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Recent Advances in Scanning Acoustic Microscopy for Adhesion Evaluation of Thin Films

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Abstract As the thin film technology has emerged in various fields, adhesion of the film interface becomes an important issue in terms of the longevity and durability of thin film devices. Diverse nondestructive methods utilizing acoustic techniques have been developed to assess the interfacial integrity. As an effective technique based on the ultrasonic wave focusing and the surface acoustic wave(SAW) generation, scanning acoustic microscopy(SAM) has been investigated for adhesion evaluation. Visualization of film microstructures and quantification of adhesion weakness levels by SAW dispersion are the recent achievements of SAM. To overcome the limitations in the theoretical dispersion model only suitable for perfectly elastic and isotropic materials, a new model has been more recently developed in consideration of film anisotropy and viscoelasticity and applied to the adhesion evaluation of polymeric films fabricated on semiconductive wafers.

Keywords: Scanning Acoustic Microscopy; Thin Film Adhesion; Surface Acoustic Wave; Dispersion

1. Introduction

In recent years, the thin film technology has emerged in various fields including material, biomedical, nuclear, acoustic, and electronic sciences. One of the primarily purposes of thin films is surface modification. Thin films serving as a passive layer to enhance the surface properties of substrates involve metallic plating, optical anti-reflective coatings, mechanical resistance against friction and wear, chemical barriers against corrosion and erosion and so on. Some thin films, on the other hand, work as active components for performing special functions. For example, metallic or semi-metallic films are utilized as main components in semiconductors based on their capability to confine electric charges. There have been extensive applications of piezoelectric and piezoresistive thin films in micro-electro-mechanical-systems(MEMS) consisting of solid-state sensors, piezoelectric actuators, and electro-acoustic transducers. Polymeric films fabricated on semiconductive substrates serve as photoresists in lithography processes that generate desired patterns with a very high resolution.

The importance of thin films is now becoming more emphasized because of their excellent quality and unique characteristics that cannot be achieved in bulk materials. In general, single or multiple film layers fabricated on a substrate are referred to as thin film structures when the substrate thickness is relatively large compared to that of the film by a factor of 50 or more. Especially from an acoustic viewpoint,

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films within a sub-wavelength range in thickness are also classified as thin films. Diverse techniques are utilized to deposit thin films with controlled thickness and material composition along with the film quality such as uniformity and adhesion. More emphasis should be placed on thin film adhesion, according to Mittal (1976), because adhesion also determines the kinetics of the film growth and the longevity and durability of thin film devices. Especially film adhesion is known to be very sensitive to the deposition parameter and the substrate surface conditions and difficult to measure compared to other film properties such as hardness, stiffness and stress. For adhesion evaluation of thin films, the development of a repeatable and reliable assessment is required and challenging. The purpose of this paper is to introduce a review of previous results on conventional acoustic methods and recent progresses in scanning acoustic microscopy(SAM) for thin film adhesion evaluation. Research directions for more complicated film structures are also proposed.

2. Overviews on Thin film Fabrication

Components for the actively functioning thin films are mostly deposited onto semiconductive substrates by means of such film-growth techniques as physical vapor deposition(PVD), chemical vapor deposition(CVD) and spin casting (Freund and Suresh, 2003). In both vapor deposition processes, film materials from a molten source or a solid undergo mass transfer in a vapor state and grow onto a substrate surface. High vacuum condition is normally required in the vapor deposition process to control the material composition and maintain a sufficiently long mean-free-path between molecules of film materials.

Widely used methods of PVD, accompanied with no chemical reaction, include physical thermal evaporation, electron beam physical vapor deposition(EBPVD) and sputtering. Thermal evaporation allows vapors of a hot source material to travel directly to the target substrate and to condense back to a solid state. In EBPVD, electron beams are utilized to transform atoms into a gaseous phase in a high vacuum environment by local boiling of the source material. In sputtering, atoms ejected by bombardment of energetic irons and accelerated through the electric field are imparted from the source with a high kinetic energy and accumulated as a film layer onto a target substrate.

For thin films of metal alloys, semiconductors, and amorphous or crystalline structures, CVD processes are mostly utilized. Thin film materials undergoing a phase change from gas to solid are accumulated onto the substrate by chemical reaction of the plasma or an ionized vapor in plasma enhanced CVD. In atomic layer deposition(ALD), the substrates are exposed to chemicals, typically called precursors, to produce extremely thin coatings.

In contrast to the vapor deposition methods, spin casting utilizes rheology of film materials in ambient environments to fabricate polymeric thin films onto metallic or semiconductive substrates. A centrifugal force applied onto a disc-shaped stage spreads the film material dissolved in a solvent onto the substrate and form a uniformly thin layer. The layer thickness of a typical film material is determined by its concentration in the solvent and the rotational speed of the stage. A curing process at high temperature above 100 °C is usually required to solidify the film material.

During the fabrication and post-fabrication processes, thin films occasionally undergo adhesion problems caused by the insufficient bond strength to the film-substrate interface (Smith, 1995). The interface may not have a strong bonding in nature due to chemical and physical dissimilarities between both materials. In addition, the interface integrity is influenced by the surface conditions of substrates. External particles may ruin the quality of substrates, and

even in a highly vacuum condition, organic materials introduced from water may contaminate substrates. Film structures grown on such substrates suffer from weak adhesion when they are exposed to internal and thermally-mismatched stresses during the deposition process or a local stress during their applications. These stresses may stimulate the weakly adhered interfaces to be debonded and result in a malfunction, degradation or even a complete failure of the film systems. Thus, thin film adhesion is a very important issue and should be necessarily evaluated by appropriate methods.

