FINAL REPORT

DOE/ER/13484--001

for

THIN-FILM CHARACTERIZATION AND FLAW DETECTION

(DE FG02-86ER 13484)

prepared by

J. D. Achenbach
Center for Quality Engineering and Failure Prevention
Northwestern University
Evanston, IL 60208

Submitted to

Office of Basic Energy Sciences
U.S. Department of Energy
Washington, D.C. 20545

Reporting Period:	February 1, 1993 - November 31, 1997
Amount of unexpended funds:	none

Principal Investigator: J. D. Achenbach
Phone 847-491-5527
Fax 847-491-5227
e-mail achenbach@nwu.edu

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

MASTER

February 25, 1998

DISCLAIMER

CONTENTS

1. Project Overview

A. Specific Project Objectives:

The objectives were to determine the elastic constants of thin films deposited on substrates, to measure residual stress and to detect and characterize defects in thin film substrate configurations.

There are many present and potential applications of configurations consisting of a thin film deposited on a substrate. Thin films that are deposited to improve the hardness and/or the thermal properties of surfaces were of principal interest in this work. Thin film technology does, however, also include high T_c superconductor films, films for magnetic recording, superlattices and films for band-gap engineering and quantum devices. The studies that were carried out on this project also have relevance to these applications.

Both the film and the substrate are generally anisotropic. A line-focus acoustic microscope has been used to measure the speed of surface acoustic waves (SAW) in the thin film/substrate system. This microscope has unique advantages for measurements in anisotropic media. Analytical and numerical techniques have been employed to extract the desired information on the thin film from the measured SAW data. Results include: (1) analytical and numerical techniques for the direct problem and for inverse methods, (2) measurements of homogeneous and superlattice film constants, (3) investigation of the effect of surface roughness and (4) measurements of residual stresses.

B. Relation of this project to the DOE mission.

Thin films enter in numerous energy producing systems, generally as thin coatings to protect surfaces of components from wear, impact, corrosion and thermal disturbances. It is important that the elastic properties of such films can be determined accurately, that residual stresses can be measured, and that defects such as delaminations can be detected and characterized. The proper functioning of thin films depends on these capabilities. Since the reliability and life extension of energy producing systems is part of the DOE mission, the work on thin films that was carried out on this project directly fits into that mission.

2. Scientific and Technical Content

A. Schedule of major research activities.

Feb.1/93 - Jan.31/94: Theoretical measurement model of V(z) curve for superlattice thin films, plus related measurements by line-focus acoustic microscopy. Comparison of theoretical and experimental results. Start of measurements of stresses by acoustic microscopy by the use of a stressing stage under the acoustic microscope. Preparation of an invited review article to be published in Advances in Acoustic Microscopy (edited by G.A.D. Briggs).

Feb.1/94-Jan.31/95: Theoretical and experimental work for characterization of residual stresses and determination of elastic constants.

Feb.1/95-Jan.31/96: Theoretical and experimental work for characterization of defects of thin films made of superlattice materials.

Feb.1/96-Jan.31/97: Use of time-resolved line-focus acoustic microscopy.

Feb.1/97-Nov.31/97: Completion of papers. Final report.

B. Scientific and technical issues.

The scientific and technical issues that have been addressed may be summarized as: (1) the measurement of elastic constants of anisotropic thin films deposited on anisotropic substrates (2) the measurement of residual stresses, and (3) the detection and characterization of flaws in thin films.

C. Experimental and theoretical approach taken.

Acoustic microscopy has been used for the characterization of a thin film deposited on an elastic substrate. The technique is based on the measurement of the V(z) curve. The V(z) curve is a record of the transducer voltage output (V) with the variation of the distance (z) between the acoustic lens and the specimen. The SAW (surface acoustic wave~~s~~) velocity can be obtained from the periodic variation of the V(z) curve. The line-focus acoustic microscope allows the measurement of SAW velocity in a single prescribed direction and hence it can be used to measure the near-surface anisotropy of elastic materials. The instrument has been used to measure the anisotropic dependence of SAW velocities on the propagation directions on single crystals, and single-crystals and superlattice thin-films grown on single-crystal substrates.

