Laser damage in silicon: energy absorption, relaxation and transport

A. Rümer, 1,* O. Osmani, 2 and B. Rethfeld 1

1 Department of Physics and Research Center OPTIMAS, University of Kaiserslautern, 67663 Kaiserslautern, Germany
2 Faculty of Physics, University of Duisburg-Essen and CeNIDE, 47048 Duisburg, Germany

Silicon irradiated with an ultrashort 800 nm-laser pulse is studied theoretically using a two temperature description that considers the transient free carrier density during and after irradiation. A Drude model is implemented to account for the highly transient optical parameters. We analyze the importance of considering these density-dependent parameters as well as the choice of the Drude carrier collision frequency. In addition, degeneracy and transport effects are investigated. The importance of each of these processes for resulting calculated damage thresholds is studied. We report fit-free damage thresholds calculations that are in very good agreement with experimental results over a wide range of pulse durations.

PACS numbers: 05.70.Ln, 79.20.Ap, 44.10.+i, 72.10.Di

I. INTRODUCTION

The interaction of laser pulses with solid matter has been subject of both experimental 1–9 and theoretical 10–17 research for many years. Especially ultrashort laser pulses, that cause minimal collateral damage to the surrounding material, are of high interest for applications in medical surgery, micromachining and nanostructuring. 18–21

For the theoretical description of laser-excited solids, both the timescale and the kind of material have to be carefully considered. 22 During and directly after the excitation, the electron system is out of equilibrium and no temperature is defined. Consequently, nonequilibrium descriptions like the Boltzmann equation 12,23–28 or kinetic Monte Carlo simulations 29–32 have to be applied.

On a timescale of about a hundred femtoseconds 3,11,27 after excitation, the electrons again follow an equilibrium distribution. A temperature can be assigned to both the electrons and the lattice. However, those temperatures will differ because the laser energy is mainly absorbed by the electrons and subsequently transferred to the lattice. Equilibration between both subsystems happens on a picosecond timescale. For metals, the well known two temperature model 10 is often applied on this timescale to describe both heat relaxation and transport within the electron and lattice subsystem.

In contrast to the excitation of metals, it is not sufficient to follow carrier and lattice temperatures when describing laser-excited semiconductors. Because the free carrier density in these materials can vary over several orders of magnitude during irradiation, its evolution has to be treated explicitly. In addition, the highly transient free carrier density causes huge changes in the optical properties of the solid. Thus, a theoretical description of laser excitation of semiconductors has to account for the transient free carrier density, transient optical properties, carrier and lattice temperature evolution as well as energy and particle transport.

This paper extends an existing two temperature description for semiconductors first presented by van Driel 11 to account for the transient optical properties during the excitation with a femtosecond-laser pulse. To that end, we will first introduce the theoretical model and the extension. Afterwards, we will investigate the influence of the transient optical parameters as well as degeneracy and transport effects on the results of the simulations. We will discuss the importance of each property and process under investigation by calculating damage thresholds and comparing with experimental data.

II. THE DENSITY-DEPENDENT TWO TEMPERATURE MODEL

In laser-irradiated semiconductors, electrons are excited from the valence to the conduction band via single or multiphoton absorption. Thus, electron-hole pairs are created. The order of the absorption process depends on the photon energy in relation to the band gap energy of the material. In our case, the irradiation of silicon with 800 nm-laser pulses, the photon energy is larger than the indirect band gap of silicon. It is therefore sufficient to consider single and two photon absorption processes. 11

Higher order processes are less probable and can, thus, be neglected. In addition, free carriers (electrons in the conduction band and holes in the valence band) can absorb further photons via free carrier absorption thereby increasing their kinetic energy. Conduction band electrons with a sufficiently high kinetic energy may ionize additional valence band electrons via impact ionization. Furthermore, free electrons and holes may recombine via Auger recombination transferring the excess energy to another free electron or hole. Meanwhile, the carrier system can couple to the lattice system. The temperatures of both systems will equilibrate until a thermal equilibrium state is reached.

All these processes as well as carrier and heat diffusion can be modeled with the density-dependent two temperature model (nTTM) first presented by van Driel 11. For sake of completeness, we will in the following derive the
original model following his work before expanding it to account for transient optical parameters during irradiation.

A. Basic Assumptions

In the framework of the nTTM, free electrons and holes are assumed to be thermalized into an equilibrium distribution function, usually a Fermi distribution

$$f_e(\varepsilon) = \frac{1}{1 + \exp \left( \frac{\varepsilon - \mu_e}{k_B T_e} \right)} , \tag{1}$$

at all times. The distribution functions of electrons and holes are assumed to possess different chemical potentials $\mu_e$, where $c$ stands for “carrier” and may be substituted by $e$ for electrons and $h$ for holes, but a common temperature $T_c$. Here, $k_B$ denotes the Boltzmann constant. The plus sign in the exponent in Eq. (1) associates with the electrons, while the minus sign associates with the holes. The assumption of thermalized carriers is, however, questionable at times shortly after laser excitation. As thermalization will take on the order of $100 \text{ fs}^{3,11,27}$ carrier temperature and distribution functions as well as chemical potentials may be interpreted as quasi-temperature, quasi-Fermi distribution functions and quasi-chemical potentials.

Conduction and valence bands are treated as parabolic bands. Consequently, the densities of states (DOS) of electrons

$$D_e(\varepsilon) = \frac{m_e^*}{h^3 \pi^2} \sqrt{2 (\varepsilon - \varepsilon_C)} , \tag{2a}$$

and holes

$$D_h(\varepsilon) = \frac{m_h^*}{h^3 \pi^2} \sqrt{2 (\varepsilon_V - \varepsilon)} , \tag{2b}$$

are applied, where $\varepsilon_C$ and $\varepsilon_V$ denote the lower edge of the conduction band and the upper edge of the valence band, respectively. A different constant effective mass $m_e^*$ is assumed for electrons and holes, respectively.