3. Adhesion Evaluation of Thin Films

3.1 Previous Results Based on Conventional Methods

To maintain the functionality of films after testing and avoid additional damage, evaluation methods for thin film adhesion are generally desired to be nondestructive and noninvasive. It is a complicated but important issue to nondestructively evaluate the integrity of thin film adhesion with appropriate methods. The challenge is that the interface of the film structure is hidden below the film. In opaque film systems it is inefficient to utilize optical methods such as microscopes and laser devices. A direct approach to the interface is very difficult if not impossible without uncovering the film from the substrate.

There have been various attempts for nondestructive evaluation(NDE) of the interfacial integrity of thin films. For instance, thermal imaging utilizing the heat loss of thermally sprayed film surfaces has been studied to visualize disbonds or delaminations in the interface(Parthasarathi et al., 1995). The thermal images, however, could offer only a relatively poor resolution compared to ultrasound techniques. Eddy current techniques have been utilized to analyze the porosity and thickness of thin films(Roge et al., 2003). However, these methods are limited to electrically conductive materials and films with limited thickness. Micro X-ray fluorescence imaging was utilized to evaluate a fused chromium disilicide coating, which served as a thermal barrier in Space Shuttle orbiter thrusters(Doering et al., 2004). Thickness and defects in the coating were detected by this technique. Other approaches on NDE of thin films were extensively reviewed in Refs.(Parthasarathi, 1996; Du, 2008).

Recent NDE techniques for film interface integrity have focused mostly on acoustic methods based on the sensitivity of acoustic waves to the interfacial conditions. Almond et al. (1981) applied an ultrasonic attenuation measurement to thermally sprayed coatings of aluminum, molybdenum and nickel aluminide at relatively low frequencies ranging from 2.5 to 15 MHz. This study presented the dependence of attenuation on film adhesion and the technical limitations resulting from the film thickness. Jiao and Rose(1991) developed an ultrasonic oblique incidence technique to detect bonding weakness. The adhesive interface of two solids was modeled as a homogeneous isotropic thin layer with particular density, thickness and elastic constants. Within relatively low frequency range (3 to 12 MHz), the frequency-dependence of reflection factors were investigated for rigid, smooth and imperfect interfaces. Xu et al.(1993) utilized leaky Lamb waves to measure the wave speeds and elastic properties of thin films and substrates. This technique was applied to titanium and aluminum samples with plasma-sprayed coatings within a sub-millimeter range. The measurement based on the modal frequency spacing could be compared with simulation results. Berndt(1989) applied acoustic emission, which occurs by crack formation and propagation, to plasma-sprayed coatings. The number of crack events and crack sizes were monitored for ceramic coatings subjected to high temperature in a qualitative manner. Thomsen et

al.(1987) adapted picosecond acoustics to characterize the properties of thin, opaque silicon and aluminum films. In this study, the optical detection of a laser-generated pulse traveling through the film and reflecting from the substrate enabled the accurate measurement of film thickness and qualitative evaluation of adhesion and homogeneity. Kinra and Zhu (1993) developed a technique for measuring the thin film thickness within a sub-wavelength range. Through the accurate measurement and inversion algorithm, the thicknesses and the wave speeds of epoxy and Plexiglas coatings were calculated and compared with theoretical prediction.

Commonly encountered issues of conventional NDE such as the poor resolution and limitations in material selection could be overcome by most acoustic methods. These methods achieved an acceptable accuracy for the quantitative evaluation of the thin film integrity. However, the relatively low frequency range below 100 MHz limited the use of the acoustic techniques for films within a sub-wavelength range in thickness. Bulk wave methods could give no access to the damage geometry of such thin film microstructures. The laser-induced wave methods are generally nondestructive in the thermo-elastic region. However, they have limitations in the generation of high frequency waves in the thermo-elastic region where the low power laser is used not to make any damage on the film structure. These limitations invoke the needs for more reliable and efficient ultrasonic NDE methods operating at a few hundred megahertz or above and guaranteeing quantitative assessment.

3.2 Scanning Acoustic Microscopy for Thin Film Characterization

Scanning acoustic microscopy(SAM) has served as an excellent technique for the thin film evaluation since its invention by Lemons and Quate(1974). Acoustic imaging of SAM operating with high frequency ultrasound ranging from a few hundred mega to gigahertz enables the qualitative evaluation of film microstructures. Surface acoustic waves(SAWs) generated by the acoustic lens with high numerical aperture(NA) are utilized for the quantitative characterization of materials. Evaluation by SAM is completely nondestructive and very efficient for thin films within a sub-wavelength range in thickness.

The operational principle of SAM for imaging is briefly explained here. As shown in Fig. 1, a tone-burst RF electrical signal with f as the central frequency excites the ZnO transducer to generate elastic waves into the buffer rod. The waves passing through a concave cavity at the end of the buffer rod converge into a highly focused spherical acoustic beam in a coupling medium(water). The waves bounce from an elastic sample placed near the focal plane and then trace back to the reversal acoustic path. At this time, the transducer converts the elastic waves into an electric signal, which is separated from the input signal by a circulator. The collected signals as mechanically scanned in the 2-D plane of the sample surface or subsurface compose acoustic images. Contrasts in the images convey information on specimens such as local variations in geometry and mechanical properties. Investigation by acoustic imaging

