Interfacial Structures Governed Plastic Deformation Behaviors in Metallic Multilayers

Ming-Zhen Wei^{1,2} · Zhen-Hua Cao¹ · Xiang-Kang Meng¹

Received: 25 November 2015 / Revised: 8 January 2016 / Published online: 8 February 2016
© The Chinese Society for Metals and Springer-Verlag Berlin Heidelberg 2016

Abstract In this work, we have investigated the mechanical properties of Cu/Ta, Ag/Cu and Ag/Nb multilayers with different heterogeneous interfaces. The results suggest that when individual layer thickness (h) is larger than 5–10 nm, the hardness/strength of three different multilayer systems has the similar length scale effect with decreasing layer thickness, while when $h \leq 5$ nm, the three multilayer systems show remarkably different plastic deformation behaviors. The strength curves exhibit the variation trends of unchanging, softening and increasing corresponding to Cu/Ta, Ag/Cu and Ag/Nb multilayers, respectively. The microstructure analysis shows that three kinds of multilayers have totally different interfacial structures, which lead to the different strengthening or softening mechanisms.

KEY WORDS: Metallic multilayers; Plastic deformation; Interfacial structures; Strengthening mechanism

1 Introduction

Metallic multilayer films have attracted much attention due to their outstanding mechanical properties [1–4]. The yield strengths of some multilayers can even approach 1/3–1/2 of the theoretical strength limit [5, 6]. The strengthening mechanisms of multilayers have been investigated extensively, and three typical mechanisms have been developed to interpret the plastic deformation behaviors in multilayers. Hall–Petch (H–P) model based on pilling-up of dislocations against the interface or grain boundaries is applicable when the individual layer thickness (h) is more

than tens of nanometers [7–9]. At smaller h of a few to tens of nanometers, the confined layer slip (CLS) mechanism involving the single dislocation loops gliding in both layers dominates the plastic deformation [10–12]. When h decreases to several nanometers, the strengthening is mainly ascribed to the interface barrier strength (IBS) to the transmission of a single dislocation [13–15].

The heterogeneous interfaces between layers play a key role in the strengthening of multilayers [16–18]. Different types of interfaces provide different barrier effects to the transmission of deformation. The interfaces can be classified as coherent, semi-coherent and incoherent, depending on the lattice parameters of the constituents of multilayer systems [19–21]. Many studies in multilayers, such as Cu/V [22], Co/Cu [23], Cu/Ni [3, 24], Cu/Zr [25, 26], Cu/Nb [27] and Cu/Fe [28] multilayers, have shown that the strength of multilayers increases with decreasing layer thickness and reaches plateau when h decreases to several nanometers. However, with further investigation, the contradictory plastic deformations have been reported when h is lower than 5 nm. Some metallic multilayers show a continuous strengthening even when h decreases to 1–2 nm [29, 30], while in other multilayers the softening occurs at

Available online at <http://link.springer.com/journal/40195>

✉ Zhen-Hua Cao
caozhenhua@nju.edu.cn

¹ Institute of Materials Engineering, National Laboratory of Solid State Microstructures, College of Engineering and Applied Sciences, Nanjing University, Nanjing 210093, China

² Department of Physics, School of Science, Linyi University, Linyi 276000, China

~5 nm. These different plastic deformation behaviors are closely related to the interfacial structures and the interaction between interfaces and partial dislocations. Hence, it is significant to investigate the interfacial structures and how the different interfaces acting as barriers to dislocation glide, which is helpful to achieve the optimum selection of multilayer systems.

In the present work, three different multilayer systems Cu/Ta, Ag/Cu and Ag/Nb have been chosen to investigate the relationship between strengthening and the interfacial structures. Three typical crystal lattices, including face-centered cubic (fcc), body-centered cubic (bcc) and tetragonal (tet) structures, are employed. Cu/Ta, Ag/Cu and Ag/Nb multilayers are the composite systems of fcc/tet, fcc/fcc and fcc/bcc, respectively. A comparison of the systems studied is shown in Table 1. In our previous work, we have reported the ultrahigh strengthening behavior in Ag/Nb multilayers [30] and the abnormal softening behavior in Cu/Ta multilayers [4, 31]. However, the further researches are still needed on how the characteristics of interfaces, including the crystallinity, the structural discontinuity and crystal orientation between layers, affect the mechanical properties of multilayers. Here we systematically studied the heterogeneous interface governed plastic deformation behaviors of metallic multilayers.