Figure 1 shows a sketch of the DOS of electrons and holes as well as their quasi-Fermi distributions and chemical potentials.

By integrating over the carrier distribution multiplied with the respective DOS, the local carrier density reads

$$n_c = 2 \left( \frac{m_e^* k_B T_e}{2 \pi h^2} \right)^{3/2} \mathcal{F}_{1/2}(\eta_e) . \tag{3}$$

Here, the function

$$\mathcal{F}_{\xi}(\eta_e) = \frac{1}{\Gamma(\xi + 1)} \int_0^\infty \frac{x^\xi}{1 + \exp(x - \eta_e)} \, dx \tag{4}$$

denotes the Fermi integral of order $\xi$. The so-called reduced Fermi levels of electrons and holes are defined as

$$\eta_e = \frac{\mu_e - \varepsilon_C}{k_B T_e} \quad \text{and} \quad \eta_h = \frac{\varepsilon_V - \mu_h}{k_B T_e} , \tag{5}$$

respectively. They indicate the position of the respective chemical potentials in relation to the band edges. If the reduced Fermi levels become positive, the respective chemical potentials are positioned inside the respective bands and the carrier system is degenerate.

B. Carrier and Energy Current

In a laser-generated free carrier system, electrons and holes basically move together as electron-hole pairs since the Dember field that builds up due to charge separation restricts carrier and current densities from becoming significantly different (ambipolar diffusion). We can therefore assume, that

$$n_c = n_h \quad \text{and} \quad \vec{j}_c = -\vec{j}_h \tag{6}$$

at each point in space. Under these conditions, the carrier current density is given by

$$\vec{j} = -D \left\{ \nabla n_c + \frac{n_c}{k_B T_e} \left[ \frac{H^{1/2}_{-1/2}(\eta_e) + H^{1/2}_{-1/2}(\eta_h)}{H^{1/2}_{-1/2}(\eta_e) + H^{1/2}_{-1/2}(\eta_h)} - 3 \right] \nabla \varepsilon_g \right\} = -D \left\{ \nabla \eta_e + \frac{n_e}{k_B T_e} \left[ \frac{H^0_{-1/2}(\eta_e) + H^0_{-1/2}(\eta_h)}{H^0_{-1/2}(\eta_e) + H^0_{-1/2}(\eta_h)} - 3 \right] \nabla \varepsilon_g \right\} \tag{7}$$

where $\varepsilon_g = \varepsilon_C - \varepsilon_V$ denotes the band gap energy, $H^{1/2}_{-1/2}(\eta_e)$ is the ratio of Fermi integrals (4) and

$$D = \frac{k_B T_e}{e} \frac{\mu_e^0 H^{0}_{1/2}(\eta_e) H^{0}_{1/2}(\eta_h)}{\mu_h^0 H^{0}_{1/2}(\eta_e) + \mu_h^0 H^{0}_{1/2}(\eta_h)} \times \left[ H^{1/2}_{-1/2}(\eta_e) + H^{1/2}_{-1/2}(\eta_h) \right] \tag{8}$$

is the ambipolar diffusion coefficient. Note that the band gap gradient $\nabla \varepsilon_g$ is non-zero because of band gap shrinkage due to increased lattice temperature and carrier density during and after excitation.\textsuperscript{5,33,34}
The heat current density \( \vec{W} = \vec{w}_e + \vec{w}_h \) is the sum of electron and hole heat current densities and reads

\[
\vec{W} = \left\{ \varepsilon_g + 2k_B \varepsilon \left[ H_0^e(\eta_e) + H_0^h(\eta_h) \right] \right\} \vec{J} - (\kappa_e + \kappa_h) \nabla T_e.
\]

(9)

The expressions for the current densities include treatment of the Peltier and Seebeck effect as well as Ohm’s and Fourier’s law. A more detailed description and background information on the transport terms is given by van Driel.\(^{11}\)

Note that whenever space charge effects cannot be neglected, for instance when studying Coulomb explosion, the assumption of ambipolar diffusion (6) is not valid anymore. Transport in this case has been studied by Bulgakova et al.\(^{14,15}\)

### C. Energy and Particle Balance

For the particle balance equation, interband laser absorption processes, namely single and two photon absorption, impact ionization, Auger recombination and carrier transport are considered:

\[
\frac{\partial n_e}{\partial t} = \frac{\alpha_{SPA} I}{\hbar \omega_L} + \frac{\beta_{TPA} I^2}{2 \hbar \omega_L} + \delta n_e - \gamma n_e^2 - \nabla \cdot \vec{J}.
\]

(10)

The first two terms describe single and two photon absorption, the third and fourth term stand for impact ionization and Auger recombination and the last term describes carrier transport. Here, \( I \) denotes the intensity of the laser pulse, \( \hbar \omega_L \) the photon energy, \( \alpha_{SPA} \) and \( \beta_{TPA} \) the single and two photon absorption coefficients, respectively, \( \delta \) the coefficient for impact ionization and \( \gamma \) the one for Auger recombination. All coefficients used in this work are given in Tab. 1.

The total carrier energy density is given by

\[
U_{e-h} = \int_{-\infty}^{e_f} \varepsilon \, D_h(\varepsilon) \, f_h(\varepsilon) \, d\varepsilon + \int_{e_c}^{e_f} \varepsilon \, D_e(\varepsilon) \, f_e(\varepsilon) \, d\varepsilon
\]

\[
= n_e \varepsilon_g + \frac{3}{2} n_e k_B \varepsilon \left[ H_1^{3/2}(\eta_e) + H_1^{3/2}(\eta_h) \right].
\]

(11)

The first term in the second line represents the potential energy of the carrier pairs, while the second term gives the kinetic energy of electrons and holes.