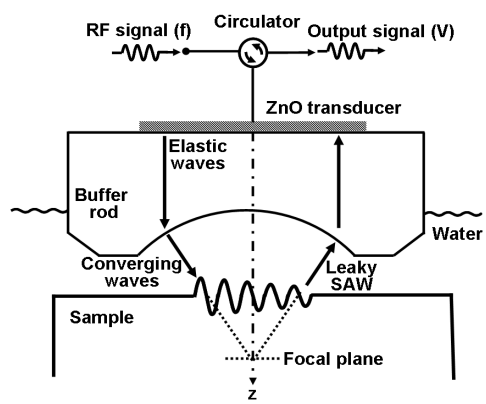

Fig. 1 Schematic illustration of an acoustic microscope operation.

enables the mapping of the elastic property variations in samples and visualizes microstructures or defects. For acoustic imaging of film structures, there have been various researches including the time-resolved technique for small components(Smith et al., 1985; Robert et al., 1999), semiconductor device packaging (Briggs and Hoppe, 1991; Gordon et al., 1993) and IC chips containing resist/metal overhang, subsurface blisters, thickness variation and microcracks(Miller, 1983, 1985).

In addition to the imaging mode, an important feature of SAM is the V(z) technique. Acoustic waves corresponding to the third critical angle of the sample generate the leaky SAWs, which interfere with longitudinal waves along the lens axis constructively and destructively. The interference generates unique oscillations in the signal amplitude (V) collected along the vertical direction (z), referred to as the V(z) response. The oscillation interval Δz of the V(z) curve is utilized to calculate the SAW velocity c_{SAW} as(Parmon and Bertoni, 1979)

$$c_{SAW} = c_W \left[1 - \left(1 - \frac{c_W}{2 f \Delta z} \right)^2 \right]^{-1/2} \quad (1)$$

where c_W represents the longitudinal wave velocity in the coupling medium. The SAW velocity is a unique property of a single material, and in case of a layered material structure represented by the thin film system, it becomes dispersive due to the waveguide characteristics of the layers with finite thickness. Analytic solutions for such a structure are generally obtained by a matrix method referred to as the Thomson-Haskell matrix model (Thomson, 1950; Haskell, 1953). Fig. 2 illustrates the matrix model for a multi-layered structure where the SAW propagates along the x-axis and exponentially decays out in the *z*-axis toward the substrate. By applying the field continuity at each layer interface, one can obtain the wave solutions. The traction-free boundary at the film surface and the infinite half-space in the substrate are also assumed.

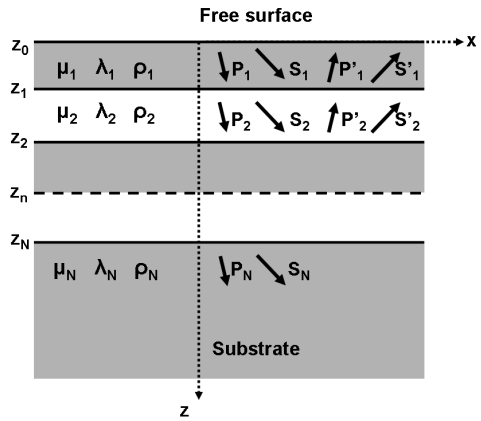

Fig. 2 A layered structure on a half space for the Thomson-Haskell matrix model. λ and μ : Lame's constants; ρ : density; P and P': amplitudes of down going and up going longitudinal waves, respectively; S and S': amplitudes of down going and up going shear waves, respectively.

It is known that the SAW velocity of the film/substrate configurations is quite sensitive to their structure and interfacial conditions. For this reason, the V(z) technique has been widely utilized to analyze the properties and integrity of thin film structures. Since Weglein(1980, 1982) pioneered the acoustic micro-metrology based on the potential of SAM, there have been extensive researches for characterization of thin films and coatings. Aizawa et al.(1992, 1993) utilized acoustic spectro microscopy as a technique for diagnosis and investigated the elastic characterization of ceramic coated tool materials such as PVD/CVD TiN coatings on WC/Co substrates. The investigation based on the dispersion measurement was extended to the evaluation of the interfacial integrity. Lee et al. (1995a, 1995b) and Achenbach et al.(1995) utilized line-focus acoustic microscopy to measure the elastic constants and mass density of films. This technique was sensitive especially to angular variations in the SAW velocity of anisotropic materials, so that it could be used

effectively for the characterization of isotropic and cubic crystalline solid and various film/substrate configurations such as Nb_2O_5/MgO and $BaTiO_3$/$LaAlO_3$.

The previous studies revealed the potential of SAM as an effective tool for the characterization of thin film structures. These studies could visualize the microstructures or flaws of films and measure the film properties such as thickness, elastic constants, and mass density. However, adhesion evaluation of thin films has been relatively unexplored compared to these film aspects.

3.3 Adhesion Evaluation by Means of Scanning Acoustic Microscopy

The film adhesion evaluation by means of SAM was first attempted by Bray et al.(1980). Patterns in chromium films on glass substrates were acoustically visualized and compared with optical micrographic images as shown in Fig. 3. Their findings indicated that the adhesion difference in the patterned film induced a variation in reflection of acoustic waves. The reflection variation led to a non-uniform contrast in the images taken at a subsurface regime, which could not be observed in the optical image. Adhesion weakness was simply modeled as a vacuum interface layer to predict the V(z) response of the film structure. Although the adhesion model was oversimplified and V(z) results were not supported by direct adhesion measurement in this study, it could offer an meaningful clue to qualitative evaluation of thin film adhesion.