2 Experimental

Cu/Ta, Ag/Cu and Ag/Nb multilayers were deposited on Si substrates at room temperature by dc magnetron sputtering. The individual layer thickness ranged from 1 to 50 nm for each multilayer system, and both of constituent layers were equal with each other. Base pressure for the sputtering chamber was kept at less than 9×10^{-5} Pa, and the sputtering pressure was 1.2 Pa. Total thickness of the multilayers was 1000 nm. For comparison, the pure Cu, Ta, Ag and Nb thin films with the thickness of 1000 nm were prepared under the same circumstances.

The cross-sectional microstructures were characterized by using a JEOL JEM-2100 transmission electron microscope (TEM) with 200-kV accelerating voltages. The nanoindentation tests were performed using a TI950

Hysitron Triboindenter with a displacement resolution of 0.1 nm and a loading resolution of 50 nN. A Berkovich tip with 50 nm radius of curvature was used in all indentation experiments. The maximum indentation depth was controlled at 150 nm (about 15% of the multilayer thickness) to minimize the effects of substrate, and the same ramp rate $\dot{P}/P = 0.1 \text{ s}^{-1}$ was adopted when the nanoindentation tests of Cu/Ta, Ag/Cu and Ag/Nb multilayers were performed.

3 Results and Discussion

The typical load–displacement curves of Cu/Ta, Ag/Cu and Ag/Nb multilayers with $h = 5$ nm are shown in Fig. 1. These curves reflect the differences in hardness of the three types of multilayers. At the same maximum indentation depth of 150 nm, the higher load means the higher hardness. From Fig. 1, we can conclude that the hardness of Cu/Ta multilayer is the highest while that of Ag/Cu multilayer is the lowest when $h = 5$ nm. All the hardness plotted as a function of h of Cu/Ta, Ag/Cu and Ag/Nb multilayers with h in the range of 1–100 nm (3–100 nm for Ag/Cu) is shown in Fig. 2a. Meanwhile, in order to explore the strengthening mechanisms of the multilayers and observe the variation trends more clearly when $h \leq 5$ nm, the strength plotted as a function of $h^{-1/2}$ is shown in Fig. 2b. The strength is estimated as indentation hardness divided by a factor of 2.7 [32]. It can be seen that the hardness/strength of three multilayer systems changes with decreasing h and shows different length scale effects. The strength of Cu/Ta multilayers increases with h and reaches a peak at $h = 10$ nm. A sharp decrease can be obviously observed when h is in the range of 5–10 nm. It comes from the phase transformation in Ta layers, which has been reported in our previous work [31]. The strength in Ag/Cu

Fig. 1 Typical load–displacement curves of Cu/Ta, Ag/Cu and Ag/Nb multilayers with $h = 5$ nm

Table 1 Comparison of three different multilayer systems

Multilayers	Crystal lattice	Interface misfit (%) ^a	Interface type
Cu/Ta	fcc/tet	–	Incoherent
Ag/Cu	fcc/fcc	12	Semi-coherent
Ag/Nb	fcc/bcc	1	Coherent

^a Interface misfit indicates the misfit of interplanar spacing between close-packed planes, i.e., Ag(111) and Cu(111) for fcc/fcc, Ag(111) and Nb(110) for fcc/bcc, respectively

Fig. 2 **a** Hardness of Cu/Ta, Ag/Cu and Ag/Nb multilayers plotted as a function of h ; **b** strength plotted as a function of $h^{-1/2}$

multilayers increases with decreasing h in the range of 5–100 nm and softens at $h < 5$ nm, while in Ag/Nb multilayers, the strength increases with decreasing h and shows a sharply rising trend even when h decreases to 1 nm. By comparing three different strength curves, we find that the differences mainly exist in the range of 1–5 nm. When h is larger than 5–10 nm, the trends are similar except that the strength of Ag/Cu multilayer with $h = 50$ nm is higher than that with $h = 20$ nm, which has been explained in the Ref. [33]. When $h \leq 5$ nm, the three strength curves have the different variation trends of unchanging, softening and increasing corresponding to Cu/Ta, Ag/Cu and Ag/Nb multilayers, respectively.

