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Research Article

Structure Transformation and Coherent Interface in Large Lattice-Mismatched Nanoscale Multilayers

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Nanoscale Al/W multilayers were fabricated by DC magnetron sputtering and characterized by transmission electron microscopy and high-resolution electron microscopy. Despite the large lattice mismatch and significantly different lattice structures between Al and W, a structural transition from face-centered cubic to body-centered cubic in Al layers was observed when the individual layer thickness was reduced from 5 nm to 1 nm, forming coherent Al/W interfaces. For potential mechanisms underlying the observed structure transition and forming of coherent interfaces, it was suggested that the reduction of interfacial energy and high stresses induced by large lattice-mismatch play a crucial role.

1. Introduction

Nanoscale metallic multilayers have been extensively studied both experimentally and theoretically for their unique mechanical properties as well as size effects [1–10]. For such multilayer systems, the interface always plays a dominant role in plastic deformation, acting as sources and sinks for dislocations [2, 7, 11]. In general, coherent and incoherent interfaces have been identified as the two main types of interface in metallic multilayers, and dislocations can cross the former but not the latter. Accordingly, the type of interface significantly affects the plastic deformation behavior of multilayer system [7, 10]. To form a coherent interface, the lattice parameter mismatch between two constitute layers should be small, and identical crystal structure is preferred. Therefore, it has been suggested that coherent interfaces hardly exist in a multilayer system possessing simultaneously different crystal structures and large lattice parameter mismatch, for example, Al/W multilayers as Al has face-centered cubic (fcc) structure with a lattice parameter of ~0.405 nm whilst W has body-centered cubic (bcc) structure with a lattice parameter of ~0.316 nm. Nonetheless, in this work, we report that coherent interface could indeed form in nanoscale Al/W multilayers, although the lattice mismatch between Al and W is as high as 27.95%. The observation of coherent interfaces in nanoscale Al/W multilayers may pave a new way for processing novel metallic multilayers having both high strength and ductility.

2. Experiments

Al/W multilayers with a total thickness of 500 nm were synthesized using the technique of DC magnetron sputtering at room temperature on oxidized Si (1 0 0) substrate. The base pressure prior to sputtering was 6.3×10^{-5} Pa whilst Ar pressure during sputtering was 0.63 Pa, with deposition rate set as 2.5 nm/min for Al layers and 5 nm/min for W layers. Under such deposition conditions, two samples were fabricated In the first sample, the individual thickness of both Al and W layers was nominally equal to 5 nm (referred to $Al_{5\,nm}/W_{5\,nm}$ hereafter); in the second sample, the thicknesses of individual Al and W layers were set separately as 1 nm and 5 nm (referred to $Al_{1\,nm}/W_{5\,nm}$ hereafter). The cross-sectional microstructures of as-deposited Al/W multilayers were investigated using JEOL 2100F transmission electron microscopy (TEM) and high resolution TEM (HRTEM) operated at 200 kV. Upon manual polishing, the TEM samples were precisely

prepared via Gatan Precision Ion Polishing System 691 by using Ar ion.

3. Results and Discussion

Figures 1(a) and 1(b) showed bright field TEM images of the cross-sectional morphologies for $Al_{5\,nm}/W_{5\,nm}$ and $Al_{1\,nm}/W_{5\,nm}$ multilayers, respectively. Both multilayers exhibited a well-defined layered structure with planar modulation layers, bright layers being Al and dark layers being W. The electron diffraction (ED) patterns of two multilayers were derived by cross-sectional observation. In Figure 1(d), no Al patterns were detected in the $Al_{1\,nm}/W_{5\,nm}$ multilayer, while were detected in the $Al_{5\,nm}/W_{5\,nm}$ multilayer as shown in Figure 1(c).

Figure 2(a) presented HRTEM images of $Al_{5\,nm}/W_{5\,nm}$ multilayers, and Figures 2(b) and 2(c) showed the Fast Fourier Transformation (FFT) images corresponding to the selected rectangular areas of 1 and 2 as marked in Figure 2(a). For $Al_{5\,nm}/W_{5\,nm}$ with individual layer thickness of 5 nm, the FFT images indicated that both Al and W retained their bulk stable crystal structures, resulting in a stacking sequence of Al in face-centered cubic structure and W in body-centered cubic structure. Further, it was evidenced from the lattice image shown in Figure 2(a) that the interfaces in the $Al_{5\,nm}/W_{5\,nm}$ multilayers were incoherent.