Addison et al.(1986) extended the study of Bray et al.(1980) and investigated the effects of different bond strengths in the SAM images of Au and Cr/Au films deposited on glass substrates. The obtained images provided the local difference of the bond strength in a qualitative manner. A supportive study was carried out to offer the correlation between the bonding compliance and fracture energy of the film through the mechanical indentation

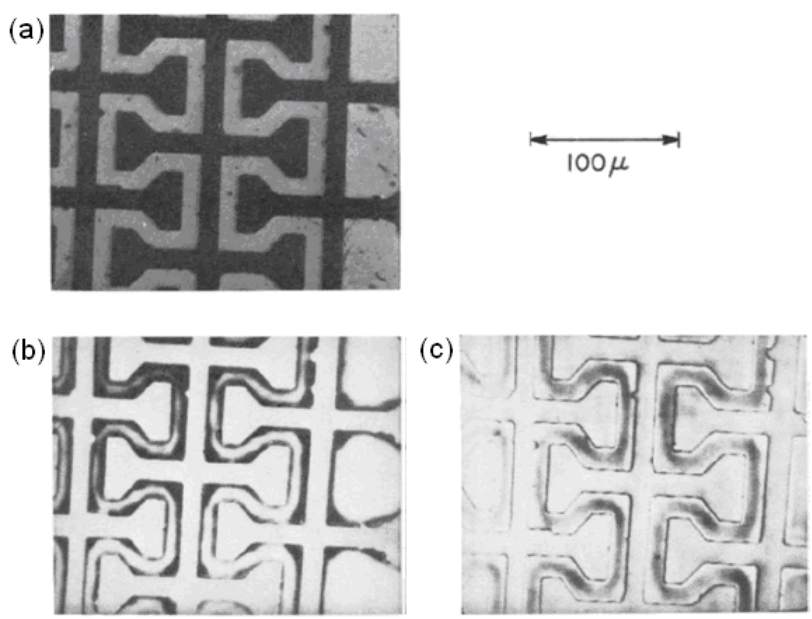

Fig. 3 Optical and acoustic microscope images of chromium poorly adhered to glass: (a) optical image; (b) acoustic image, z = 1 μm; (c) acoustic image, z = 0.5 μm. (Reprinted from Thin Solid Films, Vol. 74, R. C. Bray, C. F. Quate, J. Calhoun and R. Koch, Film Adhesion Studies with the Acoustic Microscope, pp. 295-302, Copyright (1980) with permission from Elsevier)

measurement(Addison and Marshall, 1988). In this study, Au and Cr/Au films were deposited onto sapphire substrates, and their bonding fracture energy were estimated by indentation-induced delamination sizes. This study was, however, incomplete because the correlation was only partially established between the Au/sapphire structure, and the fracture energy measurement was not supported by acoustic measurements in the same sample.

Mal and Weglein(Mal and Weglein, 1988; Weglein and Mal, 1987) investigated the influence of the bonding integrity on the Rayleigh wave velocity of a single-layered Ti/Be system. The bonding integrity was controlled by applying low deposition temperatures and by preparing the substrates with wet-machining. By operating SAM with relatively low frequencies ranging from 22 to 45 MHz, the SAW velocity of the film structures could be measured. The interfacial conditions were grouped into "good" and "bad" among the samples based on the comparisons between the measured results and theoretical dispersion curves. It was found that the SAW velocity of "good" film structures was well matched to the first higher mode, whereas that of the "bad" film structures showed somewhat irregular dispersion between the fundamental and higher modes. This result invoked the need for a virtual interfacial layer to simulate the interfacial weakness in the dispersion model. Even though the unexpected mode conversion was observed in the dispersion measurement without verifying theory, this study contributed for acoustic quantification of the interface conditions as an initial stage.

Richard et al.(1994) investigated the interfacial adhesion of a Ni film on an Al alloy substrate and an Al film on a glass substrate in the frequency range from 120 to 300 MHz. Various interfacial conditions were realized by applying paraffin oil, an intermediate Cr layer and plasma etching onto the substrate surface. The theoretical dispersion curves were predicted for perfect, totally detached and arbitrarily weakened interfaces. The measured SAW velocities of Ni/Al alloy samples agreed well with the predicted dispersion with a perfect interface, whereas the dispersion curve for the Al/glass sample matched to a single plate mode representing a complete debonding of films. However, this study compared the adhesion of different film structures without presenting any evident sources for weakened adhesion; this made the overall results unclear. A combinational study utilizing a destructive test was conducted to support the acoustic measurement(Richard et al., 1997). For Ni-SiC coatings with strong, moderate and weak adhesion, delamination induced by the scratch test was observed with SAM. The area of the delaminated region was measured for the quantification purpose, but a more systematic study directly measuring the bonding energy or interfacial strength was not available in this study.

Parthasarathi et al.(1997) studied the interfacial condition of a diamond-like carbon (DLC) film deposited on a Ti substrate. By varying deposition parameters such as the pressure of Ar and H gases and D.C. biased voltage, the adhesion strength of the interface was controlled. A correlation between the bond strength and the Rayleigh waves velocity measured by the V(z) technique was presented as shown in Fig. 4. With limited number of data measured at 200 and 600 MHz, the simulated dispersion curves with preset weakness levels was correlated to the bond strength represented by the destructively measured critical load. During the evaluation on the DLC/Ti structure classified as a hard film on a soft substrate, destructive failure modes could easily be detected. Among the similar hard-on-soft structures such as a PVD TiN film on titanium alloy and a CVD diamond film on a ceramic substrate, the delamination induced by laser spallation and mechanical indentation was visualized by SAM to characterize the interfacial

fracture(Parthasarathi et al., 1998). Acoustically, such film structures are the fast-on-slow system in many cases, in which only a single dispersion mode is detected. More care would be necessary for both the acoustic propagation theory and experiments for the slow-on-fast system, in which acoustic energy partitioning occurs during the multi-mode excitation.

