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Evaluation of residual stresses in Cu/Mo, Cu/Nb nanomultilayers with a strong fiber texture

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ABSTRACT

Residual stresses are one of the key factors for tuning the properties, microstructure, and reliability of thin films and nanomultilayers, but their measurement and evaluation are challenging. Residual stresses in nanomultilayers with {1 1 0} out-of-plane texture exhibit a dependence on the in-plane crystal orientation, which complicates their evaluation using X-ray diffraction. The texture and residual stresses were investigated for two representative nanomultilayers of immiscible materials with fcc/bcc structure: Cu/Mo and Cu/Nb grown on Si substrate with an amorphous silicon nitride layer. Both multilayered structures exhibited Cu {1 1 1} // Mo, respectively Nb {1 1 0} out-of-plane fiber texture, and showed compressive stress. A modified crystallite group method for {1 1 0}, {1 1 1} fiber texture was used to determine residual stresses in the nanomultilayers. The method was proven to be a good tool to extract the residual stress in nanomultilayers with a strong fiber texture.

1. Introduction

The stress level in thin films plays a decisive role in the reliability and material properties. High residual tensile or compressive stress may lead to buckling or delamination of the film [1]. Moreover, tuning the residual stresses of a nanomultilayer (NML) is also an effective way to tailor thermal stability and conductivity [2]. For these reasons, analyzing residual stress in NMLs is a powerful tool to understand the material properties, reliability, and microstructure of nano-scaled multilayer films.

The experimental techniques used for residual stress measurements in thin films such as curvature measurement and deformation techniques, X-ray diffraction (XRD), neutron diffraction and specialized methods are concisely reviewed [3] and have been succesfully applied to various thin films and nanomultilayers [4–9]. Nonetheless, the measurement of residual stress remains still a challenging task, especially in the case of strongly textured, complex multiphase, nanocrystalline, or amorphous materials and films [3].

In this study, XRD method was selected to approach the residual stresses in NMLs with a strong texture. Especially, the determination of residual stresses in thin films with texture along [1 1 0] is demanding, because {1 1 0} planes of a cubic crystal are mechanically non-isotropic and the residual stresses depend on in-plane crystallographic orientation. For this class of NMLs the residual stresses cannot be analyzed by a classical $\sin^2\Psi$ method, since it is applicable only for polycrystalline untextured samples [10]. In this situation, Crystallite Group Method (CGM), which assumes that all the crystallites with the same orientation define a crystallite group and can be treated as one crystal [11] offers a solution. In the present work, a modified CGM for anisotropic layers with fiber texture [12] was applied to derive the in-plane residual stress of two immiscible metal systems of fcc/bcc structures. The chosen model systems are Cu/Mo, and Cu/Nb NMLs grown on Si substrate with amorphous silicon nitride overlayer.

2. Material and methods

Cu/Mo and Cu/Nb NMLs were prepared by DC magnetron sputtering. Si (0 0 1) substrates with a 90 nm-thick amorphous silicon nitride were used as substrates for deposition. The substrates were ultrasonically cleaned with acetone, ethanol, and isopropanol for 3 min consecutively, followed by Ar drying after each solvent cleaning. RF cleaning process was performed before the deposition with Ar pressure 2 mTorr at 50 W power for 2 min. DC magnetron sputtering in a high vacuum chamber (base pressure ≤ 10^{-8} mbar) was used for deposition of Cu/Mo, Cu/Nb NMLs with gun power 80 W and Ar pressure 2 mbar. Cu_{10nm}-Mo_{10nm} and respectively Cu_{10nm}-Nb_{10nm} bi-layer structures were

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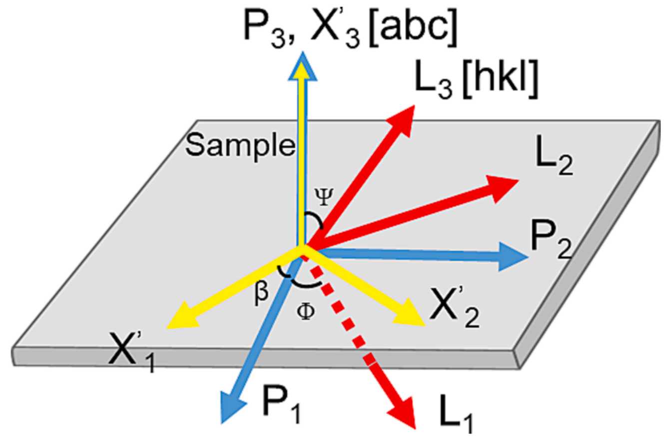

Fig. 1. Coordinates of specimen P, crystal X', and laboratory L. The 3 axes are indicated by blue, yellow, and red arrows respectively on the sample.

repeated 10 times to make total 200 nm thick layers.

