Journal of Magnetics 7(4), 151-155 (2002)

In situ Stress Measurements with Submonolayer Sensitivity As a Probe of Coherent-to-incoherent Matching at an Interface in Ultrathin Magnetic Films

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(Received 1 December 2002)

***In situ* stress changes at interfaces of ultrathin magnetic films were measured by means of a non-contact optical fiber bundle displacement detector. A bending of the substrate due to stress of a deposited film was detected in cantilever geometry. The highest sensitivity of 134 mV/μm for the displacement detector was realized with a help of computer simulation. The detector was applied to *in situ* stress measurements of Co/Pt and Ni/Pd magnetic multilayer films prepared on the glass substrates by dc magnetron sputtering. The detector turned out to have a submonolayer sensitivity that enables to observe coherent-to-incoherent transition in these mismatched multilayers and even detect the stress changes within the monoatomic coverage. This highly sensitive detector paves new way to probe the stress relaxation at an interface in ultrathin films.**

Key words: stress, multilayer, interface, fiber displacement sensor

1. Introduction

Stress inevitably generated in thin films during fabrication is known as a prime limitation to the growth of thick films, and causes mechanical deformations such as film fracture and film buckling [1]. Also, the introduction of stress into active regions of thin films can seriously affect their physical properties. For instance, film stresses can induce band gap shifts in semiconductor, transition temperature changes in superconductor, and magnetic anisotropy in magnetic films through inverse magnetostriction mechanism. Even in molecular films, such as alkanethiols, stress has fundamental information concerning the variation of mechanical properties [2]. Furthermore, from a reliability and performance point of view, stress has become an increasingly important technical issue in fabrication process, such as passivation and interconnect failures in integrated circuits [3]. Thus, accurate measurements of stress in thin films are very important for understanding a mechanical deformation mechanism and furthermore improving a reliability of thin film as devices.

2. Theory and Experiments

Various experimental techniques have been suggested for stress measurements: those based on direct measurements of elastic strains using X-rays [4, 5], and the cantilever beam techniques based on the measurement of associated curvature or deflection of the substrate, where one side of the substrate is fixed by a substrate holder and the other side of the substrate is free to move [6]. The direct X-ray techniques are most informative because they permit a determination of all stress components in the films. However, because these techniques are, in principle, based on diffraction, they cannot be used for measuring stress in noncrystalline films. Even in crystalline films these techniques are not useful in some specific circumstances, such as dynamic variation of strains. For these cases, the cantilever techniques must be used for measuring stress of the film. Several methods have been developed for the measurement of deflection: Optical interferometry [7], Laser scanning method [8, 9], and capacitance method [10]. Optical interferometry determines the curvature of the substrate by a simple count of interference fringes. This technique is highly sensitive to the curvature variation and often preferable if a more general measurement facility is not available. However, this technique is not easy to apply to *in situ* measurement of stress, so it is

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*Corresponding author: Tel: +82-82-869-8167, e-mail: midpath@mail.kaizt.ac.kr hard to analyze the stress in rapid and quantitative ways. The laser scanning method represents another method for measuring changes in substrate curvature. This method uses a position-sensitive photodetector which detects the change in angle of the reflected laser beam from the surface of a curved substrate. This laser-scanning device is very sensitive and is capable of detecting the displacement down to 1 nm. Furthermore, this technique is possibly used for *in situ* measurement of stress. However, operating this device in dynamic situation with keeping the high sensitivity, coarse signal analysis and modulation technique must be used. The capacitance method, where a change of capacitance between substrate and electrode is monitored, has been popularly adopted for *in situ* measurement of deflection. But, this method can not be applied to a sputtering system, since the capacitance is unstable due to plasma existed during sputtering process.

Here we present an *in situ* stress-measurement apparatus using an extrinsic-type reflective displacement detector. This type of detector is often referred to as a fiber optic lever sensor in the literature [11]. Several advantages of these devices are that it does not disturb the substrate, is immune to electromagnetic interference, is available in small sensor packages and may be used in inaccessible locations. It is very sensitive and possible to get accurate results when it is calibrated for a given reflecting surface. Furthermore it's sensitivity of the displacement can be improved by the effective arrangement of fibers within closely packed fiber bundles.