In order to clearly reveal the effect of the interfacial structures on the mechanical properties, we investigated the cross-sectional microstructures of three different multilayers with $h = 5$ nm. Figure 3 gives the cross-sectional TEM and high-resolution TEM (HRTEM) images of Cu/Ta multilayers with $h = 5$ nm, respectively. From

Fig. 3a, it can be seen that an alternative lamellar structure is clearly visible and the interface is sharp and straight. No obvious columnar grains can be observed. From the HRTEM image of Fig. 3b, the lattice fringes can be seen in Cu layers, indicating the Cu grains well crystallite. The fringes in Ta layers are slightly random which indicates the crystallinity is weak and only tiny nanograins form. In our previous works [4, 31], we have reported that the tetragonal β -Ta and cubic α -Ta coexist in Ta layers when $h > 5$ nm, while only α -Ta forms when $h \leq 5$ nm, which may be the reason why the hardness/strength drops sharply with decreasing h from 10 to 5 nm. In fact, the crystal α -Ta forms at some special sites with straight interface and generates exactly on the neat Cu(111) plane [31]. The selected area electron diffraction (SAED) pattern of the Cu/Ta multilayer with $h = 5$ nm is shown in the inset of Fig. 3a, and all of the diffraction rings can be identified as reflections from Cu phase and α -Ta phase, respectively.

Fig. 3 Bright field cross-sectional TEM **a** and HRTEM **b** images of Cu/Ta multilayer with $h = 5$ nm (the light gray layers indicate the Cu layers, and the dark gray layers indicate the Ta layers; the inset in **a** is the corresponding SAED pattern)

Figure 4 presents the cross-sectional TEM and HRTEM images of Ag/Cu multilayers with $h = 5$ nm. They reveal the morphology of regular interface interrupted by the periodical defect regions to reduce the mismatch strain, exhibiting a typical semi-coherent interface. The width of column grains is larger than layer thickness. The interface is a little wavy, and the columns are successive along the growth direction of multilayer. The inset in Fig. 4a is the corresponding SAED pattern of the cross section with $h = 5$ nm. The diffraction rings can be identified as reflections from Ag and Cu phases which can indicate the orientation relationship between Ag and Cu. From HRTEM image of Fig. 4b, we can see that the consecutive atomic arrangement extends from Ag layer to Cu layer across the interface, suggesting the epitaxial relationship between them. It is confirmed that the multilayer has a strong orientation relationship of a cube-on-cube growth. Meanwhile, there are many stacking faults growth from Ag layer to Cu layer across the interface, which can improve the barrier strength of interfaces.

Figure 5 shows the cross-sectional TEM and HRTEM images of Ag/Nb multilayer with $h = 5$ nm. The lamellar structure can be easily identified. The interfaces are straighter than Ag/Cu multilayers. The inset in Fig. 5a is the corresponding SAED pattern of Ag/Nb multilayer with $h = 5$ nm, which indicates the orientation relationship between Ag and Nb. From the HRTEM image of Fig. 5b, we can see that the interfacial morphology is very different from what observed in Cu/Ta and Ag/Cu multilayers. The interfaces in Ag/Nb system are more ordered than the above two, exhibiting a coherent interfacial structure. The parallel lattice fringes can be seen in both Ag and Nb layers, indicating that the grains in-plane have the strong textures. The values of the interplanar spacings in Ag (0.236 nm) and Nb (0.234 nm) layers are corresponding to Ag (111) and Nb (110) planes, respectively. Furthermore, the detailed atomic arrangement shows that Ag(111) and Nb(110) planes

are continuous across the interface. The orientation relationship had been confirmed in our previous work [30] that two different orientations exist along the interfaces, a Kurdjumov–Sachs (K–S) relationship (Ag(111)//Nb(110), Ag $\langle 110 \rangle$ //Nb $\langle 111 \rangle$) and a Nishiyama–Wasserman (N–W) relationship (Ag(111)//Nb(110), Ag $\langle 112 \rangle$ //Nb $\langle 110 \rangle$). Furthermore, some misfit dislocations can be observed at the interfaces due to the lattice mismatch between Ag and Cu layers. The stacking faults also appear in Ag layers. They both contribute to the strength of multilayers.

