Yield strength of attached copper film*

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Variation of stress in attached copper film with an applied strain is measured by X-ray diffraction combined with a four-point bending method. A lower slope of the initial elastic segment of the curve of X-ray measured stress versus applied strain results from incomplete elastic strain transferred from the substrate to the film due to insufficiently strong interface cohesion. So the slope of the initial elastic segment of the X-ray stress (or X-ray strain directly) of the film against the substrate applied strain may be used to measure the film-substrate cohesive strength. The yield strength of the attached copper film is much higher than that of the bulk material and varies linearly with the inverse of the film thickness.

Keywords: copper film, yield strength, X-ray diffraction

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1. Introduction

Interest in the mechanical properties of adherent thin films is increasing due to the demand of reliability of micro-devices made of thin films.[1] Failure of aluminum interconnects in ultra-large scale integration (ULSI) circuits, such as open circuits cracking or voiding,[2,3] or short circuits due to hillocks,[4] are believed to be associated mainly with poor mechanical properties.[5,6] Failure prevention has been a continuing problem with aluminum-based metallizations for many years. It has been claimed in recent years that copper-based metallizations are possible alternatives to aluminum-based alloys for interconnects in sub-micron technologies.[1] The obvious advantages of utilizing a copper stem are due to its lower resistivity and higher melting point expecting higher electromigration resistance. While copper offers desirable properties in terms of resistivity and melting point, several issues must be addressed before it can replace aluminum as the metallization material of choice. The mechanical behaviour of copper film is regarded as one of the key issues.

Several methods have been used to study the mechanical properties of thin films, including uniaxial tensile testing,[7,8] the bulge test,[9] nano-indentation,[10,11] and wafer curvature due to thermal strains induced during the heat cycle.[10,12] In the first two methods, the films need to be removed from the substrates prior to testing. The effects of the substrates cannot be determined and the removed films are often wrinkled and curled due to thermal stress relaxation. It is difficult to fabricate the large specimen required, and edge effects become significant for materials of very small size. A nano-indentation technique is used to test the films on the substrates. However, in order to eliminate the effects of plastic deformation in the substrate below the indenter, shallow indentation (approximately 1/7 the film thickness) must be used, which presents difficulties in accurately determining the area of contact of the indenter. So, this method is not suitable for determining the "bulk" response of the film material, and it cannot be used for thin films with an overlayer. Heating films, during wafer curvature measurement, can cause stress relaxation, recrystallization and grain growth.

2. Experiments

Pure copper films with various thicknesses are deposited by DC magnetron sputtering onto both (100)-oriented single crystal silicon wafers and high-speed steel substrates (100 mm×10 mm×3 mm). The silicon wafer is too brittle to be used for the bending test. The film thickness is controlled by the deposition time and is measured by scanning electron microscopy (SEM) in cross section view after the cleavage of the silicon wafers. The film preferred orientation or texture is detected by X-ray diffraction (XRD). Parameters used for the deposition are listed in Table 1.

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Table 1. Deposition parameters.

Base vacuum	Sputtering-gas (Ar) pressure	Target-substrate distance	Target voltage	Target current
$2.65.\times10^{-3}$ Pa	$2.64.\times10^{-1}$ Pa	10 cm	−520 V	460 mA

To measure the yield strength of deposited copper film, the polycrystalline X-ray diffraction (XRD) method of residual stress analysis is used here in combination with the four-point bending method. Compared with the methods mentioned above, this technique has the following advantages. First, it does not need to remove the films from the substrates, testing sample fabrication is easy, and the effects of the substrate can be determined. Secondly, testing is conducted at room temperature and the microstructure of the film remains unchanged. Thirdly, the penetration depth of X-rays in the material is about 10 μm, which is larger than the thickness of the films commonly used. So, this technique is suited to determining the real "bulk" response of the thin film material and can be used for materials with an overlayer.

