Vol 14 No 9, September 2005
1009-1963/2005/14(09)/1862-04

Chinese Physics

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Dependence of stresses on grain orientations in BCC polycrystalline films on substrates*

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(Received 3 December 2004; revised manuscript received 25 May 2005)

Most thin films have different thermal expansion coefficients from their substrates, thus thermal stresses will be introduced into the films when the temperature is changed during annealing and service. Calculations of these stresses for grains in various crystallographic orientations have been made for seven BCC transition metals Cr, Fe, Mo, Nb, Ta, V and W. Neglecting W, which is isotropic and the stresses are equiaxial and without grain orientation (hkl) dependence, the BCC metals may be grouped into two classes. In the first class (Cr, Mo, Nb and V), the (100)-oriented grains have the largest stresses, while the stresses σ_1 and σ_2 in other (hkl)-oriented grains decrease linearly with the increase of the angle between (hkl) and (100), and with $\sigma_1 < \sigma_2$ except in (100)- and (111)-oriented grains. In the second class (Fe and Ta), on the contrary, the (100)-oriented grains have the lowest stresses, and the stresses σ_1 and σ_2 in other (hkl)-oriented grains increase linearly with the increase of the angle between (hkl) and (100), and with $\sigma_1 > \sigma_2$ except in (100)- and (111)-oriented grains.

Keywords: BCC metals, stresses, grain orientations, calculation
PACC: 6000, 6220, 6500

1. Introduction

Thin metal films have been used extensively in microelectronics and data storage devices because of their good electronic, magnetic and optical properties. However, because of the difference in both thermal expansion coefficients and thicknesses between the film and its substrate, large biaxial stresses will be introduced into the film, when the temperature is changed during device fabrication and in service. Stress relaxation by atom migration and/or dislocation glide can cause a number of reliability problems such as delamination,[1] voiding[2,3] or cracking,[4] and hillocking.[5,6] The materials properties are not isotropic but depend on a specific direction in question. The differences of the stresses between grains can also result in the atom diffusion across grain boundary and relative grain growth for differently oriented grains in polycrystalline films.[7,8]

In bulk polycrystalline materials, isotropic behaviour is obtained when the individual grains are oriented throughout space with equal probability in directions. However, in thin polycrystalline films this is usually not the case because of their nearly two-dimensional structure. In this paper, the stresses in the plane of the film have been calculated for various (hkl)-oriented grains in polycrystalline films of body-centred cubic (BCC) metals Cr, Fe, Mo, Nb, Ta, V and W. Neglecting W, which is isotropic and the stresses are equiaxial and without grain orientation (hkl) dependence, the remaining BCC transition metals may be grouped into two classes.

2. Stresses calculations

From elastic theory, the basic equation relating stress σ_{kl} to strain ε_{ij} is the well known Hooke's Law[9]

$$\varepsilon_{ij} = s_{ijkl}\sigma_{kl}, \tag{1}$$

where s_{ijkl} are the elastic compliance constants or, shortly, the compliances of the crystal. Equation (1) stands for nine equations, each with nine terms on the right-hand side. The 81 compliances s_{ijkl} form a

*Project supported by the State Key Development Programme for Basic Research of China (Grant No 2004CB619302), and the National Natural Science Foundation of China (Grant No 50271038).

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fourth-rank tensor and transform as follows:[9]

$$s'_{mnop} = a_{mi}a_{nj}a_{ok}a_{pl}s_{ijkl}. \tag{2}$$

The rotation matrix a_{rs} depends upon crystal orientation and therefore is more properly written as $a_{rs}^{(hkl)}$, the superscripts (hkl) have been left off in Eq.(2) for simplicity. Here (hkl) denotes a particular grain orientation, that is, the grains with (hkl) plane oriented parallel to the surface of the film. For (hkl)-oriented grains, the rotation matrix a_{rs} is derived as

$$a_{rs} = a_{rs}^{(hkl)} = \begin{bmatrix} a_{11} & a_{12} & a_{13} \\ a_{21} & a_{22} & a_{23} \\ a_{31} & a_{32} & a_{33} \end{bmatrix}$$

$$= \begin{bmatrix} \dfrac{k}{\sqrt{h^2+k^2}} & -\dfrac{h}{\sqrt{h^2+k^2}} & 0 \\ \dfrac{hl}{\sqrt{h^2+k^2}\sqrt{h^2+k^2+l^2}} & \dfrac{kl}{\sqrt{h^2+k^2}\sqrt{h^2+k^2+l^2}} & -\dfrac{\sqrt{h^2+k^2}}{\sqrt{h^2+k^2+l^2}} \\ \dfrac{h}{\sqrt{h^2+k^2+l^2}} & \dfrac{k}{\sqrt{h^2+k^2+l^2}} & \dfrac{l}{\sqrt{h^2+k^2+l^2}} \end{bmatrix}. \tag{3}$$

We use a mutually orthogonal coordinate system with the "1" and "2" axes in the plane of the film and the "3" axis normal to the film surface. From Eqs.(2) and (3), we can obtain compliances s'_{mnop} in (hkl) orientation from the values s_{ijkl}, which are often expressed in (001) orientation.

