Thermomechanical properties of single crystals evaluated by impulsive stimulated thermal scattering technique

M Landa¹, B Verstraeten², J Sermeus², R Salenbien², P Sedláček¹, H Seiner¹, C Glorieux²

¹Institute of Thermomechanics, Academy of Sciences of the Czech Republic, Dolejškova 5, Prague 8, CZ 182 00, Czech Republic
²Laboratory for Acoustics and Thermal Physics, Department of Physics and Astronomy, Katholieke Universiteit Leuven, Celestijnenlaan 200D, B3001 Heverlee, Belgium
E-mail: ml@it.cas.cz

Abstract. This paper describes the application of Impulse Stimulated Thermal Scattering (ISTS) for measurement of surface acoustic wave (SAW) velocity and thermal diffusion along a free surface of a strongly anisotropic material. The motivation for this work stems from the study of thermoelastic properties of individual phases of ferroelastics; experimental results were obtained on a single crystal in the austenitic phase a Cu-Al-Ni alloy (bcc single crystal having elastic anisotropy factor of about 12). The measured SAW velocities in specific directions are in a good agreement with the values calculated for the elastic constants obtained by other ultrasonic methods. Similarly, the evaluated thermal diffusivity coefficient \((22 \pm 2) \times 10^{-6} \text{ m}^2/\text{s}\) of the austenite is consistent with the data in the literature. The proposed approach has also a potential for characterization of thin films grown on anisotropic substrates.

1. Introduction
Ferroelastic materials are materials able to form fine, geometrically ordered microstructures consisting of differently oriented regions (ferroic domains) separated by twin boundaries. When the collective orientation of such a microstructure is changed by application an external field (temperature, mechanical loadings, magnetic field) the material macroscopically exhibits a reversible pseudoplastic strain magnitude up to of 10%. The elasticity of individual phases and their behavior in the vicinity of phase transitions are fundamental characteristics of the ferroelastics. However, due to the strength of the elastic anisotropy, the determination of the all independent elastic constants of these materials fall beyond the applicability limits of conventional pulse-echo methods [1] and of the resonant ultrasound spectroscopic (RUS) method [2]. Complementarity of these two methods for bulk elasticity measurements is exploited in the combined method introduced in [3], which enables a reliable determination of the sought elastic coefficients even if the both methods fail when applied separately.

The Surface Acoustic Wave (SAW) propagation can be understood as a surface resonator [6], with properties sensitive to individual components of the elastic tensor or their combinations. This resonant approach enables the sensitivity analysis to be carried out in a similar way as in the case of RUS and the pulse echo methods [3]. Since the displacement and stress fields of SAW...
are confined to a shallow region between the free surface with controllable penetration depth, their use has a particular potential for the evaluation of thin films. It is obviously desirable that of the elastic properties of the substrate are known.

2. Description of the method

Impulse Stimulated Thermal Scattering (ISTS) is a spectral method [4], using spatially narrow band excitations (figure 1). Grating excitation is particularly useful when the short wavelengths are to be excited, which is of interest for the investigation of shallow material regions and thin coatings. In principle, grating like SAW packets can be detected using laser beam deflection. However, the requirement that the probe beam must be focused into a width that is smaller than the SAW wavelength can be quite demanding as far as the instrumentation is concerned. The heterodyne diffraction detection scheme avoids this complication, since it involves no strict requirements on the width of the probe beam. The used design scheme makes it quite easy to detect short wavelength SAW with a high sensitivity allowing to detect even very small SAW displacements. In this differential variant, the optical phase difference between the diffracted and reference probe beams is adjusted and photodiode signals detecting the respective mixed light beams are electronically subtracted. This optical system was equipped by a rotational table for angular positioning of a specimen with respect to the direction of SAW propagation. The specimen was adjusted such that the surface under study was perpendicular to the optical axis of the ISTS system.

An example of a typical ISTS signal detected by heterodyne diffraction is shown in figure 1, showing contributions of SAW (propagating in the solid surface) and Scholte-Stoneley waves (interfacial wave propagation in air along the interface) may be seen. The decay of the signal amplitude with time (as a DC part of the signal) representing characteristics of thermal diffusivity may be separated by an additional low-pass detector (figure 1).

