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Yield strengths and stress induced crackles in copper films: effects of substrate and passivated layer *

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Yield strengths in unpassivated and 530 nm TiN passivated Cu films deposited on Ti, high-speed steel and Ni substrates have been measured by x-ray diffraction (XRD) in combination with the four-point bending method. The results show that, although the texture and average grain size, investigated by XRD and transmission electron microscopy respectively, do not vary with different substrate, the yield strength of the Cu film increases obviously when a thin passivated layer is present and varies slightly with substrates. Many crackles appear in the passivated Cu film on Ti substrate but do not appear in other samples. The experimental results have been explained satisfactorily with an expression for the yield strength of thin films given previously.

Keywords: copper film, microstructure, yield strength, crackles
PACC: 6110, 6114, 7340, 7360D

1. Introduction

The primary interconnects used in large scale and ultra-large scale integrated (ULSI) circuits are Al and its alloys. In order to increase the integrated degree and working frequency of integrated circuits, the Al interconnects have been constructed in multilevel structures with various dielectric or passivated layers (such as TiN, Si_3N_4, Al_2O_3 and SiO_2) and with finer and finer dimensions.[1] Under this condition, the electrical performance and mechanical reliability of interconnects are of increasing concern. Failures of Al interconnects, such as open circuits from cracking or voiding,[2] short circuits due to hillocks,[3,4] are believed to be associated mainly with the poor mechanical properties of them. The failure prevention has been a continuing problem with aluminium-based metallizations for many years. A potential replacement material for Al is Cu, mainly due to its lower resistivity leading to lower R–C delay and Joule heating, and its higher melting point expecting to have lower electromigration.[1] However, because of the difference in the thermal expansion coefficient and thickness between Cu film and Si substrate $\left(\alpha_{\rm Cu} = 17 \times 10^{-6}\,^{\circ}{\rm C}^{-1}, \alpha_{\rm Si} = 3\times10^{-6}\,^{\circ}{\rm C}^{-1} \text{ and } \frac{h_{\rm Si}}{h_{\rm Cu}} = 400-800\right)$ and larger Young's modulus of Cu than that of Al ($E_{\rm Cu}$=127.2 GPa, $E_{\rm Al}$=70.6 GPa), large stresses are generated in the Cu film during thermal treatment required for device fabrication. Stresses as large as 210 MPa (compressive) during heating and 400 MPa (tensile) after cooling to room temperature have been measured.[5] These large stresses may cause yielding and stress-induced migration in Cu film and may result in deleterious effect on device manufacturing yield and ultimate circuit reliability. Stress-induced voids have been reported in Si_3N_4 passivated Cu lines on Ta and Al substrates,[6] indicating that stress-induced voids or crackles may be a problem with Cu metallization as it is for Al. The internal stress and microstructure of TiN film on various steels (stainless steel, tool steel (M2), cemented carbide (A30) and martensitic 420 steel) have been examined. The results showed that, for a given film thickness, the internal stress was found to vary markedly according to the substrate.[7] The experimental results of Li *et al*[8] showed that,

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for identical thickness of Al films on Si substrate, the yield stress of the Al film with a SiO_2 passivated layer was higher than that of the unpassivated film. The yield strengths of pure Al films on Si substrates measured by Doerner *et al*[9] with a substrate curvature technique were higher than that of free-standing Al-1%Si films measured by Griffin *et al*[10] with the bulge testing method. In this paper, the pure Cu film is deposited on Ti, high-speed steel and Ni substrates by magnetron sputtering method. One set of the samples is covered with 530 nm TiN passivated layer, while the other set is not. The yield strengths of the Cu films are measured by x-ray diffraction (XRD) using $\sin^2\psi$ method in combination with the four-point bending loading. The results show that, for the same substrate, the yield strengths of the Cu films with a passivated layer are higher than those without a passivated layer. They vary also according to substrates and for passivated and unpassivated Cu films. Many crackles are observed in TiN passivated Cu film on Ti substrate when the film stress is 372 MPa. However, no crackles are observed in other five samples even if the stresses are higher than 372 MPa. An expression of yield strength in polycrystalline film given previously[11] is used to explain the experimental results.

