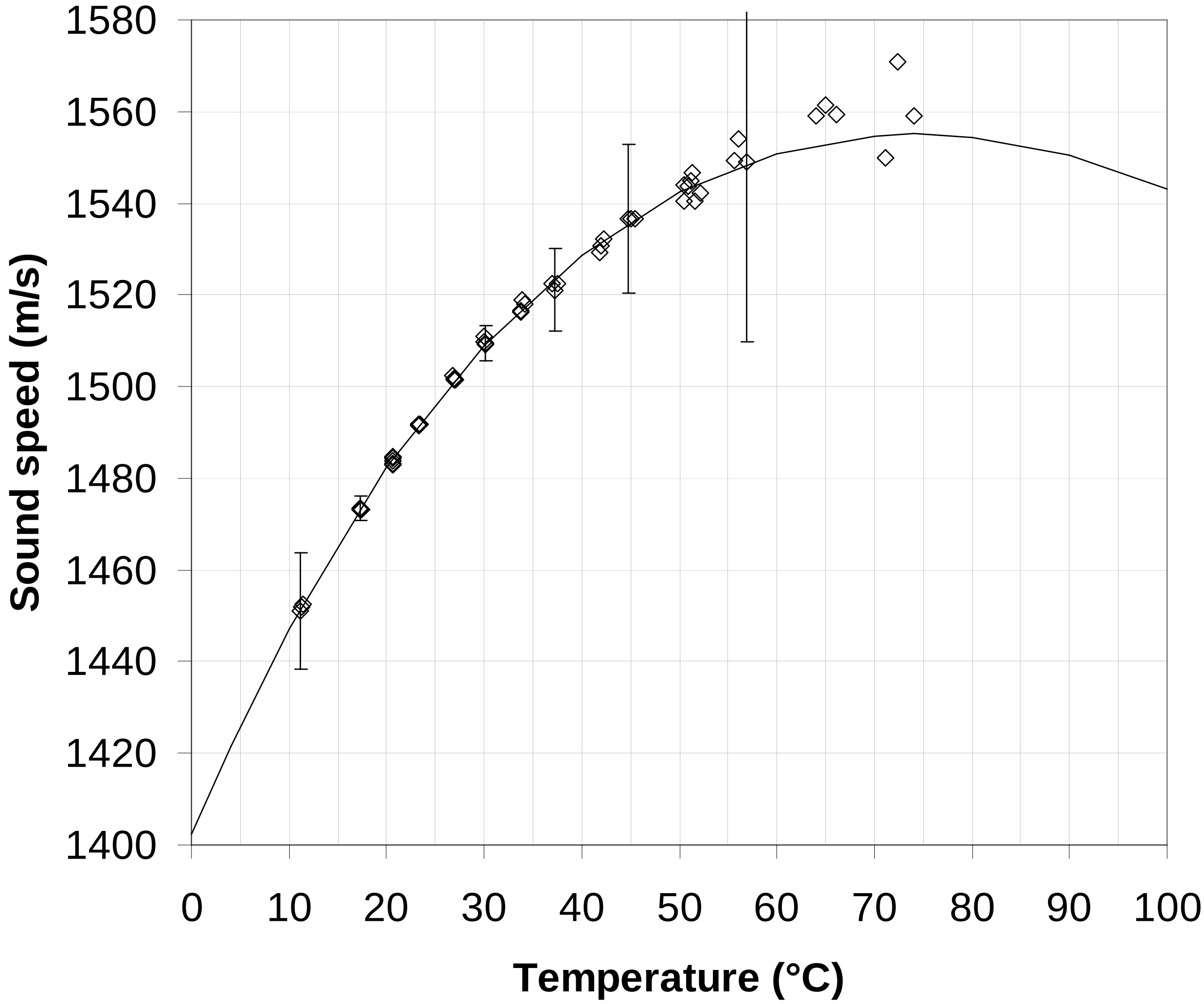

Figure 6. Graph of mean LITA measurements (diamonds) and calculated (line) sound speed vs. temperature. Sound speed of the 56th data point (about 73 °C) is outside the range of the graph.

