Temperature Dependent Elasticity of a Ni₂MnGa Film as Measured by Impulsive Stimulated Scattering

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Abstract. We report the results of an impulsive stimulated scattering measurement conducted on a 550 nm film of Ni₂MnGa on a MgO substrate, aiming at the direct experimental observation of the highly temperature dependent elasticity. The phase velocity of the surface acoustic waves excited on the surface of the coating-substrate shows an anomaly around the phase transition temperature of Ni₂MnGa, showing that it is possible to use impulsive stimulated scattering as a characterization tool to determine temperature dependent elasticity of a thin film.

1. Introduction

Impulsive stimulated scattering (ISS) is a laser ultrasonic technique used to excite and detect monochromatic surface acoustic waves (SAW) on a material. It has proven to be an useful tool in the characterization of bulk solids [1,2], free liquid surfaces [3], thin films [4] and multilayered structures [5]. A more extensive review of the technique, applications and the relationship of ISS to other laser ultrasonic methods, e.g. picosecond ultrasonics, is described in reference [6]. While it is possible to use the ISS technique in a transmission mode for the thermoelastic characterization of liquids [7], a reflection scheme was used here to characterize the opaque and highly reflective material. In this work, the phase velocity was measured for a SAW propagating along a crystal axis of a MgO single crystal substrate coated by a 550 nm Ni₂MnGa film.

Ni₂MnGa is a magnetic shape memory alloy (MSMA) with many interesting properties and with promising applications in MEMS devices. Since the phase velocity of SAW directly depends on the elastic properties of the material, anomalies in the SAW velocity reflect changes in the microstructure and are thus linked to the phase of the material. The temperature dependence of the SAW velocity was recorded in the vicinity of the magnetic and phase transition temperatures of the Ni₂MnGa film. Having experimental access to the temperature dependent elastic properties is therefore of great importance from a fundamental and application oriented point of view. A thorough temperature control of the investigated sample is required to correctly determine the transition temperatures. In this regard the question rises what effect the lasers have on the phase of the temperature sensitive coating. By comparing the observed and expected phase change temperature this question can be answered.

The conventional, thermally induced, shape memory effect (e.g. NiTi alloys) is based on a thermoelastic martensitic transformation (first-order solid-to-solid phase transition) from the cubic

parent phase (austenite) to a less symmetric low temperature phase (martensite), or vice versa. The martensitic transformation is accompanied by a high macroscopic strain (up to 10%).

In Ni_2MnGa the magnetic shape memory effect is caused by a reorientation of the martensitic variants (twins) under an external magnetic field due to the high magnetocrystalline anisotropy of the martensite phase. Only modulated phases have a twinning stress low enough for the shape memory effect. The modulated (10M) martensite is often approximated by a tetragonal structure. However, recent studies [8,9] showed that the twinning structures are more complex, since there exist two types of twinning boundaries. Only one of which has a low twinning stress and is preferred for boundary motion. Detailed structural analysis and calculations in these references revealed the mechanisms for easy mobility of twinning interfaces explaining the low reorientation stress.

The sample under investigation was an epitaxially deposited Ni_2MnGa film [10] on a MgO substrate (100) by magnetron sputtering at a deposition temperature of 700K. The deposited film was in the paramagnetic austenite phase with epitaxial relation Ni_2MnGa (100)[110] ‖ MgO (100)[100]. During cooling to room temperature, the martensite phases (mixture of 7M and NM) were reconfigured into hierarchical twin structures [10,11]. The martensitic transformation temperature for this sample was expected between 350-400K.

The Curie temperature of a bulk Ni_2MnGa crystal is a relatively stable value, $T_c = 380\text{-}385K$ for different compositions [12]. However, for thin films T_c is lower and changes with deposition conditions (358K for this particular sample [13]). As the martensitic transition temperature T_{AM} is expected to be higher than T_c, it is not possible to detect the phase transformation temperature through magnetic measurements (e.g. SQUID, see [14,15]). An acoustical approach as presented in this work can provide a complementary tool in the characterization of the martensitic transformation in shape memory alloy thin films.

2. Experimental setup

2.1. Impulsive stimulated scattering setup

In the experimental setup of ISS, as depicted in figure 1, a 10 ps pulsed laser beam (1047 nm) is split into two beams by a first-order optimized diffraction grating. These two first-order laser beams are recombined at the sample surface to create an interference pattern with a specific fringe distance that depends on the diffraction grating and the optical imaging elements that are used. The wavelength of the excited wave was confirmed by registering and analyzing the Scholte wave, an interface wave between the solid sample and the air which travels at the speed of sound in air [16]. The pulsed pump laser has a peak power of 0.5 MW, which corresponds to an average power delivered at the sample of 5mW. At the sample surface a part of the incident light is absorbed, resulting in an impulsive heating. The sudden thermal expansion accompanied by this heating launches counter-propagating SAW with a wavelength corresponding to the interference fringe distance. This results in a standing wave pattern that oscillates at a central frequency f_{SAW}, corresponding to the ratio of the dispersive phase velocity c_{SAW} and the excited wavelength. In this work, the SAW propagated along the [100] direction of the (001) plane of the MgO substrate.

The detection part of the setup consists of a CW probe beam that exactly follows the optical path of the pump beam. At the sample surface the standing SAW pattern act as a temporally modulated diffraction grating for the two probe beams, which are thus partially reflected and diffracted. Recombining the two pairs of reflected and diffracted beams results in heterodyne detection [17] of the standing wave pattern. The intensity modulations were recorded by two Si pin-photodiodes (Hamamatsu S5973) and a high speed GHz amplifier (Femto series HSA). Subtracting the two heterodyne signals results in a doubling of the signal amplitude.

