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Molecular dynamics simulation of ultrashort laser induced back-surface spallation in metallic film*

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Abstract: Using molecular dynamics methods combined with two-step radiation heating model, a complete microscopic description of the dynamic progresses involved in laser induced back-surface spallation in metallic film in stress confinement regime is provided. Different from the front-surface ejection with strongly affected mechanical stability of the front-surface by laser heating, the back-surface spallation is a disintegration of cold material. The mechanism of spallation is analyzed as a result of the interaction of the unloading wave and reflected stress wave. The propagation of laser-induced stress wave is also further investigated, and the influences of film thickness on spall thickness as well as the time when spallation begins are predicted.

Key words: Back-surface spallation; Ultrashort laser; Molecular dynamics; Metallic film

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Under laser pulse irradiation, a planar separation of material parallel to the free surface will occur at the back side of the film, which is termed as spallation[1-3]. Different from the front-surface ejection with strongly affected mechanical stability of the front-surface by laser heating[4-5], the back-surface spallation is a disintegration of cold material near the back side of the film. The analysis of the basic mechanism involved in spallation is important not only in investigating the mechanical properties of materials at a high strain rate[6-7], but also in minimizing the effects of back-spallation on front-surface ejection[8-10]. For this reason, further study on laser induced spallation is necessary.

In recent years, the damage/spallation of an irradiated bulk target in back-surface region have been studied with experiments as well as numerical simulation methods[11-12]. The instant critical fracture stress criterion[13], the stress rate criterion[14], the stress gradient criterion[15] and the cumulative damage criterion[16] have been used to determine the spallation of targets. These criterions, however, are deduced from continuum models including a number of assumptions and simplifications. Due to the nonlinear and nonequilibrium nature of the process involved in spallation, the discussion of the microscopic mechanisms and parameters of laser induced back-spallation cannot be based on direct application of continuum models or conclusions derived from back-surface spallation experiments. Molecular dynamics method can solve this problem without making any assumptions about the character of the processes and provide a detailed description of dynamic process of spallation.

Using MD methods combined with two-step radiation heating model[17], laser induced spallation in metal film is investigated. The mechanism of spallation is analyzed, and the propagation of laser-induced stress wave is also further investigated. The influences of film thickness on spall thickness as well as the time when spallation sets in are also predicted.

1 Laser energy absorption

Ultrashort laser heating of metals is achieved in two steps. First, the electron subsystem is thermalized

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quickly due to laser energy absorption, then the lattice subsystem is warmed up through energy coupling with electron subsystem. These steps can be described by the two-step radiation heating model, which has become the main model to describe the kinetics of the electron and lattice temperature evolution in a metal target irradiated with a short laser pulse and has been used widely[18-20]. The expressions are as follows

$$C_e \frac{\partial T_e}{\partial t} = k_e \frac{\partial^2 T_e}{\partial z^2} - \Phi + Q(z,t) \tag{1}$$

$$C_a \frac{\partial T_a}{\partial t} = \Phi = g(T_e - T_a) \tag{2}$$

where $Q(z,t)$ is the heat flux, C_e and C_a are the heat capacities (per unit volume) of the electron and the lattice subsystems, K_e is the electron thermal conductivity, g is the electron-phonon coupling constant, T_e and T_a are the temperature distributions of electron and lattice systems.

Laser energy absorption is achieved by adding a velocity - proportional force to the equation of each atom[21] which contributes to the increase of kinetic energy.

2 Set up of model system

A Cu film has been chosen as a model system. The atoms interact with one another through Morse potential[22], which is simple and suitable for face-centered cubic metals. The sketch map of the model is shown in Fig. 1. Periodic boundary conditions are applied in the x and y directions. Different from the previous laser ablation model[23-24], free boundary condition is applied at the bottom of the film.

