Available online at www.sciencedirect.com
ScienceDirect
Applied Surface Science 253 (2007) 6295–6300
applied surface science
www.elsevier.com/locate/apsusc

Temperature dependences of the electron–phonon coupling, electron heat capacity and thermal conductivity in Ni under femtosecond laser irradiation

Zhibin Lin, Leonid V. Zhigilei *

Department of Materials Science and Engineering, University of Virginia, 116 Engineer's Way, Charlottesville, VA 22904-4745, USA

Available online 23 January 2007

Abstract

The electron temperature dependences of the electron–phonon coupling factor, electron heat capacity and thermal conductivity are investigated for Ni in a range of temperatures typically realized in femtosecond laser material processing applications, from room temperature up to temperatures of the order of 10^4 K. The analysis is based on the electronic density of states obtained through the electronic structure calculations. Thermal excitation of d band electrons is found to result in a significant decrease in the strength of the electron–phonon coupling, as well as large deviations of the electron heat capacity and the electron thermal conductivity from the commonly used linear temperature dependences on the electron temperature. Results of the simulations performed with the two-temperature model demonstrate that the temperature dependence of the thermophysical parameters accounting for the thermal excitation of d band electrons leads to higher maximum lattice and electron temperatures achieved at the surface of an irradiated Ni target and brings the threshold fluences for surface melting closer to the experimentally measured values as compared to the predictions obtained with commonly used approximations of the thermophysical parameters.

PACS : 61.80.Az; 71.20.Be; 63.20.Kr; 79.20.Ds

Keywords: Electron density of states; Electron–phonon coupling; Electron heat capacity; Thermal conductivity; Two-temperature model; Laser melting

1. Introduction

The fast growth of femtosecond laser applications has increased the demand for realistic computational description of highly nonequilibrium processes induced in a target material by the fast laser energy deposition. The time evolution of the electron and lattice temperatures in a metal target irradiated by a femtosecond laser pulse is commonly described by the two-temperature model (TTM) [1], that accounts for the laser energy absorption by the conduction band electrons, energy transfer from hot electrons to the atomic vibrations due to the electron–phonon coupling, and the electronic heat diffusion from the irradiated surface to the bulk of the target.

The accuracy of a quantitative description of the kinetics of energy redistribution in the irradiated target in a big part relies on the appropriate choice of the temperature dependent thermophysical properties of the target material included in the TTM equations, namely, the electron–phonon coupling factor, the electron heat capacity and the heat conductivity. Due to the small electron heat capacity of the electrons, the electron temperature in the surface region of the irradiated target can be transiently brought to very high values, comparable to the Fermi temperature. At such high electron temperatures, T_e, the temperature dependent thermophysical properties of noble [2–5] and transition metals [5] can be directly affected by the thermal excitation of the lower band electrons.

The effect of the thermal excitation of electrons on the thermophysical properties is sensitive to the details of the electronic structure of the target material. In particular, in Au the d band lies ~2 eV below the Fermi level and at T_e below

* Corresponding author. Tel.: +1 434 243 3582; fax: +1 434 982 5660.
E-mail addresses: zl5c@virginia.edu (Z. Lin), lz2n@virginia.edu (L.V. Zhigilei).

0169-4332/$ – see front matter © 2007 Elsevier B.V. All rights reserved.
doi:10.1016/j.apsusc.2007.01.032

~3000 K (~0.25 eV) the d band electrons do not contribute to the electron–phonon coupling and the electron heat capacity, whereas at $\sim 10^4$ K (~1 eV) thermal excitation of d band electrons results in a significant increase of both the electron–phonon coupling factor and the electron heat capacity (e.g. the electron–phonon coupling factor exceeds the room temperature value by a factor of 8.9 at $T_e = 2 \times 10^4$ K [4]). On the other hand, in transition metals, such as Ni or Pt, the Fermi level cuts through the d band, resulting in a very high density of electron states at the Fermi level. In this case, the thermal excitation of electrons from high density of states d band to the low density of states s band can have the opposite effect on the electron heat capacity and electron–phonon coupling as compared to noble metals. In particular, the electron heat capacity of Pt has been shown to become smaller than the value given by the linear dependence on the electron temperature predicted by the Sommerfeld expansion for the electronic free energy [5]. Therefore, a detailed analysis of the connections between the electron density of states (DOS) in the target material and the temperature dependence of the thermophysical material properties is needed for a realistic description of laser material interactions.