In the work completed on this project a theoretical measurement model of the V(z) effect has been developed by following the Fourier optical approach, in which the V(z) curve is considered as a Fourier integral over the product of characteristic functions of the acoustic lens and the reflectance function of the fluid-loaded specimen. When the characteristic functions have been determined, the V(z) curve can be directly related to these functions and to the reflectance function of the specimen through a Fourier-type integral. Accurate characteristic functions are essential for the calculation of V(z) curves by this approach. Attenuation in the coupling fluid, the angular dependence of the transmission by the antireflection coating on the lens surface and the actual focal length must be carefully taken into consideration. The V(z) measurement model has been validated by comparison of its results with measurements.

With known elastic constants and mass density of the substrate and known mass density of the superlattice, three independent elastic constants of homogeneous and superlattice thin films have been determined from the inversion of the experimental SAW data. The inversion procedure consists of seeking a set of the constants that minimizes the deviations between measured and calculated velocities of SAW's propagating at incremental angles in the films deposited on substrates. The iterative calculation for minimizing the sum is carried out by a systematic function minimization algorithm known as the Simplex method.

D. Importance of solving the problem.

Thin films to improve the surface properties of components will be applied very extensively now that the techniques to deposit such films are being perfected. The capability to predict the performance of thin films requires information on mechanical properties. The results of this project are important because they provide techniques to determine such thin film properties.

3. Project Output

A. Major accomplishments on this project.

V(z) curves for a line-focus acoustic microscope have been calculated in terms of the characteristic functions of the acoustic lens and the reflectance function of the fluid-loaded specimen. More accurate expressions for the characteristic functions of the acoustic lens have been developed by taking account of attenuation in the coupling fluid, of the angular dependence of transmission by the antireflection coating on the lens surface, and by making a better estimate of the focal length. The reflectance function has been calculated for anisotropic films deposited on anisotropic substrates. The calculated V(z) curves have been compared with measurements for isotropic and anisotropic materials, and layered anisotropic materials. For thin film/substrate configurations of known elastic properties, the surface acoustic wave velocities obtained from the theoretical and the measured V(z) curves have been compared for the full range of directions of wave propagation, and excellent agreement has been observed.

The measurement model for the V(z) effect has next been applied together with an inverse method based on the Simplex method to obtain unknown elastic constants of thin films from the measured speeds of Sezawa modes in thin-film substrate systems. Results have been obtained for homogeneous nitride films and transition-metal nitride superlattice films.

In other work, line-focus acoustic microscopy has been used to determine local near-surface stresses in isotropic materials. Two surface wave modes, namely a leaky Raleigh wave and a leaky surface-skimming longitudinal wave, were excited by the acoustic microscope. It has been observed that the changes of the wave velocities are linearly proportional to the applied stress, as predicted by acoustoelastic theory. The non-uniform stress field in a loaded specimen has been determined from wave velocity measurements by the use of acoustoelastic constants obtained from a calibration test. The measured stresses are in good agreement with the results calculated by the finite element method. A self-calibrating method, which determines the stress profile directly from velocity measurements without a calibration test, has been considered and the results have been compared with experimental data.

Furthermore, a method has been presented to determine not only the elastic constants but also the mass density of isotropic and anisotropic solids and anisotropic thin films. The velocity and attenuation of leaky surface acoustic waves (SAWs) have been obtained for specified propagation directions from V(z) curves measured by line-focus acoustic microscopy (LFAM). The experimentally obtained velocities have been compared with velocities obtained from a measurement model of the V(z) curve which simulates the experiment. Since the measured and simulated V(z) curves have the same systemic errors, the material constants are free of such errors. For an isotropic solid, Young's modulus E, the shear modulus G and the mass density ρ have been determined from the leaky Rayleigh wave velocity and attenuation, measured by LFAM, and a longitudinal wave velocity measured by a pulse-echo transit-time technique. For a cubic-crystalline sold, the ratios for the elastic constants to the mass density $(c_{11}/\rho, c_{12}/\rho, c_{44}/\rho)$ have been determined from the directional variation of measured SAW velocities, using a preliminary estimate of ρ. The mass density ρ has subsequently been determined by additionally using the attenuation of leaky SAWs in crystal symmetry directions For a cubic-crystalline thin film deposited on a substrate, the elastic constants and the mass density $(c_{11}/\rho, c_{12}/\rho, c_{44}/\rho)$ of the film have been determined from the directional variation of the measured SAW velocities, and a comparison of the corresponding attenuation coefficient with the measured attenuation coefficient has been used to verify the results.

