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# Pulse laser acoustics for the characterization of inhomogeneities at interfaces of microstructures

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**Abstract**

Interfaces between neighbouring materials are often subjected to diffusion processes which cause layers having gradually varying mechanical properties—like densities, Young's moduli or shear moduli—perpendicular to the surface or interface. In this investigation particular interest is drawn on the question how the propagation characteristics of bulk acoustic waves are affected by diffusion layers. The reflection and transmission behavior of bulk acoustic waves encountering a continuum having a spatially dependent sound velocity is discussed based on numerical simulations as well as on experimental verifications. The simulated results are part of an on-going project in which material properties of MEMS devices are investigated by short pulse laser acoustic methods. Mechanical waves are excited and detected thermoelastically using laser pulses of 70 fs duration. For metals this leads to wavelengths of 10–20 nm and the corresponding frequencies amount to 0.3–0.6 THz. In contrast to previous work done in this field in which diffusion effects are generally considered as undesirable phenomena, the deliberate realization of microstructures having well defined gradually varying material properties in one or more dimensions represents a goal of this investigation. For metallic thin film multilayers thermally induced diffusion processes have shown to be an easy and reliable technique for the realization of layered structures having continuously varying mechanical properties within several 10 nm. Among the experimental methods suitable for the in-depth profiling of submicron metallic thin films providing resolutions of several nanometers, are short pulse laser acoustic methods, Rutherford backscattering spectroscopy (RBS), and glow discharge optical emission spectroscopy (GDOES). Short pulse laser acoustic methods and RBS have the advantage to be nondestructive. The short pulse laser acoustic method is described in detail and RBS measurements are presented for verification purposes. Finally potential engineering applications like micromachined spectrum analyzers, acoustic isolation layers, and band pass filters, operating at very high frequencies are presented.


*Keywords:* Functionally Graded Material (FGM); Diffusion; Laser acoustics; MEMS inspection; Wave propagation; In-depth profiling

## 1. Introduction

When a mechanical stress pulse, which is propagating in an elastic medium, encounters a material or phase interface, which generally represents a change of the acoustic impedance, it is split up into a part which propagates further into the new material and another part which is reflected. The ratio of the transmitted and the reflected stress amplitude is depending on the acoustic impedances of the neighboring materials or phases. The acoustic impedance $Z$ is defined by the product of the sound velocity $c_p$ and the density $\rho$. Provided that the acoustic impedance change is realized by a step function of infinitesimal extent, the reflection/transmission ratio is independent of the wave length i.e. the frequency of the incident pulse. However, this situation changes as soon as the acoustic impedance change takes place in a layer of finite extent. If the thickness of such a layer—which shall be called 'soft acoustic interface' in the sections below—has the same order of magnitude as the wavelength of the propagating stress wave, its reflection/transmission behavior becomes frequency dependent. This phenomenon can be utilized for a geometrical and mechanical interface characterization as well as for a new type of micro-mechanical signal filter or spectrum analyzer.

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The following numerical example shall illustrate the phenomenon:

A multilayer consisting of a pure gold layer of 5 nm thickness, embedded between two ‘soft acoustic interface’ layers and two pure aluminum layers, is exposed to a two frequency mechanical strain pulse as shown in Fig. 1. Here the ‘soft acoustic layers’ are characterized by a smooth, harmonic transition of the mechanical properties, like dilatational stiffness and density, from the aluminum value to the corresponding gold value and vice versa.

The layered structure is excited with a strain signal applied at the surface, which consists of two harmonic components (0.1 and 0.8 THz) multiplied by a hanning window as shown in the top left diagram of Fig. 1.

The total longitudinal extent of the one dimensional structure amounts to 151 nm. The thickness $d$ of the ‘soft acoustic interface layer’ amounts to 15 nm. The length scale of the diagrams of Fig. 1 is normalized with the shortest bulk wave length occurring in the entire structure for the given excitational pulse ($\lambda_{\text{bulk Au min}} = 4.016$ nm) and the time scale is normalized with the corresponding cycle of the highest frequency ($T_{\min} = 1.25$ ps).