Kosbi et al.(1998) attempted the adhesion evaluation of a gold film on a glass substrate classified as a soft-on-hard structure. Imperfect adhesion at the interface was realized by applying silicone oil diluted in acetone onto the glass substrates, and the oil concentration was systematically controlled. Differently from other conventional V(z) method, the SAW velocity was measured by the interval between the first-to-second maxima in the V(z) curve. The SAW velocity was compared with the critical load measurement by the supportive scratch test. The oil layer could effectively realize imperfect bonding conditions at the film interface, in spite of the lack of the controllability of the oil thickness. However, the measured SAW velocity dispersion showed irregularity even in the perfectly bonded film structure. In an extended study carried out for a Au/Si film structure (Pageler et al., 2000), variations in the SAW velocity dispersion were clearly observed in existence of an intermediate Cr layer applied to the substrate before the film deposition. However, rigorously quantitative evaluation of film adhesion could not be achieved through this study because physical explanations for the contribution of the Cr layer and further studies supporting the destructive evaluation were not presented.

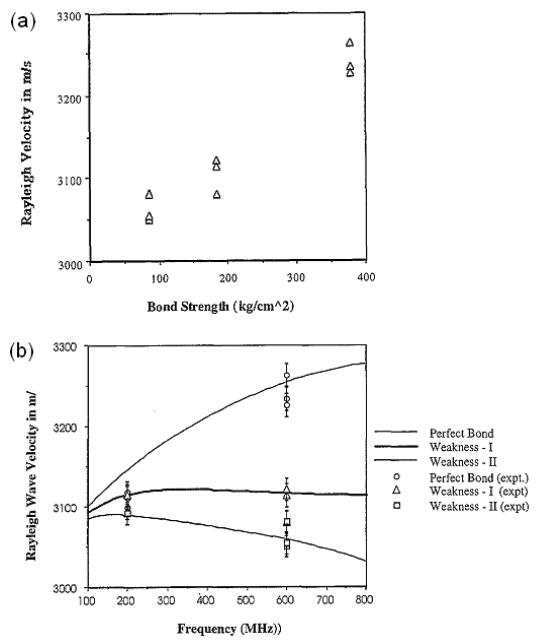

Fig. 4 (a) Measured Rayleigh wave velocity as a function of thin film bond strength. (b)Rayleigh velocity in films with varying levels of adhesion: theory and experiment (each experimental point represents a measurement in a different location) (Reprinted from Thin Solid Films, Vol. 300, S. Parthasarathi, B. R. Tittmann and R. J. Ianno, Quantitative Acoustic Microscopy for Characterization of the Interface Strength of Diamond like Carbon Thin Films, pp. 42-50, Copyright (1997) with permission from Elsevier)

Guo et al.(1999, 2001) developed a technique for adhesion evaluation of thin film structures as controlling the interfacial condition by etching substrates with different time. On the basis of line-focus acoustic microscopy, the bonding integrity of nearly isotropic films on isotropic substrates such as TiN and Ti/Al on M2 high-speed steel(HSS) were investigated. The interfacial weakness of the films was approximated as a net of springs provided by interface stiffness constants. The SAW propagation for perfectly and imperfectly bonded interfaces was predicted with a matrix method in consideration of the water-loaded boundary condition at the film surface. The corresponding reflectance functions were examined for detectability of multi-modes. The theoretical dispersion curves were compared with the V(z) measurement at 225 MHz in operating frequency. Critical normal loads were measured on both fast-on-slow(TiN on M2-HSS) and slow-on-fast systems(Ti/Al on M2-HSS) by the scratch test as a supportive destructive evaluation method. For the TiN/M2-HSS system, different deposition methods including ceramic targeting and reactive

sputtering were utilized, and the dispersion was measured to be compared with the theoretical prediction as shown in Fig. 5. Though the SAW dispersion was not distinguishable among the ceramic-targeted samples of the TiN/M2-HSS structures, the dispersion results for the reactive-sputtered samples indicated that increases in etching time softened the interface as causing poor adhesion. This result was well supported by the scratch test for the critical normal force measurement. However, in case of the Ti/Al/M2-HSS system within the category of the slow-on-fast system, the effect of etching time was found to be opposite; the increasing etching time even reduced the interfacial stiffness. Neither relevant physical explanation nor success in the destructive test was presented for such as system. For these reasons, rigorous adhesion evaluation of the slow-on-fast systems still remained unexplored and needed a support by the SAW dispersion measurement correlated to destructive tests for adhesion in terms of quantitative evaluation.

