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Time-resolved, dual heterodyne phase collection transient grating spectroscopy

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The application of optical heterodyne detection for transient grating spectroscopy (TGS) using a fixed, binary phase mask often relies on taking the difference between signals captured at multiple heterodyne phases. To date, this has been accomplished by manually controlling the heterodyne phase between measurements with an optical flat. In this letter, an optical configuration is presented which allows for collection of TGS measurements at two heterodyne phases concurrently through the use of two independently phase controlled interrogation paths. This arrangement allows for complete, heterodyne amplified TGS measurements to be made in a manner not constrained by a mechanical actuation time. Measurements are instead constrained only by the desired signal-to-noise ratio. A temporal resolution of between 1 and 10 s, demonstrated here on single crystal metallic samples, will allow TGS experiments to be used as an in-situ, time-resolved monitoring technique for many material processing applications. © 2017 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (http://creativecommons.org/licenses/by/4.0/). [http://dx.doi.org/10.1063/1.4983716]

For almost two decades, transient grating spectroscopy (TGS), also known as impulse stimulated thermal scattering (ISTS), has been implemented using an optical heterodyne detection technique in an arrangement described in Ref. 1. This methodology has been used to study the acoustic and thermal transport properties of a wide variety of systems including thin-film semiconductors, microsphere lattices, and thermal transport properties in high-Tc metals. Recently, TGS has been shown to be a useful tool in methods have been used for grain mapping in polycrystalline films, the electronic properties of 2D semiconductors, and also been applied to study the magnetic properties of thin films, the electronic properties of 2D semiconductors, and transport properties in high-Tc superconductors. Similar methods have been used for grain mapping in polycrystalline metals. Recently, TGS has been shown to be a useful tool in evaluating the effects of irradiation in metallic systems.

In this technique, a spatially periodic material excitation is generated by periodic thermal excitation, itself produced by the interference pattern of two crossed pumping laser beams at the material surface. In reflective materials, this excitation manifests itself as surface displacement and/or transient reflectivity change. To interrogate the dynamics of this excitation, a third, probing laser beam is diffracted from the excitation. The first order diffraction of this probe is directed into a photodetector such that the amplitude of the response oscillates with the frequency of the material excitation. In TGS experiments to date, a fourth laser beam, generated from the same source as the probe, is used as a reference oscillator, allowing the diffracted probe signal to be heterodyne amplified. In this configuration, the measured signal intensity has the form

\[ I_s(t) = I_R(t) + I_D(t) + 2\sqrt{I_R I_D(t)} \cos \phi, \]  

where \( I_R \) is the reference oscillator intensity, \( I_D(t) \) is the diffracted signal intensity, and \( \phi \) is the heterodyne phase difference between the reference and diffracted beams. As \( I_R \) is constant and usually of much greater amplitude than \( I_D(t) \), the result is an overall amplification of the time-dependent signal of interest.

To recover large amplitude signals suitable for the quantitative analysis of the acoustic and thermal transport properties of materials, it is a common practice to record traces at both \( \phi = 0^\circ \) and \( \phi = 180^\circ \). These two signals can be subtracted from one another to recover the total signal amplitude

\[ I_s(t) = I_R(t) - I_D(\phi = 180^\circ) = 4\sqrt{I_R I_D(t)}. \]  

In addition to further amplifying the recorded signal intensity, taking a set of measurements in this manner allows for the removal of any systematic noise from signals used for quantitative analysis. Such noise could include the impulse response of the photodetector or other electrical noise present near the experiment. In practice, the heterodyne phase is controlled by a manually adjustable, highly parallel optical flat in the path of the probe laser beam. A phase difference between probing and reference beams is generated as a function of the tilt angle of this flat due to small changes in the path length. Measurements relying on this type of manual phase control are time-limited by the actuation time between collections at different values of \( \phi \), typically in the range of tens of seconds to single minutes. This limitation must be lifted if material systems undergoing dynamic changes are to be studied using TGS. Such systems of interest may include irradiation-induced material evolution, the thermal transport properties of surfaces as oxide layer growth occurs, and the kinetics of low-temperature phase change materials.