The residual stress measuring by XRD has been well established based on the elastic theory, but the measurement of yield strength is not yet widely understood. For polycrystalline films, according to the elastic theory and Bragg's law, if a set of diffraction angles of 2θ at different ψ values are measured, the residual stress $\sigma_{\rm f}^{\rm r}$ in the plane of the film surface can be calculated from[13]

$$\sigma_{\rm f}^{\rm r} = -\frac{E\cot\theta_0}{2(1+\upsilon)}\frac{\partial(2\theta)}{\partial\left(\sin^2\psi\right)}\frac{\pi}{180}, \tag{1}$$

where ψ is the tilt angle between the normal directions of diffracted crystal plane (hkl) and sample surface; 2θ is the diffraction peak at ψ; $2\theta_0$ is the diffraction peak without stress; E and υ are the Young's modulus and Poisson's ratio of the film, respectively. If an induced stress $\sigma_{\rm f}^{\rm i}$ is exerted on the film along the direction of $\sigma_{\rm f}^{\rm r}$, the X-ray stress $\sigma_{\rm f}^{\rm r}$ should be replaced by $\sigma_{\rm f}^{\rm i}+\sigma_{\rm f}^{\rm r}$. *In situ* mechanical loading of the sample is carried out with a four-point bending device and the applied strain is monitored with a resistive strain gauge on the back of the sample, as shown in Fig. 1.

At each increment of applied strain, the X-ray stress is calculated from Eq. (1) in the Cu (222) plane from the linear regression of 2θ versus $\sin^2\psi$ data, and the X-ray stress constant is taken as

$$K = -\frac{E\cot\theta_0}{2(1+\upsilon)}\frac{\pi}{180} = -321.73\ \text{MPa}/(^\circ)$$

($E = 127.2$ GPa, $\upsilon = 0.364$). The apparatus we used was an MSF-2M X-ray stress instrument with an Fe target and power of 30 kV×10 mA.

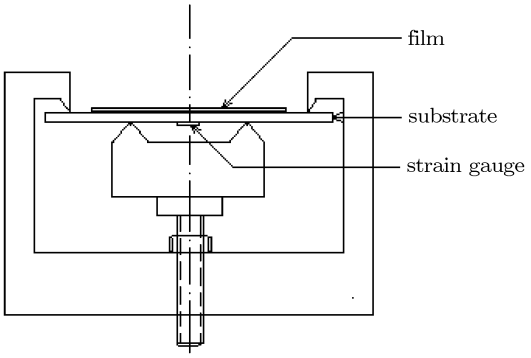


Fig. 1. Four-point bending loading device.

In order to determine the diffraction peak position 2θ accurately at each tilt angle ψ, the whole diffraction peak curve of the Cu (222) plane is recorded around 136.3° for the Fe target ($\lambda_{\rm Fe} = 1.9373$ Å, 1 Å=0.1 nm) used here and the midpoint of the full width at half maximum (FWHM) is used to determine the diffraction peak position 2θ. The side-incline method, i.e., ψ-axis is perpendicular to the 2θ-axis, is used to design the tilt angle ψ. The slope $\partial(2\theta)/\partial\left(\sin^2\psi\right)$ in Eq. (1) is calculated from the linear regression of 2θ versus $\sin^2\psi$ data.

3. Results and discussion

For convenience, the preferred orientation of deposited Cu film is analysed by XRD with a Co target. It can be seen from Fig. 2 that the relative intensities of diffraction peaks are approximately 100, 27.3, 8.36, and 10 for the (111), (200), (220), and (311) planes, respectively. By comparison with the Cu data of the Joint Committee on Powder Diffraction Standards (JCPDS) (card number 3-0932) with a Co target ($\lambda_{\rm Co} = 1.7903$ Å), the relative intensities of the diffraction peaks are 100, 46, 20, and 17 for the (111), (200), (220), and (311) planes, respectively. We find that the relative intensities of 27.3, 8.36, and 10 for the (200), (220), and (311) planes of the deposited Cu film are lower than 46, 20, and 17 for the (200), (220),

and (311) planes of the JCPDS, so that the deposited Cu film has the preferred orientation (111).

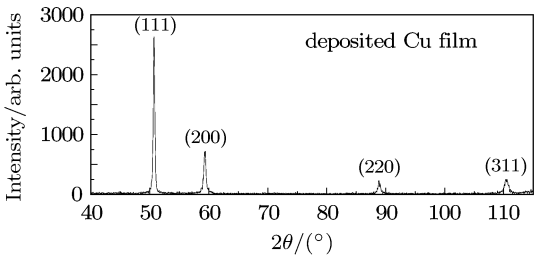


Fig. 2. The X-ray diffraction spectrum of the deposited Cu film.