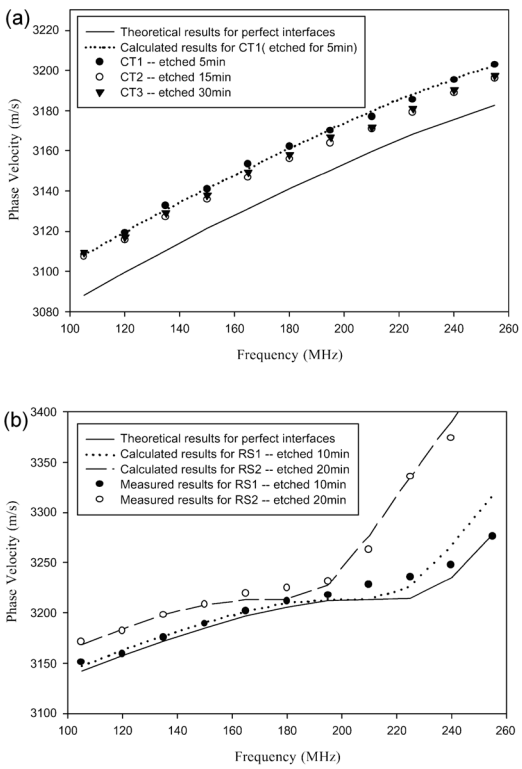

Fig. 5 Leaky SAW phase velocities calculated using the V(z) model using known material constants of the film and determined interface stiffness parameters K_L and K_T (indicated by lines), plotted together with the measured values (symbols) for the specimens: (a) ceramic targeting: CT1, CT2 and CT3; and (b) reactive sputtering: RS1 and RS2. (Reprinted from Thin Solid Films, Vol. 394, Z. Guo, J. D. Achenbach, A. Madan, K. Martin and M. E. Graham, Modeling and Acoustic Microscopy Measurements for Evaluation of the Adhesion between a Film and a Substrate, pp. 188-200, Copyright (2001) with permission from Elsevier)

Recently Du et al.(accepted) has focused on the dispersion relations of a gold film on a glass substrate classified as the slow-on-fast system to quantify the adhesion weakness. In this study, the SAW dispersion in film structures with weak adhesion was predicted by inserting a virtual layer with very low density, very small thickness and arbitrarily adjusted elastic properties. For the experiments, Au/glass film structures were fabricated by sputtering, and for realizing an imperfectly adhered interface, external impurities as organic contamination were applied before the film deposition. A broad-band measurement of the SAW velocity dispersion was enabled by modification of SAM system. As seen in Fig. 6, the result matched to the preset dispersion curves for various weakness levels. The acoustic

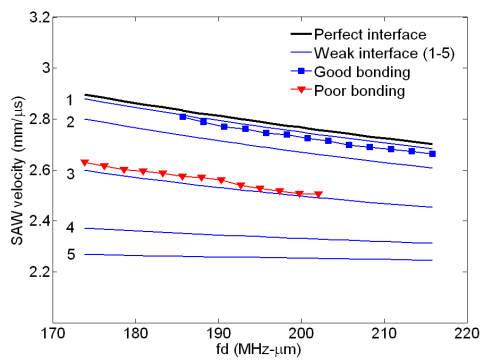

Fig. 6 Comparison in SAW velocity between good interface and poor interface. The Au sample with a good interface is close to theoretically weak interface level 1, while the poor one close to the level 3 (After Du et al., accepted)

measurement was supported by the destructive scratch test, and the resulting features were observed by the imaging mode of SAM at a slightly defocused depth near the film interface as shown in Fig. 7. The delamination in the acoustic images indicated that a plausible failure mode occurred during the scratch test for a poorly adhered film sample. A difference in the slope of the force-displacement curve measured by the scratch test at the pile-up stage could determine the bonding quality of the film interfaces in a semi-quantitative manner. This study mainly contributed to establishing a procedure of film adhesion evaluation based on the well-defined theory, V(z) measurement, supportive destructive tests and qualitative verification by acoustic images. However, the acoustic model introducing arbitrary properties to the virtual interfacial layer needed to be improved to provide a direct correlation between the bond strength and the interfacial weakness.

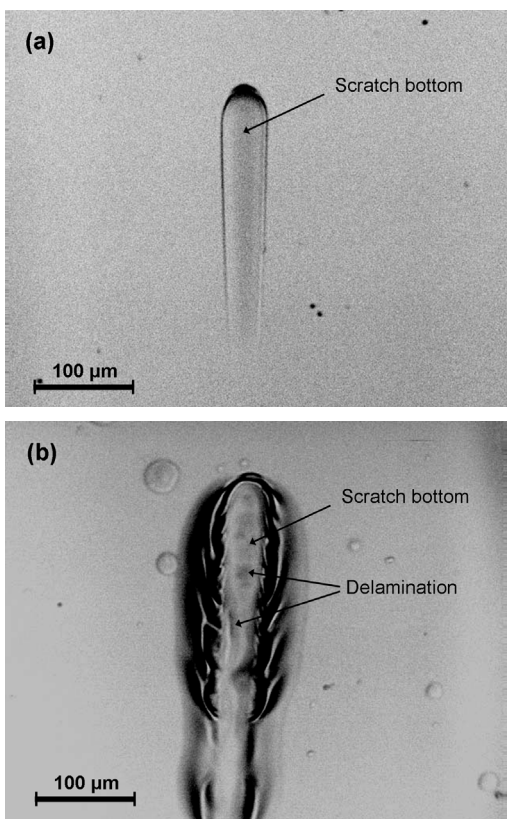

Fig. 7 Acoustic images (400 MHz) of film samples with (a) good bonding and (b) poor bonding. Non uniform brightness in the scratch bottom represents delamination. (After Du et al., accepted)

4. Research Directions for the Film Adhesion Evaluation

4.1 Goals for Future Works

As demonstrated through the previous efforts, nondestructive adhesion evaluation of thin film structures is becoming realistic by the use of the acoustic microscope. The theories for perfectly elastic and isotropic material structures were developed, and the supportive experimental procedures including qualitative and quantitative methods were established. However, applicability of these evaluation methods seems unclear for more complicated film structures in spite of their notable achievement so far. For example, in viscoelastic films, high operating frequency in SAM mode will become an issue because absorption in the films may significantly increase with increasing frequency. In films on an anisotropic substrate, acoustic wave propagation depends on the material orientation; this makes the theoretical prediction and V(z) experiments for such film structures more complex. A complete correlation between the dispersion and destructive evaluation results have not been investigated due to the lack of supporting theories and experimental procedures. In consideration of the SAM technology in the current stage, the following are meaningful goals for future research.