By recording the time signal on an oscilloscope, the central frequency f_{SAW} could be obtained after a fast Fourier transform analysis. In combination with the experimentally known wavelength of the SAW, the phase velocity could be determined for a set of temperatures ranging around the transition temperatures of the Ni_2MnGa coating.

SAW have the property that the penetration depth is comparable to the wavelength along the direction of propagation. In this work SAW with a wavelength of 10 μm were used to probe a 550 nm coating. It is clear from i.a. reference 5, where dispersion curves are shown for SAW propagating on coating-substrate systems, that even for large wavelength over coating thickness ratios the coating influences the SAW propagation.

2.2. Temperature control

The coating-substrate sample was mounted on a brass sample stage holding a calibrated Pt1000 temperature sensor and two 5W electrical resistances that served as external heaters. A PI-controller was used to automate and stabilize the temperature scan. In this way, the SAW phase velocity could be measured within a maximal uncertainty on the surrounding temperature of 0.1 K.
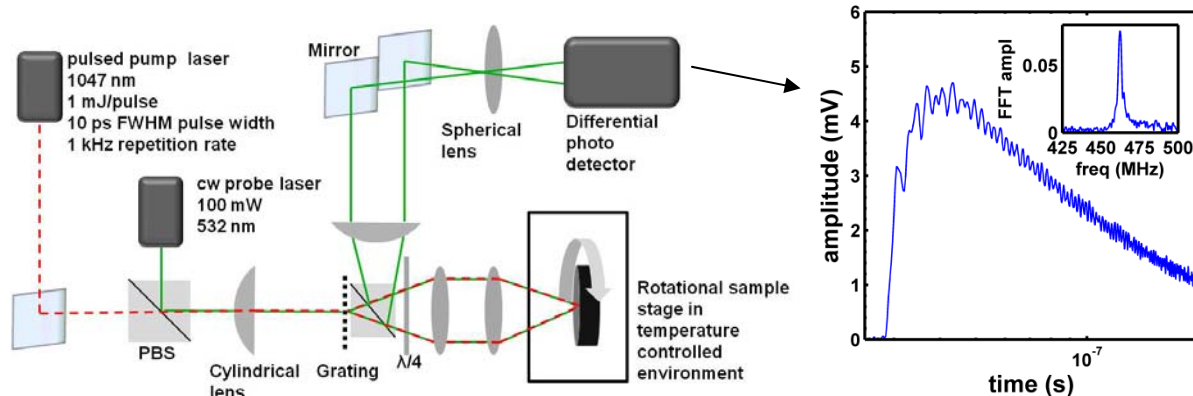

Figure 1. Schematics of the ISS setup with heterodyned detection. Pump and probe laser beams are superposed. The right figure shows a typical time signal with the FFT amplitude spectrum shown in the inset.

3. Results and discussion

In figure 2 the experimentally observed phase velocities of a SAW with a wavelength of 10 μm are shown as a function of the temperature. The transition from the ferro- to the paramagnetic state, which would show as a discontinuity in the first derivative of figure 2, was not observed as the temperature step was too rough. However, an anomaly in the phase SAW velocity around 370K is evident. This jump in the SAW velocity indicates a first order transition and it can be explained as the martensitic transformation. Moreover a narrow hysteresis between the forward (A/M) and reverse transition (M/A) is expected and observed.

The stiffening trend to the T_{AM} is an unexpected feature which might be explained by the influence of the substrate. A strain constrain, internal stresses or a rest of the austenite Ni_2MnGA might be present at the substrate-coating interface. Furhter investigation is needed.

4. Conclusion

In this article, we showed that the temperature dependence of the phase velocity of SAW propagating along the surface of a MgO substrate coated by a Ni_2MnGa film could be measured using ISS. An anomaly was observed around 370K and explained as the M/A phase transition temperature of the Ni_2MnGa coating. The effect of the lasers on the temperature and phase of the coating is therefore limited. Furthermore, this has work proven the feasibility of the exploited technique to experimentally obtain the temperature dependence of the elastic properties of magnetic shape memory alloys.

In future work the temperature dependence and magnetic field dependence of the thermal and elastic properties of the Ni_2MnGa coating will be investigated. In addition, an experimental observation of the dispersive behavior, as a function of wavenumber amplitude and direction, will be reported.
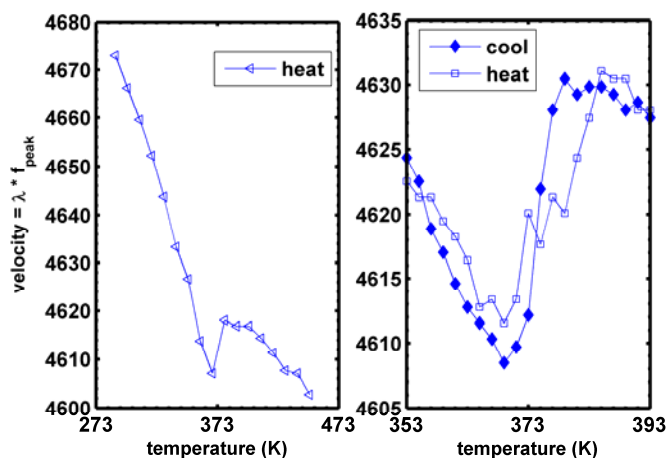
Figure 2. Temperature dependence of the phase velocity of SAW propagating on the surface of the Ni$_2$MnGa film/MgO substrate sample. The acoustic wavelength λ = 10 µm. The markers are experimental points, the lines were added to the eye.

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