An optical sensing probe was composed of 40 multimode optical fibers of 50-μm core diameter whose common end was placed behind the free end of the substrate. They were divided into 20 'illuminating fibers' where the light was transmitted to the substrate and 20 'receiving fibers' where the reflective light was guided to a photodetector. Any motion of the substrate surface modulates the intensity of the light that was seen by the receiving fibers. The variation of receiving light intensity is a measure of the displacement between the probe and the substrate surface. A cross-sectional view of the probe end is depicted in Fig. 1. When the probe was in contact with a non-deflecting substrate, no light was transmitted to the receiving fibers. As the gap distance was increased, the light transmitted to the receiving fibers was linearly increased with the distance until a maximum intensity was reached when the reflected light illuminated the entire surface of the receiving fibers. This linear range was utilized for *in situ* measurement of the gap distance.

Generally, SNR (Signal-to-Noise Ratio) of fiber displacement sensor can be improved by using the fiber bundle probe. But the characteristics of fiber bundle probe depend largely on fiber packing density, arrangement of illuminating fibers and receiving fibers, and the specifications of fibers like N.A., core size, and cladding thickness. Especially arrangement of illuminating fibers and receiving fibers is crucial parameter, which determine the characteristics of fiber bundle probe.

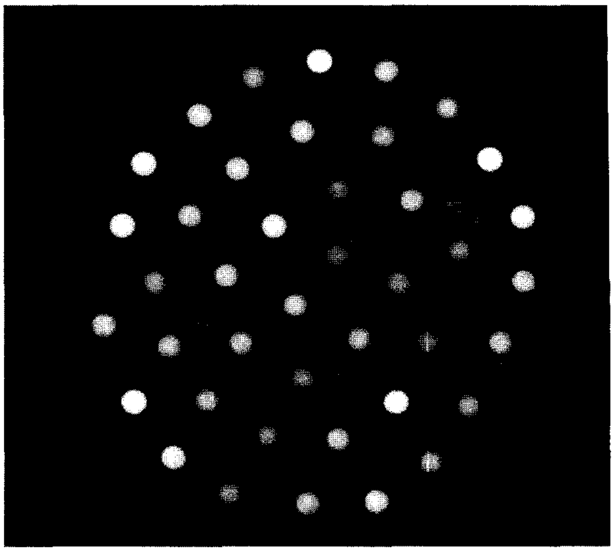


Fig. 1. A cross-sectional view of fiber bundle probe composed 40 multimode fibers close-packed in 1-mm capillary tube.

From the geometrical analysis of fiber bundle probe we could obtain the generalized formula of light intensity launched to the receiving fiber. Using this formula we could simulate the characteristics of fiber bundle probe by numerical method. The details of the algorithm will be published elsewhere. Characteristic curve, sensitivity, and dynamic range of displacement sensor as a function of core radius, N.A., and gap distance between receiving

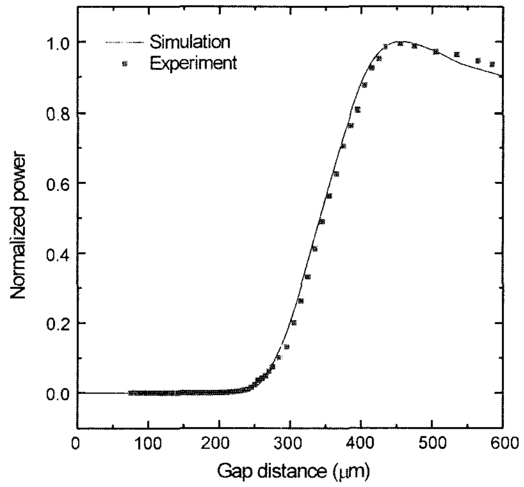


Fig. 2. Experimental characteristic curve of an effective arrangement, together with simulated one.

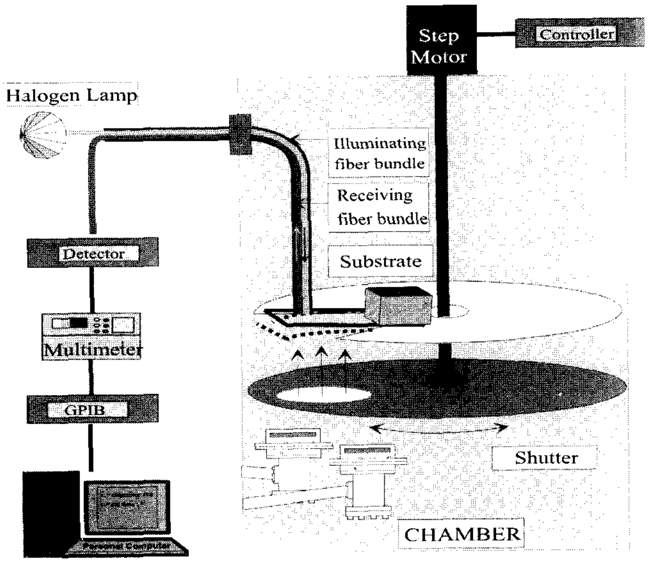


Fig. 3. A schematic diagram of an optical displacement-detection apparatus for *in situ* stress measurement of multilayers.