TEM (Transmission electron microscopy) lamellas were prepared with an FEI Helios 660 FIB/SEM (Focused ion beam / Scanning electron microscopy) with a 2, 5, 30 kV Ga ion beam. Scanning transmission electron microscopy (STEM) was performed with JEOL2200FS operating at 200 kV. A Bruker D8 Discover X-ray diffractometer was used to measure the texture and residual stress of samples. Diffraction patterns were recorded using Cu target at 40 kV and 40 mA. Since the modified method utilizes fiber texture [12], the residual stress can be measured by averaging the in-plane dependence term considering all the crystallographic orientations contributing to diffraction.

For the derivation of the equation for the residual stress, it was postulated that all the NML components have equibiaxial in-plane residual stress. Since the deposition process occurred at room temperature (RT), and the substrate temperature remained below 50 °C throughout, it is assumed that the effect of thermal stresses is negligible. In particular, for Cu thin films deposited at room temperature Pletea et al. [13] reported that the contribution of thermal stresses is approximately 4% of the total stress-thickness and thus can be neglected.

Reuss model is adopted which assumes that crystallite possesses identical stress tensors, from which an equation $<\sigma> = \sigma$ (1) can be obtained. With these assumptions, in the case of a cubic crystal, {110} out-of-plane texture, residual stress can be determined by the following equation [12]:

$$<\epsilon_{33}^{L}> = \left[\left(\frac{s_{44}}{2} + \frac{s_0(1+ <\cos 2(\Phi+\beta)>)}{4}\right)\sin^2\Psi + \frac{1}{2}(4s_{12}+s_0)\right]\sigma \quad (2)$$

where σ is in-plane residual stress, ϵ_{33}^{L} is lattice strain in laboratory coordinates, and s_{11}, s_{12}, s_{44} are compliance coefficients of cubic crystal and bracketed terms refer to an average over crystallites oriented properly to contribute to the diffraction peak, Φ, and β are rotational angles for the transformation of the coordinates (see Fig. 1). Equation (2) was derived by setting the crystal coordinates as $X_1' = [00\overline{1}]$, $X_2' = [\overline{1}10]$, $X_3' = [110]$. By setting $\Phi = 0$, $<\cos 2\beta>$ depends on the diffraction plane and inclination angle of Ψ. With the crystal coordinates set to derive equation (2), the crystallographic planes (200) and (020) have coordinates $\Psi = 45°$ and $\beta = -90°, 90°$, respectively. This can be checked by (110) stereographic projection. Then, $<\cos 2\beta> = \frac{\cos 90° + \cos -90°}{2} = 0$ is used in the equation. The elastic response of materials with a cubic crystal structure and {111} out-of-plane texture does not show this complexity, since it does not depend on in-plane orientation. The detailed methodologies for the determination of residual stresses applied in this study, including {111} fiber texture, are discussed in Supplementary Data.

3. Results and discussion

Fig. 2 shows cross-sectional bright-field scanning transmission

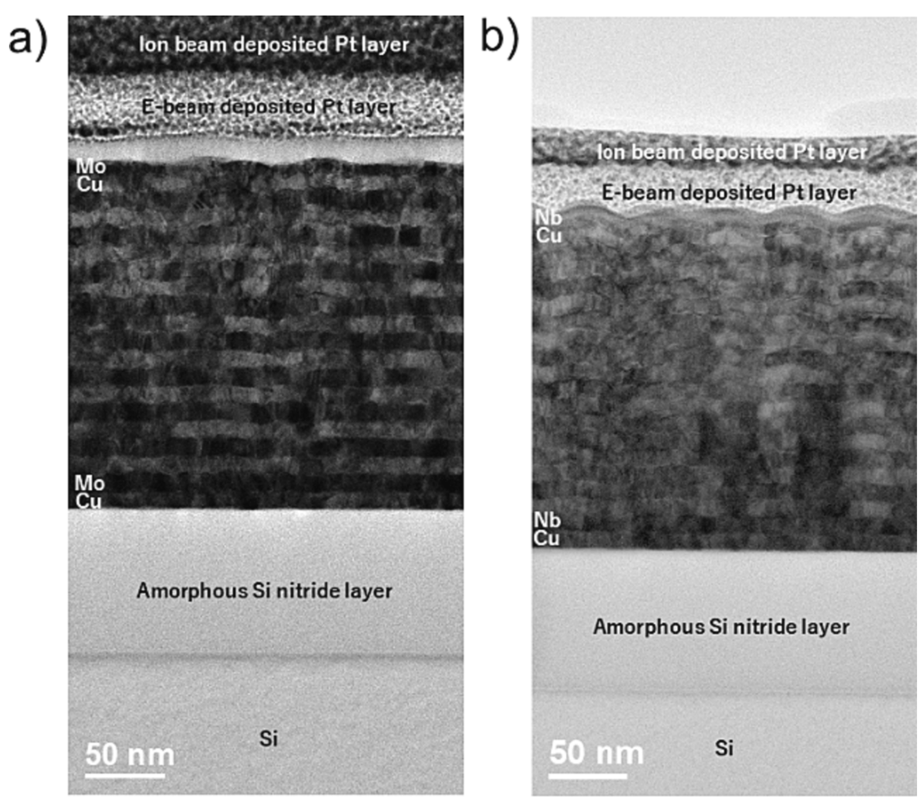

Fig. 2. Bright-field STEM images of NMLs: a) Cu/Mo b) Cu/Nb.