The total carrier energy is modified by laser absorption, transport and carrier-phonon coupling. Thus, the balance equation for carrier energy density is

\[
\frac{\partial U_{e-h}}{\partial t} = (\alpha_{SPA} + \alpha_{FCA}) I + \beta_{TPA} I^2 - \nabla \cdot \vec{W} - g (T_e - T_{ph}),
\]

(12)

where \( \alpha_{FCA} \) denotes the free carrier absorption coefficient. The carrier-phonon coupling parameter \( g \) is nonconstant in semiconductors and depends highly on carrier density. It is therefore often described by \( g = C_{e-h} / \tau_{e-ph}^{relax} \), where the carrier heat capacity \( C_{e-h} \) depends on the carrier density and \( \tau_{e-ph}^{relax} \) is the carrier-phonon energy-relaxation time.\(^{11,35,36}\)

This expression results from an analytical solution of the traditional TTM for metal films at times after laser-excitation, neglecting transport and assuming a constant heat capacity as well as a constant electron-phonon coupling parameter. Obviously, it is therefore questionable for semiconductors excited by ultrafast laser pulses. Comparison with carrier-phonon coupling parameters extracted with the Boltzmann equation, similar to what is done by Mueller and Rethfeld\(^{27}\), nevertheless shows, that the approximation is justified whenever the carrier system is not highly degenerate.\(^{37}\)

The lattice system is not directly heated up by the laser pulse but only indirectly via carrier-phonon coupling. Thus, the balance equation for lattice energy density reads

\[
\frac{\partial U_{ph}}{\partial t} = \nabla \left( \kappa_{ph} \nabla T_{ph} \right) + g (T_e - T_{ph}),
\]

(13)

where the first summand describes energy transport in the lattice system via Fourier’s law and \( \kappa_{ph} \) is the lattice thermal conductivity. The second term describes the energy exchange with the carrier system. This equation is basically identical to the one in the traditional TTM used for metals.\(^{10}\)

To calculate carrier and lattice temperature, we have to look at the temporal derivatives of carrier and lattice energy densities in more detail. Lattice energy density only depends on lattice temperature. Thus, the temporal derivative of the lattice energy density can be expressed as

\[
\frac{\partial U_{ph}}{\partial t} = C_{ph} \frac{\partial T_{ph}}{\partial t},
\]

(14)

using the lattice heat capacity \( C_{ph} = \frac{\partial U_{ph}}{\partial T_{ph}} \). For the carrier system things are more complicated because the carrier energy density does not only depend on carrier temperature but also on carrier density and band gap energy:

\[
\frac{\partial U_{e-h}}{\partial t} = C_{e-h} \frac{\partial T_e}{\partial t} + \frac{\partial U_{e-h}}{\partial n_e} \frac{\partial n_e}{\partial t} + \frac{\partial U_{e-h}}{\partial \varepsilon_g} \frac{\partial \varepsilon_g}{\partial t}.
\]

(15)

The derivative of the band gap energy can again be written as \( \frac{\partial \varepsilon_g}{\partial \varepsilon} = \frac{\partial \varepsilon_g}{\partial n_e} \frac{\partial n_e}{\partial \varepsilon} + \frac{\partial \varepsilon_g}{\partial T_{ph}} \frac{\partial T_{ph}}{\partial \varepsilon_g} \) because the band gap depends on both carrier density and lattice temperature.\(^{6,11,33}\) Furthermore, to calculate the derivatives of carrier energy density, it is necessary to calculate the derivative of the reduced Fermi levels, as can be seen in Eq. (11). This is possible by taking the derivative with respect to carrier temperature, lattice temperature and carrier density, respectively, on both sides of Eq. (3), utilizing that all three are independent properties and solving for the derivative of the reduced Fermi level. The reduced Fermi level itself can be calculated by solving Eq. (3) for the Fermi integral and comparing with tabulated data that can, for instance, be generated using the GNU Scientific Library.\(^{38}\)
D. Non-Degenerate Carrier System

If the chemical potentials of electrons and holes are located far outside the conduction and the valence band, respectively, only the Boltzmann tail of the Fermi distribution reaches inside the band. The carrier system is therefore non-degenerate and follows a Maxwell-Boltzmann distribution. Consequently, the reduced Fermi levels are large and negative and Fermi integrals of any order can be reduced to \(\exp(\eta_c)\) and \(\exp(\eta_h)\), respectively. The quotients of Fermi integrals \(H^x\) tend towards one and the carrier energy and pair current densities simplify from Eqs. (7) and (9) to

\[
\vec{J} = -D_0 \left( \frac{\hbar}{2e} \frac{1}{k_B T_e} \nabla \varepsilon_g + \frac{e}{2T_e} \nabla T_e \right),
\]

with the ambipolar diffusivity \(D_0 = \frac{2k_B T_e}{e} \frac{\varepsilon_g v_h^e v_h^e}{\hbar^2 + \nu_s^2} \) and

\[
\vec{W} = (\varepsilon_g + 4k_B T_e) \vec{J} - (\kappa_c + \kappa_h) \nabla T_e. \tag{17}
\]

The carrier energy density in a non-degenerate system is consequently given by

\[
U_{e-h} = n_e \varepsilon_g + 3 n_e k_B T_e, \tag{18}
\]

which immensely simplifies the calculation of the carrier heat capacity and other derivatives in Eq. (15).