In this letter, a modification to the optical arrangement for heterodyne amplified TGS experiments is presented which allows for the simultaneous collection of two signals with...
differing heterodyne phases, hereafter referred to as dual heterodyne phase collection TGS (DH-TGS). For materials that exhibit strong excitations in both displacement and reflectivity under an imposed transient grating, different dynamics can be probed by choosing the values of $\phi$ other than $\phi = 0^\circ$ and $\phi = 180^\circ$. Of these two excitations, it is the displacement dynamics that contains information about both acoustic and thermal properties, while the reflectivity dynamics contain information about thermal transport properties alone. However, the surface displacement response cannot be interrogated independently, causing the thermal transport dynamics of measurements sensitive to acoustic propagation to be a combination of both displacement and reflectivity responses. In light of this complexity, the performance of the DH-TGS methodology is characterized in terms of the measured acoustic response alone. For the case considered here, fixing $\phi$ to $0^\circ$ and $180^\circ$ allows for the collection of a complete TGS measurement of the excitation of interest without the constraint of mechanical actuation.

The optical arrangement for a reflective sample is shown in Fig. 1(a). The generation of the excitation grating on the sample under investigation is accomplished by passing a short pulsed laser through a volumetric diffraction grating, hereafter referred to as the “phase mask,” which is optimized to diffract most of the light into the first order. The generated grating is focused coincident with the pump source onto the phase mask at an angle $\theta_1$ (out of the plane of the page in Fig. 1(a)) such that between $L_1$ and $L_2$, there is a vertical separation of the pump and first probing paths. The reflected probing beam from the beam splitter is re-directed so it, too, is coincident onto the phase mask as the same spot as the pump but with incoming angle $\theta_2 > \theta_1$ such that the vertical separation between the second probe paths and the pump is greater than that of the first pair. For both probes, the $\pm 1$ diffraction orders are again retained, while the transmitted beam and higher orders are blocked. The vertical placement of each of the six beam paths in the plane of $L_2$ is shown in Fig. 1(b).

Between $L_1$ and $L_2$, the two reference oscillator beams are passed through a neutral density filter (ND) to avoid saturating the photodetectors downstream. Both probing paths are passed through a first phase adjust flat (PA1), which is used to uniquely control the heterodyne phase of $I_2^{180}(t)$. The extremal probing path is passed through a vertically separated, second phase adjust flat (PA2), which adds an additional path length difference and uniquely controls the phase of $I_2^{0}(t)$. Alternative geometries of the phase adjustment optics with the same independent control are possible. However, the configuration described here allows PA2 to be set and fixed to $180^\circ$ offset from PA1, necessitating only the adjustment of PA1 when tuning the response of a new sample. Upon reflection and diffraction from the sample surface, to record two heterodyne phases concurrently, two additional laser paths are added to the standard TGS implementation as a probing beam and reference oscillator pair. All of the probing beams are generated from the same continuous wave laser source. Prior to incidence onto the phase mask, the probe path is split using a non-polarizing 50:50 beam splitter (BS). The transmitted leg of this probe is focused coincident with the pump source onto the phase mask at an angle $\theta_1$ (out of the plane of the page in Fig. 1(a)) such that between $L_1$ and $L_2$, there is a vertical separation of the pump and first probing paths. The reflected probing beam from the beam splitter is re-directed so it, too, is coincident onto the phase mask as the same spot as the pump but with incoming angle $\theta_2 > \theta_1$ such that the vertical separation between the second probe paths and the pump is greater than that of the first pair. For both probes, the $\pm 1$ diffraction orders are again retained, while the transmitted beam and higher orders are blocked. The vertical placement of each of the six beam paths in the plane of $L_2$ is shown in Fig. 1(b).

FIG. 1. (a) Top-view block schematic of the DH-TGS optical geometry showing paths from laser sources to detectors. BS – beam splitter, L1 and L2 – achromatic doublets, ND – neutral density filter, PA1 and PA2 – phase adjustment flats, and APD1 and APD2 – silicon avalanche photodiodes. (b) Beam arrangement in the plane of lens L2 just before the sample surface. The positions below the dotted line represent the reflection and diffraction of the reference oscillator and probe beams from the sample surface.
the spatial coincidence of the reference oscillator and the diffracted signal for both probing paths is guaranteed by the optical geometry. In the reflection geometry, both signals pass back through L2. As these paths are now vertically separated below the plane of the pump paths, they can be picked off, separated, and directed into two fast silicon avalanche photodiodes (APD1 and APD2). The path lengths between the sample surface and APD1 and APD2 are managed such that the two signals are aligned in time when recorded on a digital oscilloscope.