2. Experiments

2.1. Sample preparation

Si wafer was usually used as a substrate in integrated circuits. However, it was too brittle to be used for bending test. Considering loading, strength, and diffraction peak position at high angles, we selected Ti, high-speed steel and Ni as substrates (100 mm×10 mm×3 mm). Pure copper films were deposited by DC magnetron sputtering onto the above substrates and a (100)-oriented single crystal Si wafer. The samples were divided into two sets. One set was covered with TiN, which was used as a passivated layer in ULSI circuits,[1] while the other set was not. The thickness of the Cu film and TiN passivated layer was measured by scanning electron microscope (SEM) in cross section view after cleavage of Si wafer. The film texture was detected by XRD. The grain size of the Cu film on different substrates was measured by transition electron microscope (TEM). The parameters used for Cu film deposition are listed in Table 1.

Table 1. Cu film deposition parameters.

Base vacuum	Sputtering-gas (Ar) pressure	Target-substrate distance	Target voltage	Target current
2.66×10^{-3} Pa	2.66×10^{-1} Pa	10 cm	−650 V	540 mA

2.2. Yield strength measurement via XRD

The residual stress measured by XRD has been well established in elastic theory, but the measurement of yield strength has not been used widely. For a homogeneous quasi-isotropic thin film, all the stresses normal to the film surface can be eliminated. Based on the elastic theory and Bragg's law, if a set of diffraction angle 2θ at different ψ are measured, the residual stress σ_f^r in the plane of the film can be calculated by

$$\sigma_f^r = -\frac{E_f \cot\theta_0}{2(1+\upsilon_f)}\frac{\pi}{180}\frac{\partial(2\theta)}{\partial(\sin^2\psi)}, \qquad (1)$$

where ψ is the tilt angle between the diffracted crystal plane (hkl) normal and sample surface normal, 2θ is the angle of diffraction peak at ψ, and $2\theta_0$ is that of the diffraction peak without stress, E_f and υ_f are the Young modulus and Poisson's ratio of the film, respectively. If a stress σ_f^a is applied to the film along the direction of σ_f^r, then the σ_f^r in Eq.(1) should be substituted by $\sigma_f^a + \sigma_f^r$. In situ mechanical loading of the sample is carried out with a four-point bending device. The applied strain is monitored with a resistive strain gauge on the back of the substrate. At each increment of applied strain, the x-ray stress is calculated using Eq.(1) for the Cu(222) plane from the linear regression of 2θ versus $\sin^2\psi$ data, and the x-ray stress constant is taken to be $K = -\frac{E_f \cot\theta_0}{2(1+\upsilon_f)}\frac{\pi}{180} = -321.73\,\text{MPa}/(^\circ)$ ($E_f = E_{Cu}$=127.2 GPa, $\upsilon_f = \upsilon_{Cu}$=0.364). The apparatus used in this work is an MSF-2M x-ray stress instrument (made in Japan) with Fe target and a power of 30 kV×10 mA.

3. Results and discussion

3.1. Microstructures

The film thickness and structure of the grains in an unpassivated Cu film were observed with SEM in cross section view after cleavage of the Si wafer. We can see that, from Fig.1, the thickness of the film is 1.34 μm and the structure of the grains is nearly columnar.

For convenience, the preferred orientation or texture of the unpassivated Cu films on Ti, high-speed steel and Ni substrate is analysed by XRD with Cu target and a power of 40 kV×40 mA. The relative intensities of the first four peaks are listed in Table 2, together with the data given by the Joint Committee on Powder Diffraction Standards (JCPDS). It can be seen that there are no obvious differences in the texture for the Cu films deposited on three different substrates. Compared with the data of JCPDS, we can see that all the samples have a slightly (111)-preferred orientation. This may result from the anisotropy of surface energy. In a previous paper,[12] the surface energies of Cu crystals have been calculated by using the modified embedded-atom method (MEAM), and the lowest surface energy corresponds to the (111) surface.