- A wave propagation theory for more general configurations such as viscoelastic layers on the anisotropic half-space should be developed. The corresponding numerical procedure should be established and implanted to a computer program to calculate the dispersion curves in such film systems.

- The model for the imperfect interfacial condition needs to be modified to quantify the interfacial weakness levels in a rigorous manner. Various conditions including discontinuities in stresses and/or displacements are worthwhile to be considered in case that the failure mechanism of the film interface is unknown.
- The acoustic field theories comprising the V(z) response need to be revisited for the use of general types of acoustic lenses. The simulated acoustic field inside the lens and the coupling medium should be suitable for characterization of the viscoelastic films on anisotropic substrates.
- The interfacial weakness in real film structures needs to be controlled quantitatively. Various methods including chemical treatments of the substrate surfaces and application of external impurities during the deposition procedure may effectively influence the interfacial quality. Through a well-controlled impurity concentration, the level of weakness can be adjusted.
- Destructive evaluation methods are required to completely support the proposed acoustic technique. A nanoindentation or pull-off test can be a candidate for measuring the adhesion strength.
- It is meaningful to visualize the film failure mode caused by the destructive test such as the indentation-induced delamination. For example, images by atomic force microscopy(AFM) or SAM could support the destructive tests and indicate their limitations in a microscopic viewpoint.

4.2 Example Study: Polymeric Films on Anisotropic Substrates

As the aforementioned research goals are pursued, the development of a novel and reliable nondestructive evaluation procedure is hoped for. There has been an advanced study aimed at these goals(Ju and Tittmann, in press) as an initial stage. In this study, acoustic propagation in thin films is extended to general anisotropic materials in consideration of material anisotropy and film viscoelasticity. The Christoffel equation and a matrix method were utilized for the layered anisotropic structures, and hysteretic absorption in ultrasonic measurements were also considered by modifying the stiffness constants into complex moduli consisting of the storage and loss terms.

A Poly-methyl-methacrylate(PMMA) film fabricated onto a silicon wafer with <100> orientation is considered as the model structure. As the acoustic impedance of PMMA polymer is not very high compared to the coupling medium (water), essentially the water-loading effect should be considered for the dispersion calculation. Reflectance functions for the viscoelastic film structure were calculated and compared with the simple elastic model as shown in Fig. 8(a). In both cases, multiple acoustic modes are observed where there are abnormal variations or dramatic changes by 2π in the phase of reflectance. The modes are denoted by arrows with and the letters H, M, and L for higher, middle, and lower modes, respectively. It is notable that the viscoelastic model indicates that the magnitudes of reflectance are small at the lower and higher modes compared to the middle mode. This implies that the middle mode may be easily detected as a main mode due to its relative large reflectance magnitude resulting in leaking acoustic waves sufficiently back to the coupling medium, although other modes are also excitable. With the systematically controlled interfacial discontinuity in displacements (ζ defined as a displacement ratio between the film and substrate), the corresponding mean reflectance functions are calculated (See Fig. 8(b)). The mean reflectance function represents the average of reflectance in all propagation directions in

anisotropic materials; this is valid when spherical acoustic lenses are used for anisotropic materials. The prediction indicates that the severity of the interface shifts the SAW velocity of the lower mode and also influences its detectability, while the higher mode becomes invisible.

Real PMMA films were coated onto a P-type silicon wafer by spin casting from Anisole 9% solution. The wafer surface was cleansed by acetone and pure water and then treated chemicals such as 6:1 buffered oxide etching (BOE) and an adhesion promoter, HMDS. The V(z) curves of each film sample were obtained and analyzed by the spatial FFT. They were converted into spectra with respect to the SAW velocity as shown in Fig. 9(a), now referred to as the SAW velocity reconstruction. The result shows that the middle mode is dominant in all samples, and its velocity does not shift much between the BOE and HMDS treated samples. However, in the case of the oil-contaminated sample, the SAW velocity in the lower mode shifts more significantly, and the higher mode becomes undetectable as seen in Fig. 9(b). Though exact matching of the SAW velocity in each mode is not available, the V(z) measurement and SAW velocity reconstruction results agree with the prediction in a qualitative manner showing the feasibility for evaluation of complicated film structures.

The adhesion evaluation attempted for the polymeric films on anisotropic substrates could partially fulfill the research directions for future works. The main difference between this study

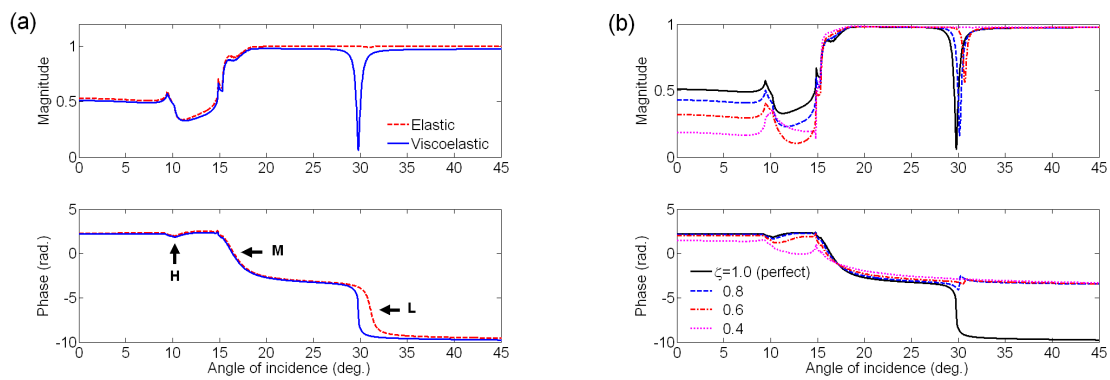

Fig. 8 Mean reflectance functions of PMMA/Si (*fd*=600MHz-μm) representing (a) the influence of viscoelasticity and (b) the interfacial weakness