The frequencies and the thickness $d$ of the ‘soft acoustic interface layer’ are chosen in a way, that the frequency dependence of the reflection/transmission ratio can be demonstrated as shown in the right diagram of Fig. 1. The length of a bulk wave in aluminum ($\lambda_{\text{bulk Al max}}$) at a frequency of 0.1 THz amounts to 64 nm. Thus, for the lower frequency the ratio of the ‘soft acoustic interface layer’ and the corresponding wave length $d/\lambda_{\text{bulk Al max}} = 0.24$ leads to a strong reflection as visible on the right diagram of Fig. 1. In the case of the higher frequency, $\lambda_{\text{bulk Al min}}$ amounts to 8 nm, $d/\lambda_{\text{bulk Al min}} = 1.9$ and the signal is dominantly transmitted.

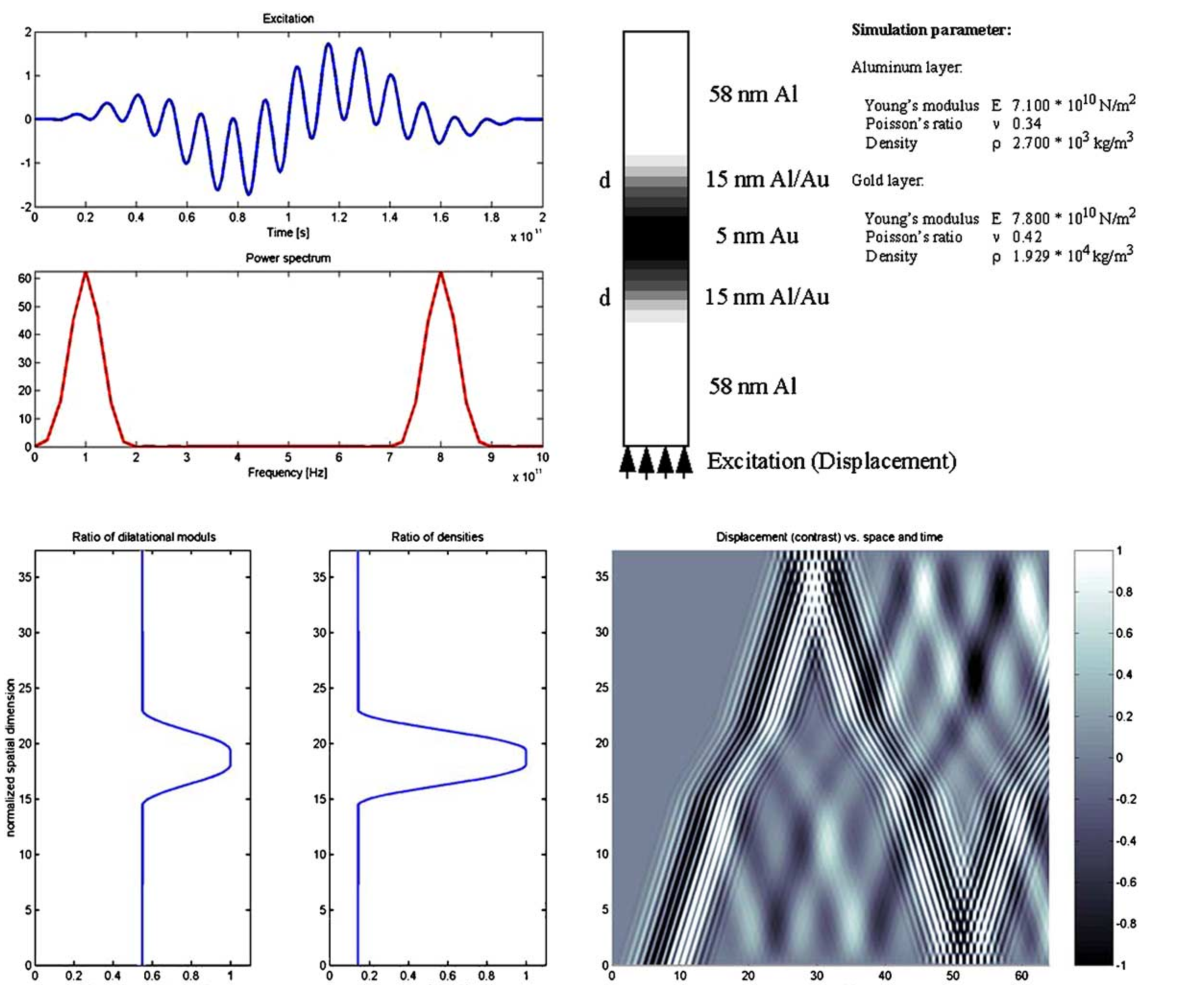


Fig. 1. Numerical simulation of the one dimensional elastic bulk wave propagation perpendicular to the surface in a Al/Au/Al thin film multilayer with ‘soft acoustic interfaces’ of thickness $d$. The structure is excited with a two frequency (0.1 and 0.8 THz) signal, multiplied by a hanning window at the surface. The thickness $d$ of the ‘soft acoustic interface’ amounts to 15 nm and lies between the two dominant wavelengths, thus allowing or impeding the transmission.

However, even for the 'long' waves the force reflection coefficient $R_f$ (see Eq. (1)) is given by the material combination and cannot be exceeded.

$$R_f = \frac{Z_{\mathrm{Au}} - Z_{\mathrm{Al}}}{Z_{\mathrm{Al}} - Z_{\mathrm{Au}}} \tag{1}$$
$$Z = c_p \rho$$

In the case of an ideal aluminum/gold interface of infinite extent, its value is 0.57, indicating that 57% of an initial stress amplitude propagating in the aluminum layer will be heading back after the reflection at the aluminum/gold interface. So in order to separate the signal by means of space or time it has to pass 'soft acoustic interface layers' several times. In the diagram on the right hand side of Fig. 1 one can see, that the more times the pulse is reflected to-and-fro between the two free ends, the more is the lower frequency component concentrated in the outer layers, whereas the high frequency component passes through all layers.