From the comparison of above three multilayer systems, we find that the differences of length scale effect on the hardness/strength arise at the scale of lower than 5–10 nm. At larger scales, the strength of the three kinds of multilayers varies linearly with $h^{-1/2}$, and hence the strengthening can be interpreted by the H–P model [7]. Usually, when h decreases to a few nanometers, a peak strength is typically reached (it can act as a characteristic scale, and in our experiments it is about 5 nm). At this small length scale, the interface barrier to the transmission of a single dislocation plays the key role in the strengthening [13, 34]. The interface factor becomes vital, and strengthening relating to the interface should be considered, such as Koehler stress and coherency stress [11, 13, 14], which results from the shear modulus mismatch and lattice mismatch, respectively. In addition, Wang and Misra [35] proposed that the interface barriers are also correlated with the defect structure of the interface and the crystallographic orientation relationships. In Cu/Ta multilayers, the crystallinity of Ta layers is weak, the interface between layers is incoherent, and coherent stress does not exist. Hence, the barrier of interface originates from Koehler stress due to great modulus mismatch. This is the reason that the strength of Cu/Ta multilayers with $h \leq 5$ nm is independent of the layer thickness.

In Ag/Cu multilayers, a softening is observed when $h \leq 5$ nm. This softening is very similar to what has been reported in Cu/Ni multilayers which results from the

Fig. 4 Bright field cross-sectional TEM **a** and HRTEM **b** images of Ag/Cu multilayer with $h = 5$ nm (the inset in **a** is the corresponding SAED pattern)

Fig. 5 Bright field cross-sectional TEM **a** and HRTEM **b** images of Ag/Nb multilayer with $h = 5$ nm (the light gray layers indicate the Nb layers, and the dark gray layers indicate the Ag layers; the inset in **a** is the corresponding SAED pattern)

superlattice structure [24]. In Ag/Cu multilayers, the superlattice forms when h is less than 5 nm. Meanwhile, the fcc/fcc crystal lattice system gives the interfaces transparent, i.e., the slip planes and slip directions of the adjacent layers are continuous. When a partial dislocation transmits from Ag to Cu layers, the interface poses a minor resistance to slip transmission, leading to a reduction of strength [17, 36]. The superlattice structure is more pronounced with the less layer thickness. It is the reason why a softening occurs when h falls from 5 to 3 nm.

In Ag/Nb multilayers, the different situations appear and the strength increases continuously until $h = 1$ nm. From the comparison of the cross-sectional images, one can find the interfacial structure of Ag/Nb multilayer is very different from that of the other two. The interface is coherent, and the hetero-epitaxial structures form due to the low lattice mismatch between Ag(111) and Nb(110) planes. At small length scale with $h \leq 5$ nm, the coherency stresses exist between Ag and Nb layers. The coherency stresses can inhibit partial dislocation motion, leading to the enhancement of strength. Furthermore, the structural difference (fcc/bcc) between Ag and Nb layers also produces a barrier to dislocation transmission across the interface due to the discontinuity of slip systems. When a dissociated glide dislocation slips from Ag layer to the opposite, dislocations pin near the interface and cannot directly penetrate the interface, which produces an additional applied stress [26, 34, 36]. In addition, the existence of stacking faults and misfit dislocations also contributes to the enhancement of multilayers, leading to a sustainable increase in strength.

4 Conclusion

It is found that the interfacial structure has a significant influence on the mechanical properties of multilayer systems. Three different plastic deformation behaviors have

been obtained at small length scales. In Ag/Nb multilayers, the strength is enhanced even when h falls to 1 nm. This high strengthening may result from the coherency stress, stacking faults and misfit dislocations. In Ag/Cu multilayers, a softening occurs when $h < 5$ nm, which is because of the formation of superlattice and the transparent interface of fcc/fcc structure, while in Cu/Ta multilayers, the barrier strength of the interface is independent of the scales, leading to the stable strength when $h \leq 5$ nm.

Acknowledgments This work was financially supported by the PAPD (No. 50831004), the Fundamental Research Funds for the Central Universities, the Innovation Fund of Jiangsu Province (No. BY2013072-06), the Natural Science Foundation of Jiangsu Province (No. 20141234), the National Natural Science Foundation of China (Nos. 51171078 and 51371096), and the State Key Program for Basic Research of China (No. 2010CB631004).