Combining the Al and W layers as a whole to form FFT images as shown in Figures 2(b) and 2(c), one could clearly identify the individual position of each spot for both Al and W layers, although some spots appeared blurrier than the others. For example, the spot of W (110) was much hazier than that of Al (111), but the positions corresponding to each spot were separated, and there was an angle of 11° between two neighboring spots. Moreover, as shown in Figure 2(d), the multilayers exhibited a strong Al {111}//W {110} growing fiber texture with a Kurdjumov-Sachs orientation [12], similar to that observed for Al/Nb multilayers having identical lattice structure (i.e., fcc/bcc) and individual layer thickness as in the present Al/W multilayers.

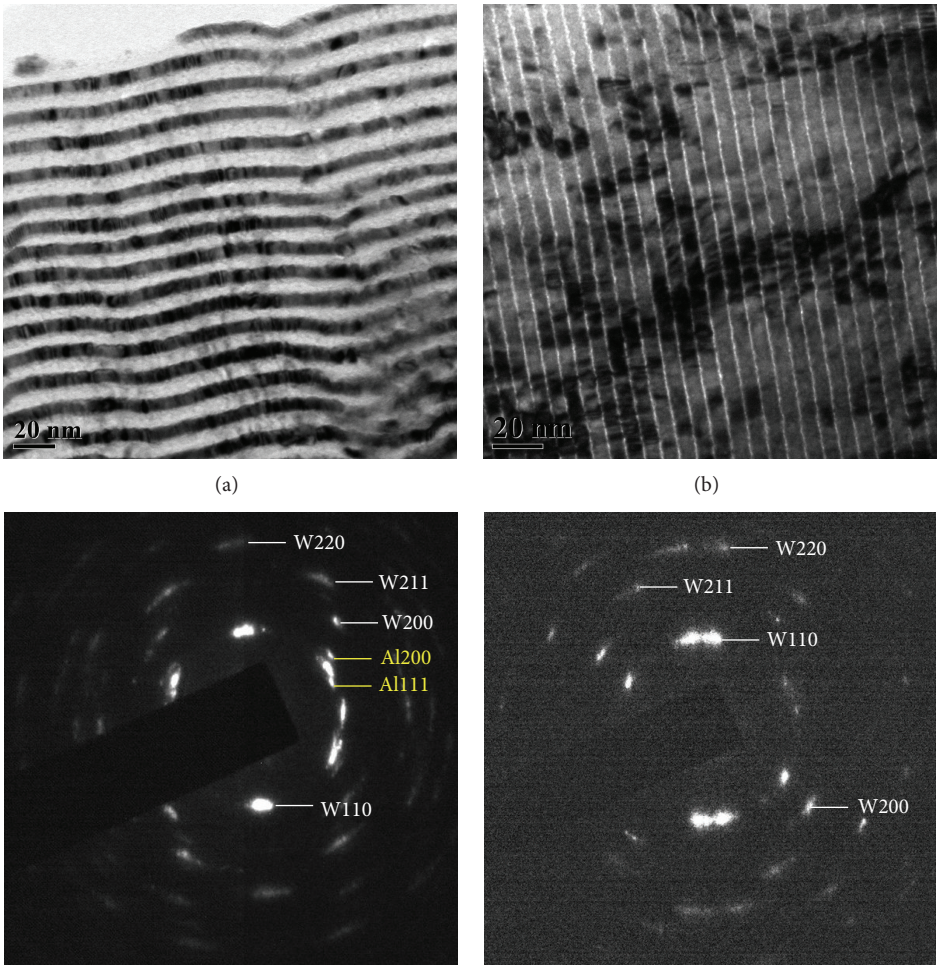
With the Al layer thickness reduced from 5 nm to 1 nm whilst that of the W layer maintained at 5 nm (i.e., $Al_{1\,nm}/W_{5\,nm}$), the HRTEM image shown in Figure 3(a) indicated that the Al layers had transformed completely from fcc lattice into bcc lattice, generating accordingly an unusual stacking sequence of bcc Al/bcc W multilayers. Moreover, Figure 3(a) demonstrated clearly that the two different metal layers possessed identical crystal structure and that the well-known lattice parameter mismatch between bulk Al and bulk W vanished in the present $Al_{1\,nm}/W_{5\,nm}$ multilayers. More importantly, it was seen from Figure 3(a) that the atomic arrangement and slip systems were continuous across the interface. In other words, coherent interface was formed between Al and W layers in $Al_{1\,nm}/W_{5\,nm}$. Further, as shown in Figure 3(c), abundant dislocations similar to those previously reported for nanoscale Cu/Nb multilayers [13] were observed in $Al_{1\,nm}/W_{5\,nm}$ multilayers. The density of dislocation is about $1 \times 10^{17}\ m^{-2}$, and dislocation of intralayer and interface ratio is about 4 : 3.

