DOI 10.5162/Ultrasonic2025/A9-a1

Thermo-Elastic Coupling in Transient Grating Spectroscopy

Jakub Kušnír[2], Kristýna Repček[2], Tomáš Grabec[1], David Mareš[1], Pavla Stoklasová[1], Petr Sedlák[1], and Hanuš Seiner[1]
[1]*Institute of Thermomechanics of the Czech Academy of Sciences, Prague, Czechia*
[2]*Faculty of Nuclear Sciences and Physical Engineering, Czech Technical University in Prague, Prague, Czechia*
kusnir@it.cas.cz

Abstract: Characterization of the thermal diffusivity on the micrometer-thick film on a substrate by transient grating spectroscopy was investigated. The experiment was performed on a thin film of NiTi at 120°C, when the film is in the austenitic phase and exhibits cubic crystallographic symmetry. Apparent anisotropy of thermal diffusivity was observed in the experimental results, as confirmed by finite-element model (FEM). However, the calculation confirmed that the measurement was only sensitive to the thermal diffusivity of the film, omitting the substrate.

Keywords: transient grating spectroscopy, elasticity, thermal diffusivity, finite elements method

Introduction

Transient grating spectroscopy (TGS) is an opto-acoustic technique used to characterize the elastic and thermal properties of solids [1, 2, 3]. TGS, also known as impulsive stimulated thermal scattering (ISTS) [4, 5] or transient thermal grating (TTG) [6], is a non-destructive and contactless method that was used successfully to determine the elastic properties of a single-crystalline thin film of NiTi on an MgO substrate [7] and to measure the thermal diffusivity of the cubic single-crystals [8].

TGS uses two pulsed laser beams interfering on the sample surface that create spatially harmonic thermo-elastic excitation. Because of the excitation pattern, thermal and acoustic transient gratings are created – forming a dynamic surface-displacement grating on which the probe laser diffracts. The probe laser is continuous, and the optical geometry ensures recombination of the probe beams in a heterodyne detection setup [1]. The relative phase shift of the probe beams (the heterodyne phase) then influences the information obtained: Without a phase shift, the signal depends solely on thermal reflectivity; with a non-zero phase shift, it reflects both thermal reflectivity and surface displacement, combining acoustic oscillations with thermal decay [9]. An ideal signal is obtained in the 'phase-grating' mode with the heterodyne phase $\theta_{\mathrm{het}} = (\pm\pi/2)$. Acoustic oscillations can be characterized directly from the frequency spectrum, while fitting the signal with a formula from analytical models allows extracting the thermal diffusivity [10, 11].

However, as described in Ref. [8], when thermal diffusivity is measured on elastically anisotropic samples, the result of the fitting formula is influenced by the elastic properties, and a correction is needed to find the true thermal diffusivity of the measured sample. To characterize the influence of the elastic anisotropy on the apparent thermal anisotropy determined by the fitting formula, a quasi-static FEM model of the TGS experiment was used [8].

In this work, we show that TGS allows us to determine the thermal diffusivity of micrometer-thick films without the influence of the substrate. The sensitivity to thermal properties is depth dependent on the wavelength λ of the interference pattern as

$$L = \frac{\lambda}{\pi}, \tag{1}$$

where L is the thermal diffusion length [11]. Therefore, thermal properties are obtained up to this depth. Here we present its potential to determine the thermal properties of a single-crystalline sample that cannot be manufactured in bulk but only as a thin film.

Methods - Transient Grating Spectroscopy

Transient grating spectroscopy, illustrated in Fig. 1, is based on diffracting pump and probe lasers on the transmission phase grating and projecting ± 1 diffraction order beams on the sample surface using the 4F imaging system. Projected pump beams interfere on the sample surface, creating a spatially harmonic excitation pattern. The fringes spacing, and thus the acoustic wavelength λ, is set by the incidence angle of the beams and the laser light wavelength. Acoustic wavelength can thus be easily changed by changing the transmission phase grating. Each interference fringe acts as thermo-elastic source, launching

DOI 10.5162/Ultrasonic2025/A9-a1

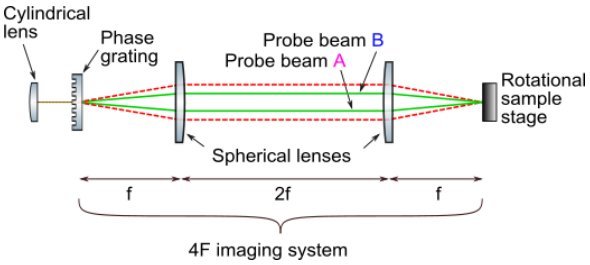

Fig. 1: Top-view schematic of the simplified optical setup of the TGS method.