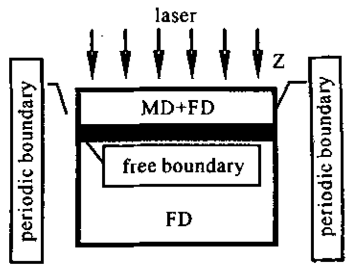

Fig. 1 Schematic sketch of the simulation model

Taking laser energy absorption , heat conduction by free electrons as well as energy exchange between electron and lattice into account, a complete model is developed and applied to investigate the phenomenon of laser induced thermal transport and spallation in the Cu film. Four models, A, B, C, and D with the same area of cross- section and different thicknesses containing 4 192, 5 472, 6 752, and 8 032 atoms respectively are used in the simulations. Infrared laser pulse enters the film in z direction with a pulse width τ_p of 1ps and a fluence of 500 MW/mm^2. When the laser pulse duration τ_p is shorter than or comparable to the characteristic time τ_s equilibrating the absorbing volume, this condition is usually referred to as inertial or stress confinement[25-26]. Here $\tau_s \approx L/V_s$ with the absorbing volume $L \approx 100$ nm and the speed of sound $v_s = 4.4$ nm/ps for Cu. This estimate gives 22. 7 ps and satisfies $\tau_p \leqslant \tau_s$, so the spallation occurs in the so-called stress confinement regime in this paper and should have a mechanical nature.

The initial velocities of atoms are given at random with a Gaussian distribution at 300 K, and follow the Maxwellian distribution at the same temperature in thermal equilibrium state through thermalizing for 50 ps before laser heating, which proves that the model is fully acceptable.

3 Results and discussion

3.1 Propagation of laser induced stress wave

In the stress confinement irradiation regime, the laser heating takes place at nearly constant volume conditions, causing a stress wave with high thermoelastic pressure buildup. Fig. 2 shows the peak values of stress wave at different time in films, which have been determined from virial. At 1 ps when pulse is just over, it is found that a compressive stress has been generated in the near surface region. The peak values first increase and then decrease at the time of reaching the back-surface except for film A, which is caused by the onset of an unloading wave and the thinner thickness of film A. The propagation of unloading wave into the film leads to the formation of tensile stress wave, which travels deep into the film leading to the decreased peak values of the front stress wave.

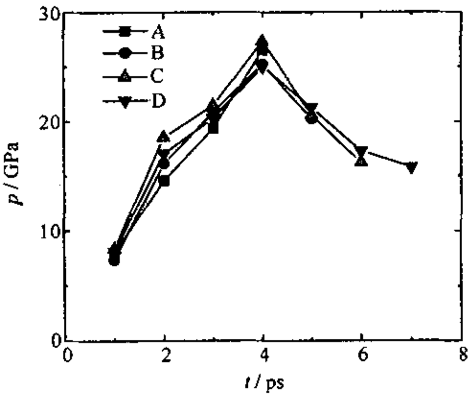

Fig. 2 Peak values of stress wave at different time

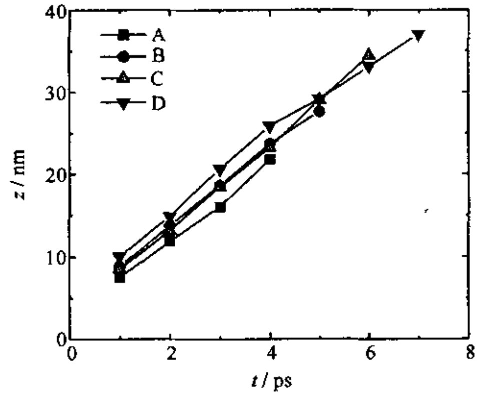

Fig. 3 Transition with time of peak positions of stress wave

Fig. 3 shows the transition with time of peak positions of stress wave. The velocities of stress wave can be determined to be about 4 400 m/s in four films based on the locations of peak values at different time. Therefore it is concluded that the propagation velocity of stress wave does not depend on the thickness of film, but is nearly equal to the sound velocity of longitudinal wave in Cu, and the laser induced stress wave is caused not by heat conduction but by lattice vibration mainly.