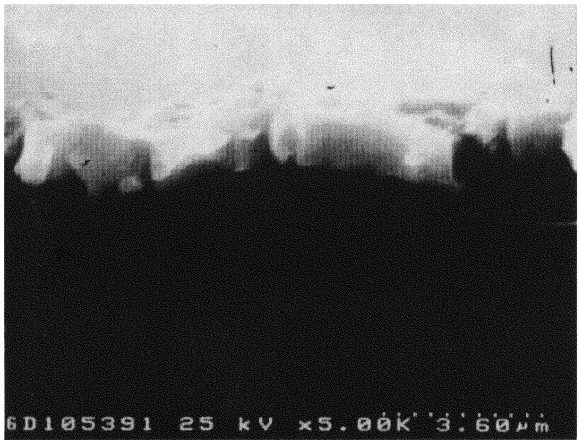


Fig.1. SEM photograph of an unpassivated Cu film on Si wafer.

Table 2. Relative intensities $\left(\frac{I(hkl)}{I(111)} \times 100\right)$ of XRD peaks for Cu films on different substrates and the data supplied by the JCPDS.

(hkl)	Relative intensity			
	Cu/Ti	Cu/Steel	Cu/Ni	Data of JCPDS
(111)	100.0	100.0	100.0	100.0
(200)	27.5	24.8	26.7	46.0
(220)	8.9	10.6	11.3	20.0
(311)	10.4	8.6	9.8	17.0

The grain sizes of the unpassivated Cu films on three different substrates are measured by TEM. The results are shown in Fig.2, in which the Cu films deposited on Ti, steel and Ni substrates are represented by Cu/Ti, Cu/steel and Cu/Ni respectively. It can be seen that the grain sizes do not vary markedly with different substrates, and the average grain size is about 0.7 μm, nearly equal to a half of the film thickness (1.340 μm).

3.2. Yield strengths

Figure 3 shows the variation of the x-ray measured stress with the applied strain for an unpassivated Cu film on Ni substrate. The residual stress (point A) is a compressive stress ($\sigma_{\mathrm{f}}^{\mathrm{r}} = -159.23$ MPa). In the elastic region (from point A to point B), the x-ray measured stress increases linearly with the applied strain, with a slope about 125 GPa which is quite close to the Young modulus of the Cu film. This indicates that the applied strain is transmitted unchanged across the interface from (top of) the substrate to (bottom of) the film.[13] As the x-ray stress goes beyond the elastic–plastic transition point (point B, termed here as yield point), it does not increase linearly with the applied strain but only increases slightly. The reason is that, the x-ray strain reflects only elastic strain, while the strain gauge shows the total (elastic and plastic) strain. It is noted that the yield strength of Cu film (351.31 MPa for point B) is much higher than that of bulk Cu (about 50 MPa).