The primary advantage of the optical arrangement described above over previous implementations of the technique is time resolution. The pumping laser in these types of experiments is commonly operated with a repetition rate in the single kHz range. Each measurement, therefore, is taken as an average over hundreds to thousands of laser pulses to reduce the system noise. Previously, a complete measurement of both heterodyne phases could be time resolution limited either by the signal collection time (in the case where many averages are taken) or by the mechanical action necessary to change the heterodyne phase (in the case where only a few averages are needed). By recording both these phases independently, the time resolution of the system becomes, in all cases, signal-to-noise ratio limited.

DH-TGS experiments using an optical arrangement as described above are carried out using a passively Q-switched, 532 nm, solid-state laser with a pulse length of 300 ps, a repetition rate of 1 kHz as a pump laser source with a spot size of 210 μm, and pulse energy at the sample surface of 2.38 μJ. The probe laser source is a 785 nm, CW diode laser modulated to a repetition rate of 1 kHz with a 20% duty cycle, a power at the sample surface of 4.7 mW for both probe/reference oscillator pairs, and a spot size of 175 μm. The probe laser is RF modulated to the frequency of the pump to reduce sample heating. The silicon avalanche photodetectors used have a 3 dB bandwidth of 1.0 GHz and manual external gain adjustment. Their outputs are concurrently recorded on a dual-band 5 GHz bandwidth digital oscilloscope.

In metallic samples investigated using this technique, $I_S(t)$ has two features of interest, a damped acoustic oscillation overlaid on a non-exponential thermal decay curve. A filtered power spectrum of $I_S^\text{tot}(t)$ can be used to identify the dominant frequencies of the surface acoustic wave (SAW) oscillations induced by the imposed transient grating. These power spectra can be used as one measure of the quality of the recorded signal. To determine a representative minimum collection time, a series of measurements is carried out on a {001} oriented sample of 99.99% pure, single crystal tungsten, varying the number of traces collected in a single measurement from 10 to 50,000. These measurements are carried out using an imposed grating wavelength of $\Lambda = 5.50 \mu$m. The time-dependent signals for a 250 trace averaged measurement from a series of three measurements at a fixed collection time.

Table 1 shows statistics for each collection time, including the signal-to-noise ratio and the 95% confidence interval of the peak position parameter from a Gaussian fit to the spectrum. As there is often a DC component retained in the power spectrum from slow thermal decay, the signal-to-noise ratio of these measurements ranged from 0.550 s to 743 s (12.5 min). As the collection time is increased, the signal-to-noise ratio of the identifiable acoustic peaks in the spectrum is increased, as expected.

![Graph](image-url)

FIG. 2. Example traces of $I_S^\text{tot}(t)$ (red), $I_S^{00}(t)$ (blue), and $I_S^{180}(t)$ (black) from DH-TGS measurements on {001} oriented single crystal tungsten using $\Lambda = 5.50 \mu$m and a collection time of 4.19 s.

![Graph](image-url)

FIG. 3. Filtered power spectra of DH-TGS measurements collected on {001} oriented single crystal tungsten using $\Lambda = 5.50 \mu$m with collection times varying from 0.550 s to 743 s. Each spectrum is a representative selection from a series of three measurements at a fixed collection time.
The signal-to-noise ratio (SNR) is calculated over the noise floor from 0.7 to 1.3 GHz as the DC component is clearly separable from the acoustic peak of interest. The entries in Table I are the average values given from three measurements of the same record length. For measurements averaging 10 and 25 laser pulses, identifiable peaks in the power spectrum were not always apparent above the noise floor. From this series of measurements, based solely on the consistency of the appearance of peaks in the acoustic spectrum, a minimum temporal resolution of between 1 and 10 s is observed.

Using this implementation of a TGS experiment, the thermal and acoustic properties of specimens subject to some external driving force can be monitored. Drivers such as temperature, voltage, or irradiation exposure can cause drastic changes in the acoustic and thermal transport properties of materials under investigation. As a simple proof of the principle experiment, a series of DH-TGS measurements is carried out on a {111} oriented sample of single crystal aluminum subject to a temperature ramp between 50 and 250 °C. Measurements were taken with a = 4.78 µm, using 1000 trace collections on a 20 s interval over a period of about 30 min. If acoustic peaks are identified in the power spectrum at dominant frequencies \( f_{TG} \), then the surface acoustic wave (SAW) speed for each measurement can be calculated as \( c_{TG} = \frac{\lambda f_{TG}}{2} \), where the imposed grating wavelength is calibrated before each experiment, as in Ref. 16. Figure 4 shows the variation in SAW speed with temperature as measured on single crystal aluminum. The apparent linear decrease in \( c_{TG} \) over this temperature range is consistent with SAW speeds calculated from temperature-dependent elastic constants of aluminum.\(^{15,17}\)

In general, collecting traces of \( P_{SC}^e(t) \) with a signal-to-noise ratio in the time domain sufficiently high enough to accurately fit thermal transport parameters requires a longer collection time than that required to identify the primary excited acoustic oscillations as discussed here. However, the difference in these time scales varies widely between different material systems. For this reason and due to contributions of the reflectivity response in the thermal decay profile, the discussion thus far has been restricted to the minimum time scale necessary to analyze the acoustic component of the DH-TGS measurement.