The symmetry of stress, strain and compliances s_{ijkl} in the first two and last two subscripts makes it possible to use the matrix notation. Both the stress components and the strain components can be written in a single subscript running from 1 to 6:

$$\begin{bmatrix} \sigma_{11} & \sigma_{12} & \sigma_{31} \\ \sigma_{12} & \sigma_{22} & \sigma_{23} \\ \sigma_{31} & \sigma_{23} & \sigma_{33} \end{bmatrix} \Leftrightarrow \begin{bmatrix} \sigma_1 & \sigma_6 & \sigma_5 \\ \sigma_6 & \sigma_2 & \sigma_4 \\ \sigma_5 & \sigma_4 & \sigma_3 \end{bmatrix}, \tag{4}$$

$$\begin{bmatrix} \varepsilon_{11} & \varepsilon_{12} & \varepsilon_{31} \\ \varepsilon_{12} & \varepsilon_{22} & \varepsilon_{23} \\ \varepsilon_{31} & \varepsilon_{23} & \varepsilon_{33} \end{bmatrix} \Leftrightarrow \begin{bmatrix} \varepsilon_1 & \frac{1}{2}\varepsilon_6 & \frac{1}{2}\varepsilon_5 \\ \frac{1}{2}\varepsilon_6 & \varepsilon_2 & \frac{1}{2}\varepsilon_4 \\ \frac{1}{2}\varepsilon_5 & \frac{1}{2}\varepsilon_4 & \varepsilon_3 \end{bmatrix}. \tag{5}$$

In the s_{ijkl}, the first two subscripts are abbreviated into a single one running from 1 to 6, and the last two are abbreviated in the same way, according to the following scheme:

Tensor notation :	11	22	33	23, 32	31, 13	12, 21
Matrix notation :	1	2	3	4	5	6

At the same time, factors 2 and 4 are introduced as follows:

$s_{ijkl} = s_{mn}$ when m and n are 1, 2 or 3;

$2s_{ijkl} = s_{mn}$ when either m or n is 4, 5 or 6;

$4s_{ijkl} = s_{mn}$ when both m and n are 4, 5 or 6.

The arrays of s_{ij} forms 6×6 squares. In the cubic system, only 3 (s_{11}, s_{12} and s_{44}) of the 36 components s_{ij} are independent.

$$s_{ij}^{cub} = \begin{bmatrix} s_{11} & s_{12} & s_{12} & 0 & 0 & 0 \\ s_{12} & s_{11} & s_{12} & 0 & 0 & 0 \\ s_{12} & s_{12} & s_{11} & 0 & 0 & 0 \\ 0 & 0 & 0 & s_{44} & 0 & 0 \\ 0 & 0 & 0 & 0 & s_{44} & 0 \\ 0 & 0 & 0 & 0 & 0 & s_{44} \end{bmatrix}. \tag{6}$$

Thus, for (hkl)-oriented grains (shown simply with prime as superscripts instead of (hkl)), Eq.(1) takes the shorter form

$$\varepsilon'_i = s'_{ij}\sigma'_j \quad (i, j = 1, 2, \cdots, 6). \tag{7}$$

using the Einstein summation convention: when a letter subscript occurs twice in the same term, summation with respect to that subscript is to be automatically understood. Solving Eq.(7) on the basis of equal strain assumption, i.e. $\varepsilon'_1 = \varepsilon'_2 = \varepsilon'$,[10] and neglecting the shear strains in the plane of the film, we find that

$$\sigma'_1 = \varepsilon' \frac{s'_{12} - s'_{22}}{(s'_{12})^2 - s'_{11}s'_{22}}, \tag{8}$$

$$\sigma'_2 = \varepsilon' \frac{s'_{12} - s'_{11}}{(s'_{12})^2 - s'_{11}s'_{22}}. \tag{9}$$

Hence it is necessary to calculate only s'_{11}, s'_{12} and s'_{22} for (hkl)-oriented grains from the values s_{11}, s_{12}

and s_{44}. It should be noted that, in spite of their appearance in matrix notation with two subscripts, the compliances s_{ij} are not the components, and so do not transform like the components, of a second-rank tensor. To transform them from (001) orientation to others it is necessary to go back to the tensor notation.