The effective elastic constants of single-crystal nitride superlattice films have been determined by calculation and by measurement methods. The calculation method was based on formulas to calculate the effective elastic constants of superlattices from the measured elastic constants of the constituent layers. The calculated effective elastic constants have been tested by comparing the corresponding surface acoustic wave (SAW) velocities calculated for thin-film/substrate systems with the corresponding SAW velocities measured by line-focus acoustic microscopy (LFAM). The measurement method determines the effective elastic constants of the superlattices directly from the SAW velocity data measured by LFAM. Two kinds of superlattice films have been considered; one has relatively flat and sharp interfaces between layers, and the other has rough interfaces with interdiffusion. The calculation method has yielded very good results for the superlattices with flat and sharp interfaces but not for the superlattices with rough interfaces. The measurement method yields results for both kinds, with the restriction that the constituent layers have similar crystal symmetries.

Also, a multiple leaky acoustic wave method to determine elastic constants from a single V(z) measurement has been presented. The V(z) curves which include contributions from different leaky acoustic waves, measured using the line-focus acoustic microscope at 225 MHz, have been compared with theoretical results predicted by a V(z) measurement model. The determination of elastic constants has been achieved numerically by seeking a set of elastic constants that leads to the best fit, in the least square sense, of the theoretical results to the experimental ones. The method has been applied to isotropic materials in bulk, and plate and thin-film configurations. Elastic constants for each of these cases have been determined. The consistency, convergence, sensitivity, and accuracy of the procedure have been investigated.

Finally, elastic constants have been determined by using time-resolved line-focus acoustic microscopy (TRLFAM). Wave forms have been measured with a time-resolved line-focus acoustic microscopy system. A measurement model which simulates the measurements has been introduced for parametric studies of the waveforms. The determination of elastic constants was achieved by systematically comparing the relative wave-mode time-delays obtained from the measurement model and the experiments. The method has been applied to determine elastic constants of both isotropic and anisotropic materials.

B. Bibliography of publications emanating from this project since January 1, 1993.

1. (with J. Kim and Y-C Lee), "Measuring Thin-Film Elastic Constants by Line-Focus Acoustic Microscopy," Advances in Acoustic Microscopy, edited by Andrew Briggs, 1:153-208, 1995.

Papers in refereed journals:

2. (with Jin O. Kim, Paul B. Mirkarimi and Scott A. Barnett), "Acoustic-microscopy measurements of the elastic properties of TiN/V_xNB_{1-x})N superlattices," Physical Review B, The American Physical Society, Vol.48, pp. 1726-1737, 1993.

3. (with Yung-Chun Lee and Jin O. Kim), "V(z) curves of layered anisotropic materials for the line-focus acoustic microscope," J. Acoust. Soc. Am. 94, pp. 923-930, 1993.

4. (with Yung-Chun Lee and Jin O. Kim), "Measurement of stresses by line-focus acoustic microscopy," Ultrasonics 32, pp. 359-365, 1994.

5. (with Yung-Chun Lee and Jin O. Kim), "Acoustic Microscopy Measurement of Elastic Constants and Mass Density," IEEE Transactions on Ultrasonics, Ferroelectrics, and Frequency Control, 42, pp. 252-264, 1995.

6. (with Y.-C. Lee, M. J. Nystrom, S. R. Gilbert, B. A. Block and B. W. Wessels), "Line Focus Acoustic Microscopy Measurements of Nb_20_5Mg0) and $BaTi0_3/LaA10_3$ Thin-Film/Substrate Configurations," IEEE Transactions on Ultrasonics, Ferroelectrics, and Frequency Control, 42, pp. 376-380, 1995.

7. (with J. O. Kim, M. Shinn and S. Barnett), "Effective Elastic Constants of Superlattice Films Measured by Line-Focus Acoustic Microscopy," Journal of Engineering Materials and Technology, 117:395-399, October, 1995.

8. (with Wei Li), "V(z) Measurement of Multiple Leaky Wave Velocities for Elastic Constant Determination," J. Acoust. Soc. of America, 100(3):1529-1537, 1996.

9. (with Wei Li), "Determination of Elastic Constants by Time-Resolved Line-Focus Acoustic Microscopy," IEEE Trans. Ultrason. Ferroelec. Freq. Control, 44(3):681-687, 1997.