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Determination of elastic properties of surface layers and coatings by resonant ultrasound spectroscopy

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Abstract. This paper deals with determination of in-plane elastic constants of thin layers deposited on substrates. Modified resonant ultrasound spectroscopy is used to measure resonant spectra before and after layer deposition. These two spectra are compared and changes in the position of the resonant peaks are associated with layer properties. It is shown that for thin layers either the elastic moduli or the surface mass density can be determined, providing the complementary information (the surface mass density for the determination of the moduli, the elastic moduli for the determination of the surface mass density) is known. As an experimental demonstration of this approach, the elastic moduli of diamond-like-carbon film deposited on a silicon substrate and the surface mass density of a thin spray paint on a silicon substrate are determined.

1. Introduction
Resonant ultrasound spectroscopy (RUS) is one of the most precise methods for experimental determination of the elastic properties of solids [1]. It has been shown recently that this method can also be used for determination of the elastic constants of thin layers [2] and [3]. In this article we present a novel approach to this technique with simplified problem modeling, and verify this approach by application to two distinctly different systems layer-substrate.

2. Method description
The RUS method for thin layers uses comparative measurements of the free vibration spectra of the same substrate with and without the layer, as shown schematically in figure 1(a). It is assumed that the measured frequency shifts of the individual resonant peaks are given by the properties of the layer which is added (or removed).

Generally, the frequency shifts depend on many factors but most important are the stiffness of the layer, its density and thickness. We can write the forward problem mathematically as

\[ \mathbf{F}(\mathbf{Q}, \rho, h, \text{geometry}) = \Delta \omega \]  \hspace{1cm} (1)

where \( \mathbf{Q} \) is the vector of in-plane layer elastic constants (plane stress is assumed), \( \rho \) and \( h \) are the layer density and thickness and \( \Delta \omega \) is the vector of angular frequency shifts. Function \( \mathbf{F} \) cannot be constructed analytically (with exception of some simple 1D geometries) but it can
be evaluated numerically by different methods. We use the Ritz method described in [4]. For thin layers, the problem given by Eq.1 can be approximated by the linear equation

\[ AQ - M\rho h = \Delta \omega \]  

where the matrix \( A \) describes the effect of the layer stiffness and the matrix \( M \) effects of the layer mass on frequency shifts \( \Delta \omega \). Both effects are multiplied by stiffness coefficients \( Q \) and the surface density \( \rho h \) of the layer. Construction of all the matrices is specified in [4]. From equation 2 we clearly see that there are two competing factors that change the resonances when the layer is added. Stiffness represented by the first term in equation 2 always tends to increase the resonant frequencies (sample gets more stiffer) and mass represented by the second term in equation 2 always shifts the resonant frequencies down. This is not surprising, though, but we must always keep in mind that in order to solve equation 2 for \( Q \) the mass density must be known.

Based on solution of the inverse problem defined by equation 2, we can distinguish two application approaches:

- **Stiffness determination**: The mass of the layer is known or it is negligible. In most cases, we cannot neglect the mass, especially if we are working with high frequencies. However, the mass density of the layer is not easy to measure when we are dealing with submicron thin films. Fortunately, the mass density of the layer can be usually estimated from bulk density measurement.

- **Mass determination**: The stiffness of the layer is known or negligible. Stiffness can be neglected, only if the material of the substrate is incomparably stiffer than the material of the layer. If deal with dust or paint particles and polymer layers we can assume \( Q \rightarrow 0 \) and calculate surface mass density from equation 2.

Both approaches are verified experimentally later in this paper.
3. Experimental setup
As it was described earlier, the key part of the RUS method is the measurement of the the spectra of the free-vibrating specimen. In order to fulfill the free boundary condition, the specimen was supported on one side by a thin optical fiber and the opposite corner was laid on a piezoelectric film (PVDF) as shown in figure 1(b). The mechanical excitation of the sample was made by the piezoelectric membrane though this corner. For point detection of the dynamical response the Microsystem Analyser Polytec MSA-500 with the laser interferometer embedded into the optical microscope was used. This Doppler vibrometer was equipped by a piezo scanner, able to measure vibration at different points of the smooth surface of the sample. By repeating excitation and vibration measurement we can get resonant spectra at each scanned point of a predetermined mesh on the specimen surface and compose the shape of vibrational eigenmodes.

Samples were plate-like parallelepipeds with the measured layer on one of their faces. Typically, their dimensions were 4.5x3.5x0.3 mm$^3$. Substrates discussed in this paper were made of silicon single crystals that exhibit very good resonant properties. From experimental point of view there were several limitations which must be considered in order to have good measurements:

- **Damping:** Both the substrate and the layer should have as low damping as possible. High damping attenuates the free vibration and makes the spectral peaks less visible.
- **Thickness:** Dimensions and geometry of both the substrate and the layer should be known, most important is the reliable determination of the layer thickness.
- **Reflectivity:** The sample must have (before and after deposition) at least one reflective surface in order to use the reflected beam for laser interferometric vibration measurement.
- **Damage:** Any damage that the sample undergoes during manipulation changes the resonant spectra and hampers the measurement. Some materials get change properties during layer deposition as well (for example if it is done at high temperature or aggressive ambiance).

4. Measurements

4.1. Stiffness measurements
were performed on diamond-like-carbon (DLC) layer on Si substrate (DLC prepared by physical vapor deposition). Silicon substrate was measured before layer deposition and then remeasured. Thickness of the layer was 375 nm (determined by AFM step) and the mass density was taken 3.5 g/cm$^3$ from bulk measurements. Layer was considered isotropic, the resulting elastic constants were: Young’s modulus $E = 581 \pm 95$ GPa and bulk modulus $K = 418 \pm 547$ GPa (the insensitivity of the resonant spectra to this coefficient is a common feature of the RUS measurement).

4.2. Mass measurements
were tested on different kinds of commercial undercoat paint sprays, were deposited upon the silicon substrates. Simultaneously, the mass of the paint was measured on static microbalance (accuracy 1 mg) using 64 cm$^2$ piece of paper with and without spray to determine change of the mass due to paint. In this way, mass measured by static and dynamic means could be independently compared and dynamic experiments verified. Static surface density was determined as $\rho_h=16.3 \pm 0.33 \mu g/mm^2$ while the dynamic method gave $\rho_h=16.2 \pm 0.39 \mu g/mm^2$. The paint thickness was unknown, from density considerations it was estimated around 5 microns.

In figure 2, the good agreement of the linear model given by equation 2 with the measured data can be seen.
5. Conclusions

Both the stiffness and the mass measurements show good accordance with the proposed linear inverse model. Moreover, the mass measurement was validated independently by static measurement using the microbalance. The main advantage of the proposed inverse modeling is that the relations between the properties of the layer and the frequency shifts are represented by linear equations, which can be easily solved and enable us to estimate the accuracy of the measurement. There are obviously some experimental constrains. Especially, the layer thickness must be known for the stiffness measurement (for the mass measurement it is not necessary).

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