Figure 3 shows one of the X-ray measured stresses with applied strain curves of pure copper film adhered to a steel substrate. The X-ray stress is calculated with Eq. (1) in the Cu (222) plane from the linear regression of 2θ versus $\sin^2\psi$, and the applied strain is monitored with a resistive strain gauge on the back of the sample. As the measured stress goes beyond the elastic-plastic transition point (open circle), it does not increase linearly with applied strain but only slightly. The main reason is that the X-ray strain reflects only elastic strain, while the strain gauge shows the total (elastic and plastic) applied strain. Another reason is that the grains are not plastically deformed simultaneously. Plastic deformation takes place first in the grains that are in the "weaker" orientations.[14] There are two important aspects that more attention should be paid to.

First, the yield strength of the copper film on steel substrate (about 212 MPa in Fig. 3) is higher than that of bulk copper (about 50 MPa),[15,16] and it varies linearly with the inverse of the film thickness, as shown in Fig. 4. These can be attributed to the substrate strengthening, grain size strengthening and film thickness effects. In previous papers,[17,18] we assumed that a polycrystalline film was composed of columnar grains with an average grain diameter d in the film plane and height h (equal to the film thickness). A simple expression for the yield strength of the polycrystalline film attached to a substrate with a passivated layer was derived from a work-energy relationship for dislocation movement,

$$\sigma_y = \frac{\sin\varphi}{\cos\varphi\cos\lambda}\frac{b(1-\upsilon_{\rm f}\cos^2\beta)}{2\pi(1-\upsilon_{\rm f})} \times\left[\frac{\mu_{\rm f}\mu_{\rm s}}{(\mu_{\rm f}+\mu_{\rm s})h}\ln\left(\frac{\beta_{\rm s}h}{b}\right)+\frac{\mu_{\rm f}\mu_{\rm t}}{(\mu_{\rm f}+\mu_{\rm t})h} \times\ln\left(\frac{\beta_{\rm t}t}{b}\right)+\frac{\mu_{\rm f}}{{\rm d}\sin\varphi}\ln\left(\frac{d}{b}\right)\right], \quad (2)$$

for which the parameters were described in our previous papers.[17,18] It is shown that the yield strength of a polycrystalline film is determined by two affecting factors (grain orientation and dislocation type, corresponding to the first and second factor, respectively) and three strengthening factors (substrate strengthening, passivated layer strengthening, and grain size strengthening, corresponding to the first, second, and third terms in square brackets, respectively). For pure copper film on a steel substrate studied here, the second term in square brackets should be omitted in Eq. (2) due to the absence of a passivated layer. Usually, the average grain size d of the polycrystalline film is not only smaller than that of the corresponding bulk material but also decreases with decreasing film thickness h,[19] so both the substrate strengthening and the grain size strengthening (the first and third terms in square brackets, respectively) are clearly related to the reciprocal of the film thickness h, and thus the yield strength of the copper film on steel substrate is higher than that of bulk copper and varies linearly with the inverse of the film thickness.

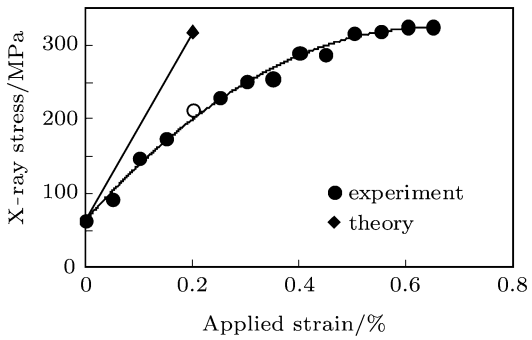


Fig. 3. The X-ray measured stress versus applied strain of pure copper film (in thickness 1.4 μm) on a steel substrate.

Secondly, the slope of the loading segment on the curve of X-ray stress versus applied strain taken below the proportional limit produces a smaller value of the modulus (about 74 GPa) than the theoretical value (127.2 GPa as shown in Fig. 3 with a straight line). Other investigators also observed a low slope of the initial loading curve in the thin films tested in both micro-tensile[8] and bulge tests.[20] They attributed this to the problem that is inherent in gripping film uniformly. The stress was underestimated, resulting in a low apparent modulus. However, this does not exist here for the adherent films. In order to

investigate the reason, the other film is deposited after vacuum Ar^+ ion bombardment cleaning for 10 min at -2 kV voltage applied to the steel substrate. The slope is raised to about 93 GPa. The lower X-ray measured stress may be caused by incomplete elastic strain transferred from the substrate to the film, owing to the fact that the film-substrate cohesion is not strong enough.