For $Al_{1\,nm}/W_{5\,nm}$ multilayers, as the pattern obtained for 1 nm thick Al layers was too vague, only the FFT image derived from both of the two constitute layers was presented in Figure 3(b). Interestingly, only one series of spots corresponding to the lattice structure of the bcc W layer showed up to be consistent with the HRTEM image of Figure 3(a) where the Al layers were seen to be transformed from fcc to bcc. It should nonetheless be mentioned that spots marked as 2, 3, 5, and 6 in Figure 3(b) were slightly larger than spots 1 and 4, indicating that the transformation appearing to be complete in Figure 3(a) may not be so complete. Slight difference may still exist between the Al and W layers in $Al_{1\,nm}/W_{5\,nm}$, as it was hard to completely eliminate the large lattice mismatch between bulks Al and W under the present constrained lattice circumstances, as evidenced along certain orientations of spots 2, 3, 5, and 6.

The present results of $Al_{1\,nm}/W_{5\,nm}$ multilayers demonstrated that the structure transformation in Al layers played a dominating role in forming coherent Al/W interfaces. Numerous models had been proposed to explain structural stabilities in multilayers [14–20]. In particular, the Redfield-Zangwill model [15] considering the influence of interfaces on stacking fault energy was found applicable in interpreting phase stability in multilayers. However, the Redfield-Zangwill model only considered phase stability of multilayers consisting of close-packed lattice structures (e.g., fcc and hcp) having similar lattice parameters (e.g., Al/Ti multilayers). In such an fcc/hcp multilayer system, by introducing stacking faults into its parent structure, it was relatively easy to transform an fcc ABCABC stacking sequence into an hcp ABABAB stacking sequence, or vice versa. For the present Al/W multilayers, however, the bulks Al and W possessed fcc and bcc lattice structures, respectively. As bcc lattice structure was not close-packed, its stacking sequence was much more complete than that of fcc and hence could not be transformed from fcc by simply introducing stacking faults into the lattice. Consequently, it became apparent that the Redfield-Zangwill model was not applicable to the present $Al_{1\,nm}/W_{5\,nm}$ multilayer system.

