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Interaction of femtosecond laser pulses with metal photocathode*

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The features of interaction of femtosecond laser pulses with photocathode are studied theoretically in this paper. The surface temperature of the metal cathode film while femtosecond laser pulses irradiation is studied with two-temperature model. With a simple photoelectric model we obtain the optimum metal film thickness for the back-illuminated photocathode. The generated ultrashort photocurrent pulses are strongly dependent on the temperature of the electron gas and the lattice during the femtosecond laser pulse irradiation on the photocathode.

Keywords: femtosecond laser, photocathode, ultrashort electron pulse time resolved electron microscopy

PACC: 7960, 8240J, 6320K

1. Introduction

The ultrafast electron dynamics and photoelectric effect when ultrashort laser pulses are incident on solid surfaces have been widely studied.[1−4] The electrons generated by the interaction of femtosecond laser pulse with metal photocathodes replicate the time duration property of the pump femtosecond laser pulse through the ultrafast photoelectric effect. A high-brightness electron beam is also necessary for free electron lasers (FEL),[5] laser accelerators,[6] setups of time-resolved electron diffraction[7,8] and generators of short pulse x-rays by means of nonlinear Thomson scattering.[9]

Could this kind of ultrashort electron pulse serve as the electron source of an electron microscope? This problem is a part of the research of what we call time-resolved electron microscopy (TREM).[10,11] We know that electron microscopy has reached a high space-resolved ability at sub-angstrom resolution, but the time-resolved ability of electron microscopy is seldom studied. Now, it is possible to capture full structural details of transition state processes with the advent of the femtosecond electron diffraction facility.[7,8] In the structure of TREM, the thermic electron gun is substituted by the femtosecond photocathode gun. When the femtosecond laser pulses irradiate the surface of the photocathode, femtosecond electron pulses are generated. With this kind of electron source, the electron microscopy will possess comparable time resolved ability. In other words, we can watch chemical reactions occurring in real time and watch the atoms moving in response to the reaction forces with TREM. This means that many scientists' dream of making the "molecular movie" will come true.

In this paper, we discuss the process of interaction of femtosecond laser pulses with photocathode. In the femtosecond scale, the nonequilibrium of electron and phonon becomes very important during the femtosecond laser pulse irradiation on metal surface. With the advent of femtosecond laser, scientists have a crucial tool to study the ultrafast nonequilibrium electron gas system, such as the transportation and scattering of the carriers in semiconductors, the electron behaviour near the Fermi surface, and the movement of electrons in superconductors. Here we devote more attention to the temperature effect of electron and lattice during the femtosecond laser pulse irradiation on the metal film surface. Then we study the ultrashort photoelectric effect when femtosecond laser pulses are incident on the metal film surface. Combining the electron temperature effects with the ultrafast photoelectric effect model, we can find the optimal copper photocathode thickness.

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2. Temperature effect

Metal photocathodes have many advantages in terms of responsibility, quantum efficiency, work function, and energy distribution of photoelectrons. The metal cathode materials are also inexpensive, have a long lifetime, and could operate at a modest vacuum quality. Therefore we choose metal photocathodes for our TREM. The back-illuminated (BI) photocathode is a kind of metal material deposited on a substrate. Compared with the front-illuminated (FI) photocathode, the BI photocathode has an advantage of easier designing of the electro-optic system for ultrashort electron beams controlling. In this paper, we pay more attention on the BI photocathode that is constructed with the metal films, such as copper, silver or gold materials, deposited on the quartz glass substrate. The schematic interaction is shown in Fig.1.

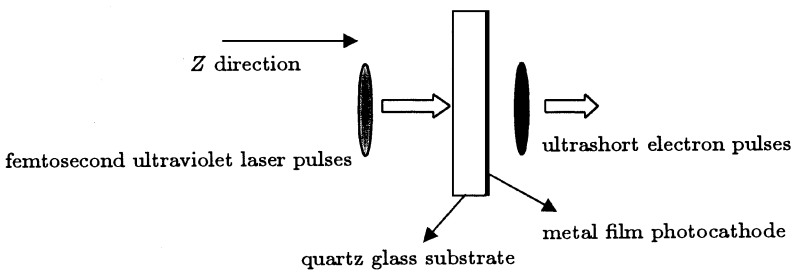


Fig.1. Schematic interaction of femtosecond laser with metal film photocathode.

