Acoustic Damping Rate Measurements in Binary Mixtures of Atomic Species via Transient Grating Spectroscopy.

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ABSTRACT

We report on an investigation of the ability of Transient Grating Spectroscopy (TGS) to accurately measure the acoustic damping rate in a pressurized gaseous medium by analyzing the temporal behavior of laser-induced gratings. Experiments were performed in a mixture of helium and argon, with a small amount of NO_2 (100 ppm level) added to generate sufficient signal levels through thermalization. Measurements were made as a function of both composition and pressure. Acoustic damping rates were determined through model fits to the acquired signals. The results were compared with theoretical calculations using both classical acoustic damping rates and a more comprehensive model that includes the additional diffusive mechanisms associated with binary mixtures of gases with widely different molecular weights. For unmixed atomic species, the classical acoustic damping rate is known to accurately model the acoustic dissipation. However, in mixtures of atomic species, additional dissipation mechanisms appear which must be accounted for. Traditional acoustic damping rate measurements require the use of an ultrasonic acoustic source and a receiver which is located at various distances from the source. This traditional technique requires a homogeneous static mixture, as well as a physically large volume. The TGS technique demonstrated here provides a nearly instantaneous measurement with reasonably high spatial resolution (probe volume on the order of cubic centimeters), and can operate in harsh environments. Experimental data agrees reasonably well with theoretical predictions, with the primary source of error being the necessary addition of NO_2.

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I. INTRODUCTION

The Transient Grating Spectroscopy (TGS) or Laser-Induced Thermal Acoustics (LITA) signal is generated by the first-order Bragg scattering of a probe beam from a laser-induced dynamic grating. The temporal behavior of the generated signal provides information on the local speed of sound and transport properties, namely viscosity and thermal conductivity. Laser-induced grating techniques have been utilized to explore time-dependent phenomena in liquids and solids (initially) and (more recently) in gases (Eichler et al., 1986 and Govoni et al., 1993). Recent efforts include continuing development of the TGS technique for thermometry and velocimetry because of its relative simplicity and because it is a frequency, rather than an amplitude, based technique, making it attractive for application to practical hardware (Brown and Roberts, 1999). Furthermore, the amplitude of the signal scales quadratically with density, a real advantage in high-pressure combustion environments where many other laser-based diagnostics encounter various difficulties such as collisional quenching of the excited state.

The decay rate of the TGS signal magnitude is a function of both thermal diffusion and acoustic damping. Employing an unfocused beam setup, acoustic damping rates from various pure gaseous species were previously investigated (Li et al., 2002). The experimental results from these pure species were compared to the classical acoustic damping rate, which models the effect of both thermal and viscous dissipation. The classical acoustic damping rate theory accurately predicts the decay rates in pure atomic species. However, the measured acoustic damping rates for diatomic and polyatomic gases are always higher than the calculated classical acoustic damping rates. The reason being the classical theory does not account for the energy transfer from the translational mode to vibrational and/or rotational modes, and these additional energy transfer mechanisms lead to a faster decay rate. Analysis shows that the acoustic damping rate increases with increasing molecular structure, pressure and acoustic frequency.

The attenuation of acoustic waves in binary monatomic gas mixtures were successfully measured in the 1950s and 1960s (Holmes and Tempest, 1960; Law et al., 1966; and Prangsma et al., 1970). The experimental results agreed well with Kohler's theory of absorption (Kohler, 1941 and 1949), where the total absorption is due to viscous dissipation, heat conduction, mass diffusion, and thermal diffusion. In most of the conventional acoustic studies (Hersfeld and Litovitz, 1959) external sound sources are used to generate traveling sound waves that are detected by receivers after propagation over fixed distances. The absorption coefficient, a, is then determined by comparing the sound wave intensities at different locations along the propagation path. Accurate distance-based measurements sometimes require a propagation path on the order of one meter (Law et al., 1966) making them difficult in confined geometries. Due to the time consuming nature of this traditional measurement technique, it can only be applied to homogeneous static environments.