Aside from the reduced Fermi levels being negative, another criterion for whether the carrier system is degenerate lies within the comparison of the carrier temperature \(T_e\) with the Fermi temperature

\[
T_F = \frac{h^2}{2 m_e^* k_B} \left( \frac{e^2}{3 \pi^2 n_e} \right)^{2/3}, \tag{19}
\]

which is different for electrons and holes because of the different effective masses. Whenever the Fermi temperature is comparable to or higher than the carrier temperature, the respective carrier system is degenerate.

E. Laser-Excitation

To obtain the intensity of the laser pulse within the material, the attenuation in the direction of propagation is calculated with the one-dimensional ordinary differential equation (ODE)

\[
\frac{dI}{dz} = - (\alpha_{\text{SPA}} + \alpha_{\text{FCA}}) I - \beta_{\text{TPA}} I^2, \tag{20}
\]

that includes linear interband or single photon absorption (SPA), linear intraband or free carrier absorption (FCA) and two photon interband absorption (TPA). Because of momentum conservation and the fact, that silicon is an indirect semiconductor, single photon processes can only happen under assistance of a phonon. Thus, the linear absorption coefficients \(\alpha_{\text{SPA}}\) and \(\alpha_{\text{FCA}}\) depend on lattice temperature. In addition, the FCA coefficient depends on carrier density because the more free carriers, the stronger the absorption. Because of the possibly steep carrier density and lattice temperature profiles within the material during irradiation, we have to account for spatial non-constant absorption coefficients when solving the ODE (20). Consequently, there is no closed-form analytical solution and the ODE (20) has to be solved numerically along with the balance equations (10), (12) and (13). The intensity transmitted through the material surface is

\[
I_0 = \sqrt{\frac{4 \ln(2) (1 - R) \Phi}{\tau_p} \exp \left\{ -4 \ln(2) \left( \frac{t - t_0}{\tau_p} \right)^2 \right\}}, \tag{21}
\]

where \(R\) denotes the reflectivity, \(\Phi\) the fluence and \(\tau_p\) the duration of the laser pulse that is shifted by \(t_0\). This shift is chosen as \(t_0 = 3 \tau_p\).

Because of the usually large spot size of the laser pulse, the radial intensity distribution can be neglected and it is sufficient to describe absorption and transport in the direction of propagation at the center of the focus.

As already indicated above, the huge changes in free carrier density during the irradiation lead to significant changes in the optical parameters, namely the reflectivity and the FCA coefficient. Nevertheless, when modeling semiconductors, the reflectivity is often assumed to depend solely on lattice temperature. In these cases the FCA coefficient is written as a solely lattice temperature dependent cross-section multiplied with carrier density. In the following, we will denote these expressions for the reflectivity and the FCA coefficient as \(T\)-expression.

To account for the influence of the transient carrier density on reflectivity and FCA coefficient, we apply a Drude model. In the framework of the Drude model, the complex dielectric function is given by

\[
\epsilon(\hbar \omega_L) = \epsilon_r(\hbar \omega_L) - \left( \frac{\omega_p^2}{\omega_L^2} \right)^2 \frac{1}{1 + i \frac{\nu_{\text{Drude}}}{\omega_L}}, \tag{22}
\]

with the intrinsic dielectric constant \(\epsilon_r\), the carrier collision frequency \(\nu_{\text{Drude}}\) and the plasma frequency \(\omega_p^2 = n_e e^2/\epsilon_0 m_{\text{opt}}^*\) with the optical effective mass \(m_{\text{opt}}^*\). From Eq. (22), the complex refractive index \(n = \sqrt{\epsilon}\), the reflectivity under normal incidence and the FCA coefficient

\[
R = \frac{\lvert n - 1 \rvert^2}{\lvert n + 1 \rvert^2} \quad \text{and} \quad \alpha_{\text{FCA}} = \frac{2 \Im (n) \omega_L}{c_0} \tag{23}
\]

can subsequently be calculated. Here, \(c_0\) denotes the speed of light in vacuum.

Concerning the carrier collision frequency \(\nu_{\text{Drude}}\), there is disagreement in literature on the question which kinds of collisions contribute (for an overview see Ref. 42 and references therein). Many authors assume
$\nu_{\text{Drude}}$ to be constant\textsuperscript{6,43–48}, while others consider non-
constant electron-electron and electron-phonon collision frequencies\textsuperscript{16,49–51}.

Electron-electron collisions, however, can not con-
tribute to the collision frequency entering the Drude
model\textsuperscript{1}. During these collisions, momentum is exchanged
between two electrons, thereby changing the velocity of
single electrons, while the collective velocity of the sys-
tem remains unchanged. The Drude collision frequency,
however, is the frequency of collisions that change the
collective velocity of the electrons\textsuperscript{60}.

In the framework of the nTTM presented here, we con-
sider carrier-ion and carrier-phonon collisions. We as-
sume the carrier-phonon collision frequency to be pro-
portional to lattice temperature:\textsuperscript{2}

$$\nu_{\text{carrier--ph}} = AT_{\text{ph}}.$$ \hspace{1cm} (24)

To our knowledge, there is no data for silicon or com-
parable semiconductors. For metals like gold, silver, copper
or aluminum, Christensen et al.\textsuperscript{52} state that the propor-
tionality constant $A$ is on the order of $1 \times 10^{11} \text{s}^{-1}\text{K}^{-1}$
to $4 \times 10^{11} \text{s}^{-1}\text{K}^{-1}$. Here, we assume that the propor-
tionality constant for silicon is comparable to those for
these metals and take $A = 1.23 \times 10^{11} \text{s}^{-1}\text{K}^{-1}$ given for
gold by Wang et al.\textsuperscript{2}