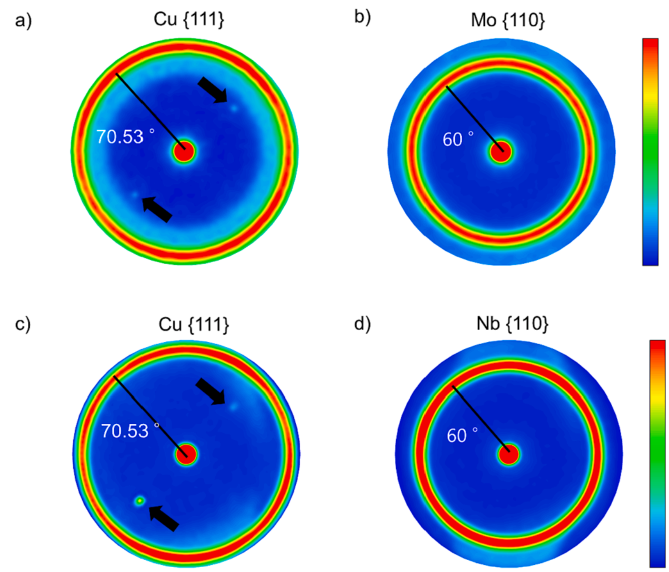

Fig. 3. Pole figure of Cu/Mo, Cu/Nb NMLs: a), b) Cu {1 1 1}, Mo {1 1 0} c), d) Cu {1 1 1}, Nb {1 1 0}.

Table1
Residual stresses in nanomultilayers.

	Cu/Mo NML	Cu/Nb NML	Cu/W NML [2]
FCC(Cu)	-0.532 ± 0.35 GPa	-0.274 ± 0.13 GPa	-1.8 ± 0.4 GPa
BCC(Mo/Nb/W)	-0.298 ± 0.37 GPa	-0.628 ± 0.39 GPa	-3.84 ± 0.09 GPa

electron microscope (BF-STEM) images of Cu/Mo and Cu/Nb NMLs. Both NMLs exhibited a 10 nm−10 nm double-layer stack repeated 10 times without a large deviation of layer thickness. The initially deposited layers are flat and planar, however, their become roughened as the NMLs grow.

Fig. 3 shows the pole figures of fcc Cu {1 1 1}, bcc Mo {1 1 0}, and bcc Nb {1 1 0}. Fig. 3a and b indicate that Cu/Mo NML exhibits Cu {1 1 1} // Mo {1 1 0} fiber texture. The amorphous silicon nitride blocked the epitaxial growth on Si. The (red) center of the two pole figures shows Cu {1 1 1} // Mo {1 1 0} out-of-plane texture, and the homogeneous (red) ring structure ($\Psi = 70.53, 60^{\circ}$ in Cu and Mo pole figures, respectively. See Figs. S2, S3) indicates the in-plane random crystallographic orientation. The black arrows in Fig. 3a result from the Si peak. Similarly, Fig. 3c, and d display the Cu {1 1 1} // Nb {1 1 0} fiber texture of Cu/Nb NML.

Table 1 lists the residual stresses of the investigated NMLs (including Cu/W NML [2] for comparison) as determined from Figs. S4, S5. All components of NMLs exhibit compressive stresses. Sufficient power and reduced Ar-pressure increase the target atom's kinetic energy, intensifying atomic peening or adatom diffusion into film grain boundaries leading to compressive residual stress [14]. The compressive stress of Mo (in Cu/Mo) and Nb (in Cu/Nb NML) is considerably lower than that of Cu in Cu/W [2], which resulted from higher target power during the deposition. Since W is low diffusion atomic species, compressive growth stress increased through the atomic peening effect. The large compressive residual stress of stiff W is then transmitted to the much softer copper nanolayers, resulting in a superposition of stresses.

4. Conclusions

This work presents a methodology to extract residual stress in anisotropic bcc materials with strong texture. Two NMLs (Cu/Mo and Cu/Nb) were prepared using magnetron sputtering. Both exhibited fiber texture with the orientation relationship: fcc Cu {1 1 1} // bcc Mo, Nb {1 1 0} out-of-plane. The NMLs show a significant variation of the in-plane compressive stress depending on the bcc metals present. This analysis is useful to extract the stress state in thin films and multilayers which is critically affecting and tailoring material properties.

CRediT authorship contribution statement

Jeyun Yeom: Conceptualization, Methodology, Data curation, Writing – original draft, Writing – review & editing, Visualization. **Giacomo Lorenzin:** Methodology, Writing – review & editing. **Claudia Cancellieri:** Methodology, Data curation, Writing – review & editing. **Jolanta Janczak-Rusch:** Funding acquisition, Supervision, Writing – review & editing.

Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Jolanta Janczak-Rusch reports financial support was provided by Swiss National Science Foundation. Giacomo Lorenzin reports financial support was provided by Swiss National Science Foundation.

Data availability

No data was used for the research described in the article.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.matlet.2023.135074.

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