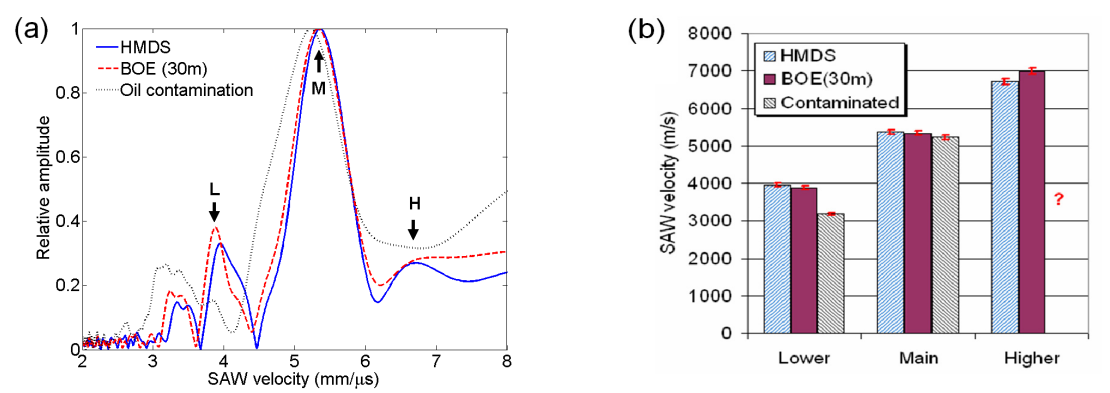

Fig. 9 (a) SAW velocity reconstruction and (b) measured SAW velocity of PMMA films on the, HMDS treated, BOE treated (30 min), and oil contaminated Si surfaces

and previous researches are presented in Table 1. The destructive adhesion tests for such a soft film are known to be very difficult in many cases due to the unexpected behaviors of the film including relaxation and swelling, so that relevant methods should be necessarily sought.

5. Concluding Remarks

In summary, the successful quantitative nondestructive evaluation of thin film adhesion has become an important and urgent need in current technology. Acoustic evaluation of the film interfacial integrity is very challenging, but it could be gradually achieved by the progresses in SAM. The acoustic wave propagation theories have been developed for thin isotropic film structures and improved upon by employing anisotropy and viscoelasticity of materials. The weak interface of the films has been approximated as a net of virtual mechanical springs and an interfacial layer and modified by discontinuities in displacement. The dispersion relation in film structures has been found to be very useful for quantification of thin film integrity. The V(z) technique for the dispersion measurement could reveal the feasibility of quantitative adhesion evaluation by means of SAM. On the basis of the notable progresses in the acoustic evaluation for metallic and ceramic films on isotropic substrates, the research directions for more complicated film structures

Table1. Comparison of film adhesion researches

Authors	Parthasarathi et al. (1998)	Kosbi et al. (1998)	Guo et al. (2001)	Du et al. (accepted)	Ju and Tittmann (in press)
Material	DLC film on Ti	Gold on glass	TiN on M2- HSS Ti/Al on M2-HSS	Gold on glass	PMMA on Si <100>
Deposition process	D.C reactive magnetron sputtering	Sputtering	Ceramic targetting / Reactive sputtering	Electroplating on substrates (prepared by PVD of Au/Cr)	Spin casting
Wave propagation model	Thomson-Haskell (Free-surface)	Farnell and Adler's model modified by Baik and Thomson.	Matrix method for isotropic layers (Water-loading)	Thomson-Haskell (Free-surface)	Matrix method for anisotropic, viscoelastic layers (water-loading)
Film Thickness	0.25 μm	100 nm	TiN (1 μm) Ti(1 μm)/Al(3.5 μm)	~0.5 μm	1.5 μm
Interface Weakness model	Stiffness constants	Slip bond	Stiffness constants	Interfacial layer	Interfacial discontinuity
Acoustic microscopy	Spherical lens (200 and 600 MHz)	Spherical lens (> 1 GHz)	Cylindrical lens (225 MHz)	Spherical lens (400 MHz)	Spherical lens (400 MHz)
Artificial weakness	Changes in deposition parameters (Ar, H pressure / dc voltage)	Silicone oil dissolved in acetone with different concentrations	Substrate etching before deposition	Applying external impurities (organic contamination)	Substrate treatments by chemicals and external impurities
Note	Fast-on-slow system. Insufficient data points	Irregularity in the measured dispersion found in the perfectly bonded films	Fail to the destructive test on the slow-on-fast system	Slow-on-fast system. Perfectly elastic isotropic materials	Viscoelastic films on anisotropic substrates
SAW velocity measurement	V(z) technique for Δz measurement	V(z) technique for first-to-second maxima interval measurement	V(z) technique for Δz measurement	V(z) technique for Δz measurement	V(z) technique for Δz measurement

such as polymeric films on anisotropic wafers were suggested. A study for adhesion evaluation of such films is now being sought as showing its partial success. Through the advance in the dispersion theory and the SAM experiments, we expect that a complete quantitative evaluation of film adhesion correlating nondestructive tests and supportive destructive measurements will be available soon.

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