The electron- and hole-ion collision frequencies are given by

$$\nu_{e--i} = \frac{\langle \nu_e \rangle}{\langle \nu_h \rangle},$$ \hspace{1cm} (25)

where $\langle \nu_e \rangle$ denotes the mean free path of electrons and
holes for Coulomb collisions and $\langle \nu_h \rangle = \sqrt{2(U^\text{kin})/m_e^*}$
the mean electron and hole velocity. The mean electron
and hole energies ($U^\text{kin}$) can be calculated by splitting the
second summand in Eq. (11), that represents the carrier
kinetic energy, into the different parts for electrons and
holes and dividing by carrier density. The mean free
path of electrons and holes, respectively, in a screened
Coulomb potential is given by$^{53} \langle \ell_e \rangle = \frac{\kappa_e^2}{n_e \pi}$, where the inverse
screening length $\kappa_e$ for a free carrier gas can be calculated following Refs. 27, 54, and 55:

$$\kappa_e^2 = \frac{e^2}{\epsilon_0} \int \frac{\partial D_e(\varepsilon)}{\partial \varepsilon} f_e(\varepsilon) \, d\varepsilon$$ \hspace{1cm} (26)

$$= \frac{e^2}{\epsilon_0} \left( \frac{m_e^*}{\pi \hbar^2} \right)^{3/2} \sqrt{\frac{k_B T_e}{2}} \mathcal{F}_{-1/2}(\eta_e),$$

using Eqs. (1), (2) and (4). Inserting this into Eq. (25) and summing up the contributions of electrons and holes
leads to the carrier-ion collision frequency

$$\nu_{\text{carrier--ion}} = \frac{\sqrt{3} \pi_0}{e^2} (k_B T_e)^{3/2} \left[ H_{1/2}^{1/2}(\eta_e) \sqrt{\frac{H_{1/2}^{3/2}(\eta_e)}{m_e^*}} ight]$$ \hspace{1cm} (27)

including collisions of both types of carriers.

The total carrier collision frequency can now be calcu-
lated by summing up the carrier-ion and carrier-phonon
contributions (24) and (27).

## III. RESULTS

The balance equations (10), (12) and (13) are solved
in one dimension using a finite differences scheme. While
an explicit forward time centered space (FTCS) scheme
is used to obtain lattice temperature and carrier density,
the carrier temperature is calculated using an iterative
Crank Nicolson scheme.\textsuperscript{56} The equations are solved on
a staggered grid, where the grid points for carrier and
energy currents are positioned in between the grid points
for temperatures and density. As initial conditions, we
choose an equilibrium at $T_e(z,0) = T_{\text{ph}}(z,0) = 300 \text{K}$
and, consequently, $n_e(z,0) = 10^{12} \text{cm}^{-3}$. As energy-
conserving boundary conditions, we use $J_z(z,t) = 0$, $W_z(z,t) = 0$ and $\kappa_{\text{ph}} \partial T_{\text{ph}}/\partial z = 0$ at the surface ($z = 0$)
and at $z = 10 \mu\text{m}$ assumed as the thickness of the ma-
terial. The model parameters used for the calculations
presented below are listed in Table I. It should be noted
here that the nTTM like the commonly used TTM re-
quires the use of various material parameters as well as
approximations. While some of these parameters are af-
fected with certain inaccuracies, numerous studies have
proven the usefulness of the TTM in the past.\textsuperscript{52,57–60} Ob-
vviously, it is possible to choose material parameters in
such a way that the simulation gives the best agreement
with the experiment, e.g. by fitting certain material pa-
rameters. However, we would like to point out, that no
fitting was performed in the present simulations.

The material parameters used here (cf. Tab. I) are all com-
monly accepted for the case of silicon. The sole exception
is the carrier-phonon collision frequency (24). No value
for the proportionality constant $A$ is known for silicon.
The choice to use the value given by Wang et al.\textsuperscript{2} is ar-
bitrary and should not be understood as a fit.

For all figures showing temporal evolutions, a 100 fs-
laser pulse with a fluence of 130 mJ/cm\textsuperscript{2} at 800 nm is
applied.

Figure 2 shows the temporal evolution of carrier den-
sity (green dashed curve) and temperature (red solid
curve) as well as lattice temperature (blue dotted curve)
at the surface of laser-irradiated silicon. The data pre-
cented in the figure was calculated using the full model
considering the transient optical parameters obtained
with the Drude model, a Fermi distributed carrier system
and transport.

The carrier temperature starts to increase early at very
low intensities until it reaches a plateau of approximately
1680 K at about 100 fs. In the following 100 fs it only
increases slightly before it ascends rapidly when the laser
pulse approaches its maximum. The carrier temperature
reaches its maximum of about 32 100 K slightly after the
maximum of the laser pulse and then decreases mainly
TABLE I. Model parameters for silicon.