The nonequilibrium dynamics and temperature effect of the electron gas and the lattice are the potential problems that are not fully understood when ultrashort laser pulses irradiate the solid material. The energy transportation of electron and phonon in a metal, subjected to an ultrashort laser pulse, is extensively studied within the two-temperature model (TTM).[12]

When femtosecond laser pulses are incident on a solid, the temperature of the electron gas and the lattice will change rapidly. For the laser pulse duration shorter than a few picoseconds, the electron gas and the lattice in the solid are in a nonthermodynamic equilibrium during femtosecond laser pulse irradiation.[2] During the process of the interaction of nanosecond laser pulses with solid, the temperatures of the electron gas and lattice in it are characterized by a common temperature T. In the TTM, the electron and phonon subsystems are described by two separate heat baths with temperatures T_e and T_l as denoted respectively by subscripts e and l. C_e and C_l are the electronic and phonon heat capacities respectively.

The TTM is described by

$$C_e(T_e)\frac{\partial T_e}{\partial t} = K \cdot \frac{\partial^2 T_e}{\partial z^2} - g \cdot (T_e - T_l) + G(r,t), \quad (1a)$$

$$C_l(T_l)\frac{\partial T_l}{\partial t} = g \cdot (T_e - T_l), \quad (1b)$$

$$G(r,t) = \alpha(1-R)I_0(r,t)e^{-\alpha z}, \quad (2)$$

where K is the thermal conductivity, R is the surface reflectance, α is the absorption coefficient and g is the electron–phonon coupling coefficient. These equations have been used to explain the laser ablation of metal material[13] and the ultrafast non-equilibrium electron dynamics in the solid.[1,2,14] The electron–phonon coupling coefficient, thermal conductivity, electronic and lattice capacities of several typical metals are shown in Table 1.

Table 1. Parameters in the two-temperature model for some typical metal materials.

Metal material	$g/10^{16}\mathrm{Wm^{-3}K^{-1}}$	$C_e/\mathrm{Jm^{-3}K^{-2}}$	$K_0/\mathrm{Wm^{-1}K^{-1}}$	$C_l/10^6\mathrm{Jm^{-3}K^{-1}}$
gold	2.1	71	318	2.5
molybdenum	13	350	135	2.8
nickel	36	1065	91	4.1
copper	10	96.6	385	3.5
silver	2.3	–	429	–

For the effect of the ballistic electron transportation, the electron mean free path has been determined to be more than 100nm in these films at room temperature. As for most photocathodes, the metal film is deposited on the substrate with a thickness ranging from 10 to 80nm. Because the metal film thickness is much smaller than the electron mean free path, we neglect the heat transportation term in Eqs.(1). We neglect the thermal conductivity also for the reason of ultrashort pump laser pulse duration and the feature of photocathode thickness.

Corkum *et al*[14] estimated the electron diffusive features by neglecting the electron–phonon coupling terms in Eqs.(1), and gave a critical length for the diffusion electrons, which is $L_c = \sqrt{2K_{e,0} \cdot \tau_R / C'_e T_l}$ with $\tau_R = C'_e T_e(z=0)/g$ being the electron–phonon relaxation time. Using the parameters in Table 1, we can estimate τ_R which is about several picoseconds. The equilibration time for electrons and the lattice depends on the electron–phonon coupling coefficient, which will determine the diffusive penetration depth of electrons.

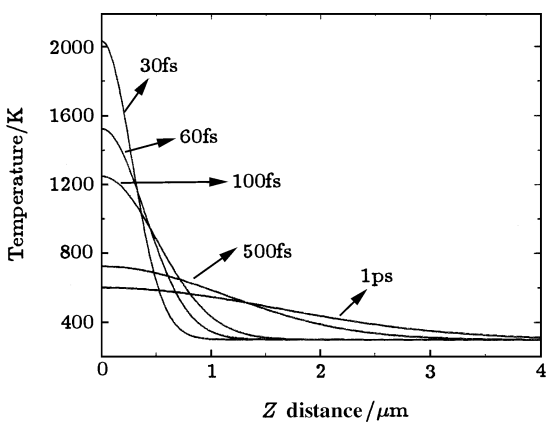


Fig.2. Electron temperature decay as function of time and distance.

