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Adhesion energy and related plastic deformation mechanism of Cu/Ru nanostructured multilayer film

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Sputter-deposited Cu/Ru nanolaminate composites with individual layer thickness as small as 1.5 nm were tested by nanoindentation probing to initiate and drive delamination. Focused ion beam (FIB) observations show the layer buckling and interface delamination with lateral crack along the interface between the multilayer and substrate. The buckling behaviors, dependent on the critical length scale, are rationalized in the light of the repeating structural unit in the coherent multilayer. In addition, due to the presence of the interfaces and constraints between the hetero-layers, the condition for plastic dissipation in multilayers shifts significantly from those of single layer films, therefore a modified energy-dissipative model has been employed to obtain the quantitative adhesion energy for Cu/Ru multilayer, which agrees well with previous reports on the Cu-based films adherent on rigid substrates. The present result provides meaningful adhesion energy estimates and helps to understand the underlying deformation mechanism in metallic multilayers.

1. Introduction

Recent years, nanostructured multilayer films, which offer an excellent opportunity to study the effects of diminishing length scales on material properties, particularly at nanostructure length scales less than 5 nm, has gathered much interest in the materials community [1,2]. The use of multilayered films has proliferated in the fields of protecting structural materials in harsh environment, thermal and irradiation resistance coating, hydrogen energy storage, X-ray optics properties and micro-electromechanical systems (MEMS) [1–5]. Layers applied in these devices are as thin as 1–2 nm [6,7]. In all of these applications, performance, reliability and durability are tied directly to good adhesion between film and substrate [8,9]; this fact is reflected in the numerous numbers of tests that have been developed to quantitatively estimate film adhesion [10–15]. The test methods include indentation tests, uniaxial tensile, stressed overlayers, scratch tests, four-point bending, etc., suggesting that interfacial adhesion is material, geometry and even industry specific [10–15]. In these methods, indentation methods are generally preferred to evaluate interfacial adhesion owning to these reasons (i) it is relatively easy to perform, (ii) it requires no difficult sample preparation steps, and (iii) it causes only negligible thermal processing steps that may affect the adhesion characteristics of the system involved. The relevant theoretical models are mainly based on the elastic buckling of films, as proposed by Marshall and Evans [16,17].

The indentation fracture test in the as-deposited film creates a plastic impression on the film surface, triggering axisymmetric buckling upwards and subsequent fracture along the film/substrate interface. The center is constrained, giving rise to a double buckle configuration as shown schematically in Fig. 1. Lateral cracks generate beneath the indent and propagate initially almost parallel to the film/substrate interface. The Marshall and Evans model [16,17] provides a relationship between the strain energy release rate for the interface crack, *G*, and the observed circular delamination of radius, *a*, thus allowing the determination of driving force in the form of the strain energy release rate. However, Evans model is often impractical for soft or strongly adhering films, because of the ductile and strongly adhering films tend to simply deform plastically before the development of sufficient elastic strain energy for delamination [18]. To overcome this limitation, researchers have used superlayers deposited over the film to restrict tremendous plastic deformation prior to delamination [19].

Since many of the mechanisms have been identified and

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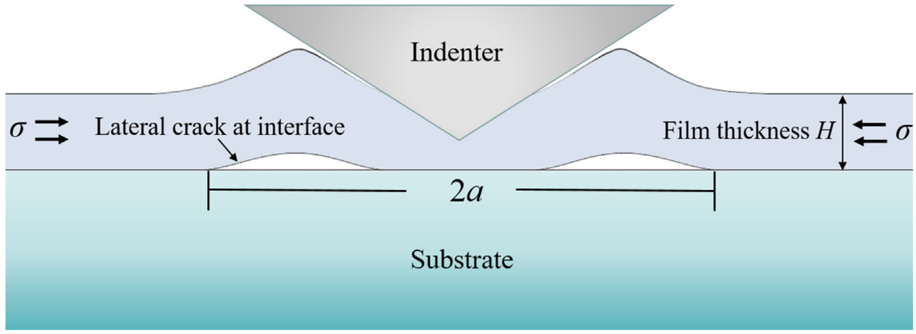

Fig. 1. A schematic illustration of indentation induced delamination and lateral cracks at film/substrate interface. Indentation causes dilated plastic zone which drives buckling and delamination.

explored for single and bilayer film/substrate interface systems, few of adhesion energy measurements have been developed for multilayered structures with a large number of hetero-interfaces [1]. The buckling behaviors of nanostructured multilayers under indentation are poorly understood and evaluations of interfacial adhesion still need a proliferation of research, especially under indentation [12,13]. In view of its extraordinarily high specific strength and low solubility in Cu, Ru was considered as an excellent candidate as a barrier material for advanced integrated circuits and MEMS. Recently, we have reported the outstanding mechanical properties of Cu/Ru multilayers [20,21]. To understand the fundamentals of such contact-induced behavior, we reported the results of nanoindentation testing that enables measurement of adhesion energy of Cu/Ru multilayer on rigid substrate in this paper. As crack formation inside films will make the stress distribution much more complicated and no longer fit the assumptions of the Marshall and Evans model [14]. Due to the fact that crack formation will release the elastic strain energy, multilayer with layer thicknesses as small as 1.5 nm (no crack formation inside film [21]) is chosen to obtain the adhesion energy and a modified energy-dissipative model has been proposed for multilayers.