| Quantity                        | Symbol | Value | Reference          |
|---------------------------------|--------|-------|--------------------|
| Indirect band gap               | \( \varepsilon_g \) | \( 1.16 - \frac{7.02 \times 10^{-4} \cdot R^2}{T_{ph} + 1108 \ K} - 1.5 \times 10^{-8} \ cm \ n_e^{1/3} \) | eV | Refs. 6, 11, and 33 |
| Electron effective mass         | \( m_e^* \) | 0.33 \( m_e \) | Ref. 35 |
| Hole effective mass             | \( m_h^* \) | 0.81 \( m_e \) | Ref. 61 |
| Optical effective mass          | \( m_{\text{opt}}^* \) | 0.15 \( m_e \) | Ref. 6 |
| Lattice heat capacity           | \( C_{ph} \) | \( 1.978 + 3.54 \times 10^{-4} \ K^{-1} T_{ph} - 3.68 K^2 T_{ph}^{-2} \) | J/(cm\(^3\) K) | Refs. 11 and 62 |
| Lattice thermal conductivity    | \( \kappa_{ph} \) | \( 1585 K^{1.23} T_{ph}^{-1.23} \) W/(cm K) | Refs. 11 and 62 |
| Melting temperature             | \( T_{\text{melt}} \) | 1685 K | Ref. 6 |
| Carrier thermal conductivity    | \( \kappa_{e-h} \) | \( (-3.47 \times 10^{16} + 4.45 \times 10^{14} K^{-1} T_e) \) eV/(scm K) | Refs. 35 and 63 |
| Carrier ambipolar diffusivity   | \( D_0 \) | \( 18 (300 K/T_{ph}) \) cm\(^2\)/s | Ref. 11 |
| Auger recombination coefficient | \( \gamma \) | \( 3.8 \times 10^{-31} \) cm\(^6\)/s | Refs. 11 and 64 |
| Impact ionization coefficient   | \( \delta \) | \( 3.6 \times 10^{10} \exp[-1.5 \varepsilon_g/(k_B T_e)] \) s\(^{-1} \) | Ref. 11 |
| Carrier-phonon relaxation time  | \( \tau_{e-ph}^{\text{relax}} \) | 0.5 ps | Ref. 11 |
| Optic properties at 800 nm      | \( \alpha_{\text{SPA}} \) | \( 1.12 \times 10^3 \exp(T_{ph}/430 \ K) \) cm\(^{-1} \) | Ref. 39 |
| Two photon absorption coefficient| \( \beta_{\text{TPA}} \) | 9 cm/GW | Ref. 39 |
| Free carrier absorption coefficient | \( \alpha_{\text{FCA}} \) | \( 2.56 \times 10^{-18} \) cm\(^2\) (\( T_{ph}/300 \) K) \( n_e \) | Ref. 39 |
| Reflectivity                    | \( R \) | \( 0.329 + 5 \times 10^{-5} K^{-1} (T_{ph} - 300 \ K) \) | Ref. 39 |
| Optical properties at 800 nm    | \( \alpha_{\text{FCA}} \) | calculated using Eq. (23) | Ref. 41 |
| Reflectivity                    | \( R \) | calculated using Eq. (23) | Ref. 41 |
| Intrinsic dielectric constant   | \( \varepsilon_r \) | \( 13.631 + 0.041 i \) | Ref. 65 |

due to carrier-phonon coupling.

The first increase in carrier temperature is due to SPA. The few electron-hole pairs excited on this timescale have an excess energy of \((\hbar \omega_L - \varepsilon_g)\). In the approximation of a non-degenerate carrier system, which is valid on this timescale (cf. Sec. III C), this excess energy corresponds to a quasi-temperature of about 1690 K. Due to carrier-phonon coupling the actual carrier temperature is slightly lower. The second increase in carrier temperature happens later when, on the one hand, TPA becomes important for far higher intensities near the maximum of the pulse and, on the other hand, FCA becomes important because of an increasing free carrier density.

Due to carrier-phonon coupling, carrier and lattice temperatures tend to equilibrate. Consequently, the lattice heats up at later times than the carriers. Total equilibration is not achieved for several tens of picoseconds because, even on this timescale, the carrier system is still heated up slightly as a result of Auger recombination.

The carrier density reaches its maximum shortly after

FIG. 2. (Color online) Characteristic temperature and density evolution at the surface of laser-irradiated silicon obtained with the nTTM. The laser intensity is sketched in arbitrary units as a gray area.
the laser pulse and slightly earlier than carrier temperature. The maximum carrier density of $1.7 \times 10^{21} \text{cm}^{-3}$ is nine orders of magnitude larger than the initial carrier density. The increase is mainly due to photon absorption, while impact ionization appears to be insignificant. After reaching its maximum, the free carrier density starts to decrease due to Auger recombination.

In the following subsections, we will investigate the influence of several processes and properties on the outcome of our simulations. To further check the importance of the process or property under investigation and at the same time validate our model against the experiment, we present and discuss calculated damage thresholds at the end of each subsection. As a criterion for damage, we choose that the maximum lattice temperature reaches melting temperature $T_{\text{melt}} = 1685 \text{K}$. The damage threshold curve for the full model as well as experimental data published by Allenspacher et al.\textsuperscript{8} and Pronko et al.\textsuperscript{66} is always shown as a reference.

A. Influence of Density-Dependent Optical Parameters

In the last section, we demonstrated that the free carrier density in silicon may increase by as much as nine orders of magnitude during the irradiation with an ultrashort laser pulse. In the following, we will investigate the importance of the transient, density-dependent optical parameters. For simplicity, we assume here that the carrier system is non-degenerate and neglect carrier and energy transport.

Figure 3(a) depicts the reflectivity during the irradiation of silicon calculated using the $T$-expression (solid curve) and the Drude model (dashed curve), respectively, to determine both the reflectivity and the FCA coefficient as denoted in Tab. I. We immediately notice major differences: While the reflectivity calculated with the $T$-expression increases mirroring the behavior of lattice temperature, the reflectivity calculated with the Drude model decreases roughly reflecting the inverse behavior of carrier density. The reflectivity calculated with the Drude model already shows significant changes during irradiation, while the increase calculated with the $T$-expression is delayed and less pronounced. After the initial steep decrease calculated using the Drude model, the reflectivity increases again and shows a second slight minimum, after which it increases further.

This behavior will be discussed in more detail in the next subsection. But first, let us have a look at the impact of the drastically different reflectivities shown in Fig. 3(a) on carrier and lattice temperatures as well as carrier density.