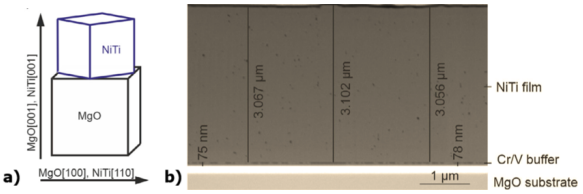

Fig. 2: a) Schematic showing B2 austenite unit-cell orientations of the film with respect to the substrate, b) cross-sectional SEM thickness measurement of the supported 3 µm thick NiTi layer on MgO substrate.

acoustic waves with a well-defined wave vector in a direction perpendicular to the excitation lines.

The probe beams go through the same optical setup as the pump beams. The beams are diffracted on the transient grating created on the sample surface after the excitation. The optical setup is in the Littrow configuration, where each probe beam diffracts in the reflection direction of the other, enabling heterodyne detection. Detection is carried out in the 'phase-grating' mode (heterodyne phase $\theta_{\mathrm{het}} = (\pm\pi/2)$), which maximizes sensitivity to out-of-plane displacement.

The measurement setup was similar to that used in the previous work of the authors [2]. The pump laser was a Nd:YAG pump laser (1064 nm, 0.54 ns, 200 µJ, 1 kHz) where the mean power on the sample surface was 17 mW. The probe beams were from the continuous-wave laser at 532 nm (120 mW) with mean power on detector around 15 mW. The optical setup was such that the resulting acoustic wavelength was $\lambda = 7\mu$m. The signal-to-noise ratio and sensitivity are further enhanced by measuring the intensity using differential Si photodiodes with a 60 dB amplifier. The differential setup further enhances the signal quality by removing phase-insensitive influences [4, 11].

The heterodyne probe signal was recorded by WaveRunner 640Zi oscilloscope (4 GHz bandwidth, vertical resolution of 8 bits, sampling rate 5 GS /s). The signal was averaged 50 thousand times in the time-domain for enhanced resolution and further reduced noise. Fourier analysis then reveals the dominant SAW frequencies, from which the SAW velocities were obtained using $v = f\lambda$, where λ is the excitation grating period, and were used to determine elastic constants [2].

To determine the thermal diffusivity, the fitting of an analytical function in the time-domain was used [10, 11]:

$$I(t) = A\left[\mathrm{erfc}\left(q\sqrt{\alpha t}\right) - \frac{\beta}{\sqrt{t}}\exp\left(q^2\alpha t\right)\right] + B\sin\left(2\pi f t + \varphi\right)\exp\left(-\frac{t}{\tau}\right) + C, \quad (2)$$

with A, B and C being constants, $q = 2\pi/\lambda$ is the acoustic wave number, α is thermal diffusivity, β describes the displacement and reflectivity contribution ratio, f frequency of the acoustic wave, τ the acoustic decay and ϕ the phase of the acoustic wave.

The experimental results were compared with the simulated results from the FEM simulation of the TGS experiment described in detail in [8].

Study case

The sample used in this study was a thin film of a NiTi shape memory alloy. The film was epitaxially grown by DC magnetron sputtering on a single-crystalline MgO(100) substrate [12]. To reduce epitaxial stress, buffer layers of vanadium and chromium were used, with a combined thickness of around 50 nm.

The epitaxial relationship between the NiTi austenite (B2 structure) and the MgO substrate was MgO(100)[001] ∥ V/Cr(100)[011] ∥ NiTi B2(100)[011]. This indicates that the austenite unit cell is rotated by 45° around the substrate normal relative to the film, such that of MgO aligns with of the austenite (as shown in Fig. 2a). The film thickness was measured by scanning electron microscopy (SEM) on a cross-section prepared by focused ion beam (FIB) milling using a FEI Helios NanoLab 600i, with the determined value of 3060 nm (Fig. 2b).