Here an investigation of acoustic damping rates of helium-argon mixtures in a pressurized environment (up to 7.4 atm) using TGS is reported. In executing the TGS technique, an instantaneous ultrasonic sound wave is generated by a pair of crossed laser pulses. The intensity of the dynamic sound wave decaying with time in the probe volume is "read out" by a cw laser beam. Analysis of the resulting signal beam reveals the sound wave attenuation. The non-intrusive TGS technique is able to measure the acoustic wave attenuation at a single point making it useful in environments with spatial gradients. The lifetime of the TGS signal is usually on the order of a microsecond or less; therefore, the technique yields useful measurements in turbulent environments. The TGS signal intensity grows quadratically with density making it very attractive in pressurized environments. Since the sound wave is generated by crossed laser pulses, the acoustic wavelength is determined by the laser's optical wavelength and the beam crossing angle. This typically produces acoustic wavelengths on the order of a few tens of microns restricting TGS measurements to the hypersonic regime. Because it is very difficult to generate detectable TGS signals in pure helium, a trace amount of NO_2 was added to help generate signals in the present study. Overall, the TGS measurement of acoustic damping rate in He-Ar mixtures fits the theory well, within the tested pressure range.

II. TRANSIENT GRATING SPECTROSCOPY

The principal of the TGS technique has been discussed elsewhere (Eichler et al., 1986; Brown and Roberts, 2002; Cummings, 1995; Paul, 1995). Here, only a brief description is given to illustrate the fundamentals of the technique. An optical intensity grating can be generated when two laser beams with parallel, linear polarization are spatially and temporally overlapped. The grating spacing, L, is given by:

$$\Lambda = \frac{l}{2\sin\left(\frac{q}{2}\right)} \cdot \qquad (1)$$

The wavelength of the grating-inducing beams is given by l while the full angle between the beams is denoted by q. Through various processes, e.g., electrostriction, thermalization, etc., the light intensity grating produces a modulation in the local density, $\Delta\rho$, and therefore in the local index-of-refraction, Δn. The hydrodynamic response of the gas to the local density disturbance is the production of two counter-propagating acoustic waves and a non-propagating, but diffusing, entropy or thermal wave. The alternating constructive and destructive interference of the two counter-propagating waves produces a standing sound wave. Therefore, the local index-of-refraction oscillates at a frequency (the Brillouin frequency) solely determined by the grating spacing and the local speed of sound. The pulsed pump beams, which form the grating (ideally delta functions in time) have a pulse width of approximately 10 ns in the experiments presented here. After the grating-inducing pulses have passed through the sample volume, the grating structures themselves will be washed out in time due to dissipation of both the acoustic and thermal waves. If a long-pulse or continuous-wave probe beam illuminates the grating under the Bragg scattering (coherent scattering) condition, the optical grating can be efficiently read out. The Bragg-scattered signal is detected with a photomultiplier tube and stored on a fast-sampling oscilloscope. The TGS signal appears as an exponentially damped oscillation. For excitation by an infinitely short laser pulse at $t = 0$, the signal will be given approximately at $t > 0$ as (Brown and Roberts, 1999):

$$I_{signal}(t) \propto \left\{A\exp\left(-D_{th}q^2t\right) + B\exp\left(-\Gamma q^2t\right)\cos\left(mc_sqt + f\right)\right\}^2 \qquad (2)$$

In the above, D_{th} denotes the thermal diffusivity, G denotes the acoustic damping coefficient, c_s denotes the speed of sound, and $m = 1$ or 2 for thermal or electrostrictive signals respectively. The grating wave vector q is defined as $q = 2p/L$. The coefficients A, B and phase angle j are simply used as fitting parameters. The ratio of A to B determines the relative amounts of electrostriction and thermalization present in the calculated signal. The parameter f is useful in fitting noisy data in which there is ambiguity in setting the zero point on the timeline. It should be noted that Eqn. (2) presumes an infinitely fast thermalization rate, a pump laser with an infinitely narrow pulse width as well as pump and probe beams with plane wave fronts of infinite extent. In Eqn. (2), the exponential factor of the second term represents the absorption of the acoustic wave intensity.