Figure 3(b) clearly shows that the maxima of carrier density as well as carrier and lattice temperatures are all higher when using the Drude model than when using the $T$-expression. This is most prominent for carrier temperature but also noticeable for the final lattice temperature.

FIG. 3. (Color online) Reflectivity (a), carrier density as well as carrier and lattice temperature (b) calculated using either the $T$-expression or the Drude model.

The densities calculated using the Drude model and the $T$-expression, respectively, deviate at their maximum but tend towards similar values for later times. The reason is that Auger recombination, as a three particle process, is much stronger for higher densities.

The significant differences, especially in temperature, shown in Fig. 3(b) indicate that the choice of using either the $T$-expression or the Drude model to calculate reflectivity and FCA coefficient might strongly influence the damage thresholds estimated applying the nTTM.

In Fig. 4 the damage thresholds calculated using the $T$-expression (blue triangles) and the full model including density-dependent optical parameters (red diamonds) are shown in comparison to experimental data published by Allenspacher et al.\textsuperscript{8} and Pronko et al.\textsuperscript{66} as well as theoretical predictions presented by Chen et al.\textsuperscript{35}. Both our calculations consider transport and a Fermi distributed carrier gas. For their calculations, Chen et al.\textsuperscript{35} solved an nTTM in combination with the $T$-expression. As damage criterion, they used a critical density fitted to match the experimental determined threshold for a pulse duration of 500 fs.

Figure 4 clearly shows, that our calculation using the $T$-expression significantly overestimates the damage thresholds while the damage threshold calculated using the Drude model are in very good agreement with the
FIG. 4. (Color online) Damage thresholds calculated using either the T-expression or the full model and thereby the Drude model in comparison to experimental data obtained by Allenspacher et al. and Pronko et al. as well as thresholds simulated by Chen et al.

experimental data even over a wide range of pulse durations from 80 fs up to 6 ps. The thresholds calculated by Chen et al. agree reasonably well with the experiment. However, the threshold decreases for long pulses, whereas the experimental data does not show any decrease. For these pulse durations in the picosecond range, the thresholds calculated with our full model considering density-dependent optical parameters better resemble the behavior of the experimental data.

We can therefore conclude, that it is highly important to account for the transient optical parameters changing due to the strongly non-constant free carrier density in laser-excited silicon.

B. Influence of Collision Frequency

Following this conclusion, we will now look at the reflectivity obtained with the Drude model, especially the importance of the choice of carrier collision frequency, in more detail. We will again neglect transport and assume the carrier system to be non-degenerate at all times.

Figure 5(a) depicts the reflectivity calculated with the Drude model for different assumptions on the carrier collision frequency together with the carrier density obtained considering both types of collisions. The reflectivity considering both carrier-ion and carrier-phonon collisions (red solid curve) is the same as shown in Fig. 3(a). If only carrier-ion collisions are considered (green dash-dotted curve), the reflectivity minimum is slightly lower than when both types of collisions are considered. In addition, there is no second minimum but only a shoulder during the time. When, on the other hand, only carrier-phonon collisions are considered (blue dashed curve), there is not even a hint of a second minimum but a noticeably lower single minimum when carrier density is maximal and a continuous increase in reflectivity afterwards. For a constant collision frequency of 1 PHz (orange dotted curve) the behavior is basically the same. The only difference is that the reflectivity minimum is higher and comparable to the reflectivity minima considering only carrier-ion collisions or both types of collisions.

Explanations for these different behaviors of reflectivity can easily be found by looking directly at the carrier collision frequency depicted in Fig. 5(b). For a constant collision frequency, the reflectivity reflects the inverse behavior of the carrier density. If only carrier-phonon collisions are considered, the collision frequency (24) increases only slightly (within the same order of magnitude) reflecting lattice temperature. This increase is quite slow and so small that it does not noticeably influence reflectivity. Therefore, the behavior of the reflectivity when considering only carrier-phonon collisions is the same as when assuming a constant collision frequency. The reflectivity minimum is, however, significantly lower because the collision frequency considering carrier-phonon collisions is lower than the constant frequency at all times, while the carrier densities are compa-
rable. For the irradiation of silicon with an 800 nm-laser, the Drude model predicts a lower reflectivity minimum for a lower collision frequency whenever the carrier density is below $7 \times 10^{21} \text{cm}^{-3}$ which is the case in all calculations presented here.

When only carrier-ion collisions are considered, the initial collision frequency is about one order of magnitude lower than when only carrier-phonon collisions are considered. The collision frequency then reflects carrier temperature. The behavior similar to the one of carrier temperature is to be expected because for a non-degenerate carrier system, the screening for carrier-ion collisions is Debye screening and Eq. (27) reduces to

$$
\nu_{\text{carrier-ion}} = \left( \frac{1}{m_e} + \frac{1}{m_h} \right) \frac{\sqrt{3\pi\varepsilon_0} \varepsilon}{e^2} (k_B T_e)^{3/2}.
$$

During the calculation, the collision frequency changes over more than two orders of magnitude.

While for a constant or nearly constant collision frequency the carrier density dictates the behavior of reflectivity, in this case of a strongly transient collision frequency both, collision frequency and carrier density, compete for dominance over reflectivity. Therefore, let us remember the influence of carrier density and collision frequency on reflectivity before analyzing the reflectivity in detail: According to the Drude model, an increase in carrier density causes a decrease in reflectivity. For the carrier density range considered here, the Drude model additionally predicts a lower reflectivity for a lower collision frequency.

On basis of these two statements, let us now analyze the reflectivity when only carrier-ion collisions enter the collision frequency (green dash-dotted curve in Fig. 5).