Results and Discussion

We carried out a 45° angular scan (with a step of 1°) of TGS measurement with $\lambda \approx 7\mu$m. The sample was measured at an elevated temperature of 120 °C at which the film was in a single crystalline austenite phase. The obtained time-domain signal is shown in Fig. 3 (top) with a strong acoustic and thermal

DOI 10.5162/Ultrasonic2025/A9-a1

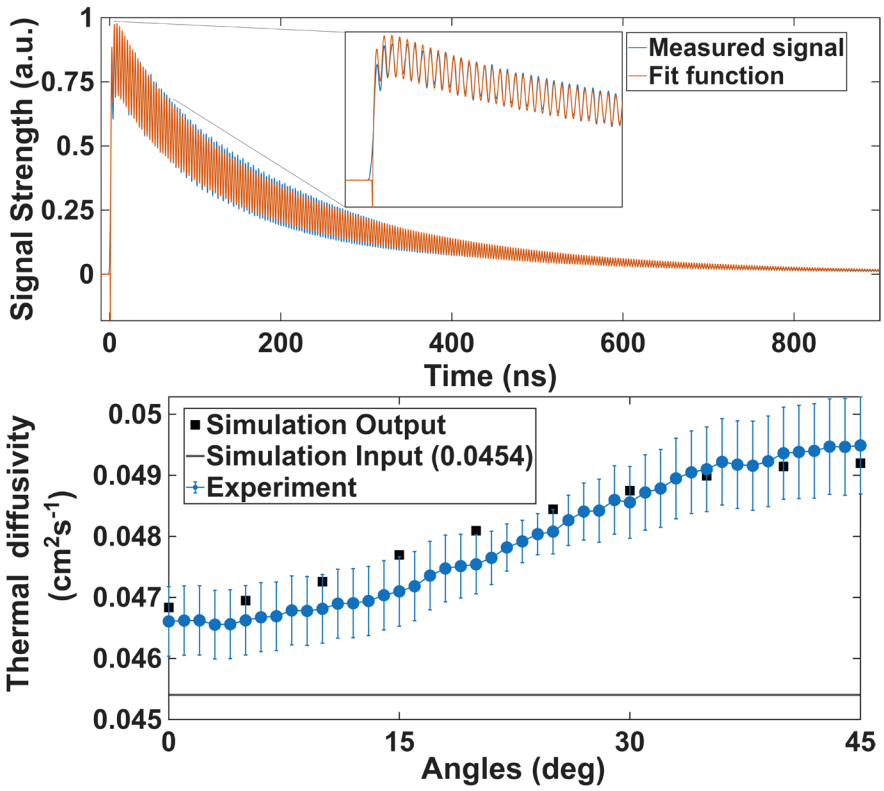

Fig. 3: Top: Time-domain signal in the 'phase-grating' measurement mode for epitaxial NiTi(001)[100] ($3\mu m$) on MgO(001)[110], with the measured signal in solid blue and calculated signal from the fitting function Eq. (2)*. Bottom: Comparison of experimentally measured (blue circles), simulated (black squares) and simulation input (black line) thermal diffusivity of thin film of NiTi on MgO. The black line is the true thermal diffusivity of the austenite phase of NiTi at 120 °C [8].*

response. The measured signal was fitted by Eq. (2), orange line, to obtain the thermal diffusivity.

Although the film is crystallographically cubic and therefore the thermal properties should be isotropic anisotropy of the thermal diffusivity was measured as shown in Fig. 3 (bottom) as blue circles with error bars (obtained from the fitting formula). This anisotropy results from the elastic anisotropy, which the derivation of the fitting formula (2) omits, and it is thus only apparent – as described in detail in Ref. [8]. However, quasi-static FEM simulation of the TGS experiment was used to find the true isotropic thermal diffusivity of the studied material by comparing the results of the FEM simulation with the experimental results, where isotropic thermal diffusivity was used as input. The input value for the FEM simulation is the considered the sought thermal diffusivity of the austenite phase of the NiTi at 120 °C, resulting in $\alpha = 0.0454\ \mathrm{cm^2 s^{-1}}$.