3.2 Formation process and mechanism of laser induced spallation

During laser-material interaction, a stress wave is generated and travels towards the film. The stress distribution of lattice for film *B* before spallation is shown in Fig. 4. At the back side of the film with a free surface, the stress wave is reflected completely, and the compressive stress becomes a tensile stress with negative values. Simultaneously, an unloading pressure wave propagating from the front-surface leads to the development of tensile stress with negative values. The spallation occurs when the unloading wave from the front face and the reflected stresses wave generate tensile stresses exceeding the strength of the material. Obviously, there is a peak near the location of spallation with more concentrated tensile stress.

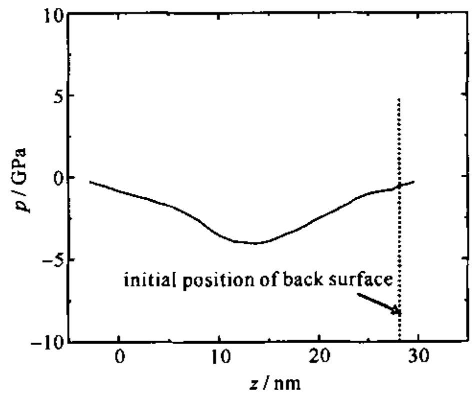

Fig. 4 Stress distribution of lattice in film *B* before spallation

The detailed formation process of laser induced spallation for film B is shown in Fig. 5, where the atoms are darkened according to their local temperature, which is calculated from the average kinetic energy of local atoms. As it can be seen from Fig. 4 and Fig. 5, the breakup occurs at a position which has the maximal temperature, tensile stress and the minimal number density of atoms. The formation process of spallation experiences a three-stage process: void nucleation, growth and coalescence. Firstly, there is a strong tensile concentration near the spall plane before spallation resulting in the consecutive broken of metallic bond and voids in the central of the films. In the meantime, the local temperature strongly increases due to the transformation of potential energy of atoms into kinetic energy. Then some of the voids collapse, others grow, coalesce, eventually leading to the disintegration of the film. No gas-phase atoms have been ob-

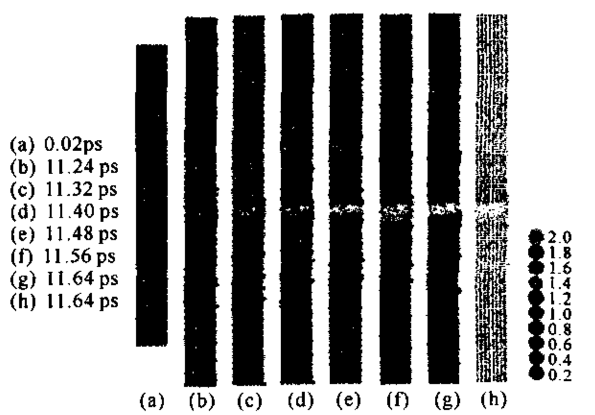

Fig. 5 Snapshots of spallation progress, atoms are darkened according to their local temperature, in units of the melting point of copper T_m

served inside the growing voids, indicating that the processes of void nucleation and growth are not related to boiling but have a mechanical nature, which proves spallation occurs under stress confinement condition.

At the same time of front-surface expansion, the back surface is deviated from its initial position and moves downwards in the direction of depth, which was also found by Elizer[27]. It takes no more than 1ps for spallation and void distribution is convergent, resulting in a narrow zone of spall damage. Such a narrow zone, in which the coalescing voids concentrate, is the characteristic of high strain rate. Fig. 5(g) and Fig. 5 (h) are the snapshots of the same time with different sizes of atoms painted. In Fig. 5(h), there are a few disordered atoms near the spallation surface supporting the statement of small damage area. In addition, the regular lattice structure near the spall plane is still preserved without melting, indicating that the spallation is a result of dynamic tensile stress perpendicular to this plane and independent of the surface ejection accompanied by melting or evaporation under laser irradiation.

3.3 Influence of film thickness on spallation thickness and start time of spallation

Fig. 6 shows the evolution of spallation thickness and the start time of spallation *vs* film thickness. The spallation start time delays and the spallation thickness increases with thicker film. This is caused by a rapid hydrodynamic attenuation: the thicker the film is, the lower the stress amplitude at the back-surface is. So the longer the over tension to achieve spallation, the later the spall starts. As a result, the thicker the spallation will be. Compared to the films with micron thickness[28-29], the ratio of spallation thickness to film thickness is bigger and the position of spall plane is found to be located farther away from the back-surface. This observation is attributed to the strong temperature dependence of the material's tensile strength, which decreases with the increasing temperature. For thicker film at micron scale, the influence of tensile strength changing with temperature is weaker than that for thinner film.