2. Experimental procedures

Epitaxial Cu/Ru multilayers were deposited on Si (1 0 0) substrates by means of radio frequency and direct current magnetron sputtering. A total of 167 bilayers of Cu and Ru with a nominal individual layer thickness of 1.5 nm were alternately deposited to a total film thickness of 500 nm. Residual stresses σ_r in the multilayers were recorded by a flatness and wafer stress analyzer (BGS 6341 IC). Microstructural characterization was carried out by high resolution transmission electron microscopy (HRTEM) with a JEOL-2100F operated at 200 keV. Sample preparation was conducted via mechanical polishing to a final thickness of approximately 30 μm, and thinning to electron transparency with a Gatan Precision Ion Polishing System 691. The Cu/Ru multilayer was indented to a displacement of 400 nm with the MTS Nanoindenter XP system (MTS, Inc.) equiped with Berkovich tip. Experimental details of the sample preparation and nanoindentation technique were reported previously [20,21]. In order to reveal the interfacial failure mechanism, some indentations were cross-sectioned by dual beam focused ion beam (FIB) and characterized by HRTEM.

3. Experimental results and discussion

The cross-sectional HRTEM image are depicted in Fig. 2, showing that a polycrystalline multilayered morphology of alternating layers of Cu and Ru grows epitaxially on each other and

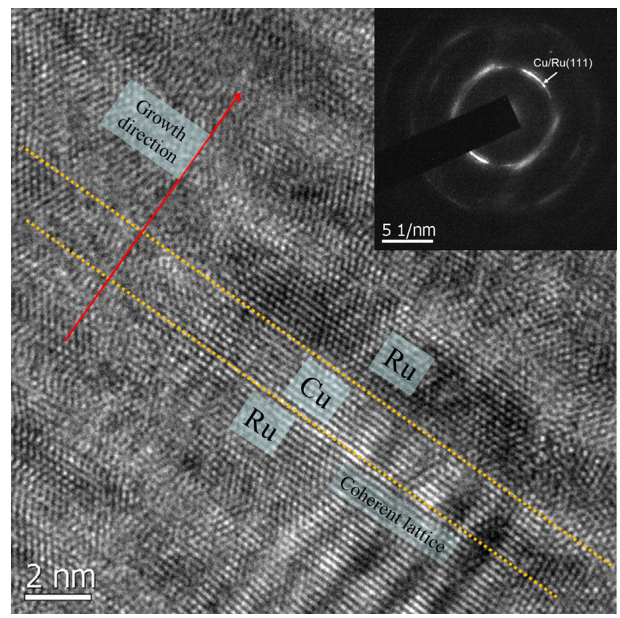

Fig. 2. HRTEM bright-field micrograph of as-deposited 1.5 nm Cu/Ru multilayer. Chemically sharp interfaces between epitaxial layers of Cu and Ru are clearly seen. Note selected area diffraction pattern showing coherent (111) Cu/Ru interface texture.

forms superlattice columnar crystal structure. The layers are textured in the growth direction, with (1 1 1) plane of fcc Cu/Ru forming the coherent interfaces between the two phases, as shown in the embedded selected area diffraction pattern.

Upon performing nanoindentation tests, load–displacement curves are obtained. For a range of bulk materials, the relationship $P = K_m h^2$ was found to describe the penetration depth, h, in terms of the applied force P, where K_m is the Loubet's equivalent elastic plastic parameter [22,23]. But, for a film/substrate system, the relationship between P and h^2 will change [22]. Fig. 3 shows the load-displacement squared data in the Cu/Ru multilayer. The red dashed lines represent the relationship $P = K_m h^2$ and the black solid lines are the varying P as a function of h^2 during penetration. The load-displacement squared plots of Fig. 3 show an initial straight line segment to the turning point which is identified as 'coating only' behavior [22] reflecting buckling in compression instability. Beyond the turning point, substrate effect emerges and significant tensile stresses are being generated in an approximately circular pattern around the outer periphery of the contact zone, and the film is forced to buckle to conform to the film/substrate interface delamination and pile-up (schematically show in Fig. 1). The