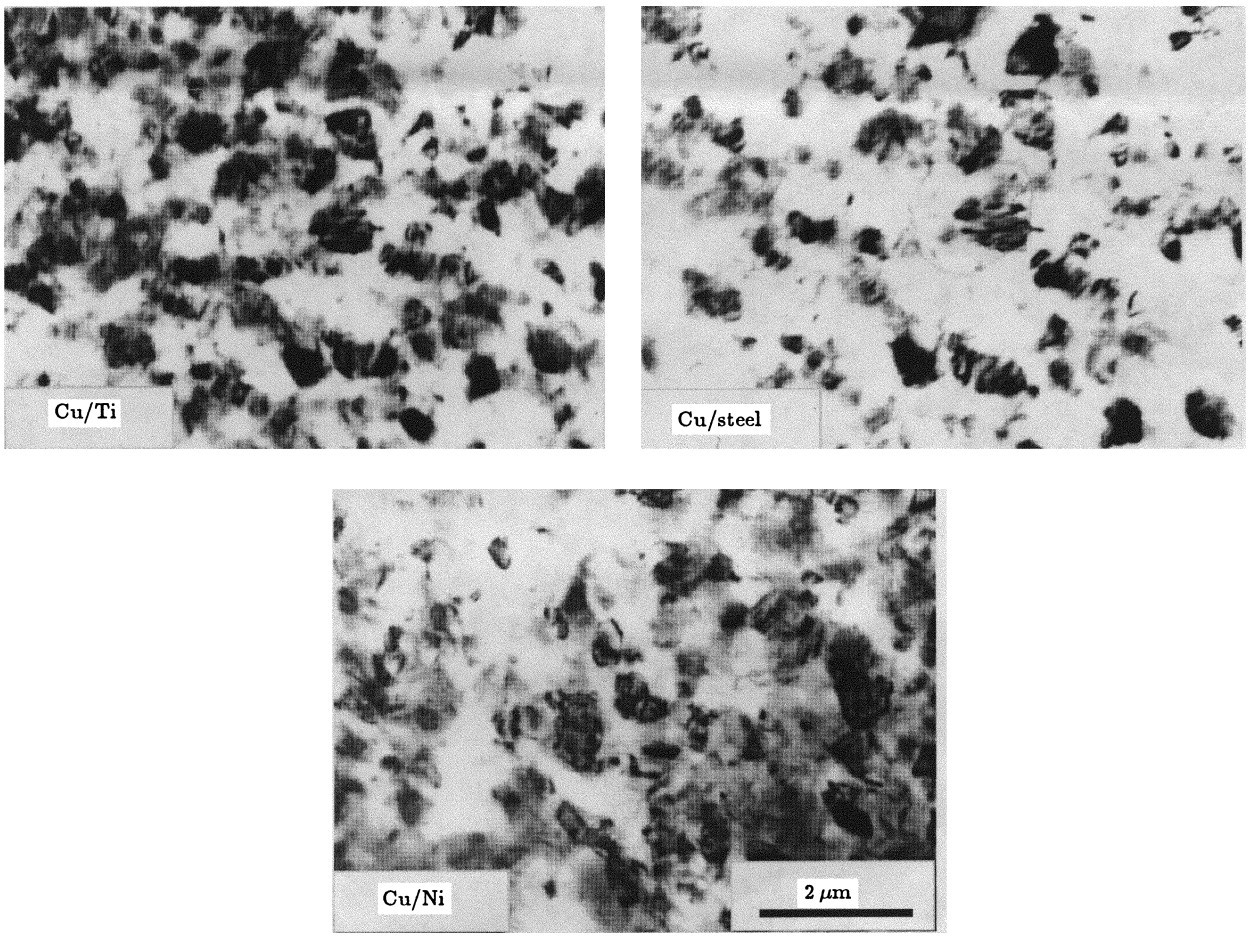


Fig.2. TEM micrographs for Cu films deposited on Ti, steel and Ni substrates.

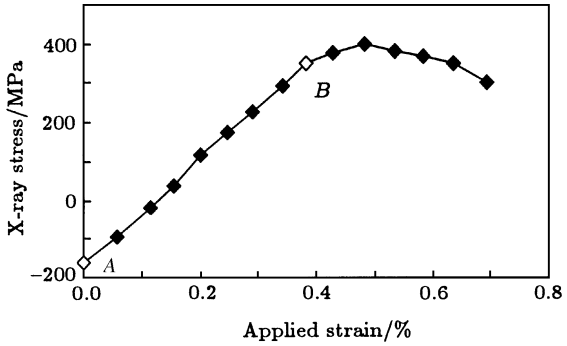


Fig.3. X-ray measured stress versus applied strain for an unpassivated Cu film on Ni substrate.