In this paper, we investigate the effect of the thermal excitation of the d band electrons on the thermophysical properties of Ni. In the next section, the connections between the electron DOS of Ni and the temperature dependence of the electron–phonon coupling, electron heat capacity and thermal conductivity are analyzed. The qualitative differences between the temperature dependences obtained for Ni and earlier predictions for Au [4] are related to the differences in the electron DOS of the two metals. In Section 3, practical implications of the thermal excitation of the d band electrons are investigated by performing a series of TTM simulations with the modified temperature-dependent thermophysical properties. The evolution of the kinetics of the electron–phonon equilibration, the electron and lattice temperatures, and the fluence thresholds for surface melting are investigated for Ni films of various thicknesses and a bulk Ni target. A brief summary of the results is given in Section 4.

2. The effect of thermal excitation of electrons on thermophysical properties of Ni

The effect of the thermal excitation of electrons on thermophysical properties of Ni is investigated here based on the electron DOS obtained from the electronic structure calculation performed within the density functional theory using the Vienna Ab-initio Simulation Package (VASP) [6]. The Projector Augmented Wave (PAW) potential [7] is used in the calculation, where the exchange correlation term is treated within the Generalized Gradient Approximation (GGA). The calculations are done for nonmagnetic Ni at the equilibrium lattice constant of 3.53 Å. The electron DOS for Ni obtained from VASP at $T_e = 0$ K is shown in Fig. 1, together with the Fermi distribution function plotted at three different values of the electron temperature.

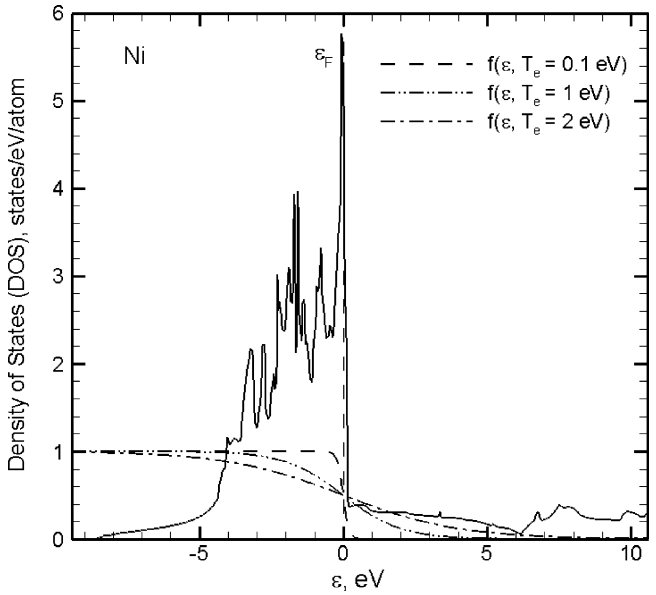

Fig. 1. Electron DOS of Ni obtained in electron structure calculations performed with VASP (solid line). The Fermi distribution function is also shown for three different values of the electron temperature. The energy is shown with respect to the Fermi level at zero temperature, ε_F.

The electron heat capacity dependence on the electron temperature can be expressed as [8]:

$$C_e(T_e) = \int_{-\infty}^{\infty} (\varepsilon - \varepsilon_F) \frac{\partial f(\varepsilon, \mu, T_e)}{\partial T_e} g(\varepsilon)\, d\varepsilon \quad (1)$$

where $g(\varepsilon)$ is the electron DOS at the energy level ε, μ the chemical potential at T_e and $f(\varepsilon, \mu, T_e)$ is the Fermi distribution function, defined as $f(\varepsilon, \mu, T_e) = \{\exp[(\varepsilon - \mu)/k_B T_e] + 1\}^{-1}$. The determination of the chemical potential μ, required in Eq. (1), is done through setting the result of the integration of the product of DOS and the Fermi distribution function at T_e over all energy levels to be equal to the total number of valence electrons. It can be seen from Figs. 1 and 2(a) that as the electron temperature increases, the excitation from high density of states below ε_F to the states above ε_F increases, leading to the increase in the chemical potential, whereas the chemical potential of the free electron model, calculated from the Sommerfeld expansion at low temperatures [8], decreases.

In Fig. 2(b), the temperature dependence of the electron heat capacity, calculated from Eq. (1) with the DOS determined from VASP calculations, is shown together with the commonly used linear approximation, $C_e(T_e) = \gamma T_e$, obtained from the Sommerfeld expansion with $\gamma = 1065$ J m^{-3} K^{-2} measured at low temperatures [9]. The heat capacity calculated with the realistic DOS is lower than the one predicted by the linear dependence, $C_e(T_e) = \gamma T_e$, at all temperatures and exhibits a steadily increasing deviation from the linear dependence. This deviation can be explained by analyzing the characteristics of the Ni DOS shown in Fig. 1. The high density of electron states at the Fermi level ensures that the 3d band electrons can be easily excited to the 4s band. The 4s band has a much smaller density of states as compared to the density of states at the