Note that the magnitude of λ was chosen so that the thermal properties would be probed only in the NiTi film – based on Eq. (1), the thermal-diffusion depth should be approximately $2.2\mu\mathrm{m}$. This was confirmed by the agreement of the experiment with the simulation, which was performed for considering bulk NiTi and omitting the substrate altogether.

Conclusion

Characterization of thermal diffusivity of epitaxial NiTi film was performed by TGS. The acoustic wavelength of 7 microns allowed us to probe only the film (with the thickness of 3 micrometers), omitting the influence of the MgO substrate. Although the film was crystallographically cubic and should be isotropic in thermal properties, anisotropic results were obtained by fitting the TGS signals. This is in agreement with earlier findings of the influence of elastic anisotropy on the TGS signals and the rate of the slow decay which corresponds with the thermal diffusivity. These results were confirmed by FEM simulations, allowing us to determine the true value of the thermal diffusivity of an elastically anisotropic thin supported film.

References

[1] A. A. Maznev, K. A. Nelson, and J. A. Rogers. "Optical heterodyne detection of laser-induced gratings". In: *Opt. Lett.* 23.16 (1998), pp. 1319–1321. DOI: 10.1364/OL.23.001319.

[2] P. Stoklasová et al. "Laser-Ultrasonic Characterization of Strongly Anisotropic Materials by Transient Grating Spectroscopy". In: *Experimental Mechanics* (2021). DOI: 10.1007/s11340-021-00698-6.

[3] K. Repček et al. "Compliant Lattice Modulations Enable Anomalous Elasticity in Ni–Mn–Ga Martensite". In: *Advanced Materials* 36.39 (2024), p. 2406672. DOI: 10.1002/adma.202406672.

[4] B. Verstraeten et al. "Determination of thermoelastic material properties by differential heterodyne detection of impulsive stimulated thermal scattering". In: *Photoacoustics* 3.2 (2015), pp. 64–77. DOI: 10.1016/j.pacs.2015.05.001.

[5] T. Grabec et al. "In-situ Characterization of Local Elastic Properties of Thin Shape Memory Films by Surface Acoustic Waves". In: *Smart Materials and Structures* 25.12 (2016). DOI: 10.1088/0964-1726/25/12/127002.

[6] J. A. Johnson et al. "Non-diffusive thermal transport in GaAs at micron length scales". In: *Journal of Applied Physics* 118.15 (2015), p. 155104. DOI: 10.1063/1.4933285.

[7] T. Grabec et al. "Guided acoustic waves in thin epitaxial films: Experiment and inverse problem solution for NiTi". In: *Ultrasonics* 138 (2024). DOI: 10.1016/j.ultras.2023.107211.

[8] J. Kušnír et al. "Apparent anisotropic thermal diffusivity measured in cubic single crystals by transient grating spectroscopy". In: *Journal of Applied Physics* 133.12 (2023), p. 125108. DOI: 10.1063/5.0136850.

[9] J. A. Johnson et al. "Phase-controlled, heterodyne laser-induced transient grating measurements of thermal transport properties in opaque material". In: *Journal of Applied Physics* 111.2 (2012), p. 023503. DOI: 10.1063/1.3675467.

[10] C. A. Dennett and M. P. Short. "Thermal diffusivity determination using heterodyne phase insensitive transient grating spectroscopy". In: *Journal of Applied Physics* 123.21 (2018), p. 215109. DOI: 10.1063/1.5026429.

[11] O. W. Käding et al. "Transient thermal gratings at surfaces for thermal characterization of bulk materials and thin films". In: *Applied Physics A* 61.3 (1995), pp. 253–261. DOI: 10.1007/BF01538190.

[12] K. Lünser et al. "How to grow single-crystalline and epitaxial NiTi films in (100)- and (111)-orientation". In: *Journal of Physics: Materials* 6.3 (2023), p. 035002. DOI: 10.1088/2515-7639/acd604.

Acknowledgments

This work was financially supported by the Czech Science Foundation [Project No. 22-13462S], Operational Program Johannes Amos Comenius of the Ministry of Education, Youth and Sport of the Czech Republic, within the frame of project Ferroic Multifunctionalities (FerrMion) [Project No.CZ.02.01.01/00/22_008/0004591], co-funded by the European Union and by the Grant Agency of the Czech Technical University [No. SGS25/168/OHK4/3T/14].