The measured results of tensile yield strengths for different samples are shown in Fig.4 together with the values calculated theoretically (see next section). It can be seen that, firstly, the experimental results are nearly consistent with the theoretical values, especially for passivated Cu films. Secondly, for the same substrate, the yield strength of the Cu film with a passivated layer is higher than that without a passivated layer. Similar results were also noted by Li *et al.*[8] They found that, for same thickness of Al film on Si substrate, the yield stress of the Al film with a 640 nm SiO_2 passivated layer was higher than that of the unpassivated Al film. Thirdly, for both of the passivated Cu films and unpassivated Cu films, the yield strength increases slightly for Ti, high-speed steel, and Ni substrates successively.

In a previous paper,[11] we assumed that a polycrystalline film was composed of columnar grains with an average grain diameter d in the plane of the film and height h (equal to the film thickness). An expression for the yield strength of the polycrystalline film on a substrate and that with a passivated layer is given by

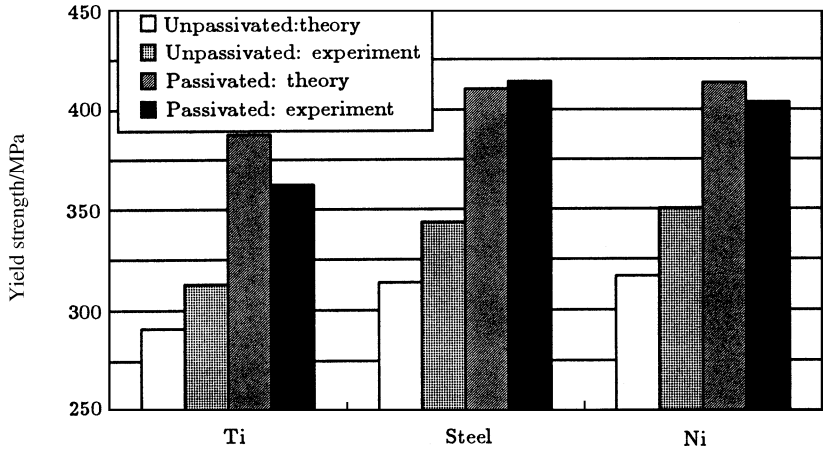


Fig.4. The yield strengths of passivated Cu films and unpassivated Cu films on Ti, high-speed steel, and Ni substrates.

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

where φ and λ are the included angles made by the glide plane normal direction with the glide direction of the dislocation and the film normal direction, respectively. b is the Burgers vector of the dislocation, υ_{f} is Poisson's ratio of the film, β is the included angle between the Burgers vector and the dislocation line; μ_{f}, μ_{s} and μ_{t} are the elastic shear moduli of the film, the substrate and the top (or passivated) layer, respectively, β_{s} and β_{t} are numerical constants depending on the materials that constitute the interfaces, and t is the thickness of the passivated layer.

This expression shows that the yield strength of a polycrystalline film attached to a substrate with a passivated layer depends on the following five factors.

(1) Orientation factor, $c_{(hkl)} = \dfrac{\sin\varphi}{\cos\varphi\cos\lambda}$, the subscripts (hkl) denote a particular orientation of the grain, that is, the grain with (hkl) plane oriented parallel to the film surface.

(2) The types of dislocation involved predominantly in the film, $c_{\mathrm{disl}} = \dfrac{b(1-\upsilon_{\mathrm{f}}\cos^2\beta)}{2\pi(1-\upsilon_{\mathrm{f}})}$, $c_{\mathrm{disl}} = \dfrac{b}{2\pi(1-\upsilon_{\mathrm{f}})}$, and $c_{\mathrm{disl}} = b$ correspond to the mixed, edge, and screw dislocations respectively.

(3) Substrate strengthening, $c_{\mathrm{sub}} = \dfrac{\mu_{\mathrm{f}}\mu_{\mathrm{s}}}{(\mu_{\mathrm{f}}+\mu_{\mathrm{s}})h} \times \ln\left(\dfrac{\beta_{\mathrm{s}}h}{b}\right)$.

