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Elastic Microanalysis at Lower Temperatures

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Abstract. Acoustic microscopy is wellknown nowadays as a powerful non-destructive tool to investigate the local elastic and structural properties of compact materials, composites and films. However, the application of commercial microscopes is limited to temperatures near ambient, because aqua dest. as a quite suitable immersion fluid is usually preferred. Here we report on our recent investigations, which are governed by the aim to extend elastic microanalysis to temperatures well below room temperature. All measurements are done between 90K and 230K by use of liquid propane as acoustic bonding medium. It is shown that elastic microanalysis at 1GHz can be successfully performed but is considerably affected by the strong temperature dependent properties of the coupling fluid chosen.

1. INTRODUCTION

A quite promising practical application of ultrasonic microscopy benefits from the possibility to analyse the elastic behaviour of materials under investigation also in a quantitative manner. Such measurements of elasticity can be performed with an acoustic microscope rather locally at very high sound frequency by means of the so-called *V(z)-method* [1]. Essentially this method is based on sound velocity measurements of Rayleigh-type sound waves, which can be excited on the sample surface within a small region of only several 10 μm in diameter. In this sense the information received is named *local* and justifies the term *elastic microanalysis* often used.

2. EXPERIMENTAL

2.1 Low Temperature Chamber

A low temperature chamber has been recently developed in our lab, which especially allows microscopic investigations by means of ultrasound over a broad range well below room temperature [2] and, above all, is compatible with a commercial acoustic microscope (ELSAM / Leitz). The temperature is monitored with a platinum sensor and can be stabilized within 0.1 K by means of an electrical heater. Gases after become liquid are more or less suited to serve as acoustic coupling fluid at temperatures below ambient and must be supplied from outside as well as the cooling agent, for which liquid nitrogen is preferred.

2.2 Coupling Fluid

For all measurements presented in this paper, propane of quite high purity (99.95%) is used as bonding medium, which under ambient pressure become liquid between 83K and 231K. Its suitability as an acoustic coupling fluid depends much on its viscoelastic properties and their dependence on temperature and sound frequency in particular. Both sound velocity and dynamic viscosity are important parameters characterizing the acoustic transmission properties of a fluid. The sound velocity c_L determines the resolution obtainable whereas the viscosity η causes the sound attenuation α in a fluid and limits the signal-to-noise ratio, which can be achieved with an acoustic microscope at a given frequency ω .

Shown in Fig.1 (left) is the attenuation factor $\alpha / \omega^2 \propto \eta$ experimentally obtained at 1 GHz as described by us earlier [2] in more detail and the sound velocity (right) as a function of temperature. Obviously, both are strongly temperature dependent and optimal conditions are not achieved for the entire liquid range. In

comparison with room temperature measurements by means of aqua dest., however, the signal-to-noise ratio is quite high for a temperature interval of at least 100 K (shown hatched in Fig.1).

Figure 1: Ultrasonic attenuation factor (left) determined at 1GHz and sound velocity (right) of liquid propane as a function of temperature at ambient pressure. The temperature range where the signal-to-noise ratio is comparable with or better than that achieved at room temperature by means of aqua dest. is shown hatched.

3. RESULTS

3.1 Velocity of Surface Waves

Numerous examples given in literature confirm the practical use of the $V(z)$ -method at room temperature demonstrating its feasibility for a sound frequency up to more than 1GHz in principle [1]. On the other hand, only a few of such investigations at a temperature different from ambient and, to our knowledge, none temperature dependent with GHz-sound over a broader range are performed until now.

Considering the lack of experience, materials with well defined elastic properties are preferentially suited to verify how accurate temperature dependent velocity measurements can be done by means of our device. For this purpose single crystalline aluminum of high purity and a borsilicate glass of known specification are chosen for our experiments. The surfaces of the aluminum crystals are oriented along various axis and are electrochemically polished in particular. The surface roughness of the samples used is lower than about 20 nm at least and should be therefore of minor importance in all cases. For two selected specimen the sound velocity data obtained microscopically are shown in Fig.2 as an example.

Figure 2: Temperature dependent surface wave velocity of a borsilicate glass (left) and an [111]-oriented aluminum single crystal (right) obtained microscopically at 1GHz by use of liquid propane as coupling fluid. Additionally shown by a straight line is the expected temperature dependence of the velocity of the free Rayleigh-wave.

Bearing in mind that during microscope operation the samples under investigation are always covered with a coupling fluid, their surfaces are not free but are, strictly speaking, interfaces between a solid and the fluid chosen. Hence, any motion of the solid surface is more or less affected by the liquid loading. In consequence, surface waves excited microscopically are *leaky* sound waves and their dispersion differs somewhat from that of *free* Rayleigh-waves. Generally, the leaky wave is expected to be a little faster than the respective Rayleigh-wave [3].

Taking furthermore into account the temperature dependent density and sound velocity of liquid propane, the velocity of the leaky wave in our case should become increasingly higher in comparison with the Rayleigh-velocity when the temperature goes down. The findings of Fig.3 are in fact consistent with the theoretical expectation but quantitatively the agreement is not quite satisfying, because the increase of the velocity experimentally obtained is significantly larger than that calculated. Thus a certain discrepancy still remains due to reasons not yet completely clarified but originates most probably in the *non-ideal* character of the fluid. In particular, it is believed that the disagreement results from the finite dynamic shear elasticity of liquid propane which has been neglected in the theoretical description so far.

