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Intraband and interband absorption of femtosecond laser pulses in copper*

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Abstract

We investigated the optical properties of pure copper irradiated by a femtosecond laser pulse. Self-absorption of 50-fs laser pulses at 800 nm and 400 nm wavelengths (below and above the interband absorption threshold, respectively) is studied for peak laser intensities up to 10^{15} W/cm^2. Theoretical description of laser interaction with copper target is developed, solving numerically the energy balance equations for electron and ion subsystems together with Maxwell equations for laser radiation field inside the target. The theory accounts for both intraband and interband absorption mechanisms. We treated in detail the changes in electron structure and distribution function with an increase in electron temperature, as well as the ensuing changes in thermodynamic properties, collision frequencies, optical and transport coefficients. Experimental work on self-absorption of femtosecond laser pulses in copper targets at 800 nm and 400 nm wavelengths is ongoing. Results for 800 nm wavelength are reported. Theory and experiment are in good agreement.

Keywords: Copper; Core state ionization; Femtosecond; Interband; Metal plasma transition

Interaction processes of high intensity short laser pulses with matter currently cover a large field of experimental and theoretical investigation (Gavrilov *et al*., 2004; Limpouch *et al*., 2002, 2004; Shokri *et al*., 2004; Shorokhov & Pukhov, 2004). In this paper, we studied absorption of femtosecond laser radiation by a metal target surface as a function of pulse wavelength, intensity, and duration, which provides important information on the electron properties of the metal. During femtosecond pulse irradiation, the laser energy is absorbed by electrons in intraband and interband transitions, raising the electron temperature T_e up to tens of electron-volts, while the ion lattice remains cold throughout the laser pulse duration (Eliezer, 2002). Sampling of electron properties in that domain, mostly inaccessible at present by any other means, allows testing theoretical predictions for electron mobility and collision frequencies (Milchberg *et al*., 1988), transition from solid-state to plasma electron behavior (Price *et al*., 1995), and temperature effects on the band structure and the interband absorption spectra (Fisher *et al*., 2001).

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Band structure of metals for low T_e (T_e much smaller than Fermi energy E_F or an energy scale W of large variations of the electron density of states (DOS), typically a few electron-volts) is well known. However, changes in band structure and in electron properties of an ordered system at $T_e \sim min\{E_F, W\}$ and higher have not been studied in detail, and present a great interest both for fundamental research and for applications (for example, in fast optoelectronics). The focus of the present work is a detailed theoretical and experimental study of the evolution of band structure and electron properties of copper under femtosecond laser irradiation.

Copper (or noble metals in general) is a natural choice for such a study due to the presence of a fully-occupied d-band, a few electron-volts below the Fermi level. The Fermi level is located in the half-occupied, nearly free-electron (NFE) s-band, 7–8 eV above the bottom of the conductivity band. In copper, the d-band is 3–4 eV wide, and its upper edge lays 2.0–2.2 eV below the Fermi level. Thus, at room T_e the presence of the d-band does not affect the NFE character of the metal (except for a slight increase in the effective s-electron mass due to screening by d-electrons).

At room T_e, excitation of a d-band electron into the conductivity band is only possible for the photon energy above 2 eV. Indeed, the state into which the electron is excited must be vacant, that is, above the Fermi level. Thus, at room T_e, there exists an interband absorption edge in copper at 2.0–2.2 eV. The absorption coefficient for 1.5 eV photons is 0.036 while the absorption coefficient for 3.0 eV photons is 0.53. When T_e increases, vacancies appear in the lower part of the conductivity band, so interband absorption from d-band into conductivity band becomes possible at lower photon energies, as well. As T_e reaches a few electron-volts, thermal depopulation of the d-band results in a reduced screening of the nucleus by the d-electrons, so the binding energy of the remaining d-electrons increases and the d-band "sinks" relatively to the bottom of the conductivity band (Price *et al.*, 1995). This result is quenching of the interband absorption at $T_e > 10$ eV. Sinking of the d-band is accompanied by d-state localization, which occurs at $T_e = 5$–10 eV. At room T_e removal of an electron from the d-band does not produce a lattice defect, however, as d-states become increasingly localized, removal of a d-electron leaves a higher-charge ion at the given lattice site. Therefore, as the d-states localize, ionization stage of ions starts to vary at random between lattice sites, the lattice periodicity fails, and the Bloch picture breaks down. This transition from a periodic lattice regime to a solid plasma regime produces a sharp increase in the energy transfer rate between electrons and ions. It can also result in a non-thermal melting. Indeed, at any instant, for a given ion, among the six pairs of its nearest neighbor ions at least one pair of ions likely has a dissimilar charge. In that case, the local potential minimum no longer coincides with the lattice site position.