A comment is required concerning the difference in the angles \( \theta_1 \) and \( \theta_2 \) which control the vertical separation of the two probe paths in Fig. 1(b). In practice, these two angles should be set as close as possible to each other while maintaining the ability to independently control the heterodyne phase of each path. The lenses L1 and L2 used in the current iteration of this experiment are achromatically, but not aspherically, corrected doublets. A small spherical aberration present in the lens system causes a focal length shift between the two probing paths. The overlap volume of the two pump beams is large enough with the spot sizes used here to allow for the simultaneous collection of both \( I_{S}^{1}(t) \) and \( I_{S}^{100}(t) \) with careful positioning of the sample surface in the beam path direction. Upon initial alignment, the photodetectors must be tuned to compensate for differences in signal amplitude occurring due to lens aberration and differences in intrinsic gain. The response amplitudes can be matched by setting the heterodyne phase to \( \phi = 0 \)° for both probe paths, taking the real-time Fourier transform of the photodetector response on the acquisition oscilloscope, and manually tuning the photodetector gain such that the SAW peak in the transform of each detector has the same power. Future implementations of this technique should utilize both achromatically and aspherically corrected lens systems for L1 and L2 to reduce the sensitivity to this aberration.

The removal of any long-timescale systematic error by subtracting \( P_{SC}^e(t) \) from \( I_{S}^{100}(t) \) to find \( I_{S}^{100}(t) \) provides several improvements in the analysis of elastic properties through the measured acoustic response. As the signal intensity measured on the photodetectors is intrinsic shot-noise limited, the method proposed here relying on the upstream splitting of the same probe laser source to create both probing paths should, in theory, not result in an improvement in the overall acoustic signal intensity compared to a single-phase collection using the same probe laser source. However, as the reference oscillator needs to be attenuated to avoid saturating the photodetector,
splitting the probe laser source allows for the ratio of the reference oscillator intensity to the diffracted signal intensity, \( I_0/I_D(t) \), to be increased across two detectors compared to the single detector in traditional TGS experiments. Additionally, the power spectra of short time scale collections with traditional TGS experiments which do not correct the systematic component of \( I_0^{ref}(t) \) may be more complicated to analyze in the low frequency region. This complication becomes detrimental for measurements at long acoustic wavelengths or for particularly compliant materials.

In contrast to the acoustic analysis, the determination of the thermal transport properties from \( I_0^{ref}(t) \) relies on model fitting, for which the removal of the systematic error by subtracting \( I_0^{th}(t) \) and \( I_0^{180}(t) \) is a necessary step.\(^2,11\) Given this requirement and the presence of a systematic component to the captured signals, which is common, DH-TGS will allow for thermal transport properties to be measured on much shorter time scales than traditional experiments. Converting existing TGS experiments to DH-TGS experiments is possible as the optical arrangement described here requires only the addition of a small number of optics and a second photodetector. The true advantage using the methodology described in this letter will be the ability to near-continuously characterize both the thermal and elastic properties of dynamic material systems undergoing some external forcing. In contrast, traditional TGS experiments have largely been limited to static material systems. This static-to-dynamic shift will allow the transient grating method to be used in \textit{in-situ} applications which have not yet been practical with single heterodyne phase collection.

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\(^1\) A. A. Maznev, K. A. Nelson, and J. A. Rogers, “Optical heterodyne detection of laser-induced gratings,” \textit{Opt. Lett.} \textbf{23}, 1319–1321 (1998).

\(^2\) J. A. Johnson, A. A. Maznev, M. T. Bulsara, E. A. Fitzgerald, T. C. Harman, S. Calawa, C. J. Vineis, G. Turner, and K. A. Nelson, “Phase-controlled, heterodyne laser-induced transient grating measurements of thermal transport properties in opaque material,” \textit{J. Appl. Phys.} \textbf{111}, 025303 (2012).