fiber and illuminating fiber were investigated by this simulation program. Also, effective arrangement of fiber bundle exhibiting the highest sensitivity was determined by considering all possible combinations of receiving and illuminating fiber arrangement. According to this highest sensitive arrangement we made fiber bundle probe. Fig. 2 shows the simulated characteristic curve of a displacement probe for the highest sensitive arrangement, together with the actual characteristic curve for the fabricated probe. We confirm that the simulation precisely predict the actual characteristic curve. This actual characteristic curve was obtained for cantilever geometry where the substrate surface was coated by 1000-Å-thick Al. The sensitivity of the probe was 134 mV/μm, and minimum detectable displacement was 7.5 Å using a voltmeter of 100-μV resolution.

In Fig. 3, we depict a schematic diagram of an optical displacement-detection apparatus for *in situ* measurement of stress of a film. The power applied to targets was 30 W each. The substrate to target distance was 127 mm. The dwelling time of the substrates under the target was controlled using a microprocessor controlled stepping motor. The probe was guided outside the vacuum chamber through a 3.5" del-seal fringe feedthrough. Quartz tungsten halogen lamp (12 V, 100 W) was used as a light source. Any ac noise received by a photodetector was filtered out with a cutoff frequency of 20 Hz. The displacement of the substrate with deposition of a film could be seen in a real time on the computer monitor using a GPIB interface every 0.051 sec. The backside of the substrate was coated by 1000-Å-thick Al to enhance the sensitivity of the detector. A change in the gap distance between the probe and the substrate due to the stress of a film, was measured by linear change in the intensity of the reflecting light from the substrate. The stress was determined from the change of the gap distance Δd using a Stoney's formula as follows [12]:

$$\sigma=\frac{E_s t_s^2}{3l^2(1-\nu_s)}\frac{\Delta d}{\Delta h}, \qquad (1)$$

where E_s, ν_s, t_s, and l are Young's modulus, Poisson's ratio, thickness, length of the substrate, respectively, and Δh is the change of the film thickness. So, using E_s=1.51 $\times 10^{12}$ dyne/cm^2, ν_s=0.3, t_s=130 μm, l = 4 cm for glass substrate and assuming a monoatomic layer deposition of Δh = 2 Å, a minimum detectable stress was estimated to be 2.9 × 10^7 dyne/cm^2.

3. Results and Discussion

We have applied the apparatus to several multilayered systems, where two dissimilar materials were alternately deposited. For these systems, two regimes should be distinguished considering the lattice mismatch between two adjacent layers. If the lattice mismatch between material A and B is not too large, minimizing the total energy leads to a situation whereby, below a critical thickness t_c, the misfit can be accommodated by introducing a tensile stress in one layer and a compressive stress in the other such that ultimately the two materials A and B adopt the same in-plane lattice parameter. This regime is called the coherent regime.

The elastic energy associated with the coherent situation is proportional to the stressed volume. Increasing the thickness of one of the layers will therefore increase the elastic energy. At a certain critical thickness t_c, it becomes energetically more favorable to introduce misfit dislocations which partially accommodate the lattice misfit, allowing the uniform stress to be reduced. The lattice-registry is then lost and the layers become partially coherent, or briefly "incoherent". The critical thickness t_c, where a coherent-to-incoherent transition occurred could be estimated by the following relation [13]

$$\frac{t_c}{b}=\frac{Gx}{4\pi|\eta|E}\ln\left(\frac{t_c}{b}+1\right), \qquad (2)$$

where b=Burger's vector, G=shear modulus, E=elastic constant in the film plane, and η is lattice mismatch.