## 2. Numerical simulation of bulk waves in inhomogeneous solids

In this section the bulk wave propagation in an elastic continuum with a spatially dependent sound velocity is described. In the case of a linear elastic homogeneous continuum the one dimensional propagation of bulk waves is described by Eq. (2).

$$(\lambda + 2\mu)\frac{\partial^2 u_x}{\partial x^2} = \rho \frac{\partial^2 u_x}{\partial t^2} \tag{2}$$

In which $\lambda$ and $\mu$ denote the Lamé constants and $u$ denotes the displacement. In this case the speed of sound is constant; Eq. (3):

$$c_p = \sqrt{\frac{\lambda + 2\mu}{\rho}} \tag{3}$$

In an inhomogeneous material, however, the dilatational modulus and the density become functions of the spatial variable $x$, i.e. $(\lambda + 2\mu) := M(x)$ and $\rho := \rho(x)$. Eq. (2) needs to be rewritten and can no longer be solved by an approach which separates time- and space harmonic components.

$$\frac{\partial}{\partial x} M(x) \frac{\partial u_x}{\partial x} + M(x)\frac{\partial^2 u_x}{\partial x^2} = \rho(x)\frac{\partial^2 u_x}{\partial t^2} \tag{4}$$

Eq. (4) can be solved numerically only by replacing the derivatives with central finite differences as shown in Eq. (5):

$$\left(\frac{M_{j+1} - M_{j-1}}{2\Delta x}\right)\left(\frac{{}^{n}u_{j+1} - {}^{n}u_{j-1}}{2\Delta x}\right) + M_j\left(\frac{{}^{n}u_{j+1} - 2\,{}^{n}u_j + {}^{n}u_{j-1}}{\Delta x^2}\right)$$
$$= \rho_j\left(\frac{{}^{n+1}u_j - 2\,{}^{n}u_j + {}^{n-1}u_j}{\Delta t^2}\right) \tag{5}$$

In which $\Delta x$, $\Delta t$ represent the spatial and the time increment, respectively. $j$ denotes the spatial index and $n$ denotes the time index. Eq. (5) can be rearranged and the time boundary problem can be solved numerically.

## 3. Micro-mechanical realization of soft acoustic interfaces

Whereas the numerical example presented in the introduction follows some kind of an 'if we could' approach, the sections below deal with the micro-mechanical realization on the one hand and with quantitative in-depth profiling methods on the other hand. The material combinations and the dimensions of the specimen are chosen in accordance with the parameters for an optimal detectability of a short pulse laser acoustic set-up which was developed at the Center of Mechanics, ETH Zürich. The light-matter interaction of this set-up is optimized for aluminum thin films and therefore at least the surface layer needs to be aluminum. The set-up is described in Section 4.1.