The frequency of the standing acoustic wave is solely determined by the ratio of the local sound speed and the grating spacing, c_s/L. The frequency of the TGS signal generated through electrostriction is equal to twice this ratio. During electrostriction, the local fluid experiences compression and rarefaction during every grating period and thus the density variation, Dr, as well as the index-of-refraction, Dn, changes its sign during every period. As a consequence, the reflectance of the fluid has two peaks within every period. If the grating is generated by thermalization, the fluid only experiences rarefaction. The reflectance reaches its maximum when Dr reaches its minimum value. Therefore, the frequency of the TGS signal generated through thermalization is the same as that of the standing acoustic wave frequency.

III. ACOUSTIC DAMPING RATE *G*

The attenuation of acoustic waves can be caused by various kinetic processes in the fluid which may include the transport of heat and momentum, vibrational and rotational relaxation, and chemical or structural changes on a molecular scale (Trusler, 1991). For single-atom species, the acoustic wave attenuation is caused by friction and heat conduction which can be described by the classical acoustic damping rate. For multi-atom species with internal structures, translational energy is transferred to vibrational/rotational energy along with the redistribution of translational momentum. Hence, the acoustic waves attenuate faster than predicted by the classical acoustic damping rate. This phenomena has been observed in earlier TGS experimental investigations (Li et al., 2002). For mixtures of atomic species, the density and temperature gradients caused by the sound wave will also generate species concentration gradients that will be damped by diffusion. Therefore, the effect of mass diffusion should be included in the description of acoustic wave absorption.

In previous traditional studies of acoustic wave attenuation in binary monatomic mixtures, the measured absorption coefficient, a, was defined by the decay of the sound wave amplitude with distance as $\exp(-az)$. This definition of absorption is well-suited for explaining the results of measurements of acoustic amplitude at various

distances from a transmitter. In TGS measurements, the acoustic damping rate, G, gives the sound wave absorption in time, as defined in Eqn. (2). The absorption coefficient, a, and acoustic damping rate, G, are related in a simple way:

$$\Gamma = \frac{a\, c_s}{q^2} \tag{3}$$

In the classical view, the energy of the sound wave is dissipated due to both friction and heat conduction; therefore, the damping rate of the amplitude of an acoustic wave is a function of the transport properties of viscosity and thermal conductivity. The classical acoustic damping coefficient in a gas can be expressed as (Trusler, 1991):

$$\Gamma_c = \frac{1}{2r_0}\left[\frac{4}{3}m + (g-1)\frac{k}{c_p}\right], \tag{4}$$

where m is the dynamic viscosity (with both sheer and bulk contributions), k the thermal conductivity, c_p the heat capacity at constant pressure, g is the ratio of specific heats and r_0 is the gas density.

For a mixture of two monatomic gases, the additional acoustic wave absorption arising from diffusions between two species can be evaluated as (Trusler, 1991):

$$\Gamma_d = \frac{1}{2}gx_1x_2D\left[\frac{M_2 - M_1}{M} + \frac{g-1}{gx_1x_2}K_T\right]^2, \tag{5}$$

where M_1 and M_2 are the molecular weights of the two component gases, x_1 and x_2 are their mole fractions in the mixture, $M=x_1M_1+x_2M_2$, D is the binary mass diffusion coefficient and K_T is the thermal diffusion ratio, which in tern is a function of temperature, concentration, and molecular weights. For a single atomic species, $M_1 = M_2$, the thermal diffusion rate is zero and Eqn. 5 is identically zero; thus only viscous dissipation and heat conduction contribute to acoustic wave attenuation.