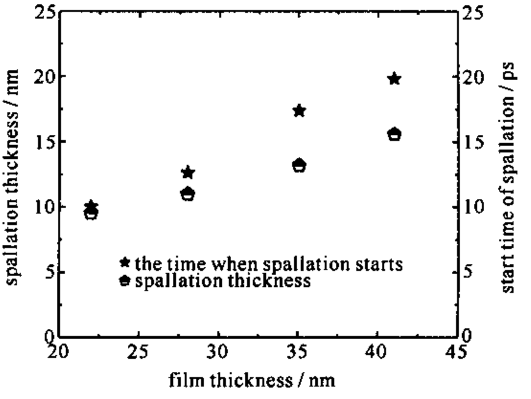

Fig. 6 Evolution of spall thickness and the time when spallation starts *vs* film thickness

4 Conclusion

The dynamics of picosecond laser pulse induced spallation in Cu film in stress confinement regime is studied by means of molecular dynamics method. The main conclusions are as follows: Spallations are observed at the positions of 9.52, 11, 13.2 and 15.6 nm and the spallation start time is determined to be 10.014, 12.64, 17.38 and 19.82 ps respectively; The snapshots of atoms provide a detailed description of spallation process, which proves the three-stage process of the spallation: void nucleation, growth and coalescence; The mechanism of spallation is analyzed and the spallation is caused by the interaction of the unloading wave and reflected stresses wave; The spallation thickness as well as the time when spallation sets in are observed to increase with the film thickness.

References:

[1] Barbee T W, Seaman L, Crewdson R, et al. Dynamic fracture criteria for ductile and brittle metals[J]. *J Mater*, 1972, **7**:393-401.

[2] Johnson J N. Dynamic fracture and spallation in ductile solids[J]. *J Appl Phys*, 1981, **52**:2812-2825.

[3] Kanel G I, Razorenov S V, Bogatch A, et al. Spall fracture properties of aluminum and magnesium at high temperatures[J]. *J Appl Phys*, 1996, **79**:8310-8317.

[4] Zhidkov A G , Zhigilei L V, Sasaki A, et al. Short-laser-pulse driven emission of energetic ions into a solid target from a surface layer spalled by a laser prepulse[J]. *Appl Phys A*, 2001, **73**:741-747.

[5] Zhigilei L V, Leveugle E, Garrison B J, et al. Computer simulations of laser ablation of molecular substrates[J]. *Chem Rev*, 2003, **103**:321-348.

