Coherent Atomic Motion in Nano-crystal Film

Junjie Li, Xuan Wang, Shouhua Nie, Richard Clinite, and Jianming Cao (FSU, Physics)

Introduction

The structural dynamics of thin films induced by the ultrafast laser excitation has been a subject of extensive study for many years. Some recent measurements using ultrafast diffraction reveal a large in-plane (along the film surface) atomic motions [1], which can not be explained by the commonly used one-dimensional (along the film normal) model. Here, we report a theoretical study of the structural dynamics in metallic films in response to ultrafast laser heating using a two-dimensional model, which incorporates the atomic motions in both in-plane and normal directions. The results show that the surface shape and the orientations of nano-crystal grains are essential to determine the modes of lattice motions. Moreover, a large projection of coherent lattice oscillation in the in-plane direction is found, which was thought to be very small and was neglected in one-dimensional models.

Results and Discussion

Fig. 1 (a) Two typical lattice configurations. (b) Calculated result of Bragg Peak oscillation in the in-plane direction. The temperature change of 10-nm Aluminum film is 20 degree, corresponding to pump laser power of 3 mJ/cm². (c) Ultrafast electron diffraction experimental results of 20-nm Aluminum film with pump power 3 mJ/cm² [1]. The difference in vibrational period in (b) and (c) is due to the difference in film thickness.

In the calculation, the orientation of nano-crystal particles is assumed to be isotropic and connected by grain boundaries. The Fermi-Pasta-Ulam model is used to calculate the lattice expansion of each nano-particle, which assumes that atoms only interact with their closest neighbors via harmonic potential and the temporal evolution of atomic position is calculated by:

\[
H = \sum_{i,j=1}^{N} \left( \frac{\tilde{p}_{i,j}^2}{2m} + U_{i,j}(x, y, T) \right), \quad \frac{\partial \tilde{p}_{i,j}}{\partial t} = -\nabla U_{i,j}(x, y, T) = -(k_{y} \Delta \tilde{x}_{i,j} + k_{x} \Delta \tilde{y}_{i,j}) \quad \text{[1]}
\]

Considering that electron-phonon thermalization time (< 2 ps) is much shorter than the period of film coherent vibration (~ 7 ps), we assume an instant build-up of thermal stress. The temporal evolution of Bragg (311) peak position is shown in the figure 1 (b). Our simulation produces large amplitude of coherent lattice motion in the in-plane direction, which is totally ignored in the one-dimensional model. In addition, the simulation results exhibit very good agreements in the oscillation amplitude with that of experimental results.

Acknowledgements

The project is partially supported by the National Science Foundation under the grant number DMR-0606431.

References