(4) Passivated (or top) layer strengthening, $c_{\mathrm{top}} = \dfrac{\mu_{\mathrm{f}}\mu_{\mathrm{t}}}{(\mu_{\mathrm{f}}+\mu_{\mathrm{t}})h}\ln\left(\dfrac{\beta_{\mathrm{t}}h}{b}\right)$.

(5) Grain size strengthening, $c_{\mathrm{grain}} = \dfrac{1}{d}\dfrac{\mu_{\mathrm{f}}}{\sin\varphi} \times \ln\left(\dfrac{d}{b}\right)$.

In thin films with a face-centred cubic (FCC) structure, such as the Cu film considered here, the plastic deformation occurs as the slip of the dislocation occurs on the close-packed {111} or octahedral planes along the close packed $< 110 >$ direction. There are four equivalent octahedral planes each with three $< 110 >$ slip directions, giving 12 equivalent slip systems, as listed in Table 3.

Table 3. Twelve slip systems of the metals with a FCC structure.

$[\bar{1}10](111)$	$[1\bar{1}0](11\bar{1})$	$[110](1\bar{1}1)$	$[110](\bar{1}11)$
$[10\bar{1}](111)$	$[101](11\bar{1})$	$[10\bar{1}](1\bar{1}1)$	$[101](\bar{1}11)$
$[0\bar{1}1](111)$	$[011](11\bar{1})$	$[011](1\bar{1}1)$	$[01\bar{1}](\bar{1}11)$

For a thin film, the plastic deformation occurs when the film stress produces a resolved shear stress that surpasses the critical resolved shear stress (CRSS). Crystallographic orientation determines the resolved shear stress for a given film stress. The values of the $c_{(hkl)}$ have been calculated for the 12 low-index grain orientations, and the average values $C_{(hkl)}$ are listed in Table 4. Here the average values $C_{(hkl)}$ have been calculated from the 12 slip systems (Table 3) except infinite values.

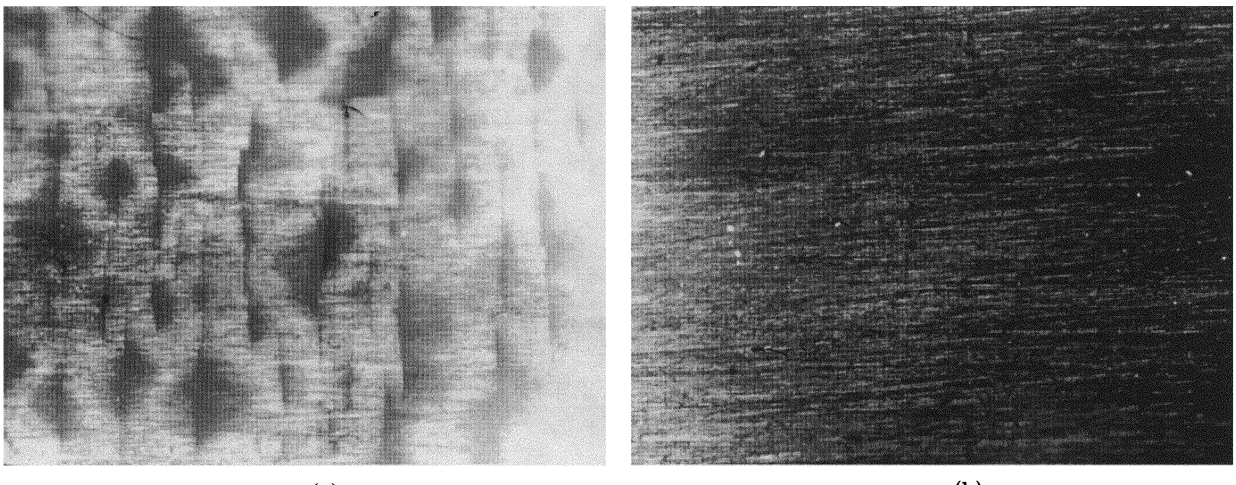
Table 4. Average values of the orientation factor for 12 grain orientations in FCC films.