The total acoustic damping rate in a He-Ar mixture will be the sum of the classical acoustic damping rate, G_c, and the diffusive acoustic damping rate G_d. To estimate the acoustic damping rate, the parameters in Eqns. (4) and (5), namely the density r_0, dynamic viscosity m, heat capacity ratio g, thermal conductivity k, heat capacity at constant pressure c_p, binary mass diffusion coefficient D and thermal diffusivity K_T all need to be evaluated for a binary mixture. For atomic gases, $\gamma = 1.667$ at room temperature. The density r and heat capacity at constant pressure c_p were computed through simple mole fraction relations:

$$r = xr_{Ar} + (1-x)r_{He}, \text{ and} \tag{6}$$

$$c_p = xc_{p,Ar} + (1-x)c_{p,He}, \tag{7}$$

where x is the mole fraction of Ar and implicitly, $(1-x)$ is the mole fraction of He. The mixture transport coefficients m, k, D and K_T can be calculated from kinetic theory, using the Lennard-Jones potential as a molecular model (Hirschfelder et al., 1964). The force constants of the mixture for this potential were obtained from the force constants of the pure species by:

$$r = \frac{r_{He} + r_{Ar}}{2} \text{ and } \frac{e}{k} = \sqrt{\frac{e_1}{k}\frac{e_2}{k}}, \tag{8}$$

where r is the collision diameter and e/k is the potential parameter.

IV. EXPERIMENTS

The optical setup for the TGS experiments presented here is shown schematically in Fig. 1. The frequency-doubled output of a Q-switched Nd:YAG laser was used to generate the gratings. This short-pulse beam (~10 ns) was divided into two pump beams of equal energy using a 50/50 beam splitter. For the experiments reported here, the pump energy was varied from 10 to 150 mJ per pulse for both beams. The 514 nm output (~1W) of an Ar ion laser was used as the probe beam to read out the gratings. To trace out the path of the generated TGS signal beam, ~4% of the probe beam was split off and steered through the probe volume at the Bragg angle. This false-signal beam was used during the alignment of the detector and eliminated during actual experiments. The three (or four when including the false signal) laser beams were directed into the sample volume inside the high-pressure gas cell with a three-dimensional phase matching arrangement. The signal beam was directed by a mirror through spatial and spectral filters and then onto a photomultiplier tube (PMT). The PMT was connected to a digital oscilloscope to acquire the TGS signals. The signals were typically acquired and saved into a computer for off-line analysis. The high-pressure gas cell, 12.7 cm in diameter, was built to operate at pressures up to 35 atm. To provide optical access, two 25.4 mm thick BK-7 windows are mounted along the optical path. Measurements were made at room temperature over a range of pressures extending from 1 to 7.4 atm.

