Nuclear Instruments and Methods in Physics Research B 342 (2015) 300–304

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research B

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

Electron emission from a double-layer metal under femtosecond laser irradiation

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ARTICLE INFO

Article history:
Received 11 July 2014
Received in revised form 15 October 2014
Accepted 17 October 2014
Available online 11 November 2014

Keywords:
Electron emission
Double-layer structure
Femtosecond laser

ABSTRACT

In this paper we theoretically investigate electron emission during femtosecond laser ablation of single-layer metal (copper) and double-layer structures. The double-layer structure is composed of a surface layer (copper) and a substrate layer (gold or chromium). The calculated results indicate that the double-layer structure brings a change to the electron emission from the copper surface. Compared with the ablation of a single-layer, a double-layer structure may be helpful to decrease the relaxation time of the electron temperature, and optimize the electron emission by diminishing the tailing phenomenon under the same absorbed laser fluence. With the increase of the absorbed laser fluence, the effect of optimization becomes significant. This study provides a way to optimize the electron emission which can be beneficial to generate laser induced ultrafast electron pulse sources.

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1. Introduction

The rapid development of ultrafast laser techniques, such as chirped-pulse amplification, makes it possible to obtain powerful femtosecond laser pulses [1,2]. Studies of the interaction between a femtosecond laser and metal become more important to many applications, and among these studies, that of the electron emission relaxation dynamics attracts intense interest [3–7]. The femtosecond laser pulse induced photoelectron emission and thermoelectron emission are widely used in basic researches and practical applications [8–10]. The ultrafast electron pulse generated by a femtosecond laser pulse can be used to diagnose the plasma [11–13], and confirm the lifetime of excited electrons [14–16], etc. In addition, it can also be used in the generation of laser induced electron pulse sources [17] and ultrashort X-ray pulses [18,19].

According to published studies of electron emission, it is known that photoelectron emission is the main body of the ultrashort electron pulse source. When the femtosecond laser is applied in the ablation, photoelectron emission occurs in the course of laser pulse duration. However, the thermoelectron emission with high electron temperature, takes place in the thermal relaxation period of metal, and influences the profile of the produced electron pulse. From solid state physics, we know that the electrons are bound to the band of energy levels and can move within the metal. However, they can be released after absorbing enough energy. Therefore, the electron temperature is a key factor in determining the number of the emitted electrons which can be a character of the electron emission current. Along with the deepening of the studies about electron emission physical mechanism, the details of the electron emission process have been known [20–24].

Femtosecond laser induced ultrafast electron emission consists of thermoelectron emission and photoelectron emission. In practical applications, the researchers hope that the duration of the ultrafast electron beam is narrow. If the temperature of electron on the metal surface is high enough, these electrons will obtain thermal energy to overcome the bound potential, whereas an undesirable feature of thermoelectron emission – tailing phenomenon comes along at the same time [25,26]. The tailing phenomenon enlarges the duration of the emitted electron pulse, and the direct way to reduce the tailing phenomenon of thermoelectron emission is to decrease the relaxation time of electron temperature on the metal surface. Consequently, decreasing the relaxation time helps to decrease the duration of thermoelectron emission.

In this paper, we use a double-layer metal structure as our investigation model due to the fact that some papers suggest that the substrate layer can help to reduce the temperature relaxation time of the surface layer during femtosecond laser irradiation

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http://dx.doi.org/10.1016/j.nimb.2014.10.014
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[27,28], which optimizes the duration of the electron emission. We take advantage of a theoretical method to investigate the electron temperature. As we introduce the double-layer structure, the two-temperature function is revised by considering the boundary between surface and substrate layers. After calculating the temperature of electron and lattice, the Richardson–Dushman equation which describes the rate of electron emission is applied. The samples in the calculation are a single-layer Cu and a double-layer structure composed of Cu and substrate metals, such as Au and Cr. The predicted results show that the substrate layer can influence the electron emission.

2. Mathematical model

2.1. Two-temperature model

The two-temperature model (TTM) is used to calculate the temperature of electrons for it considers the variation of the temperature of lattice at the same time during the calculation. What is more, the response time of the metals heated by femtosecond laser pulse is much shorter than the relaxation time of electron–lattice coupling [29,30].

It is known that the radiation energy is initially absorbed by the free electrons of metals and then transferred to the lattice. The TTM helps to describe the process of temperature increase caused by the interaction between laser pulse and metal, and also model the energy transfer between electrons and lattice at the same time.

The one-dimension TTM is given below:

$$C_e\frac{\partial T_e}{\partial t}=\frac{\partial}{\partial x}\left(k_e\frac{\partial T_e}{\partial x}\right)-G(T_e-T_l)+S \tag{1}$$

$$C_l\frac{\partial T_l}{\partial t}=\frac{\partial}{\partial x}\left(k_l\frac{\partial T_e}{\partial x}\right)+G(T_e-T_l) \tag{2}$$

where t is the time, x is the depth, C_e and k_e are the heat capacity and the heat conductivity of electron, C_l is the lattice heat capacity, k_l is the lattice heat conductivity, T_e and T_l are the temperature of electron and lattice respectively, G is the electron–lattice coupling factor, and S is the laser heat source term. The heat source S can be modeled with a Gaussian temporal profile [31]:

$$S=\sqrt{\frac{\beta}{\pi}}\frac{I_{ab}}{t_p\alpha}\exp\left[-\frac{x}{\alpha}-\beta\left(\frac{t-2t_p}{t_p}\right)^2\right] \tag{3}$$

where I_{ab} is the absorbed laser fluence, $I_{ab}=I_{in}(1-R)$, I_{in} is the incident laser fluence, and $R=0.962$ (at wavelength of 800 nm) is the reflection coefficient of target [32], t_p is the full-width at the half maximum (FWHM), $\alpha=\alpha_p+\alpha_b$ is the sum of the penetration depth ($\alpha_p=13$ nm) and the ballistic range ($\alpha_b=15\pm4$ nm) [33], and from many articles we learn that the ballistic energy transport can influence the lattice temperature [34,35], and $\beta=4\ln(2)$.

