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Molecular dynamics simulation of thin film interfacial strength dependency on lattice mismatch

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ABSTRACT

Laser-induced thin film spallation experiments have been previously developed to characterize the intrinsic interfacial strength of thin films. In order to gain insights of atomic level thin film debonding processes and the interfacial strength dependence on film/substrate lattice structures, in this study, molecular dynamics simulations of thin film interfacial failure under laser-induced stress waves were performed. Various loading amplitudes and pulse durations were employed to identify the optimum simulation condition. Stress propagation as a function of time was revealed in conjunction with the interface structures. Parametric studies confirmed that while the interfacial strength between a thin film and a substrate does not depend on the film thickness and the duration of the laser pulse, a thicker film and a shorter duration do provide advantage to effectively load the interface to failure. With the optimized simulation condition, further studies were focused on bulk Au/Au bi-crystals with mismatched orientations, and Ni/Al, Cu/Al, Cu/Ag and Cu/Au bi-crystals with mismatched lattices. The interfacial strength was found to decrease with increasing orientation mismatch and lattice mismatch but more significantly dominated by the bonding elements' atomic structure and valence electron occupancy.

1. Introduction

Thin film plays an increasingly important role in micro/nano-electro-mechanical systems for various technological applications. For design and applications of thin film devices, it is crucial to understand the fundamental governing mechanisms of the interfacial strength and failure structure. Introduced by Yang [1] and Vossen [2], and further developed by Gupta et al. [3–5], Wang et al. [6–9], laser-induced thin film spallation experiments have been proved to be a versatile technique to characterize the intrinsic interfacial strength of thin films under tensile [1–7], mixed-mode [8,9] and pure-shear loading [10]. Fig. 1(a) shows the schematic of the tensile laser spallation experiment, in which a high-energy laser pulse is impinged on a thin absorbing layer confined between the back surface of the substrate and a confining material. Upon absorbing the laser energy, the sudden expansion of the confined layer generates a high-amplitude compressive stress wave directed towards the test film. When the compressive stress wave reaches the free surface of the test film, it becomes a tensile wave that loads the film-substrate interface in tension. Provided with sufficiently high laser pulse energy, the tensile interfacial stress could cause delamination failure of the film-substrate interface. By measuring the free surface displacement of the test film at the onset of interfacial failure, the intrinsic strength of the film–substrate interface can be inferred using elastic wave theory.

While laser spallation technique has been very versatile for experimentally measuring the interfacial strength of a variety of material systems [11–15], the short duration of the stress wave presents challenges in directly visualizing the stress wave propagation and the interfacial failure evolution. Various numerical simulations have been performed to study the fundamental mechanisms of the laser shock induced deformation. Numerical simulation is particularly beneficial in cases that important processes occur at the extreme conditions or inaccessible for direct experimental investigation. Finite element simulation has been used mostly to investigate laser induced surface ablation [16] and interface debonding [17–20]. In recent years, molecular dynamics (MD) simulations have been widely adopted to enhance the understanding of fundamental physical phenomena, to capture detailed deformation of material deformation in short time and to reveal the dislocation activities. MD methods have been widely used for study of various basic mechanisms of laser shock compression [21], surface ablation [22], cluster aggregations and molecular ejections excited by pulse lasers in the recent past [23,24]. These results can contribute to better understanding of interactive dynamic behaviors in atomic scale between the laser and materials, and the deformation mechanisms in materials. However, the interfacial strength dependence and the thin film/substrate debonding structure dependence on different types of interface structures have not been fully investigated. Furthermore, while numerous MD simulations of tension tests [25,26] have been performed to study the debonding features and interfacial strength of double material interfaces, the investigation of stress wave induced interfacial failure has not been studied yet.

In order to gain insights of the thin film debonding processes and the interfacial strength dependence on film/substrate lattice mismatch and orientation mismatch, in this study, MD simulations of stress wave

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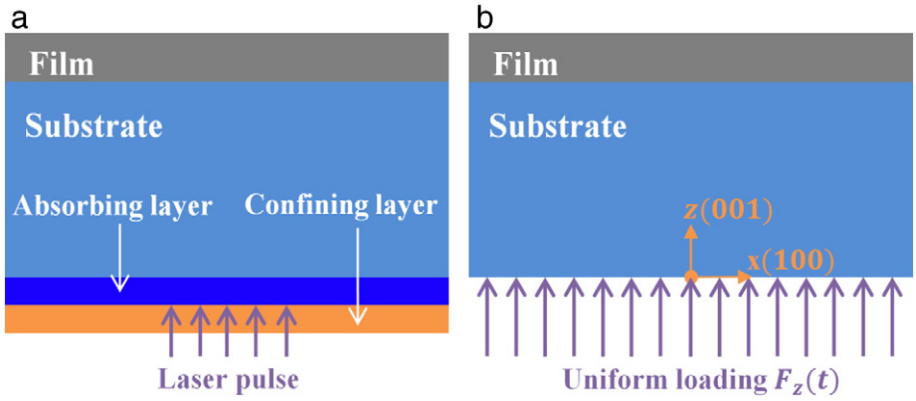

Fig. 1. (a) Schematic of laser-induced thin film spallation experiment; (b) Schematic of the MD model used in this study.