Grain orientation (hkl)	(100)	(110)	(111)	(210)	(211)	(221)	(310)	(311)	(320)	(321)	(331)	(511)
Average values $C_{(hkl)}$	2.000	1.414	3.464	4.691	3.277	7.546	4.287	3.507	6.579	3.659	9.053	3.006

The effects of grain orientations on the stress, strain energy density, abnormal grain growth and texture in thin films have been discussed in previous papers.[14–21] For simplicity, taking $\beta_s = \beta_t = 1$ and $\beta = \frac{\pi}{2}$ (for edge dislocation), we have calculated the yield strengths of unpassivated Cu films and TiN passivated Cu films on Ti, steel and Ni substrates, and the results are shown in Fig.4. The following constants are used in this calculation: $C = \overline{C}_{(hkl)}$=4.374 (average values of Table 4), $b \approx a_{Cu} = 0.36\,\text{nm}$, $\upsilon_f = \upsilon_{Cu} = 0.364$, $\mu_f = \mu_{Cu} = 54.6\,\text{GPa}$, $\mu_{Ti} = 42.92\,\text{GPa}$, $\mu_{steel} \approx \mu_{Fe} = 86\,\text{GPa}$, $\mu_{Ni} = 94.7\,\text{GPa}$, $\mu_t = \mu_{TiN} = 258\,\text{GPa}$ (these values are taken from Ref.[22]), h=1.34 μm (Fig.1), d=0.7 μm (Fig.3).

3.3. Crackles

It is interesting to note that many crackles oriented perpendicular to the loading direction are observed in TiN passivated Cu film on Ti substrate, as shown in Fig.5, when the film stress is 372 MPa. However, the crackles are not observed in the passivated Cu films on steel and Ni substrates even if the film stress is as large as 400 MPa. Vinci *et al*[6] also found that for Si_3N_4 passivated Cu lines on Ta and Al substrates, after heating to 350 °C and then being cooled to 150 °C, no voids appeared in Cu lines on Ta substrate (Cu/Ta), but heavy voids were observed in Cu lines on Al substrate (Cu/Al). They thought that this was caused by a larger stress in Cu/Al (321 MPa) than in the Cu/Ta (180 MPa). According to their point of view, the crackles should be observed in the passivated Cu films on steel and Ni substrates since the film stress is 400 MPa (larger than 372 MPa, at which many crackles appeared). We think that the forming and expanding of crackles in thin films are decided by both the stress level and the shear modulus of the substrate. The higher the stress in thin films and the lower the shear modulus of the substrate, the easier the forming and expanding of the crackles. Appearance of the voids or crackles only in the passivated films is that the voids or crackles form first in the brittle passivated layer and result in a stress concentration in the underlying film. However, this process is restricted by the substrates, especially those with higher shear modulus. From this point of view together with the shear moduli of the substrates (26.5(Al), 42.92(Ti), 70.7(Ta), 86(Fe), 94.7(Ni), in units of GPa),[22] the appearance of voids in Si_3N_4 passivated Cu lines on Al substrates [6] and crackles in TiN passivated Cu film on Ti substrate can be explained.



(a) (b)

Fig.5. Optical photograph (×32) of the crackles in TiN passivated Cu film on Ti substrate (a), which are not seen on steel and Ni substrates (b).

4. Conclusions

The yield strength of the Cu film is found to increase obviously when a thin passivated layer is coated on its top surface, and to vary slightly for different substrates. The reason is that the substrate and the passivated layer have certain strengthening action, the higher the shear modulus, the stronger the strengthening action. The appearance of voids or crackles only in passivated films is because the voids or crackles form first in the brittle passivated layer and result in stress concentration in the underlying thin film. However, this process is restricted by the substrates, especially those with higher shear modulus.

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