Several methods and techniques have been tested and compared in order to obtain a more or less defined continuous transition of the mechanical properties of one layer to the properties of the neighboring layer within some 10 nm. Among the methods are an electron beam vapor deposition chamber with two beams and targets which can individually be controlled, an alternating sequence of various layers of different thicknesses, ion beam implantation, and thermally induced diffusion. For the problem outlined above, thermally induced diffusion turned out to be the best solution since this method allowed the realization of a series of varying thicknesses d by varying the temperature. A standard specimen consisting of a vapor deposited 30 nm Au layer embedded between two 60 nm Al layers on a $Al_2O_3$ substrate is chosen. The Al/Au combination has the advantages of a strong acoustic contrast ($R_f = 57\%$) and high diffusion tendency of the gold atoms.

A series of specimens has been exposed to 300/200/100 °C in a vacuum oven during 30 min (30′ temperature rise; 30′ to keep the temperature constant, 30′ cooling to room temperature). In parallel, a series of reference specimens consisting of 150 nm Al films on $Al_2O_3$ substrates has been exposed to the same temperature cycles in order to exclude potential effects of the grain structure or the optical surface reflectivity on the photo acoustic detection.

## 4. In-depth profiling methods

The majority of the measurements for the analysis of the acoustic properties of interfaces between neighboring metallic layers presented in this investigation, has been carried out on a short pulse laser acoustic set-up.

For verification purposes and in order to receive more detailed information about the buried gold layer and its diffusion into the aluminum layers, RBS measurements have been performed on the series of multilayers which has been exposed to the different temperature histories.

Glow discharge optical emission spectroscopy (GDOES), shall be briefly described here as a further method for in-depth profiling. However, the equipment available to the authors could not fulfill the resolution demands of the specimen presented above.

### 4.1. Short pulse laser acoustics

The laser acoustic method presented below, works in the thermoelastic region which means that there is no ablation and therefore the technique is nondestructive. A short laser pulse (800 nm, 70 fs), the pump pulse, is absorbed at the metallic thin film surface and is initiating an elastic pulse which propagates into the film. Echoes, occurring at discontinuities of the acoustic impedances, are heading back to the surface and are causing a slight temporary modification of the optical reflectivity. The optical reflectivity at the surface is scanned versus the relative time shift with respect to the initial pump pulse with a probe pulse, which is created by a partly reflecting mirror and which follows a different path in order to allow the control of the time shift between the excitational and the detecting pulse. The experiment is repeated at a repetition rate of 81 MHz while the time shift is changed.

The method was first presented by Thomsen et al. [1,2]. A detailed analysis of the light-matter interaction for the excitation as well as for the photo acoustic detection is given by Profunser et al. [3]. As visible on Fig. 2 several additional features apart from the laser source, the beam splitter, and the variable delay line are necessary. The main challenge lies in the reduction of optical and electronical cross-talk between the excitational and the detecting signal path. Therefore two modulation frequencies, cross-polarization, and balanced photo detection is used. A more detailed description of the set-up and of the signal processing is given by Vollmann et al. [4].

The diagram of Fig. 3 shows two independently measured reflectivity curves compared with a simulation (top). The thicknesses of the multilayer are: 60 nm Al/30 nm Au (80%)/60 nm Al on a $Al_2O_3$ substrate.

Each curve represents an average of 50 individually measured curves. The parameters used for the numerical simulation of Fig. 3 of the embedded gold layer are determined by Rutherford Backscattering Spectroscopy measurements which are presented in the following section. For lack of a more sophisticated approach, the mechanical properties like the dilatational modulus and the density of the 'diffusion alloy' consisting of 80% Au and 20% Al, are calculated by a linear interpolation of the values of the corresponding components. Most likely the intermetallic diffusion occurred during the manufacturing process.

