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Laser-excitation of electrons and nonequilibrium energy transfer to phonons in copper

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ABSTRACT

After the irradiation of a copper sample with an ultrashort laser pulse, electrons do not follow a Fermi distribution anymore but instead are in a nonequilibrium state. In contrast, the lattice cannot be excited directly by the laser pulse, due to the frequency mismatch. The energy increase in the phononic system only happens due to electron–phonon scattering. We investigate the initial electron dynamics using full Boltzmann-type collision integrals, including material-dependent characteristics by implementing a realistic density of states. We show results on the absorbed energy, details of the electronic nonequilibrium and the resulting electron–phonon coupling parameter in dependence on the photon energy. Our results show a counteracting dependence on the photon energy, which, on the one hand, enables the *d*-band electrons to absorb high-energy photons and on the other hand, increases the probability of multi-photon absorption.

1. Introduction

Since the invention of the laser, laser-matter interaction has become an important topic of fundamental as well as applied research. With help of ultrashort laser pulses in the femtosecond regime, scattering processes in solid materials can be induced and studied on their intrinsic timescales. Particularly, the electron–phonon coupling strength is of broad interest for a large variety of phenomena ranging from superconductivity [1,2] over plasmonics [3,4] and spintronics [5–9] to the observation of new transient states of matter [10,11]. Dynamical processes, like ultrafast laser-induced demagnetization or nanostructuring of solids, are strongly influenced by the interplay of electron–electron and electron–phonon scattering processes [8,9,12,13]. The electron–phonon coupling parameter, describing the strength of the energy transfer from the initially heated electrons to the crystal lattice, is often assumed to be constant. However, different publications have shown, that it depends on macroscopic parameters like the electron and phonon temperature [14,1,15,16] or the specific density of states (DOS) [16–18,15,19,20]. In this work, we investigate the influence of different photon energies and fluences on the energy gain of the electrons and on the electron–phonon coupling parameter after an ultrashort laser excitation. We focus our calculations on the effects of nonequilibrium electron distributions.

In the next section, we introduce the basic equations and assumptions, necessary to understand the analysis of the results presented thereafter. These are, on the one hand, considerations of the energy deposition in dependence on the laser fluence and photon energies applied, and on the other hand, peculiarities in the simulations of the nonequilibrium electron distribution for the excitation with different photon energies. Finally, we present the influence of this nonequilibrium on the electron–phonon coupling strength. The latter again depends on the photon energy, as well as on the energy absorbed by the electrons. The results are presented for copper. They are compared to the case of gold, which was discussed in Ref. [15], revealing commonalities of the behavior of nobel metals.

2. Theory

One of the basic models to describe the electron and phonon dynamics during and after the laser excitation is the two-temperature model (TTM) [21]

$$\frac{\partial u_e}{\partial t} = -\alpha(T_e - T_p) + S(t), \tag{1a}$$

$$\frac{\partial u_p}{\partial t} = +\alpha(T_e - T_p). \tag{1b}$$

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It traces the time development of the internal energies u_e and u_p of the electrons and the phonons, respectively. The electron energy is increased due to laser excitation, represented by the source term $S(t)$. This energy increase leads to an elevated electron temperature T_e. The energy transfer from the electrons to the phonons is described by a term proportional to the difference of their respective temperatures, T_e and T_p, and an electron–phonon coupling parameter α.

The source term, $S(t)$, describes the energy deposition into the material in dependence on time and is proportional to the intensity in the material. The intensity

$$I = I_0(1 - R) \tag{2}$$

inside the material itself depends on the external intensity I_0 and the reflectivity R of the material. The TTM has been widely and successfully applied to the description of a large variety of experimental measurements [22–26]. Moreover, different expansions facilitate the study of heat transport [14,27,28], excitation of semiconductors or distinct phonon modes [17,29,30] and magnetization dynamics [5,31,32].

However, during and directly after the laser excitation, the electrons are driven out of equilibrium and do not follow a Fermi distribution anymore. Thus, no temperature is defined and the application of temperature-dependent calculations is questionable [33,18,15,34]. In order to follow this nonequilibrium dynamics, we apply full Boltzmann-type collision integrals, described in Ref. [18]. The Boltzmann equation

$$\frac{df(\vec{k}, t)}{dt} = \Gamma_{\text{el–el}} + \Gamma_{\text{el–ph(-phot)}} + \Gamma_{\text{absorp}} \tag{3a}$$