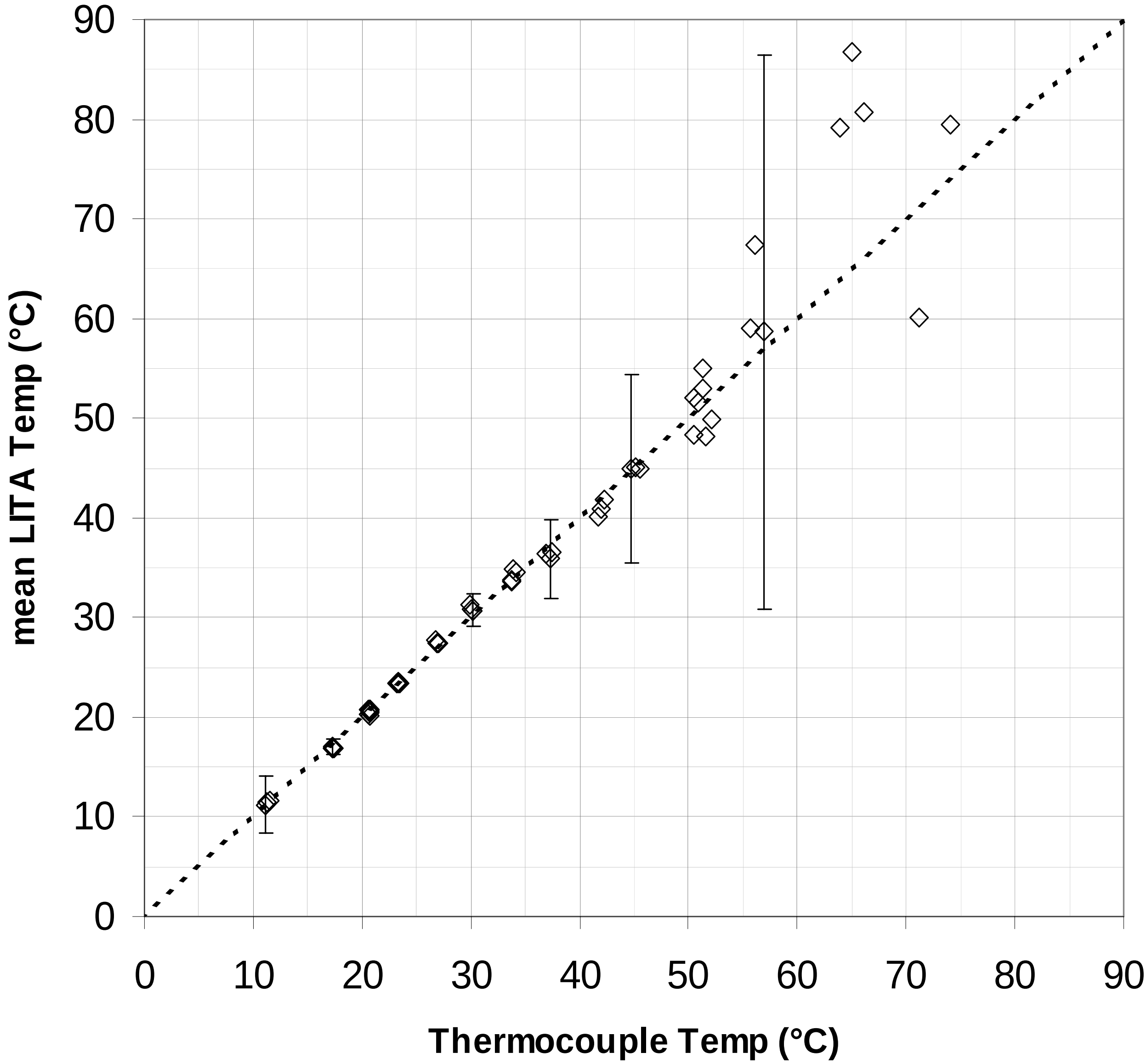

Figure 7. Inferred mean LITA temperature plotted against type T thermocouple temperature measurements. Dashed line represents perfect agreement. The 56th inferred mean LITA temperature point is outside the range of the graph and is about 125 °C.