3. Sample preparation

A single crystal of Cu-14.3% Al-4.1% Ni (wt.%) alloy was prepared by the Bridgman method. The crystal structure at room temperature is b.c.c. (cubic austenite β-phase), because the transition temperature (\(M_s \sim 193\text{K}\)) to the martensite phase is much lower. A cube-like specimen (dimensions equal to approximately \(10 \times 10 \times 10\text{ mm}^3\)) was cut with faces perpendicular to the [110], [1\(\bar{1}\)0] and [001] crystallographic axes. By cooling below \(M_s\), the austenite phase
Table 1. Elastic constants of the CuAlNi alloy

| $\rho_o$ | $C_{11}$ | $C_{12}$ | $C_{44}$ | $A$ | $C_L$ | $C''$ |
|---------|----------|----------|----------|-----|-------|-------|
| [g/cm$^3$] | [GPa] | [GPa] | [GPa] | [1] | |
| 7.055 | 142.80 | 126.84 ±0.15 | 95.90±0.30 | 12.02 | 230.73 | 7.98 |

$A = 2C_{44}/(C_{11} - C_{12})$, $C'' = (C_{11} - C_{12})/2$, $C_L = (C_{11} + C_{12})/2 + C_{44}$

can be transformed into the martensite of orthorhombic 2H structure. The elastic constants of this material were measured by conventional pulse echo method [5] and the results (table 1) were then used for the following SAW analysis.

Figure 2. Results of ISTS measurements on CuAlNi single crystal.

4. Experimental results and analysis

Experiments were carried out on the laser ultrasonic system designed in ATF KUL, Belgium. The ISTS signals were obtained during sample rotation on both the (001) and (110) surfaces of the CuAlNi single crystal at ambient conditions (RT = 25°C). The nominal optical wavelength of the grating was 50 µm. The fast component of each signal, obtained from a fast (AC) photodetector was averaged 4096 times, while the longer time evolution of the signal, obtained by a slower (DC) photodetector was averaged 2024 times. The angular step was 2.5 or 5°.

The frequencies of the periodical SAW and Scholte wave packets were determined from the peaks in the frequency spectra of the recorded signals obtained by FFT. The effective (imaged) wavelength ($\lambda_{eff} = 55$ µm, typically) of the SAW grating waves, which can slightly deviate from the nominal value of the phase mask, was calibrated by comparing the found Scholte velocity with the literature value of the sound velocity of air at the temperature of the ambient. In figure 2, the resulting SAW velocities are represented by points, the size of which reflects the relative magnitude of the spectral peak. Using material properties from Table 1, the phase velocity of quasi transverse waves and planar SAW were calculated and plotted in the diagram. The ”complete” theoretical angular dependence of SAW was calculated step by step from a numerical model of a surface resonator of length 2A with periodical boundary conditions [6]. For each step, the matrix of elastic constants was recalculated for a given general crystallographic orientation. The results of this model were verified by the solutions of the analytical formulas [7].
for the SAW velocities in special directions. The "analytical" points are denoted by squares and labeled by "VR" with the propagation direction. In spite of the strong anisotropy, good agreement was found between the experiment and the theoretical prediction. The dependence of the SAW penetration depth on the orientation can be evaluated from the displacement fields $(u_x, u_z, u_y)$ mapped for $\theta = 0, 10, 20, 30, 45^o$ in the plane (001) and for $\theta = 0, 15, 30, 60, 90^o$ in the (110) plane. The penetration depth significantly increases far from the principal crystallographic directions. Near the [-110] direction in the (110) plane, a transition from the SAW mode to the $q_T$slow mode can be observed.

Since the examined material is cubic, the thermal diffusivity (as a second rank tensor [8]) should be isotropic, i.e. representable by a single scalar quantity. This quantity can be estimated from the DC part of the ISTS signal (figure 1). When fitting this signal by $\text{erfc}(-\beta t)$, the time decay coefficient $\beta$ can be expressed as $\beta = D_T(2\pi/\lambda_{eff})^2$, which enables the thermal diffusivity coefficient $D_T$ to be calculated [9]. The averaged value of the thermal diffusivity obtained from the all angular measurements was $D_T = (22 \pm 2).10^{-6}$ m$^2$/s. This value is slightly higher than estimates (about 16.10$^{-6}$ m$^2$/s) obtained from typical thermal properties of CuAlNi alloy [10], indicating a good crystalline quality. The given measurement uncertainty is based on the standard deviation between measurements taken under different angles. As expected, no systematic variation of the thermal diffusivity with angle was found for this cubic crystal.

5. Conclusion
Thermomechanical properties of a strongly anisotropic cubic single crystal were determined by ISTS methods. The angular dependence of both the SAW velocity and the thermal diffusivity was determined in the (001) and (110) planes. The experimentally obtained SAW velocities were compared with theoretical predictions based on a numerical model. In spite of the strong anisotropy, a satisfactory agreement between experimental and theoretical results was found, which proves the reliability of the numerical algorithm used for the calculation of the SAW velocities, and shows that this algorithm could be possibly used for inverse determination of the elastic constants. For the thermal diffusivity measurements, the results are, as well, in an acceptable agreement with the literature data. Both the experimental determination and theoretical analysis of thermal diffusivity in single crystals need, however, further investigation.

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