Two effects are governing the shape of a reflectivity change curve versus the time shift between the pump pulse and the probe pulse, which are presented in Fig. 3: The dominant effect is the initial jump of the reflectivity change caused by the local heating at the surface. Proportionally to the heat conduction into the surrounding area, the reflectivity change decays nearly exponentially. This is due to the fact, that the heating pump pulse is much shorter than the thermal relaxation time. Superimposed to this thermally caused effect, one can see the (more or less) periodic alternation of the optical reflectivity change which is caused by the stress pulse echoes reaching the surface. The polarities of two consecutive echoes are inverted, which indicates that the acoustic

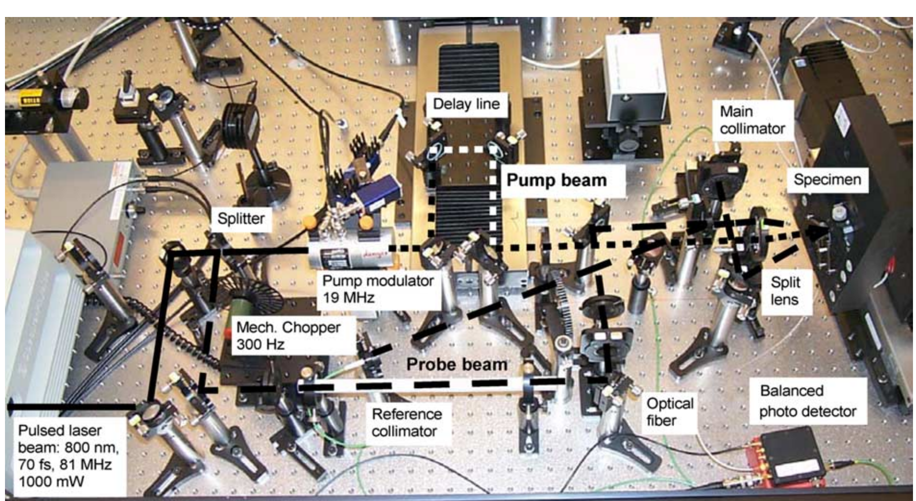


Fig. 2. Experimental set-up as used for the photo acoustic thin film profiling.

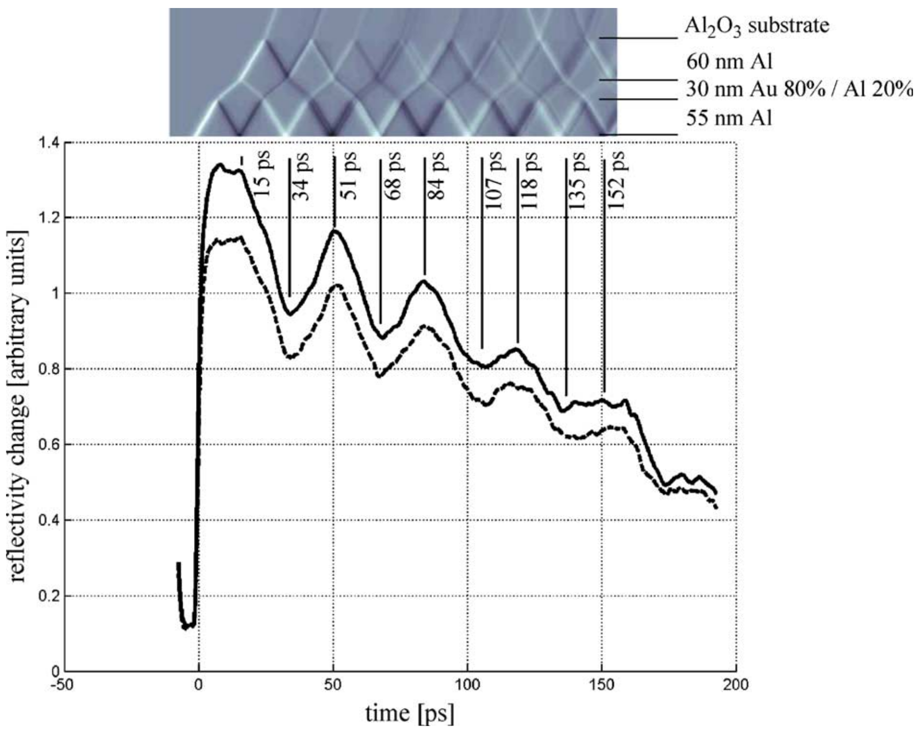


Fig. 3. Two photo acoustic measurements of a 60 nm Al/30 nm Au/60 nm Al multilayer on a $Al_2O_3$ substrate compared with a numerical simulation.

impedance of the embedded layer (i.e. the gold layer) is higher than the one of the surrounding layers. Thus a compressive stress pulse is converted into a tensile stress pulse and vice versa, whenever it reaches the free surface.