As an example, in the copper metal the electron temperature distribution decays with distance larger than several microns and with time longer than several picoseconds if we take into consideration the thermal conductivity term, as shown in Fig.2. The thickness of photocathode is usually about several tens of nanometre, so we can neglect the thermal conductivity during the femtosecond laser irradiation on the metal film. We numerically resolve the TTM model equations without taking into consideration the thermal conduction at the copper photocathode surface. As shown in Fig.3, the electron temperature rises in a very short time during the laser heating process, and then the temperatures of electrons and phonons become the same through the electron–phonon coupling process. During this process, the electron temperature distribution will show non-thermal equilibration.[2] On solving the equations, we consider that the laser intensity I is of an order about $10^{10}\mathrm{W/cm^2}$ and the duration of laser pulses is about 30fs.

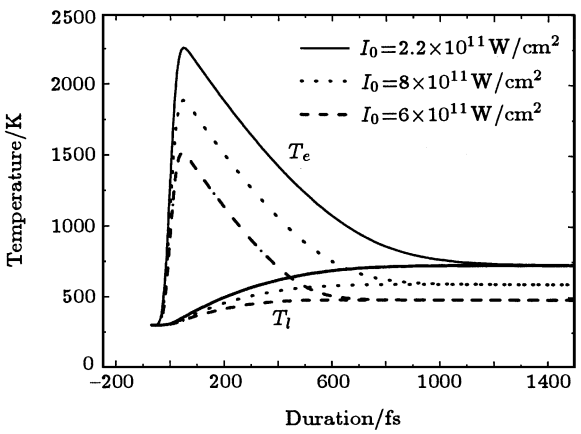


Fig.3. Electron and phonon temperature distributions during the femtosecond laser pulse irradiation on the metal film.

3. Ultrashort photoelectric effect

We know that if the photon energy is higher than the work function of the metal material, photoelectrons are emitted from the metal surface. Since the birth of femtosecond lasers, the ultrafast photoelectric effect has attracted scientists' attention.[14–20] With a simple analytical photoelectric model, we can obtain the ultrashort photoelectron current generated from the metal photocathode irradiated by the femtosecond laser pulses. In this simple model, we find that the maximum photoelectron current will be dependent on the thickness of metal film.

The competing factor that limits the photoelectron current in thick films is the difficulty of electrons escaping from the surface after being excited to energies greater than the work function. Even though a large number of electrons are liberated as photons penetrate into a very thick film, the probability of these electrons propagating through multiple layers of metal atoms and projecting into the vacuum region becomes smaller as the film thickness increases. It is reasonable to expect a maximum photocurrent at a particular thickness for a given competition between the probabilities of photon interaction and electron escaping.

When a laser pulse with an ultrashort duration of t propagates in the direction z, its intensity I_0 atten-

uated to an amount I_z is given by

$$I_z = I_0(t)(1-R)\mathrm{e}^{-\alpha z}, \tag{3}$$

where R is the metal film reflectivity and α is the absorption coefficient of the metal material. What we are interested in is the photoelectrons emitted in the direction of the laser propagation. We assume that the photoelectron yield is isotropic in metal film. In the infinitesimal distance $\mathrm{d}z$ within the film, the absorbed laser energy $\mathrm{d}I_z$ and the generated electron current density $\mathrm{d}J_{z0}$ can be respectively expressed as

$$\mathrm{d}I_z = -\alpha I_0(t)(1-R)\mathrm{e}^{-\alpha z}\mathrm{d}z, \tag{4}$$

$$\mathrm{d}J_{z0} = \frac{\eta e \mathrm{d}I_z}{h\nu} = \frac{\alpha\eta e I_0(t)(1-R)\mathrm{e}^{-\alpha z}\mathrm{d}z}{h\nu}, \tag{5}$$

where η is the photoelectric quantum efficiency, $h\nu$ is the pump photon energy, and e is the fundamental unit of electric charge. Those photoelectrons generated having a certain energy, i.e. only the electrons at a distance to the front side of cathode δ smaller than the mean free path, can escape from the metal film surface. Considering the metal as a collection of atoms per unit volume, the photocurrent at a distance z could be expressed as $\mathrm{d}J_z(t) = \mathrm{d}J_{z0}\mathrm{e}^{-\beta(\delta-z)}$, here β is the photoelectron current decay coefficient. Substituting Eq.(5) into the expression of $\mathrm{d}J_z$, we obtain

$$\mathrm{d}J_z(t) = \frac{\alpha\eta e(1-R)I_0(t)}{h\nu}\exp[-\beta(\delta-z)-\alpha z]\mathrm{d}z. \tag{6}$$