In the stronger cohesive case and prior to yielding, the induced film strain $\varepsilon_{\rm f}^{\rm i}$ is equal to the applied strain $\varepsilon^{\rm A}$. The induced film stress $\sigma_{\rm f}^{\rm i}$ is given by Hooke's law, as follows,

$$\sigma_{\rm f}^{\rm i} = E_{\rm f}\varepsilon_{\rm f}^{\rm i} = E_{\rm f}\varepsilon^{\rm A}. \tag{3}$$

The X-ray stress $\sigma_{\rm f}^{\rm x}$ (or total stress) is the algebraic sum of induced stress $\sigma_{\rm f}^{\rm i}$ and residual stress $\sigma_{\rm f}^{\rm r}$ in thin film, i.e.,

$$\sigma_{\rm f}^{\rm x} = \sigma_{\rm f}^{\rm i} + \sigma_{\rm f}^{\rm r} = E_{\rm f}\varepsilon^{\rm A} + \sigma_{\rm f}^{\rm r}. \tag{4}$$

It is shown that the elastic segment of the curve for X-ray stress versus applied strain is a straight line with the slope $E_{\rm f}$ and intercept on the stress axis of residual stress $\sigma_{\rm f}^{\rm r}$.

When the interface cohesion is not strong enough, only partial strain can be transferred from the substrate to the film. We introduce a parameter, $\alpha(<1)$, termed the strain transfer factor or cohesive strength factor, to describe the interface cohesive strength, thus

$$\varepsilon_{\rm f}^{\rm i} = \alpha\varepsilon^{\rm A}, \tag{5}$$

$$\sigma_{\rm f}^{\rm x} = \alpha E_{\rm f}\varepsilon^{\rm A} + \sigma_{\rm f}^{\rm r}. \tag{6}$$

A lower slope may be obtained experimentally, as in Fig. 3. In the strong cohesive case, as discussed above, $\alpha = 1$. In the other extreme case, the interface is completely decoupled (i.e., a free standing film lying on a substrate), so the transferred strain in the film will be zero and $\alpha = 0$. One can use the slope of the elastic regime of X-ray stress (or X-ray strain directly) for the film against the substrate applied strain to measure the film-substrate cohesive strength. Considering the selective diffraction and a certain penetration depth of the X ray, we also expect that the cohesive strength at each interface for multi-layer systems will be able to be determined by measuring the strain transfer rate across each interface.

Figure 4 shows that the tensile yield strength of the pure copper film on the steel substrate varies linearly with the inverse of film thickness. A similar relation was also obtained by Venkatraman *et al.*[21] for aluminum film on a silicon substrate.

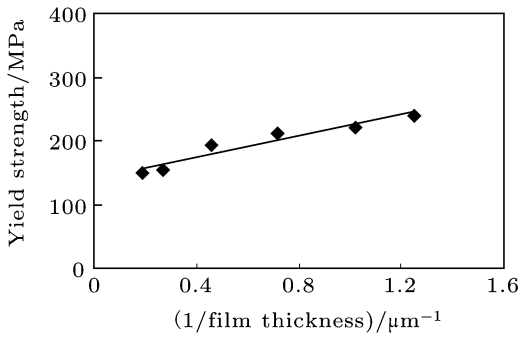


Fig. 4. Variation in yield strength of pure Cu film on a high-speed steel substrate with the inverse of film thickness.

4. Conclusion

The curve for stress versus strain of pure copper film attached to high-speed steel substrate is obtained by XRD in combination with the four-point bending method. A lower slope of initial elastic segment of the curve for X-ray measured stress versus applied strain results from incomplete elastic strain transferred from the substrate to the film due to insufficiently strong interface cohesion. So, the slope of the initial elastic regime or cohesive strength factor α $(0 \leq \alpha \leq 1)$ can be used to represent the film-substrate cohesive strength. The yield strength of pure copper film on a high-speed steel substrate is much higher than that of the bulk material, and it varies linearly with the inverse of the film thickness. This is consistent with theoretical analysis.

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