[6] Gilath I, Eilezer S. Brittle-to ductile transition in laser-induced spall at ultrahigh strain rate in 6061-T6 aluminum alloy[J]. *Appl Phys Lett*, 1988, **52**(15):1207-1209.
[7] Cottet F, Boustie M. Spallation studies in aluminum targets using shock waves induced by laser irradiation at various pulse duration[J]. *J Appl Phys*, 1989, **66**(9):4067-4073.
[8] Zhigilei L V, Garrison B J. Microscopic mechanisms of laser ablation of organic solids in the thermal and stress confinement irradiation regimes[J]. *J Appl Phys*, 2000, **88**:1281-1298.
[9] Zhigilei L V, Garrison B J. Computer simulation study of damage and ablation of submicron particles from short-pulse laser irradiation[J]. *Appl Surf Sci*, 1998, **127** - **129**:142-150.
[10] Zhidkov A G, Zhigilei L V, Sasaki A, et al. Short-laser-pulse-driven emission of energetic ions into a solid target from a surface layer spalled by a laser prepulse[J]. *Appl Phys A: Mater Sci Process*, 2001, **73**:741-747.
[11] Paltauf G, Schmidt-Kloiber H. Laser-induced micro bubble formation at a fiber tip in absorbing media: Experiments and theory[J]. *Appl Phys A*, 1996, **62**:303-311.
[12] Li Y C, Tan F L, Guo Y, et al. Determination of the damage evolution equation and spallation criterion of metals[J]. *Journal of Ningbo University (NSEE)*, 2003, **16**(4):442-446.
[13] Rinehart J S. Some quantitative data bearing on the scabbing of metals by an explosive attack[J]. *J Appl Phys*, 1951, **22**:555-562.
[14] Whiteman P. Preliminary report on the effect of stress rate on the dynamic fracture in steel, brass and lauminium[R]. AWRE SWAN-10/611, 1962.
[15] Thurstan R S, Mudd W L. Spallation criterion for numerical computational data[R]. LA-4013 TID-4500, 1968.
[16] Tuler D R, Butcher B M. A criterion for the time-dependent of dynamic fracture[J]. *J Fra Mech*, 1968, **4**:431-440.
[17] Qiu T Q, Tien C L. Mass transfer in turbulent impinging slot jets[J]. *Journal of Heat Transfer*, 1993, **835**:115-124.
[18] Wellershoff S S, Hohlfeld J, Güdde J, et al. The role of electron-phonon coupling in femtosecond laser damage of metals[J]. *Appl Phys A: Mater Sci Process*, 1999, **69**:99-107.
[19] Antaki P J. Importance of nonequilibrium thermal conductivity during short-pulse laser-induced desorption from metals[J]. *Int Heat Mass Transfer*, 2002, **45**:4063-4067.
[20] Schmidt V, Husinsky W, Betz G. Ultrashort laser ablation of metals: pump-probe experiments, the role of ballistic electrons and two-temperature model[J]. *Appl Surf Sci*, 2002, **197**:145-155.
[21] Häkkinen H, Landman U. Superheating, melting, and annealing of copper surfaces[J]. *Phys Rev Lett*, 1993, **71**:1023-1026.
[22] Girifalco L A, Weizer V G. Application of the Morse potential function to cubic metals[J]. *Physical Review*, 1959, **114**:687-690.
[23] Liu X, Wang Y. Femtosecond laser ablation of metals: an molecular dynamics simulation study[J]. *Chinese Optics Letters*, 2005, **3**:57-59.
[24] Liu X, Wang Y. Molecular dynamics study on the mechanism of ultrashort laser ablation of metals[J]. *High Power Laser and Particle Beams*, 2005, **17**(4):500-504. (in Chinese)
[25] Zhigilei L V. Dynamics of the plume formation and parameters of the ejected clusters in short-pulse laser ablation[J]. *Appl Phys A*, 2003, **76**:339-350.
[26] Zhigilei L V, Leveugle E, Garrison B J, et al. Computer simulations of laser ablation of molecular substrates[J]. *Chem Rev*, 2003, **103**:321-348.
[27] Eleezer S, Gilath I. Laser-induced spall in metals: experiment and simulation[J]. *J Appl Phys*, 1990, **67**(2):715-724.
[28] Wang Y G, Boustie M, He H L, et al. Numerical simulation of damage evolution on mechanical behavior and tensile spallation of pure aluminium under laser shock loading[J]. *High Power Laser and Particle Beams*, 2005, **17**(9):1281-1285. (in Chinese)
[29] Wang Y G, Boustie M, He H L, et al. Experimental study on mechanical behavior and tensile spallation of pure aluminum under laser shock loading[J]. *High Power Laser and Particle Beams*, 2005, **17**(7):966-970. (in Chinese)

超短激光诱导金属薄膜后向层裂的分子动力学模拟

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摘 要: 采用结合双温模型的分子动力学方法详尽描述了应力约束区域内部金属薄膜后向层裂的动力学过程。与辐照表面在激光加热作用下机械稳定性受到强烈影响而发生的前向喷射不同,后向层裂是冷材料的断裂。分析了层裂机制,得出靶材是在卸载波及被反射的压力波的共同作用下发生层裂;探讨了激光诱导压力波的传播规律,预测了不同靶厚下的层裂厚度及其对层裂开始时间的影响。

关键词: 后向层裂; 超短激光; 分子动力学; 金属薄膜