Integrating Eq.(6) with respect to distance from 0 to δ, we obtain the emitted photocurrent as

$$J(t) = \frac{\alpha\eta e}{(\beta-\alpha)h\nu}(1-R)I_0(t)\mathrm{e}^{-\beta\delta}[\mathrm{e}^{(\beta-\alpha)\delta}-1]. \tag{7}$$

From Eq.(7), we can deduce the optimal thickness of the metal film $\delta_{J=\max} = \ln(\beta/\alpha)/(\beta-\alpha)$. If the film is thicker than $\delta_{J=\max}$, the liberated electron should traverse through a longer path length to reach the metal film surface and its motion would not be so easy.

Theoretical calculations of β and α are very difficult. The coefficients β and α are all related with the electron and phonon temperatures that have been predicted in Section 2. The coefficient β must be larger than α because the generated photocurrent will decay rapidly due to electron collisions. The typical absorption coefficient α for some copper at wavelength of 266 nm is about 5×10^7/m. The coefficient β is also connected with the photoelectron energy distribution generated at z. The energy distribution of photoelectron is related with the pump photon energy and work function. The photoelectron energy distribution is non-equilibrium Fermi–Dirac distribution during the femtosecond laser pulse heating. The empirical coefficient β we give is about 5.03×10^7/m for copper at wavelength of 266 nm. The value of α is the ratio of transmission and that of β is the ratio of charge decay. The experimental work of Brogle *et al*,[5] measured the back-illumination photocathode photoelectron emission. The copper film thickness they used is larger than 27 nm, while the optimum thickness of film is actually smaller than 27 nm for copper at wavelength of 266 nm. Actually, when the metal film thickness decreases, no sufficient free electrons are present around the Fermi energy surface of the metal volume. The calculated photocurrent is shown in Fig.4, indicating that the optimum thickness is about 20 nm for copper film. The calculated photocurrent agrees very well with the related quantum efficiency (Q. E.).

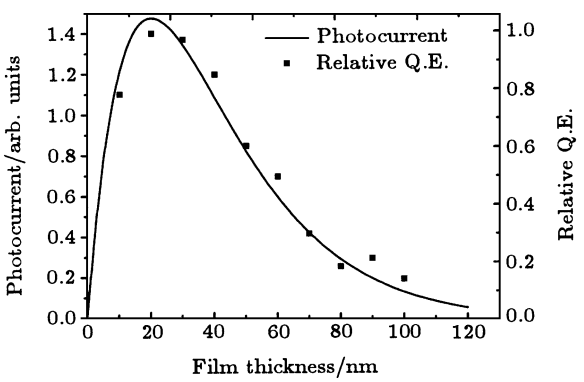


Fig.4. Photoelectron current and relative quantum efficiency versus different thickness of copper film.

Because of the surface reflectivity, the optical absorption and the photoelectron photocurrent decay coefficient are all related to the temperature of the electron gas and lattice. The coefficient of α, β and R should be expressed as

$$\alpha = \alpha_0 + a_1(T_e - T_0) + b_1(T_e - T_0),$$
$$\beta = \beta_0 + a_2(T_e - T_0) + b_2(T_e - T_0),$$

and

$$R = R_0 + a_3(T_e - T_0) + b_3(T_e - T_0)$$

respectively. In order to fully understand the physical process of the ultrashort photoelectric effect, the coefficient of a_i and b_i can be determined experimentally.