Using Eqs.(2) and (6) and the rules for transforming between the 2-subscript and the 4-subscript notation given above, one can find that

$$\begin{aligned} s'_{11} =& s_{11}[(a_{11})^4 + (a_{12})^4 + (a_{13})^4] + (2s_{12} + s_{44}) \\ & \times [(a_{11})^2(a_{12})^2 + (a_{11})^2(a_{13})^2 \\ & + (a_{12})^2(a_{13})^2], \end{aligned} \tag{10}$$

$$\begin{aligned} s'_{12} =& s_{11}[(a_{11})^2(a_{21})^2 + (a_{12})^2(a_{22})^2 + (a_{13})^2(a_{23})^2] \\ & + s_{12}[(a_{11})^2(a_{22})^2 + (a_{11})^2(a_{23})^2 \\ & + (a_{12})^2(a_{21})^2 + (a_{12})^2(a_{23})^2 \\ & + (a_{13})^2(a_{21})^2 + (a_{13})^2(a_{22})^2] \\ & + s_{44}(a_{12}a_{13}a_{22}a_{23} + a_{11}a_{13}a_{21}a_{23} \\ & + a_{11}a_{12}a_{21}a_{22}), \end{aligned} \tag{11}$$

$$\begin{aligned} s'_{22} =& s_{11}[(a_{21})^4 + (a_{22})^4 + (a_{23})^4] + (2s_{12} + s_{44}) \\ & \times [(a_{21})^2(a_{22})^2 + (a_{21})^2(a_{23})^2 \\ & + (a_{22})^2(a_{23})^2], \end{aligned} \tag{12}$$

where rotation matrix a_{rs} depends upon the grain orientation (hkl) as expressed in Eq.(3). By substituting for a_{rs} and the values of compliances s_{11}, s_{12} and s_{44} (listed in Table 1 together with anisotropy factor $A = 2C_{44}/(C_{11} - C_{12})$ used to analyse the calculated results)[11] into Eqs.(10)–(12), then into Eqs.(8) and (9), the stresses σ'_1 and σ'_2 in the plane of the film are obtained for various (hkl)-oriented grains in a polycrystalline film of BCC metals Cr, Fe, Mo, Nb, Ta, V and W. The results of the calculations are listed in Table 2 by assuming the thermal strain $\varepsilon' = 0.1\%$. Variations of stresses σ_1 (dashed line) and σ_2 (solid line) in various (hkl)-oriented grains with the including angle between (hkl) and (100) are shown in Fig.1 for Cr, Fe, Mo, Nb, Ta and V.

Table 1. Values of compliances s_{11}, s_{12} and s_{44} (in units of $10^{-11}\mathrm{m}^2/\mathrm{N}$) used in the calculation and the anisotropy factor A for BCC metal Cr, Fe, Mo, Nb, Ta, V and W[11].

materials	Cr	Fe	Mo	Nb	Ta	V	W
s_{11}	0.300	0.760	0.280	0.660	0.686	0.683	0.244
s_{12}	–0.043	–0.287	–0.078	–0.233	–0.258	–0.234	–0.068
s_{44}	0.992	0.892	0.910	3.480	1.212	2.348	0.624
A	0.690	2.360	0.775	0.510	1.560	0.780	1.000

Table 2. Biaxial stresses $\sigma_1^{(hkl)}$ and $\sigma_2^{(hkl)}$ (in units of $10^7\mathrm{N}/\mathrm{m}^2$) in the plane of the film as a function of grain orientation (hkl) in thin polycrystalline film composed of BCC metals.

