Mechanics of Materials 42 (2010) 491–501

Contents lists available at ScienceDirect

Mechanics of Materials

journal homepage: www.elsevier.com/locate/mechmat

Thermomechanical wave propagation in gold films induced by ultrashort laser pulses

Yong Gan [a], J.K. Chen [b,*]

[a] Department of Civil & Environmental Engineering, University of Missouri, Columbia, MO 65211, USA
[b] Department of Mechanical & Aerospace Engineering, University of Missouri, Columbia, MO 65211, USA

ARTICLE INFO

Article history:
Received 25 September 2009
Received in revised form 23 January 2010

Keywords:
Ultrafast thermomechanics
Ultrashort pulse laser
Metal thin film
Two temperature model
Molecular dynamics

ABSTRACT

The thermomechanical behavior of gold films caused by ultrashort pulse lasers is investigated using a combined continuum-atomistic approach with the inclusion of the hot electron blast force. During the very early laser-metal interaction, a nonequilibrium thermal state and a significant hot electron blast wave are generated in the films. The laser heating leads to an initial compressive stress with the peak occurring near the irradiated side of the films. For a 100-nm film, a conversion from compression to tension starts in the mid-portion, the resulting tensile stress converts back to compression, and then the above stress conversions repeat. For the thicker films of 500 nm and 1 μm, the tensile stress starts from the irradiated side due to the free surface stress reflection, and the resulting two-fold shock wave comprising tension and compression propagates towards the rear surface and then reflects back to the front-side. The effect that the thermal stress decreases with increase of the pulse duration is more pronounced in thinner films. Based on the simulated dynamic stress responses, nonthermal damage to the 100-nm film could occur in its mid-depth region while the material could be nonthermally removed from either the front or rear-side of the 500-nm and 1-μm films.

1. Introduction

Ultrashort laser pulse has been attracting great attention due to their wide range of applications in the areas of physics, chemistry, biology and engineering. For example, ultrafast laser pulse could be used to generate high-density plasmas (Murnane et al., 1989), demagnetize ferromagnetic metallic thin films (Bigot et al., 2009), observe fundamental processes of chemical reaction dynamics such as bond formation and breaking (Zewail, 2000; Frischkorn and Wolf, 2006), deliver foreign gene into cells *in vitro* (Tirlapur and König, 2002), image and manipulate biological systems (Sheetz and Squier, 2009), and synthesize metal and semiconductor nanomaterials (Lu et al., 2007; Besner et al., 2008; Liu et al., 2008).

On the other hand, throughout the years, the extensive applications of structures at the micro- and nanoscale level have stimulated tremendous efforts to develop robust techniques for micro- and nanostructuring (Madou, 2002). Among those efforts, ultrashort laser pulses have been demonstrated to be a promising and powerful tool for precisely processing metal thin films (Shirk and Molian, 1998; Preuss et al., 1995). This material processing can be categorized into two main regimes: nanostructure fabrication and high-precision machining. The former includes changing the film surface topography and forming nanojets and nanobumps on the film surface in a controllable way (Nakata et al., 2003; Korte et al., 2004; Koch et al., 2005; Kuznetsov et al., 2009). The latter is to machine the films by removing material with minimal burr formation and collateral damage (Momma et al., 1996; Perry et al., 1999). Under the ultrashort laser irradiation, a metal target would experience a highly nonequilibrium thermal state and thus

* Corresponding author. Tel.: +1 573 882 8159; fax: +1 573 884 5090.
E-mail address: chenjnk@missouri.edu (J.K. Chen).

0167-6636/$ - see front matter © 2010 Elsevier Ltd. All rights reserved.
doi:10.1016/j.mechmat.2010.01.006

exhibits very different thermomechanical behaviors from those caused by conventional pulses longer than a few hundred ps (10^{-12} s). Therefore, further studies toward a thorough understanding of ultrafast laser-induced thermomechanical behaviors of metal thin films are important for the exploration and optimization of ultrafast laser processing of metal thin films.

The present work focuses on the thermomechanical response of gold thin films caused by ultrashort pulse lasers. Numerical simulation complements analytical theory by enabling the extension of its scope beyond strict limits and experiments by facilitating measurements of quantities of theoretical interest and cost-effective parametric studies. So far, the continuum-based approach has been largely used to model the ultrashort laser-metal interaction (Chen et al., 2002, 2004; Wang and Xu, 2002; Meshcheryakov and Bulgakova, 2006; Lee and Tsai, 2008). It is well known that an ultrashort laser processing could result in extremely high temperature (e.g., 1000 K) and strain rate (e.g., 10^9 s^{-1}). The main challenge of the continuum-based approach is the availability of thermomechanical material properties at such extremely high strain rates and elevated temperatures. To circumvent this difficulty, a combined continuum-atomistic modeling method without requiring *a priori* knowledge of size, temperature and rate dependent thermomechanical properties is proposed in this study.