The emission of photoelectron current is dependent on the temperature of the electron gas and the lattice. The photoemission model of Fowler[15] and Dubridge[16] (F-D) was extended by Bechtel[17] to include N-photon photoemission. Here we just discuss

the linear and first-order photoelectric effect. The first-order photocurrent density could be expressed as

$$J(t) = a_0(e/h\nu)AI_0(t)(1-R) \times T^2(t)F\left(\frac{h\nu-\psi}{kT(t)}\right), \tag{8}$$

where $F(x)$ is the Fowler function, a_0 is the Richardson constant, k is the Boltzmann constant, R is the reflection coefficient, T is the mean temperature of the surface, and ψ is the work function of metal. Actually the F-D and Bechtel theory only gives the calculated photoelectron current emission from the surface of photocathode, it cannot give what the optimal thickness is for the BI photocathode. From the extended F-D photoelectric theory, the excited ultrashort electron current density depends on T_e^2. In our theory, we consider the collective effect and obtain a relation between the photoelectron current and the surface temperature. The temperature of electron and lattice and the electron–phonon coupling should be incorporated into the coefficients α and β for more precise consideration. From this simple photoelectric model, we can also find that the duration of the emitted photocurrent pulses is similar to the time factor of laser pulses duration.

4. Conclusions

In this paper, we put forward a simple ultrashort photoelectric effect model combined with the TTM model. From our model the optimized thickness of the BI copper metal photocathode is obtained. The optimum thickness is related to the optical absorption coefficient and the photoelectron current decay coefficient, which can be expressed as $\delta_{J=\max} = \ln(\beta/\alpha)/(\beta-\alpha)$. This could account for the rapid increase of photoelectron current as the film thickness approaches $\delta_{\max}$, and the exponential decay when the film thickness is larger than $\delta_{\max}$.

References

[1] Alsayed-Ali H E, Norris T B, Pessort M A and Mourou G A 1987 *Phys. Rev. Lett.* **58** 1212

[2] Fann W S, Storz R, Tom H W K and Bokor J 1992 *Phys. Rev. Lett.* **68** 2834

[3] Schmidt V, Husinsky W and Betz G 2000 *Phys. Rev. Lett.* **85** 3516

[4] Schoenlein'S R W *et al* 1996 *Science* **287** 5461

[5] Brogle R, Muggli P, Davis P, Harrapetian G and Joshi C (On the internet web page: WWW. epaper.kek.jp/p95/ARTICLES/WPC/WPC20.PDF)

[6] Tang Y Q, Zhao K, Hao J K, Wang L F, Quan S W and Yang X 2000 *Acta Phy. Sin.* **54** 1002 (in Chinese)

[7] Ihee H, Lobastov V A, Gomez U M, Goodson B M, Srinivasan R, Ruan C Y and Zewail A H 2001 *Science* **291** 458

[8] Cao J, Hao Z, Park H, Tao C, Kau D and Blaszczyk L 2003 *Appl. Phys. Lett.* **83** 1044

[9] Rose-Petruck Ch, Jimenez R and Guo T 1999 *Nature* **398** 310

[10] Liu Y Q, Zhang J, Li Y T, Zhang J and Qiu Y 2005 *Physics* **34** 287 (in Chinese)

[11] Liu Y Q, Zhang J, Liang W X and Wang Z H 2005 *Acta Phys. Sin.* **54** 1593 (in Chinese)

[12] Anisimov S I *et al* 1974 *Eksp. Teor. Fiz.* **66** 776
Anisimov S I *et al* 1974 *Sov. Phys. JETP* **39** 375

[13] Wellershoff S S, Hohlfeld J, Güdde J and Matthias E 1999 *Appl. Phys.* A **69** S99-S107

[14] Corkum P B, Brunel F, Sherman N K and Srinivasan-Rao T 1988 *Phys. Rev. Lett.* **61** 2886

[15] Fowler R H 1931 *Phys. Rev.* **38** 45

[16] Dubridge L A 1933 *Phys. Rev.* **43** 727

[17] Bechtel J H 1975 *J. Appl. Phys.* **46** 1585

[18] Girardeau-Montaut J P and Girardeau-Montaut C 1995 *Phys. Rev.* B **51** 13560

[19] Yang H, Hu B X and Wu J L, 2001 *Chin. Phys.* **10** 1062

[20] Wang X F, Jia T Q and Xu Z Z 2005 *Acta Phys. Sin.* **54** 3451 (in Chinese)