(hkl)	Cr		Fe		Mo		Nb		Ta		V		W	
	$\sigma_1^{(hkl)}$	$\sigma_2^{(hkl)}$	$\sigma_1^{(hkl)}$	$\sigma_2^{(hkl)}$	$\sigma_1^{(hkl)}$	$\sigma_2^{(hkl)}$	$\sigma_1^{(hkl)}$	$\sigma_2^{(hkl)}$	$\sigma_1^{(hkl)}$	$\sigma_2^{(hkl)}$	$\sigma_1^{(hkl)}$	$\sigma_2^{(hkl)}$	$\sigma_1^{(hkl)}$	$\sigma_2^{(hkl)}$
(100)	38.91	38.91	21.14	21.14	49.50	49.50	23.42	23.42	23.36	23.36	22.27	22.27	56.82	56.82
(110)	30.87	37.76	39.24	27.97	41.68	47.32	13.51	19.92	32.77	26.90	18.36	20.93	56.82	56.82
(111)	32.47	32.47	36.67	36.67	42.67	42.67	14.10	14.10	31.71	31.71	18.70	18.70	56.82	56.82
(210)	33.35	38.11	29.99	24.48	44.19	48.03	15.94	20.78	28.62	25.34	19.60	21.36	56.82	56.82
(211)	33.65	34.09	31.37	30.62	44.02	44.38	15.47	15.84	29.31	28.92	19.40	19.57	56.82	56.82
(221)	31.36	35.07	38.98	31.19	41.87	45.01	13.51	16.56	32.71	28.88	18.38	19.81	56.82	56.82
(310)	35.58	38.43	25.35	22.73	46.37	48.63	18.53	21.69	26.06	24.38	20.68	21.73	56.82	56.82
(311)	35.49	35.68	26.52	26.30	45.96	46.11	17.76	17.95	26.74	26.60	20.40	20.47	56.82	56.82
(320)	31.85	37.90	34.83	26.31	42.68	47.60	14.42	20.24	30.93	26.21	18.85	21.10	56.82	56.82
(321)	32.08	35.96	35.10	28.62	42.69	45.91	14.28	17.69	31.08	27.64	18.80	20.27	56.82	56.82
(322)	32.69	33.20	35.10	33.92	42.97	43.41	14.41	14.81	31.05	30.48	18.87	19.07	56.82	56.82
(331)	31.10	36.33	39.17	29.49	41.75	46.11	13.48	18.03	32.77	27.86	18.36	20.34	56.82	56.82
(332)	31.69	33.91	38.54	33.18	42.06	43.97	13.62	15.36	32.53	30.00	18.45	19.31	56.82	56.82
(410)	36.79	38.61	23.55	22.05	47.53	48.95	20.15	22.26	24.95	23.96	21.27	21.93	56.82	56.82
(411)	36.64	36.73	24.33	24.24	47.16	47.23	19.44	19.53	25.43	25.38	21.03	21.06	56.82	56.82
(421)	33.42	36.67	30.43	26.17	44.10	46.75	15.71	18.80	28.86	26.40	19.51	20.72	56.82	56.82
(510)	37.47	38.70	22.69	21.73	48.17	49.13	21.13	22.61	24.40	23.75	21.59	22.04	56.82	56.82
(511)	37.33	37.38	23.22	23.19	47.88	47.91	20.54	20.59	24.74	24.72	21.41	21.42	56.82	56.82

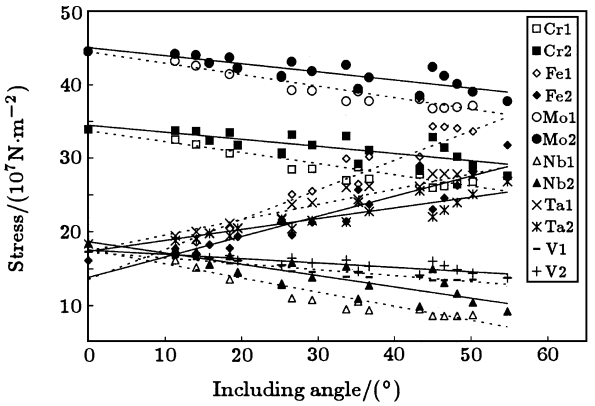


Fig.1. Variation of the film plane stresses σ_1 (dashed lines) and σ_2 (solid lines) in various (hkl)-oriented grains with including angle between (hkl) and (100) for BCC metal Cr, Fe, Mo, Nb, Ta and V.

3. Conclusions

The following conclusions can be obtained from the calculated results:

(1) For W, which is completely isotropic ($A = 1$), the stresses in the plane of the film are equiaxial and without grain orientation (hkl) dependence.

(2) For Cr, Fe, Mo, Nb, Ta and V metals, only in (100)- and (111)-oriented grains, the stresses in the plane of the film are equiaxial and this is not the case in most other oriented grains. The difference between σ_1 and σ_2 in other oriented grains increases with increasing deviation of the anisotropic factor A of the metal from 1, being small in V ($A = 0.78$) and large in Fe ($A = 2.36$).

(3) The remaining BCC metals other than W may be grouped into two classes. In the first class (Cr, Mo, Nb and V, with anisotropy factor $A < 1$), (100)-oriented grains have the largest stresses in the plane of the film and the stresses σ_1 and $\sigma_2 (> \sigma_1)$ in most other (hkl)-oriented grains decrease linearly with the increase of the angle between (hkl) and (100). In the second class (Fe and Ta, with $A > 1$), on the contrary, the (100)-oriented grains have the lowest stresses in the plane of the film and the stresses σ_1 and σ_2 $(< \sigma_1)$ in most other (hkl)-oriented grains increase linearly with increasing angle between (hkl) and (100).

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