In contrast to the numerical example presented in the introduction (see Fig. 1), an acoustic pulse, which consists of two frequencies only cannot be realized with the present photo acoustic set-up (Fig. 2). The strain pulse, created by a pump laser pulse for the parameters used in the photo acoustic set-up, is calculated according to Profunser et al. [3] and shown in Fig. 4.

So in order to demonstrate the dependence of the reflection/transmission ratio on $\lambda/d$ experimentally, the thickness of the 'soft acoustic interface' $d$ needs to be varied. This has been realized by the exposure of a series of equally manufactured specimen to different temperature treatments, which leads to different grades of thermally induced diffusion.

Fig. 5 shows the photo acoustic measurements of three standard specimen (60 nm Al/30 nm Au/60 nm Al on $Al_2O_3$) which were heated up to 100/200/300 °C in a vacuum oven, in comparison with the measurement of the untreated specimen previously shown in Fig. 3. For better visibility the curves are shifted in the vertical axis. Each curve represents an average of 50 measurements.

The measured reflectivity change curves of the thermally treated specimen (Fig. 5) clearly indicate that the broadening of the diffusion zone i.e. the broadening of the 'soft acoustic interface' suppresses the acoustic

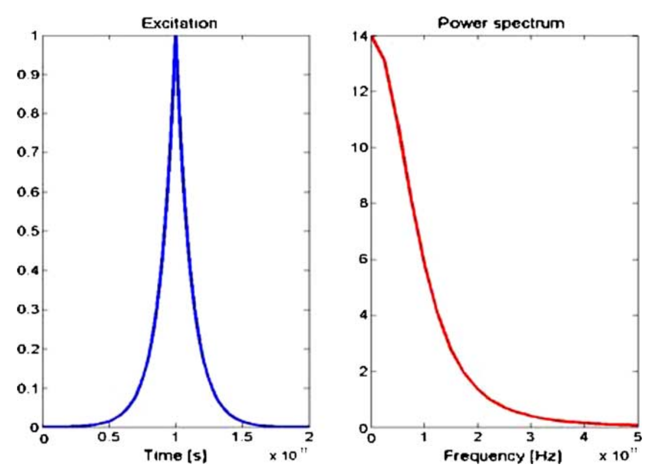


Fig. 4. Excitation (displacement) as used for the numerical simulation of Fig. 3.

contrast. One can also see, that the echo which occurs at the Al/$Al_2O_3$ substrate interface remains detectable even after the thermal treatment of 300 °C.

The inverse problem, i.e. the quantification of the thickness of the 'soft acoustic interface' as a function of the amplitude decay of the echoes, has not been performed yet.

### 4.2. Rutherford backscattering spectroscopy

In order to verify the hypothesis of a thermally induced intermetallic diffusion, Rutherford backscattering

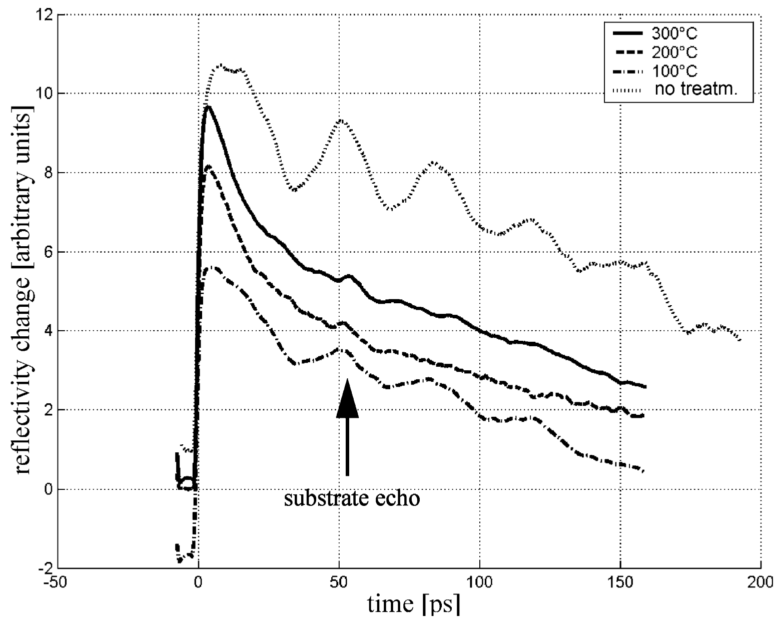


Fig. 5. Photo acoustic measurements of four 60 nm Al/30 nm Au/60 nm Al multilayers on a $Al_2O_3$ substrate which were exposed to different temperatures in order to 'smoothen' the acoustic interfaces by thermally induced intermetallic diffusion.

spectrometry (RBS) measurements are performed at the PSI/ETH Laboratory of Ion Beam Physics, Zürich.