The numerical simulations are performed for three gold films of various thicknesses subject to femto- and picosecond laser pulses to study ultrafast thermomechanical response in metal films. The remaining paper is organized as follows. Section 2 gives the coupling of the continuum-level two temperature model (TTM) and the classic molecular dynamics (MD) with the hot electron blast force for the ultrafast laser-metal interaction. Then, the detailed simulation procedure, results and discussion are presented in Section 3. Conclusions are drawn in Section 4.

2. Modeling of ultrafast laser-metal interaction

For metals, the deposited laser energy is firstly absorbed by electrons, and then transferred to the lattice via collision between electrons and phonons. Since ultrashort laser pulses have a duration less than or comparable to the time required for the electron–phonon equilibration, a thermal nonequilibrium between electrons and lattice occurs. At the continuum-level, the nonequilibrium thermal transport can be described by the TTM (Chen and Beraun, 2001).

$$C_e\frac{\partial T_e}{\partial t} = \nabla(K_e\nabla T_e) - G(T_e - T_l) + S \tag{1}$$

$$C_l\frac{\partial T_l}{\partial t} = \nabla(K_l\nabla T_l) + G(T_e - T_l) \tag{2}$$

where T is temperature, C is heat capacity, K is thermal conductivity, G is the electron–phonon coupling parameter, S is the volumetric laser heat source, t represents time, and the subscripts e and l denote the quantities of the electron and lattice, respectively.

Essentially, the thermomechanical response of metals caused by ultrashort laser pulses can be investigated by coupling the TTM with thermomechanical equations. However, for the analysis of ultrafast laser processing of metal films, this continuum-based approach has two main drawbacks: (1) the required lattice thermal and mechanical properties at extremely high rates and elevated temperatures, such as thermal expansion coefficients, moduli, heat capacity, and thermal conductivity, are barely available, and (2) the progressive lattice information, e.g., crack formation and phase change due to superheating, can not be accurately described. To overcome these limitations, an MD model is introduced to couple with the energy equation of electrons, Eq. (1), in this work. It has been shown that MD is an efficient tool for modeling ultrafast thermomechanical behavior of materials under extremely high temperature and strain rate loading (e.g., Gan and Chen, 2009a). In the MD simulation, the lattice properties are considered through the interatomic potential and the lattice deformation can be easily tracked and examined by the atom's trajectory.

The equations of motion for a MD system are written as

$$m_i\frac{d^2\mathbf{r}_i}{dt^2} = -\sum_{\substack{j=1\\ j\neq i}}^{N}\frac{\partial U(\mathbf{r}_{ij})}{\partial \mathbf{r}_{ij}} \tag{3}$$

in which m_i and $\mathbf{r}_i$ are the mass and position of atom i, respectively, $U(\mathbf{r}_{ij})$ is the interatomic potential between atoms i and j separated by the distance $\mathbf{r}_{ij}$, N is the total number of atoms in the system, and the subscript i runs over all the atoms in the system. Due to the vast number of atoms, the equations of motion are usually integrated by numerical algorithms (Allen and Tildesley, 1987), e.g., Velocity Verlet and Gear Predictor-Corrector. Integrating the equations of motion yields the atom's trajectory, from which the properties of interest can be extracted. It should be noted that only the lattice system is considered in the classic MD simulation.

In the combined TTM-MD approach, the electron–lattice interaction $G(T_e - T_l)$ in Eq. (1) is coupled into the MD equation so that the lattice temperature evolution can be presented. The coupling of the TTM and MD can be fulfilled in two ways. The first method is to add one damping force term to the equations of motion (Ivanov and Zhigilei, 2003),

$$m_i\frac{d^2\mathbf{r}_i}{dt^2} = -\sum_{\substack{j=1\\ j\neq i}}^{N}\frac{\partial U(\mathbf{r}_{ij})}{\partial \mathbf{r}_{ij}} - \xi m_i\boldsymbol{v}_i^T \tag{4}$$

$$\xi = \frac{\frac{1}{n}V_c\sum_{m=1}^{n}G(T_l - T_e^m)}{\sum_{k=1}^{N_V}m_k(\boldsymbol{v}_k^T)^2} \tag{5}$$

In this work, the explicit finite difference method (FDM) is used to solve Eq. (1) for the electron temperature. The MD system is discretized into a collection of volumes corresponding to the FDM cells. The temperatures of electrons and lattice are thus associated with the FDM cells. The maximum time step for the FDM determined by the von Neumann stability criterion is typically smaller than the time step used in the integration of the MD equations of motion. Hence, the MD time step is chosen to be multiple times of the FDM one. In Eq. (5), $\boldsymbol{v}_i^T$ is thermal velocity of atom i, V_c is the volume of the FDM cell in which atom i