induced thin film interfacial failure were carried out. In the laser-induced thin film spallation experiments such as those by Wang et al. [6–9], high energy laser pulses were applied, which led to the subsequent thermal expansion of the absorbing layer to generate compressive stress waves. In the current study, since the primary interest is in the stress wave propagation and the interfacial failure process of thin film/substrate interface of different orientation and lattice mismatch, the mechanisms of stress generation from the deposited laser energy were omitted. Fig. 1(b) shows the schematic of the MD model for this study. A time-dependent spatially uniform loading was applied on the film/substrate system. The load function took a similar form to that measured by experiment but with a shorter timescale. In this case, the major simulation problem became the investigation of a high strain-rate and time-dependent compressive stress wave-induced thin film/substrate interfacial failure. Various loading amplitudes and pulse durations were employed to characterize the dynamic behaviors of the interfaces. Stress propagation as a function of time was investigated in conjunction with the interface structures in order to reveal the fundamental mechanisms of thin film interfacial debonding process. The interfacial strength dependence on orientation mismatch was investigated by using Au bi-crystals with different grain orientations, while Cu/Al, Cu/Ag, Cu/Au and Ni/Al bi-materials were used as the film/substrate in lattice mismatch cases. In materials that contain pre-existing defects or plastic deformations, the interaction between dislocations and interface also plays important role in interfacial strength [27]. However, in this study, pure elastic deformation is involved during the stress propagation before the interfacial failure. Thus, effect from interaction between dislocations and the interfaces is not considered. In both the orientation mismatch and lattice mismatch studies, compressive load with varying magnitude was applied to the substrate surface which is away from the film. It is aimed to find the minimum compressive load that induces the interfacial failure, so that the interfacial strength can be calculated accordingly.

2. Simulation methods

In the following work, numerical experiments of interfacial structure behaviors under the excitation by short duration compressive stress waves were carried out via MD simulation. In this approach, the film/substrate system consists of two blocks as illustrated in Fig. 2. Thin film/substrate models (where the substrate thickness is about six times of film thickness) were used for parametric studies to find out the optimal simulation condition and the two bulk systems (where the film and substrate have similar thickness) were used for interfacial strength dependence study based on the parametric study result. The atomic views shown in the figures are x–z cross-sections. Centrosymmetry parameter method [28] was used to visualize the local lattice disorder around an atom in Fig. 2(a) and (b). Different colors were assigned to atoms according to their range of the centrosymmetry parameter. The blue atoms represent perfect lattice structures, while atoms with other colors represent dislocations formed in the grain boundary. The interfaces formed during simulation can be observed clearly between the film and substrate as in Fig. 2(a) and (b). In Fig. 2(c) and (d), the different colors of atoms represent different types of atoms. In simulations, the z direction is normal to the interface and is the direction of the prescribed displacement as well. Periodic boundary conditions (PBC) were applied in lateral x and y directions, while in the loading z direction the boundary was fixed. In order to achieve an equilibrium configuration, energy minimization with conjugate gradient method [29] was first applied, by holding the top and bottom surfaces along the z direction fixed. The structure was then annealed at a temperature of 300 K and relaxed. After the relaxation, the top surface of the structure was fixed, and a one-dimensional time-dependent compressive load was applied onto the bottom surface of the structure. The one-dimensional compressive load $F(t)$ has a Gaussian distribution and can be expressed by:

$$F_z(t) = F_{\max} \exp\left\{-2\frac{(t-t_0)^2}{T^2}\right\} \tag{1}$$

where T is the period of the Gaussian distributed stress wave, t_0 is the time shift, F_{max} is the peak value of the stress wave, ranging from 0.1 to 0.4 nN. Different T-values indicate different durations of the applied Gaussian stress wave pulse. In simulation, the time shift value and the loading duration change accordingly with the loading period T. For example, the whole Gaussian distributed wave has a $t_0 = 1.5$ ps for $T = 1$ ps, and the loading lasts for 3 ps in total.

Parametric study of the thin film/substrate system was conducted first with the goal to investigate the effect of the film thickness and the duration of the stress wave pulses. Au/Au bi-crystal model (Fig. 2(a)) with orientation mismatch [001]/[111] and Cu[001]/Al[001] model (Fig. 2(b)) were chosen for these studies, with total model size around 8 nm × 8 nm × 24 nm in x, y and z directions. In film thickness effect studies, film thickness was varied from 15 Å to 40 Å. Various amplitude stress waves were applied to induce the interface failure. Different durations of the stress wave pulse have different wavelengths, and large wavelength induces difficulty to interfacial failure for films with small thicknesses due to cancellation between the incident compressive stress wave and the reflected tensile stress wave. Various stress wave duration (T), ranging from 0.1 ps to 3 ps, were applied in our study.

Based on the findings from the parametric study, bi-crystals were used to study the effect of the orientation and lattice mismatch on interfacial strength. For orientation mismatch, Au bi-crystals were constructed with different grain orientations. The orientation mismatch between the two blocks forms a grain boundary. Fig. 2(c) shows one representative simulation structure of an Au bi-crystal with gain boundary normal (z direction) oriented in [001] and [111] respectively. In order to study the interfacial strength relation with orientation mismatch, three different Au bi-crystals with grain boundary normal (z direction) in [001]/[110],