First, we have measured *in situ* stress of Co/Pt multilayers whose critical thickness of Co is about two atomic layers. In Fig. 4, we demonstrate a typical plot of *in situ* measurement of the gap distance between the substrate

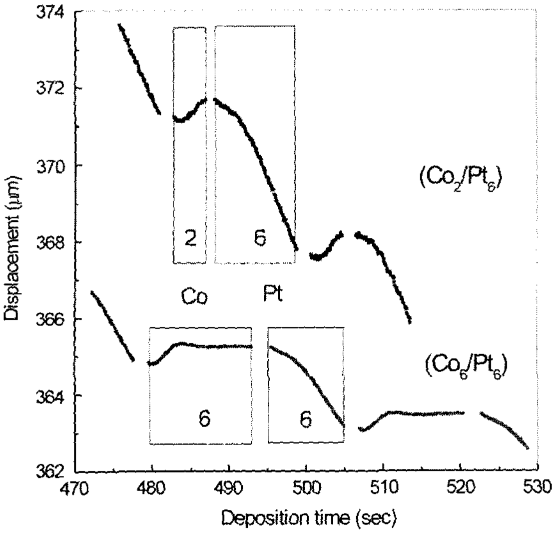
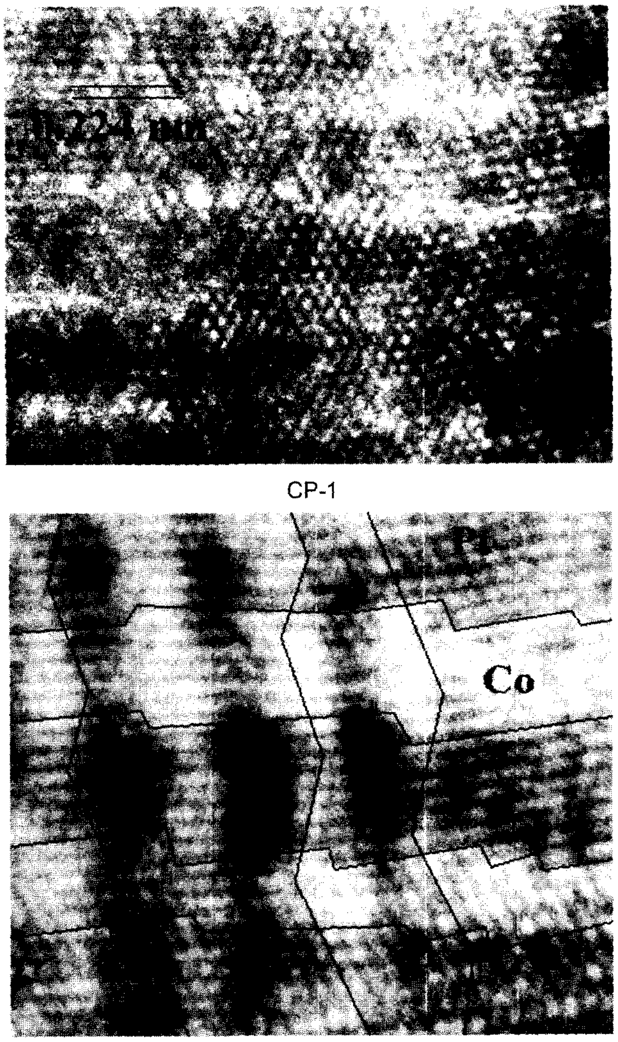


Fig. 4. A typical plot of *in situ* measurement of the gap distance between the substrate and the optical probe with the deposition time for two samples. One (CP-1) is the multilayer having 2 atomic layers of Co and 6 atomic layers of Pt, and the other (CP-2) is 6 atomic layers of Co with the same Pt layers.

and the optical probe with the deposition time for two samples. One(CP-1) is the multilayer having 2 atomic layers of Co and 6 atomic layers of Pt, and the other(CP-2) is 6 atomic layers of Co with the same Pt layers. The base pressure before the sputtering was below 8×10^{-7} Torr and the Ar sputtering pressure of 2 mTorr were used. The positive slope for the Co sublayer implies the existence of a tensile stress in the layer. While, the negative slope for the Pt sublayer implies a compressive stress in the layer. This result is as expected, since the d spacing of the (111) matching plane of Co is 9.8% smaller than that of Pt. However, it is noteworthy that the slopes of the Co sublayers are different for two samples. As expected in estimating the critical thickness, CP-1 has nearly constant tensile stress in the Co sublayer, whereas CP-2 shows two-step like behavior as the Co thickness increase. That is, if Co is deposited more than 2 layers on Pt, Co subject to the incoherent matching with Pt and relax its stress via dislocation making.