$$\frac{dg(\vec{q}, t)}{dt} = \Gamma_{\text{ph–el(-phot)}} \tag{3b}$$

does not trace a macroscopic parameter such as the internal energy, but the microscopic distribution function f of the electrons and g of the phonons. The internal energies can be calculated with help of these distribution functions as will be described below. The change of the distribution function is calculated by applying collision terms for electron–electron $\Gamma_{\text{el–el}}$, electron–phonon $\Gamma_{\text{el–ph(-phot)}}$ and phonon–electron $\Gamma_{\text{ph–el(-phot)}}$ scattering. The energy deposition of the laser pulse in the material is achieved either by the electron–phonon(-photon) collision or by inverse Bremsstrahlung. The latter is represented by the absorption term Γ_{absorp}. All collision integrals, Γ, depend on the density of states $D(E)$ of the participating particles, Pauli-factors ensuring Pauli-blocking within the electron system, matrix elements representing the collision probability. For further details of the collisions integrals see Eqs. (10), (15), (17) and (19) in Ref. [18]. In particular, electron–phonon–photon collisions and inverse Bremsstrahlung require a Bessel function $\bar{J}^2_\ell(\gamma, \omega)$, which represents the absorption of a certain number of photons ℓ [35,36]. The parameter $\gamma = (eE_0/m_e)\Delta p$ depends on the electrical laser field E_0, the effective band mass of the electrons m_e and the exchanged momentum during the process Δp. For low intensities ($\gamma \ll \hbar\omega_L^2$) it can be approximated as

$$\bar{J}^2_\ell(\gamma, \omega) \approx \frac{1}{(\ell!)^2}\left[\frac{\gamma}{2\hbar\omega_L^2}\right]^{2|\ell|} \propto \left(\frac{I}{\omega_L^4}\right)^{|\ell|} \tag{4}$$

for $|\ell| \geq 1$ [35,36]. Without an external laser-field, the Bessel function for ℓ=0 equals unity, while all other orders vanish, thus no photons are absorbed. If an external laser field is applied, the zero-order Bessel function decreases, while all other orders start to increase and absorption processes take place. For the intensities analyzed here, the absorption of higher photon orders is much less probable then of lower orders, which is reflected in the fact that $\bar{J}^2_{\ell+1}$ is always much smaller then $\bar{J}^2_\ell$. The Bessel function not only enters the collision term of the inverse Bremsstrahlung, but also the collision term of the electron–phonon interaction. This reflects the fact that photon absorption can also take place during electron–phonon collisions [35]. We indicate this in the notation $\Gamma_{\text{el–ph(-phot)}}$ for this process. The major part of the absorption is, however, determined by the inverse Bremsstrahlung Γ_{absorp} [33].

The probability of single- or multi-photon processes increases with decreasing photon energy $\hbar\omega_L$ and increasing intensity I, which can be connected to the later used fluence of the laser pulse F, which is given by

$$F = \int_{-\infty}^{\infty} I(t)dt. \tag{5}$$

In our work, we assume a laser pulse that is rectangular in time. Thus the intensity is constant over the whole pulse duration and the fluence $F = I\tau_L$ within the material is proportional to the intensity and the pulse duration τ_L. This assumption was made to better distinguish between excitation and relaxation processes. Furthermore, we only fix the intensity inside the material, I, in order to enable a better comparison between results obtained for different photon energies. The electrons are described by an effective one-band model [18]. Moreover, only longitudinal phonon modes are considered and described within the Debye model. For the comparison with the two-temperature model, the internal energies of electrons and phonons,

$$u_e = \int dE\, f(E) D_e(E) E, \tag{6a}$$

$$u_p = \int dE\, g(E) D_p(E) E, \tag{6b}$$

can be determined from the corresponding distributions f and g and density of states $D_{e/p}(E)$ for electrons and phonons, respectively. The electron density of states is taken from Lin et al. [16] and is assumed to be constant during the calculations. To further compare our results with the input parameters of the TTM, we calculate an electron–phonon coupling parameter

$$\alpha[f(t), g(t)] = \frac{du_p[g(t)]/dt}{T_e[f(t)] - T_p[g(t)]}, \tag{7}$$

based on Eq. (1) [15], with help of the phonon–electron collision term. Either laser-excited (nonequilibrium) electrons or hot, thermalized (equilibrium) electrons interact with cold phonons. For the nonequilibrium case, we calculate "corresponding temperatures" $T_p[g(t)]$ and $T_e[f(t)]$ by comparing the internal energy and number of particles of the nonequilibrium distribution function to a Fermi–Dirac or Bose–Einstein distributed system, respectively. This yields well-defined "corresponding temperatures", chemical potentials and equilibrium distribution functions. The equilibrium distribution, on the other hand, can be used to calculate an energy change of the equilibrated system using our Boltzmann approach, to extract an equilibrium coupling parameter.

3. Energy deposition

There are two important factors that influence the total internal energy gain of the electrons at the end of laser excitation. These are the fluence and the photon energy of the laser pulse applied to the sample [35,18]. In our work, we do not consider the reflectivity explicitly. Thus, all fluences shown here are inside the material. The probability for the absorption of ℓ photons increases with increasing fluence and decreasing photon energy [35], see Eq. (4). Moreover, single-photon processes are more probable than multi-photon processes.

In our simulations, we assume a 10 fs-laser pulse with rectangular pulse shape, to better distinguish between excitation and

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