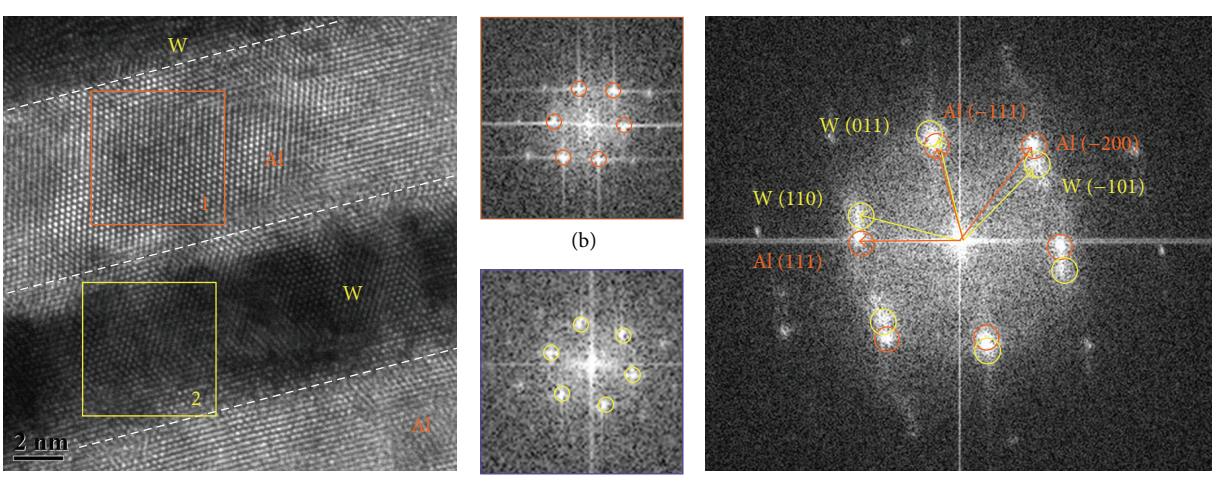
For some nonequilibrium structured multilayers such as Co/Cr [21] and Cu/Fe [22], it had been found that Co and Cu could be transformed from close-packed lattice structure to nonclose-packed one. However, given that the atomic radii of the two constitute layers in both Co/Cr and Cu/Fe multilayer systems were almost equal, it was quite different from the phase transition occurring in the present Al layers because of the large lattice mismatch between Al and W.

From the viewpoint of thermodynamics, the balance between the volumetric and interfacial components of total free energy [16, 17, 19, 20] had been widely used to interpret the phase stability of crystalline materials. For the present Al/W multilayer system, when the thickness of Al layers was smaller than a critical value, the crystal structure of Al tended to adopt the crystal structure of its substrate, that is, W, resulting in a crystalline structure different from the equilibrium structure of Al. Once Al was deposited onto W, the volumetric free energy gained from accommodating the large lattice-mismatch between the two constitute layers was stored in the multilayers. For $Al_{1\,nm}/W_{5\,nm}$ multilayers, the



(a) (b) (c) (d)

Figure 1: Cross-sectional TEM images of (a) $Al_{5\,nm}/W_{5\,nm}$ and (b) $Al_{1\,nm}/W_{5\,nm}$ multilayers showing well-defined layered structures in nanoscale and ED images of (c) $Al_{5\,nm}/W_{5\,nm}$ and (d) $Al_{1\,nm}/W_{5\,nm}$ multilayers.



(a) (b) (c) (d)

Figure 2: (a) Presents HRTEM image of $Al_{5\,nm}/W_{5\,nm}$ multilayers; (b) and (c) are Fast Fourier Transformation (FFT) images of selected rectangular areas "1" and "2" marked in (a); and (d) is a combined FFT image derived from Al and W layers simultaneously.