RBS relies on the fact that the energy of a MeV ion backscattered from a target material is a function of the target atomic mass and the depth at which the scattering took place. A detailed description of the method is given by Chu et al. [5]. Therefore, quantitative depth profiles of the elemental composition of the target can be obtained. The method is nondestructive and a depth resolution of 2 nm can be obtained in the near surface region.

Fig. 6 shows the Al and Au atom concentrations versus depth of a standard specimen in the untreated case and after a 300 °C exposure. It turns out, that even for the untreated specimen an Au concentration of 100% is not achieved in the middle layer. This might be caused by diffusion processes occurring during the manufacturing, by impurities in the vacuum chamber, or by the fact that the surface and interface is not perfectly plane within the region to be analyzed. The profiles presented in Fig. 6 are used for the numerical simulation of the bulk wave propagation shown in Fig. 4 and correspond very well with the photo acoustic measurements.

### 4.3. *Glow discharge optical emission spectroscopy (GDOES)*

Seeking for further, independent, in-depth profiling metrology, the standard specimen has also been analyzed by GDOES. An improved version of this method

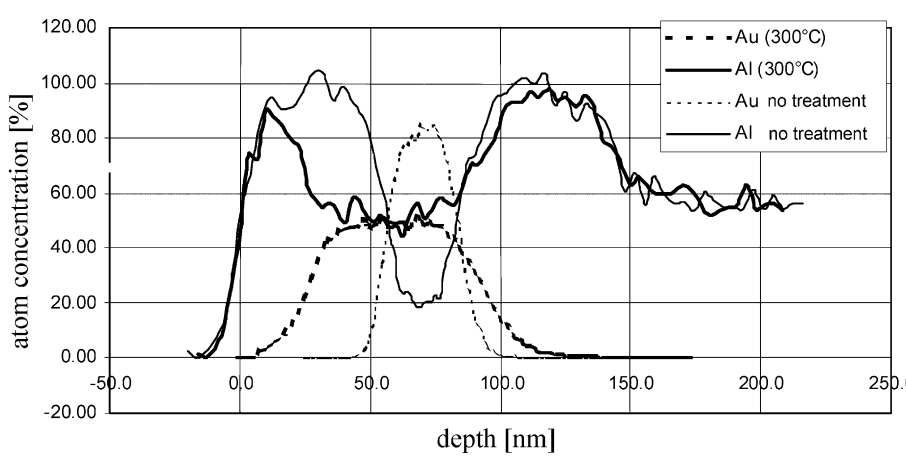


Fig. 6. Atom concentration versus depth calculated from RBS measurements for an untreated and a 300 °C treated standard specimen (60 nm Al/30 nm Au/60 nm Al multilayers on a $Al_2O_3$ substrate).

is presented by Payling et al. [6]. However, the resolution demands of the present specimen was to high for the equipment available.

## 5. Conclusions, outlook, and potential applications

Frequency dependent reflection/transmission phenomena of bulk waves propagating perpendicular to the surface of metallic thin film multilayers are presented based on a numerical simulation and on photo acoustic measurements. Steps towards a quantitative, nondestructive, photo acoustic determination of intermetallic diffusion effects at a resolution of few nanometers have been demonstrated. The hypothesis, that the acoustic interface between neighboring Au/Al layers can be 'smoothed' by thermally induced diffusion processes, has been reinforced by quantitative Rutherford backscattering spectroscopy measurements, by photo acoustic experiments, and by numerical simulations.

Future directions of the on-going research project are the extension to two dimensional wave propagation phenomena occurring at 'soft acoustic interfaces', the improvement of the resolution of the photo acoustic experiments, and the realization of prototypes of engineering applications like micro-mechanical band pass signal filters and spectrum analyzers.

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