To check this scenario, we have investigated the interfacial structure of two samples using cross-sectional TEM. Fig. 5 shows the TEM images of two samples. In CP-1, we found that the lateral planes are in relatively good lattice-registry. However, in CP-2, instead of undissociated misfit dislocation we found many steps and stacking faults near the interface. Unlike our expectation, the stress relaxation comes from the partial dislocation rather than the apparent undissociated dislocation.

This mechanism is quite reasonable because the introduction of the partial dislocation is energetically favorable compared to the formation of the undissociated dislocation. In this way, we could successfully demonstrate the coherent-to-incoherent transition in Co/Pt multilayers with *in situ* stress measurements, and it was confirmed by the TEM images.



CP-1

CP-2

Fig. 5. Cross-sectional TEM images of CP-1 and CP-2.

We have applied this apparatus to other multilayer system that has a large difference in lattice parameters between two dissimilar materials, and have small critical thickness. Nickel and palladium was an interesting candidate for satisfying these conditions. For Ni/Pd multilayer system, the critical thickness of Ni estimated using the reported values of b=2.5 Å, G/E = 1.639, η = 10.2, and x = 2, is about 1 Å. From this we could expect that a large misfit and small Young's modulus in Ni/Pd multilayer system precipitates an immediate incoherent growth within a monolayer of nickel.

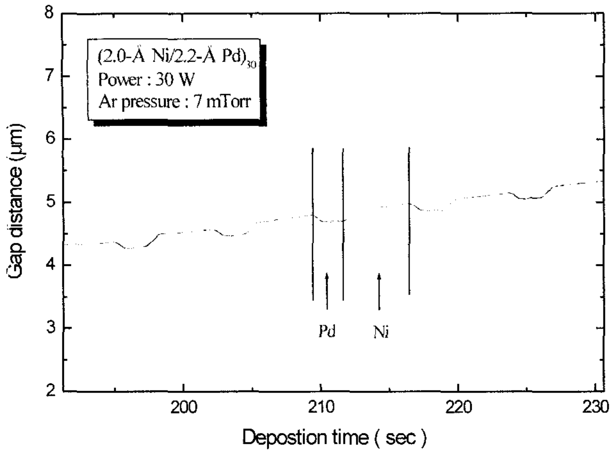


Fig. 6. A result of *in situ* measurement for the (2.0-Å Ni/2.2-Å Pd)$_{30}$ which is fabricated at an Ar pressure of 7 mTorr.

Fig. 6 shows a typical result of *in situ* measurement for the (2.0-Å Ni/2.2-Å Pd)$_{30}$ which is fabricated at an Ar pressure of 7 mTorr. This corresponds to one-monolayer Ni and one-monolayer Pd. The deposition rates achieved under this process conditions were 0.4 Å/sec for Ni and 0.9 Å/sec for Pd. At the start of Ni sublayer deposition on Pd, the stress in Ni sublayer is observed to be large and tensile in nature, and before a half monolayer is deposited it relaxes to tensile stress of moderate value. The stress in Pd sublayer on Ni was large and compressive at the very beginning of deposition and relaxes in successive deposition. The tensile stress after incoherent growth indicates that the intrinsic stress of Ni is tensile at an Ar sputter pressure of 7 mTorr. This two step behavior of stress near the interface was possibly due to a coherent-to-incoherent transition in the matching planes of Ni and Pd. This result is well collaborated with the theoretical estimation that expects a critical thickness of 1 Å in Ni/Pd multilayers. This successful observation of the variation of stress within a monoatomic layer confirms that this tool has submonolayer layer sensitivity in measuring the *in situ* stress.

4. Conclusions

Interestingly, this fiber optic sensor turned out to be sensitive enough to measure the variation of stress near the interface. It implies that this tool is capable of detecting the displacement caused by submonolayer deposition although this simple apparatus doesn't use the modulation technique nor the coarse signal analysis. We conjectured that this outstanding performance of observing interfacial stress was from the effective arrangement of fiber bundles in the sensing probe and the stabilized amplifying circuits with a low pass filter. When observing the coherent-to-incoherent transition with increasing the film thickness, one can easily determine the critical thickness using this high sensitive stress measuring technique without knowing the detailed microstructure and calculating a dislocation density. We strongly believe that this tool has an ultra high sensitivity compared to other techniques and it could be served as another powerful tool for investigating the interfacial state such as RHEED and LEED.

Acknowledgement

This work is supported by the Korean Ministry of Science and Technology through the Creative Research Initiatives Project.

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