When the electron temperature is lower than the Fermi temperature, the electron heat capacity is proportional to T_e as the equation [36]: $C_e=\gamma T_e$, where $\gamma=\pi^2n_ek_B/2T_F$, n_e is the density of the free electrons, and k_B is the Boltzmann constant. The lattice heat capacity is set as a constant because it changes tinily at different temperatures. The electron heat conductivity can be expressed as $k_e=k_{e0}BT_e/(AT_e^2+BT_l)$ [37], where k_{e0}, A and B are the material constants. $G=G_0(A(T_e+T_l)/B+1)$ is the electron–lattice coupling factor, where G_0 is the coupling factor at room temperature [38]. Since the mechanism of heat conduction in metal is mainly determined by the electrons, we consider the lattice heat conductivity as a constant.

The interface of the double-layer metal film is $x=l$, then for the double-layer film, TTM can be expressed as [39]:

$$C_e^I\frac{\partial T_e^I}{\partial t}=\frac{\partial}{\partial x}\left(k_e^I\frac{\partial T_e^I}{\partial x}\right)-G(T_e^I-T_l^I)+S^I \tag{4}$$

$$C_l^I\frac{\partial T_l^I}{\partial t}=\frac{\partial}{\partial x}\left(k_l^I\frac{\partial T_l^I}{\partial x}\right)+G(T_e^I-T_l^I) \tag{5}$$

$$C_e^{II}\frac{\partial T_e^{II}}{\partial t}=\frac{\partial}{\partial x}\left(k_e^{II}\frac{\partial T_e^{II}}{\partial x}\right)-G(T_e^{II}-T_l^{II}) \tag{6}$$

$$C_l^{II}\frac{\partial T_l^{II}}{\partial t}=\frac{\partial}{\partial x}\left(k_l^{II}\frac{\partial T_l^{II}}{\partial x}\right)+G(T_e^{II}-T_l^{II}) \tag{7}$$

2.2. Electron emission model

When the temperature of electrons is enough high, the thermo-electron emission will take place, which causes the tailing part of the Fermi-Dirac distribution leak into the vacuum level [40]. The total electron emission flux J_{em} is consisted of the thermionic emission flux and the photoemission flux, and the rate of electron emission is described [41–43].

$$J_{em}=A_0T_e^2\left[\exp\left(\frac{\varphi_w}{k_BT_e}\right)+\sum_{N=1}^{\infty}a_NF(X_n)\left(\frac{e}{h\nu}\right)^NI_{ab}^N\right] \tag{16}$$

where A_0 is the Richardson–Duschman constant, φ_w is the work function of the metal which is set as 4.56 eV, h is the Planck constant, ν is the laser frequency, T_e is the electron temperature, the parameter a_N is proportional to the N-photon ionization coefficients. The multiphoton emission of Cu is dominated by three-photon photoelectron emission, so we set N as 3 and a_3 as $1\times10^{-46}\,(\mathrm{m^2\,A^{-1}})^3$. The Fowler function $F(X_n)$ can be learned from Refs. [42,44]. All the parameters are listed in Table 1.

2.3. The initial and boundary conditions

The initial conditions to solve the above equations are:

$$T_e^I(x,0)=T_l^I(x,0)=T_0 \tag{8}$$

$$T_e^{II}(x,0)=T_l^{II}(x,0)=T_0 \tag{9}$$

Before the irradiation, the electron and lattice temperature are considered to be at the same initial temperature: $T_0=300$ K. The boundary conditions are:

$$k_e\frac{\partial T_e^I}{\partial x}\bigg|_{x=0}=-(E_f+e\phi)J_{em} \tag{10}$$

$$\frac{\partial T_e^{II}}{\partial x}\bigg|_{x=L}=0 \tag{11}$$

$$\frac{\partial T_l^I}{\partial x}\bigg|_{x=0}=\frac{\partial T_l^{II}}{\partial x}\bigg|_{x=L}=0 \tag{12}$$

where $E_f+e\phi$ is the potential barrier for an electron to escape from the metal surface. As the double-layer structure is in perfect thermal contact, the interfacial thermal resistance is ignorable and is

Table 1
Thermal physical parameters [21,45,46].

	Cu	Au	Cr
G (10^{17} J m^{-3} s^{-1} K^{-1})	1.0	0.22	4.2
γ (J m^{-3} K^{-2})	96.6	68	194
k_{e0} (J m^{-1} s^{-1} K^{-1})	401	315	95
C_l (10^6 J m^{-3} K^{-1})	3.43	2.5	3.3
A (10^7 s^{-1} K^{-2})	1.28	1.18	7.9
B (10^{11} s^{-1} K^{-1})	1.23	1.25	13.4

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