Quantitative theoretical study of the laser radiation absorption in copper is based on the solution of energy balance equations for electron and ion subsystem,

$$C_e(T_e)\,\frac{\partial T_e}{\partial t} = \frac{\partial}{\partial x}\left(\kappa(T_e)\,\frac{\partial T_e}{\partial x}\right) - U(T_e,T_i) + Q(x,t), \quad (1)$$

$$C_i(T_i)\,\frac{\partial T_i}{\partial t} = U(T_e,T_i) \equiv \gamma(T_e,T_i)(T_e - T_i), \quad (2)$$

together with Maxwell equations for radiation propagation in the medium,

$$\bar{\nabla}\times\bar{H} + i\,\frac{\omega}{c}\,\varepsilon(x,t)\bar{E} = 0, \quad (3a)$$

$$\bar{\nabla}\times\bar{E} - i\,\frac{\omega}{c}\,\mu\bar{H} = 0, \quad (3b)$$

$$Q(x,t) = \frac{1}{2}\,\mathrm{Re}\{\sigma(x,t)\}\cdot|E(x,t)|^2, \quad (4a)$$

$$\varepsilon(\omega,x) = 1 + i\,\frac{4\pi\sigma(x,t)}{\omega}. \quad (4b)$$

Here, x is the distance from the target surface inward, t is time, T_i is ion temperature, C_e and C_i are electron, and ion heat capacities, κ is electron heat conductivity, $U = \gamma(T_e - T_i)$ is the energy transfer rate between subsystems, Q is radiation energy deposition rate, ε and σ are permittivity and conductivity of copper at laser frequency ω under given conditions $T_e(x,t)$ and $T_i(x,t)$. All extensive quantities are defined per unit volume. Equations are solved numerically using a finite differences scheme. Methods utilized for determination of the equation parameters $C_e(T_e)$, $C_i(T_i)$, $\gamma(T_e,T_i)$, and $\kappa(T_e,T_i)$ will be described separately. In Cu, the number of conductivity electrons changed significantly during the laser pulse even at modest intensities ($\sim 3\cdot 10^{13}$ W/cm^2 and higher in 50 fs pulse). Thus, ensuing changes in free electron density, ion charge, electron effective mass, electron-electron and electron-ion coupling, and intraband (Drude) absorption had all to be taken into account. Extensive use has been made of INFERNO code (Liberman 1979, 1982) for determination of equilibrium electron properties in the T_e range between 0.01 and 100 eV. To model the interband absorption, DOS in d-band $g(E)$ was approximated by several straight segments $a + bE$ to form continuous profile with $\int g(E)dE$ giving 10 d-electron states per ion at any T_e. Band width was scaled to the distance from the s-band bottom, to reproduce the narrowing of the sinking band. Finite state lifetime was also accounted for. Transition from periodic lattice to solid plasma regime at $T_e = 5$–10 eV was introduced explicitly. The calculated electron temperature at irradiated target surface, as a function of time, is shown in Figure 1.

Experimental work is carried out at Soreq 10 TW short-pulse laser facility. A comparison between the experimental results and the theoretical predictions for the absorption of 800 nm wavelength, 50 fs FWHM duration laser pulses in high-quality commercial Cu targets (Janos Technology uncoated copper plano-mirrors) is presented in Figure 2. Experiments on 400 nm wave-length pulses absorption are ongoing.

The absorption coefficient values shown are averaged both over temporal profile of the laser pulse and over surface laser intensity profile. Theory and experiment are in

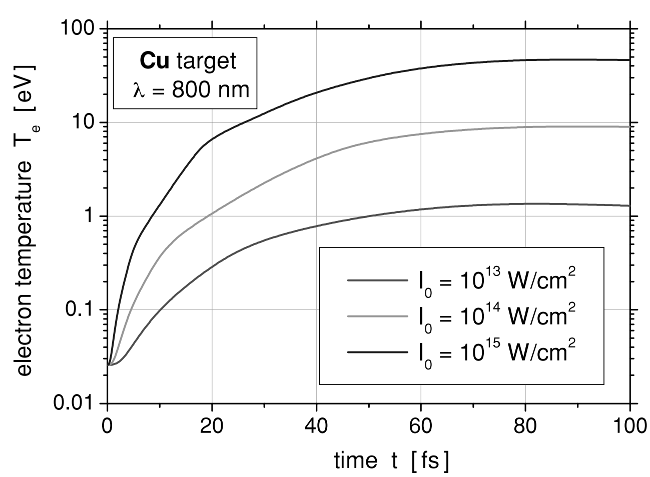

Fig. 1. Calculated electron temperature at target surface.

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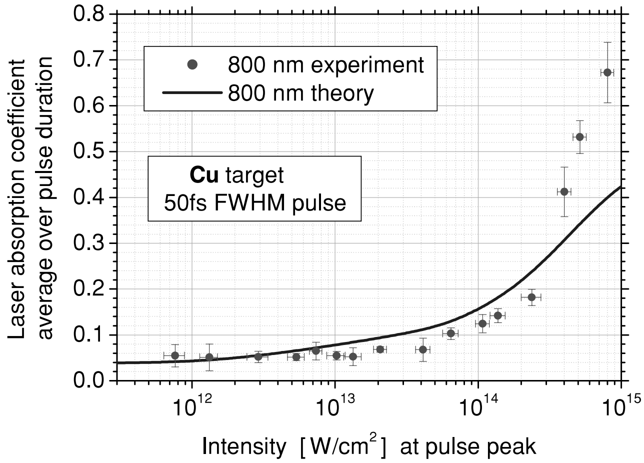

Fig. 2. Results for average absorption coefficient.

good agreement. Excessive absorption in experiment above 3×10^{14} W/cm^2 is due to the laser prepulse effect.

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