\(^3\) N. Boechler, J. K. Eliason, A. Kumar, A. A. Maznev, K. A. Nelson, and N. Fang, “Interaction of a contact resonance of microspheres with surface acoustic waves,” \textit{Phys. Rev. Lett.} \textbf{111}, 036103 (2013).

\(^4\) J. A. Johnson, A. A. Maznev, J. Cuffe, J. K. Eliason, A. J. Minnich, T. Kehoe, C. M. S. Torres, G. Chen, and K. A. Nelson, “Direct measurement of room-temperature nondiffusive thermal transport over micron distances in a silicon membrane,” \textit{Phys. Rev. Lett.} \textbf{110}, 025901 (2013).

\(^5\) A. Rogers, A. A. Maznev, M. J. Banet, and K. A. Nelson, “Optical generation and characterization of acoustic waves in thin films: Fundamentals and applications,” \textit{Annu. Rev. Mater. Sci.} \textbf{30}, 117–157 (2000).

\(^6\) J. Janusonis, T. Jansma, C. L. Chang, Q. Liu, A. Gatilova, A. M. Lomonosov, V. Shalagatskiy, T. Pezeril, V. V. Tmnov, and R. I. Tobey, “Transient grating spectroscopy in magnetic thin films: Simultaneous detection of elastic and magnetic dynamics,” \textit{Sci. Rep.} \textbf{6}, 29143 (2016).

\(^7\) L. Yang, J. D. Koralek, J. Orenstein, D. R. Tibbetts, J. L. Reno, and M. P. Lilly, “Measurement of electron-hole friction in an n-doped GaAs/AlGaAs quantum well using optical transient grating spectroscopy,” \textit{Phys. Rev. Lett.} \textbf{106}, 247401 (2011).

\(^8\) N. Gedik, J. Orenstein, R. Liang, D. A. Bonn, and W. N. Hardy, “Diffusion of nonequilibrium quasi-particles in a cuprate superconductor,” \textit{Science} \textbf{300}, 1410–1412 (2003).

\(^9\) R. A. Duncan, F. Hofmann, A. Vega-Flick, J. K. Eliason, A. A. Maznev, W. Liu, D. E. J. Armstrong, K. A. Nelson, and S. L. Dudarev, “Lattice swelling and modulus change in a helium-implanted tungsten alloy: X-ray micro-diffraction, surface acoustic wave measurements, and multiscale modelling,” \textit{Acta Mater.} \textbf{89}, 352–363 (2015).

\(^{10}\) F. Hofmann, D. R. Mason, J. K. Eliason, A. A. Maznev, K. A. Nelson, and S. L. Dudarev, “Non-contact measurement of thermal diffusivity in ion-implanted nuclear materials,” \textit{Sci. Rep.} \textbf{5}, 16042 (2015).

\(^{11}\) R. A. Duncan, F. Hofmann, A. Vega-Flick, J. K. Eliason, A. A. Maznev, A. G. Every, and K. A. Nelson, “Increase in elastic anisotropy of single crystal tungsten upon he-ion implantation measured with laser-generated surface acoustic waves,” \textit{Appl. Phys. Lett.} \textbf{109}, 151906 (2016).

\(^{12}\) O. W. Kading, H. Skurk, A. A. Maznev, and E. Matthias, “Transient thermal gratings at surfaces for thermal characterization of bulk materials and thin films,” \textit{Appl. Phys. A} \textbf{61}, 253–261 (1995).

\(^{13}\) J. Fizev, “The determination of the elastic constants of isotropic solids by means of transient thermal surface gratings,” \textit{J. Appl. Phys.} \textbf{119}, 015301 (2016).

\(^{14}\) D. Gerlich and E. S. Fisher, “The high temperature elastic moduli of aluminum,” \textit{J. Phys. Chem. Solids} \textbf{30}, 1197–1205 (1969).

\(^{15}\) C. A. Dennett, P. Cao, S. E. Ferry, A. Vega-Flick, A. A. Maznev, K. A. Nelson, A. G. Every, and M. P. Short, “Bridging the gap to mesoscale radiation materials science with transient grating spectroscopy,” \textit{Phys. Rev. B} \textbf{94}, 214106 (2016).

\(^{16}\) A. G. Every, A. A. Maznev, W. Grill, M. Pluta, J. D. Comins, O. B. Wright, O. Matsuda, W. Sachse, and J. P. Wolfe, “Bulk and surface acoustic wave phenomena in crystals: Observation and interpretation,” \textit{Wave Motion} \textbf{50}, 1197–1217 (2013).