References

- [1] S.L. Lehoczky, J. Appl. Phys. **49**, 5479 (1978)
- [2] W.M.C. Yang, T. Tsakalakos, J.E. Hilliard, J. Appl. Phys. **48**, 876 (1977)
- [3] A. Misra, M. Verdier, Y.C. Lu, H. Kung, T.E. Mitchell, M. Nastasi, J.D. Embury, Scr. Mater. **39**, 555 (1998)
- [4] M.Z. Wei, Z.H. Cao, J. Shi, G.J. Pan, L.J. Xu, X.K. Meng, Mater. Sci. Eng. A **598**, 355 (2014)
- [5] B.M. Clemens, H. Kung, S.A. Barnett, MRS Bull. **24**, 20 (1999)
- [6] G.S. Was, T. Foecke, Thin Solid Films **286**, 1 (1996)
- [7] E.O. Hall, Proc. Phys. Soc. Lond. B **64**, 747 (1951)
- [8] N.J. Petch, J. Iron Steel Inst. **174**, 25 (1953)
- [9] P.M. Anderson, C. Li, Nanostruct. Mater. **5**, 349 (1995)
- [10] L.H. Friedman, D.C. Chrzan, Phys. Rev. Lett. **81**, 2715 (1998)
- [11] J.D. Embury, J.P. Hirth, Acta Metall. Mater. **42**, 2051 (1994)
- [12] M.A. Phillips, B.M. Clemens, W.D. Nix, Acta Mater. **51**, 3157 (2003)
- [13] J.S. Koehler, Phys. Rev. B **2**, 547 (1970)
- [14] A. Misra, J.P. Hirth, H. Kung, Philos. Mag. A **82**, 2935 (2002)
- [15] S.I. Rao, P.M. Hazzledine, Philos. Mag. A **80**, 2011 (2000)
- [16] R.C. Cammarata, K. Sieradzki, F. Spaepen, J. Appl. Phys. **87**, 1227 (2000)

- [17] R.G. Hoagland, R.J. Kurtz, C.H. Henager, *Scr. Mater.* **50**, 775 (2004)
- [18] Y.P. Li, G.P. Zhang, W. Wang, J. Tan, S.J. Zhu, *Scr. Mater.* **57**, 117 (2007)
- [19] J.A. Bain, L.J. Chyung, S. Brennan, B.M. Clemens, *Phys. Rev. B* **44**, 1184 (1991)
- [20] B. Shoykhet, M.A. Grinfeld, P.M. Hazzledine, *Acta Mater.* **46**, 3761 (1998)
- [21] F. Appel, U. Christoph, *Intermetallics* **7**, 1173 (1999)
- [22] E.G. Fu, N. Li, A. Misra, R.G. Hoagland, H. Wang, X. Zhang, *Mater. Sci. Eng. A* **493**, 283 (2008)
- [23] A. Tokarz, P. Wiczorek, T. Frączek, Z. Nitkiewicz, *Phys. Status Solidi* **5**, 3526 (2008)
- [24] Y. Liu, D. Bufford, H. Wang, C. Sun, X. Zhang, *Acta Mater.* **59**, 1924 (2011)
- [25] J.Y. Zhang, X. Zhang, G. Liu, G.J. Zhang, J. Sun, *Mater. Sci. Eng. A* **528**, 2982 (2011)
- [26] J.Y. Zhang, Y. Liu, J. Chen, Y. Chen, G. Liu, X. Zhang, J. Sun, *Mater. Sci. Eng. A* **552**, 392 (2012)
- [27] X.Y. Zhu, J.T. Luo, F. Zeng, F. Pan, *Thin Solid Films* **520**, 818 (2011)
- [28] Y. Chen, Y. Liu, C. Sun, K.Y. Yu, M. Song, H. Wang, X. Zhang, *Acta Mater.* **60**, 6312 (2012)
- [29] D. Bufford, Z. Bi, Q.X. Jia, H. Wang, X. Zhang, *Appl. Phys. Lett.* **101**, 223112 (2012)
- [30] M.Z. Wei, J. Shi, Y.J. Ma, Z.H. Cao, X.K. Meng, *Mater. Sci. Eng. A* **651**, 155 (2016)
- [31] M.Z. Wei, Z.H. Cao, J. Shi, G.J. Pan, L.J. Xu, X.K. Meng, *Mater. Sci. Eng. A* **646**, 163 (2015)
- [32] M. Mata, M. Anglada, J. Alcala, *J. Mater. Res.* **17**, 964 (2002)
- [33] M.Z. Wei, L.J. Xu, J. Shi, G.J. Pan, Z.H. Cao, X.K. Meng, *Appl. Phys. Lett.* **106**, 011604 (2015)
- [34] J.W. Yan, G.P. Zhang, X.F. Zhu, H.S. Liu, C. Yan, *Philos. Mag.* **93**, 434 (2013)
- [35] J. Wang, A. Misra, *Curr. Opin. Solid State Mater. Sci.* **15**, 20 (2011)
- [36] G.P. Zhang, X.F. Zhu, *Acta Metall. Sin.* **50**, 148 (2014). **(in Chinese)**