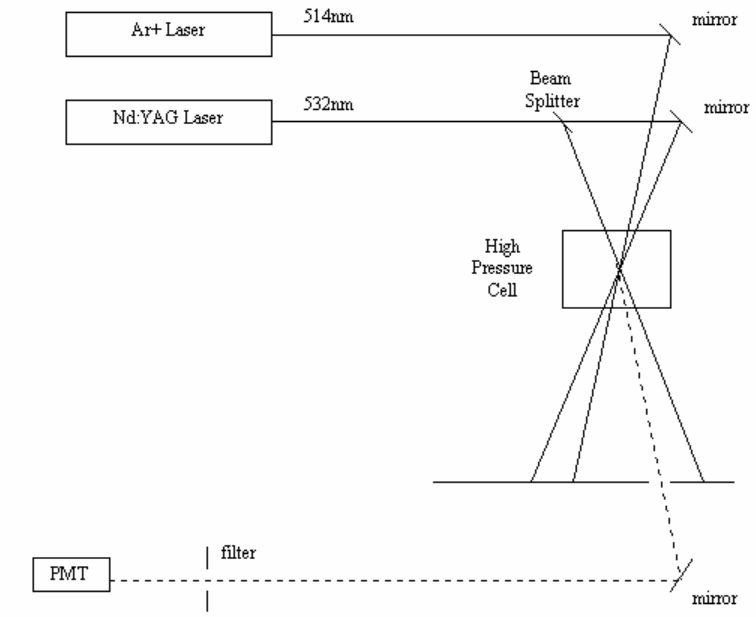

Figure 1 Schematic of the TGS optical setup. The beam-crossing angle is typically on the order of a few degrees and is greatly exaggerated in this schematic. During alignment, 4% of the probe beam was picked off and used to trace the path of the signal beam.

When the probe volume diameter is small (as with focused pump beams), the effect of the sound waves propagating out of the probe volume provides a significant contribution to the apparent grating decay rate. Therefore, it is more difficult to accurately extract the acoustic damping rate from TGS signals generated using focused laser beams where the probe volume is typically a few hundred microns in diameter at the waist and the TGS signals decays to zero after a few hundred nanoseconds. For measurements in reacting flows, where high temperatures result in low density and hence low signal levels, focused beams are necessary for detectable signal generation. In the present TGS experiments unfocused laser beams were used to investigate the acoustic damping rates. The diameter of the probe volume is ~8 mm, resulting in TGS signals which persist for up to a few microseconds. Because these measurements were made at room temperature, the pump intensity provided by unfocused beams was sufficient to generate high signal strengths (recall that signal strength scales quadratically with density). When high spatial resolution is required, such as in inhomogeneous environments, focused pump beams should be used and attention paid to the effect of sound wave propagation out of the probe volume. A comparison

between sample TGS signals generated by focused and unfocused pump beams under similar experimental conditions is shown in Fig. 2.

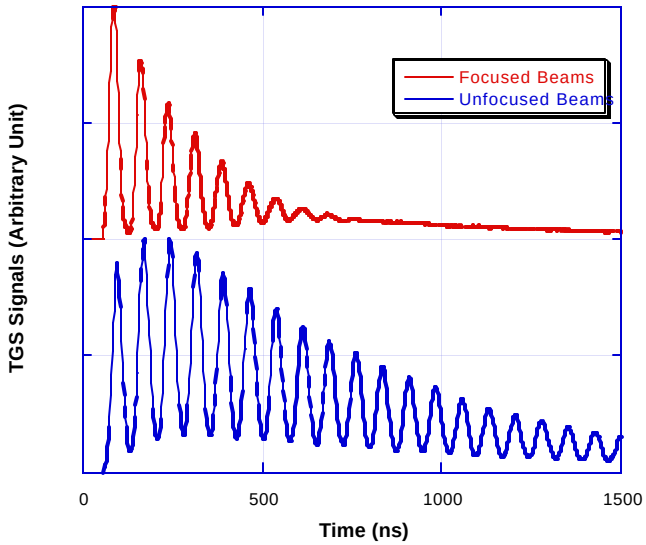

Figure 2 TGS signals from pure argon at 1.7 atm through thermalization. Probe volume diameter of 200 μm for the focused beam geometry and 8 mm for the unfocused beam geometry. Pump energy = 13 mJ/pulse, probe power = 80 mW, grating spacing Λ = 22.4 μm, and 64 shot average.

During the experiments, Ar and He were introduced into the high-pressure chamber separately. The TGS signals were acquired when the macroscopic flowfield motions were completely dissipated and the gases well mixed. The generation of an electrostrictive grating in He is very difficult, especially with unfocused pump beams due to He's very low electrostrictive coefficient. To generate a detectable signal, a small amount of NO_2 (less than 100 ppm) was seeded into the gas samples to generate resonant TGS signals through the thermalization process. The mole fractions of Ar and He in the mixtures were calculated by analyzing the oscillation frequencies of the TGS signals. From an analysis presented elsewhere (Brown and Roberts, 1999), there exists a relationship between temperature and frequency of TGS signal generated through the thermalization process:

$$T = f^2 \Lambda^2 \left(\frac{M}{g} \right) \frac{1}{R} \tag{9}$$

In a binary mixture of Ar, with mole fraction x and He, with mole fraction (1-x), the molecular weight of the mixture is:

$$M = xM_{Ar} + (1 - x)M_{He} \tag{10}$$

where M_{Ar} and M_{He} are the molecular weights of Ar and He, respectively. Combining Eqns. (9) and (10) the mole fraction of Ar in an He-Ar mixture can be accurately calculated by measuring the TGS signal frequency:

$$x = \frac{gRT - M_{He}\Lambda^2 f^2}{(M_{Ar} - M_{He})\Lambda^2 f^2} \tag{11}$$

In Eqn. (10), the temperature T is known (room temperature), and the grating spacing L can be determined from calibration using TGS signals from a single species (Ar in this case). Therefore, the mole fractions of Ar and He can be easily evaluated by analyzing the oscillation frequency f of the acquired TGS signals.

V. RESULTS AND DISCUSSIONS

The acoustic damping rates of He-Ar mixtures under various pressures and mole fractions were investigated. All signals analyzed in the present work are due to the resonance response of the seeded NO_2. The acoustic wave frequency ranges from ~14 MHz in pure argon to ~45 MHz in pure helium at the grating spacing of 22.4 μm. In this frequency-pressure region, dispersion of the acoustic absorption rate is negligible (Prangsma et al., 1970). The acoustic damping rate is extracted by curve-fitting the TGS signal with Eqn. (2). Commercial software (KaleidaGraph®) which employs the Levenberg-Marquardt algorithm was utilized in the nonlinear least squares fit of the TGS signals. A sample of a TGS signal and curve-fitting result is shown in Fig. 3. The curve fitting error is usually less than 5%, as described elsewhere (Li et al., 2002).

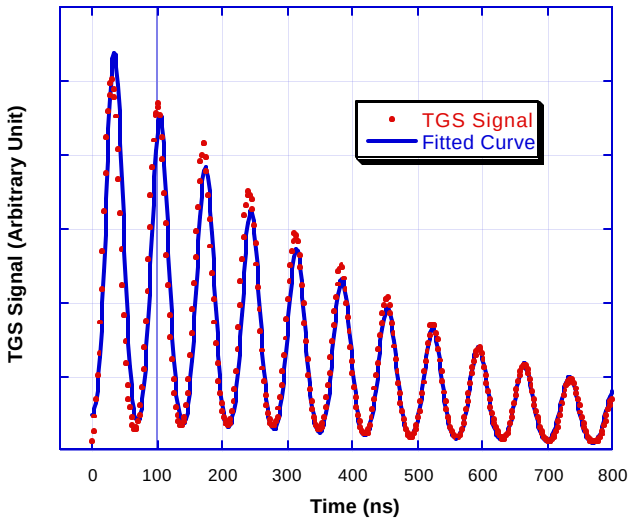

Figure 3 TGS signal in pure argon with curve fitting. Pressure = 1.0 atm, pump energy = 105 mJ/pulse, probe power = 0.6 W, grating spacing Λ = 22.4 μm, and 64 shot average. Acoustic damping rate derived from curve fitting is 1.68×10^{-5} m^2/s, while theoretical calculations yields 1.44×10^{-5} m^2/s.

Since G_c and the mass diffusion coefficient D in Eqn (5) are both proportional to the inverse of the pressure while other transport properties like viscosity m, thermal conductivity k, and thermal diffusivity K_T are not significant functions of pressure (Hirschfelder et al., 1964), the total acoustic damping rate, G, will scale with the inverse of pressure. Therefore, the reduced acoustic damping rate, Gp, is independent of pressure. Figure 4 shows the experimental and theoretical values of the reduced acoustic damping rate from He-Ar mixtures at 1 atm, 2.4 atm, 4.1 atm and 7.4 atm. Overall, the experimental results fit theory well and show the trend of Gp changing with species concentrations.