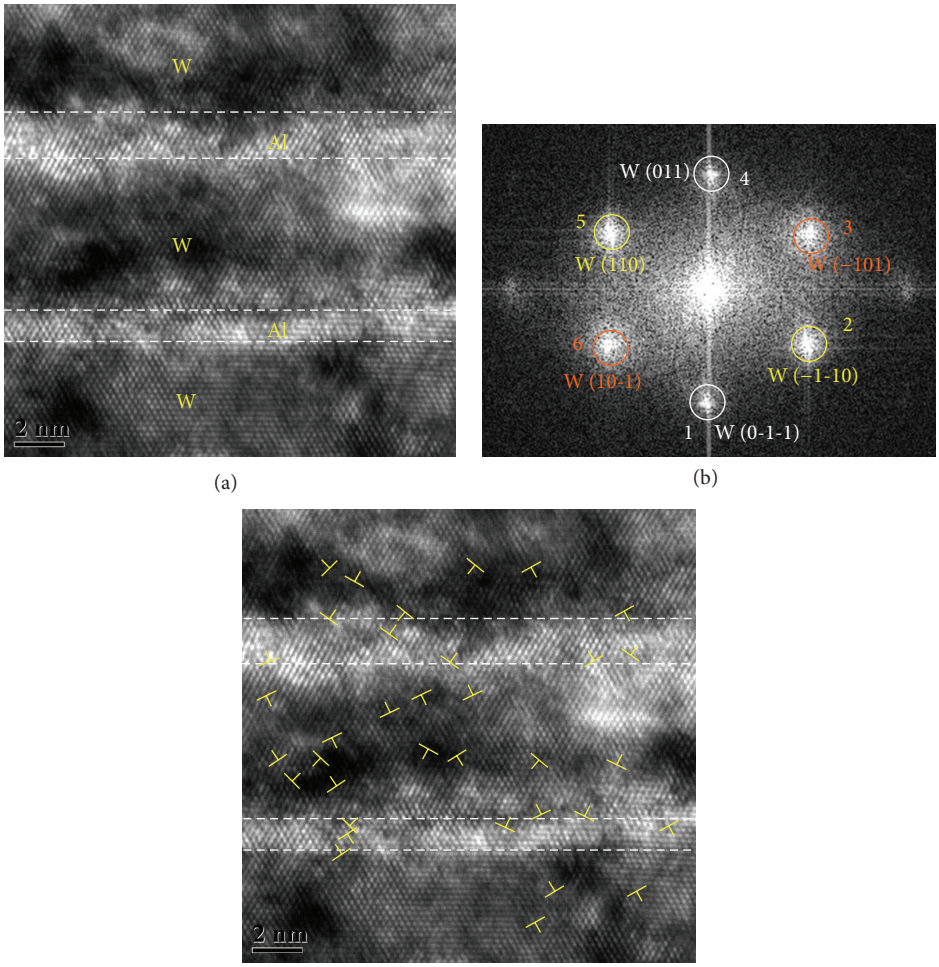


Figure 3: (a) Presents HRTEM image of $Al_{1\,nm}/W_{5\,nm}$ multilayers; (b) is a combined FFT image derived from both Al and W layers; and (c) is a dislocation distribution in the multilayer system.

W layers deposited prior to the deposition of Al served as the template and could confine the lattice structure of the much thinner Al layer, transforming it into bcc Al. This structure transition was driven by a reduction in overall interfacial energy which should be more than compensating the increase of energy from forming metastable phase [23], that is, bcc Al herein. Moreover, as shown in Figure 3(c), dislocations with high density were present in the $Al_{1\,nm}/W_{5\,nm}$ system, suggesting that the stresses induced by coherency strain were sufficiently large as the existence and maintaining of dislocations at such small length scale required extremely high stresses [24]. The misfit dislocation is an effective way to decrease volumetric free energy. However, dislocation prefers to nucleate at free surface than interface [25, 26]. Once dislocation nucleated, it propagates or glides to the interface to relieve misfit strain. Nucleation of dislocations at the free surface during deposition and propagation of these dislocations to the interface are critical steps in the relaxation of coherency strain, as shown in Figure 1. However, if lattice frictional stress in the layer is high, most dislocations may not reach the interface, depending upon the growth temperature and deposition rate. And it will take a higher temperature and longer time to expedite the lattice relaxation process. Appearance of dislocations at interface and in the W layer also indicated the relaxation of coherency strain and minimization of energy. As the thickness of Al layer is much thinner and softer than that of W layer, the dislocations easily glide to the interface from inner area of the Al layer, which could be harder in the W layer. Therefore, dislocations mostly appeared at interface and inside the W layers as observed in Figure 3.

As the individual layer thickness of Al was increased to 5 nm, the energy derived from lattice fit dropped below the volume energy of the strained Al film. Consequently, the nonequilibrium structure of the Al layer (i.e., bcc Al in the present study) broke down so that it would grow in its equilibrium structure (i.e., fcc), forming incoherent Al/W interfaces as observed in $Al_{5\,nm}/W_{5\,nm}$. In addition,

few dislocations were observed in the $Al_{5\,nm}/W_{5\,nm}$ system, indicating that its internal stresses were much smaller than those present in the $Al_{1\,nm}/W_{5\,nm}$ system.

4. Summary

In summary, despite the large lattice mismatch and quite different stacking sequences between Al and W, we demonstrated in this paper that structural transformation and coherent interface could occur in nanoscale Al/W multilayers, depending upon the thickness of the constitute layers. A critical layer thickness appeared to exist, below which an Al layer would adopt the structure of its template (W layer) driven by the reduction of interfacial energy and high stresses induced by large lattice mismatch; otherwise, the Al layer would transform to its equilibrium structure as both lattice distortion and high internal stresses were released.

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