Ideal fluids are characterized by the absence of any shear stress. In *real* fluids, however, shear stresses usually appear as a result of the finite shear viscosity especially at higher frequency. Consequently, shear elasticity cannot be neglected in general, and at a liquid-solid boundary the lateral motion of a solid surface will be additionally influenced by the occurrence of viscous shear stress in the fluid.

Evidently, a finite shear elasticity also allows the excitation of shear waves. But shear waves in real fluids are of diffusive type, because the stress components σ_{ij} are proportional to the time derivative of the shear strains - and not to the strains themselves - i.e.

$$\sigma_{ij} = \eta_s \left(\partial \varepsilon_{ij} / \partial t \right) \quad \text{and} \quad i \neq j. \quad (1)$$

As is wellknown, the shear viscosity η_s usually dominates the absorption of longitudinal sound waves in fluids. Therefore it is commonly justified to approximate

$$\eta_s \approx \eta \propto \rho c_L^3 \cdot (\alpha_L / \omega^2). \quad (2)$$

Assuming plan waves, the equation of motion then yields a dispersion relation of the type

$$c_s \propto \sqrt{\omega \cdot \eta}, \quad (3)$$

i.e. the propagation of viscous shear waves is strongly dispersive and their velocity depends much on the sound frequency excited. Viscous waves are strongly damped by nature and from the proportionality (3) it becomes immediately clear that their excitation is generally favoured at higher frequency by the increasing acoustic impedance ρc_s . This is the exact reason, why the shear viscosity of liquid propane should be responsible by part for another phenomenon unexpected, which is subject of the following section and the particular cause for the striking restriction in temperature of the aluminum data in Fig.2.

3.2 Excitation of Surface Waves

On cooling the samples, not only the dispersion of the leaky waves but also their intensity excited at constant incident power shows a pronounced temperature dependence. Qualitatively, it can be established that the latter always tends to decrease and it seems that this phenomenon is only little dependent on the respective sample investigated. The collection of $V(z)$ -curves presented in Fig.3 (left) is exemplary and shows what happens when the temperature decreases. Below about 150 K the intensity of the surface wave becomes rapidly smaller although its attenuation, i.e. the decrease in amplitude per period, has only moderately changed. The temperature dependence of the surface wave intensity normalized to its high temperature level is shown for two examples at the bottom of Fig.3 (right).

Lens aperture and refraction index are functions of temperature due to their dependence on the sound velocity in the fluid. Recalling this, one might suspect that the observed intensity decrease simply results from the fact that the angle of surface wave excitation steadily increases on cooling and will finally leave the lens aperture. However, as can be easily verified (see Fig.3 at the top) this explanation has to be dropped. Nevertheless, the comparison of quite different materials clearly points out that the efficiency loss of surface wave generation always intensifies for an excitation angle anywhere between 30° and 35°. In other words, this phenomenon depends primarily on the inclination angle of the incident sound wave, suggesting that a more accurate treatment of sound reflection at liquid-solid boundaries is required.

Regarding inclined incidence onto such boundaries, a longitudinal sound wave obviously possesses a non-vanishing velocity component parallel to them. Consequently, a large gradient of the tangential component may arise in the liquid boundary layer, because it seems reasonable to assume that at the boundary surface itself the fluid totally adheres to the solid. Viscous shear stresses which simultaneously occur then may lead to a large energy dissipation in the fluid just at the boundary.

However, this interpretation is based on a condition which is opposite to that what is often assumed for convenience - namely that a fluid can move *freely* parallel to a boundary and normal velocity components

Figure 3: V(z)-curves of borsilicate glass (left) obtained microscopically at 1GHz and temperatures as indicated. Shown on the right are the temperature dependence of the angle of aperture and surface wave excitation (at the top) in liquid propane as well as the surface wave intensity (at the bottom) normalized to the level at higher temperature.

need to be considered only. In contrast to this, a more realistic treatment of sound wave reflection at liquid-solid boundaries has to include the possible generation of viscous shear waves in the fluid as well. The theoretical description is, of course, a little more elaborate and depends much on the boundary conditions chosen (see for example [4], p.91f). But let us assume for simplicity that the solid can be fairly represented by a rigid wall. Calculating the transformation coefficient of the incident longitudinal into a reflected transverse wave in the liquid medium then yields

$$\nu_{LT} \propto \frac{(c_s/c_L) \sin \theta}{\sqrt{1 - (c_s/c_L)^2 \sin^2 \theta} + (c_s/c_L) \sin \theta \tan \theta} \xrightarrow{c_L \gg c_s} (c_s/c_L) \sin \theta, \quad (4)$$

where the approximation on the right is totally sufficient in our case since c_s is of the order of 100 m/s and thus significantly smaller than c_L . The incident angle θ relevant to our investigations is the excitation angle of surface waves θ_{sw} for which $\sin \theta_{sw} = c_L/c_{sw}$ is given by Snells law. Neglecting furthermore the rather weak variations of c_{sw} and of the attenuation factor α/ω^2 within the temperature region of interest then the acoustic power dissipated by the generation of viscous shear waves becomes

$$P_{LT} \propto |\nu_{LT}|^2 \propto \omega \cdot c_L^3. \quad (5)$$

Hence, the acoustic energy loss right at the excitation angle of surface waves is quite sensitive to changes of the longitudinal sound velocity c_L . Since the latter in liquid propane strongly increases with decreasing temperature, the generation of surface waves should become increasingly affected as well. In short, theory essentially predicts what is experimentally found.

Summarizing, elastic microanalysis at 1GHz, though successfully done here over a broader temperature range, suffers considerably from the temperature dependent properties of the coupling fluid chosen.

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