The addition of a very small amount of impurity gas into a pure sample or gas mixture will alter the sound speed and other intrinsic properties such as the compressibility to a degree commensurate with the mole fraction of the impurity. However, the collisional relaxation time of the total mixture may be altered in a disproportionate way with respect to the mole fraction of the impurity (Zuckerwar, 2002). This is particularly true when a trace amount of a molecular gas is added to an atomic gas. The ro-vibrational states of the molecular species are coupled to the translational states through collisions and provide another means for damping out the density perturbations. The disproportionate influence on relaxation is dependent on the host gas and is strongest for He (Zuckerwar, 2002).

Due to relaxation, the acoustic damping coefficient is modified by a term that scales as the inverse of the product of c_s, q, and τ_a where τ_a denotes the collisional relaxation time of the molecular species (Zuckerwar, 2002). This additional term is often folded into the bulk viscosity for computational convenience (Berne and Pecora, 1990). Acoustic attenuation is increased in the atomic gas with the addition of the trace molecular species. In the experiments described here, gases were introduced into the test cell sequentially and partial pressures were used in part to assess mixture fractions. The TGS sound speed measurement was also used to accurately assess the relative amounts of Ar and He. While the mole fraction of NO_2 was held below 100 ppm, the exact value for each measurement shown in Fig. 4 is unknown. In affect, there is some uncertainty in the calculated bulk viscosity and consequently the acoustic damping coefficient. This uncertainty is reflected in the difference between the calculated and measured damping coefficient at low Ar concentrations. The small amount of scatter in the measurements for all Ar concentrations is in part due to relative differences in the amount of NO_2 present.

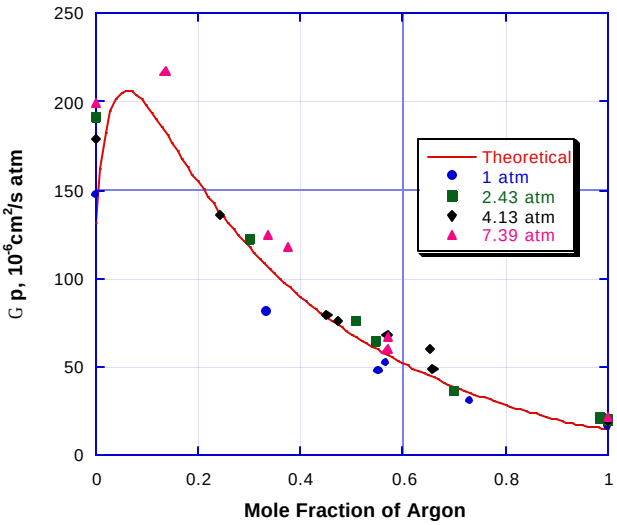

Figure 4 Comparison with theory of experimental results for acoustic damping rate in He-Ar mixtures at 1 atm, 2.43 atm, 4.13 atm and 7.39 atm. Grating spacing Λ = 22.4 μm.

VI. SUMMARY

In summary, we have shown that Transient Grating Spectroscopy can be used to measure acoustic damping coefficients in gas mixtures without the need for extended propagation distances. TGS measurements can be made in small volumes and harsh environments that are not amenable to acoustic transducer/receiver setups. TGS has been shown to be capable of single-shot measurements, rendering the technique nearly instantaneous (on the order of a few hundred ns) compared to most other time scales. The ability to measure acoustic damping rates in inhomogeneous and unsteady flows make TGS an attractive alternative to traditional measurement techniques. The study indicates the role of mass diffusion in determining acoustic damping when the local density field in a fluid is optically perturbed. Consistent with previous work, it also indicates the strong influence of a trace molecular species on the acoustic damping in an atomic host.

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