The reflectivity first decreases because of increasing carrier density but then reaches an early minimum when the collision frequency increases significantly, causing in turn an increase in reflectivity. This increase is first steep and then slowing down. The steep increase is due to a still increasing collision frequency at that time. Then the collision frequency passes its maximum and begins to decrease again. Consequently, the reflectivity should decrease. At that time the carrier density is, however, already decreasing which should lead to an increasing reflectivity. Hence, the increase in reflectivity caused by the decreasing carrier density is hindered by a decrease in reflectivity due to a decreasing collision frequency. Overall, the reflectivity increases first very slowly then faster once the decrease in collision frequency becomes less steep and the influence of carrier density begins to dominate.

When both carrier-ion and carrier-phonon collisions are considered, the behavior of the reflectivity is basically the same as when only carrier-ion collisions are considered, but with slight differences. The carrier-phonon collisions cause a larger maximum collision frequency which leads to a higher minimum reflectivity. In addition, after the collision frequency passes its maximum, the reflectivity reaches a slight second minimum despite the, at that time, decreasing carrier density. This is because, for a short time, the influence if the decreasing collision frequency dominates over the influence of the decreasing carrier density. Afterwards, the influence of carrier density dictates the behavior of reflectivity which then increases.

The influence of the different collision frequencies and, consequently, different reflectivities on carrier and lattice temperature is depicted in Fig. 6. It is obvious that the different assumptions on the collision frequencies cause drastic differences in carrier and lattice temperature. While the lowest final lattice temperature, reached when only carrier-phonon collisions are considered, is about 700 K, the highest final lattice temperature, reached under the assumption of a constant collision frequency of 1 PHz, is about 1900 K and therefore above melting temperature (see Tab. I).

This is interesting because the lowest final lattice temperature is reached in the case of the lowest reflectivity. In this case the intensity within the material is highest. For identical absorption coefficients, the absorption and, hence, the final lattice temperature should be highest. The absorption coefficients are, however, not identical for the four different collision frequencies considered here. Especially the FCA coefficient (23), that is proportional
to the imaginary part of the refractive index depends on the choice of the collision frequency. It is lower for a lower collision frequency. The collision frequency considering only carrier-phonon collisions is significantly lower than in the other cases during most of the laser pulse, especially at the pulse maximum (cf. Fig. 5(b)). During that interval, free carrier absorption is consequently weakest when only carrier-phonon collisions are considered. This explains why the final lattice temperature is lowest in this case although the intensity passing into the material is highest.

We conclude, that the calculated temperatures depend highly on the assumption on carrier collision frequency. Consequently, this assumption might significantly alter the prediction of damage thresholds.

Figure 7 depicts the damage thresholds calculated with the full model with a dynamical carrier collision frequency as well as two different constant collision frequencies in comparison to experimental data published by Allenspacher et al.\textsuperscript{8} and Pronko et al.\textsuperscript{66}.

As has been discussed in Sec. II D, the system of equations drastically simplifies when the carrier system is assumed to be non-degenerate (Maxwell-Boltzmann distributed). In the following, we will investigate, whether this assumption is justified in laser-excited silicon. For that purpose, we use a dynamical carrier collision frequency and again neglect transport.

The distribution functions of electrons and holes enter the equation for carrier energy density (11) and consequently the calculation of carrier temperature. Thus, when abandoning the assumption of a classical non-degenerate carrier gas, the expression for carrier energy (18) becomes invalid and the full equation (11) has to be used instead. Consequently, the calculation of carrier temperature following Eq. (15) also becomes more laborious.

Figure 8 shows carrier densities and temperatures as well as lattice temperatures calculated assuming a non-degenerate carrier gas and allowing the carrier gas to become degenerate, respectively. While the choice of carrier distribution function has almost no influence on carrier density, there are significant differences in carrier temperature and, especially, lattice temperature (Fig. 8(b)).

To check whether and when the carrier system becomes degenerate, carrier temperature and Fermi temperatures of electrons and holes (19) are depicted in Fig. 9(a). If the Fermi temperature of electrons and holes is larger than carrier temperature, the respective carrier system is degenerate and should be described by a Fermi distribution. While the electron system is degenerate for a short time during the laser pulse and then again after the laser pulse, the hole system only becomes degenerate after the pulse at later times than the electrons. While the hole system becomes non-degenerate again after about 33 ps, the electron system is still degenerate at the end of the calculation at 50 ps.
FIG. 8. (Color online) Carrier density and temperature (a) as well as lattice temperature (b) calculated assuming either a Maxwell-Boltzmann distributed or a Fermi distributed carrier system.

Alternatively, we can check whether the chemical potential of the electrons (holes) is positioned inside the conduction (valence) band, which is another indication for degeneracy. It is equivalent to the question whether the reduced Fermi levels of electrons and holes (5) are negative (chemical potential is positioned outside the band) or positive (chemical potential is positioned inside the band). Figure 9(b) shows the reduced Fermi levels of electrons and holes. It is obvious that this criterion indicates a degenerate electron and hole system in the same time intervals as the comparison with Fermi temperature. In addition, we see that the degeneracy of electrons is more pronounced than the degeneracy of holes because the reduced Fermi level of the electrons is larger due to different effective masses. This means in turn that the chemical potential of the electrons is positioned deeper within the conduction band than the one of the holes is in the valence band.

Figure 8(b) clearly shows that the assumption on the distribution function influences final lattice temperature. Consequently, it might also affect the estimation of damage thresholds.

To investigate the influence of the assumed distribution function on the predicted damage thresholds, Fig. 10 depicts the thresholds estimated applying either a Maxwell-Boltzmann distribution (blue triangles) or using the full model including a Fermi distribution (red diamonds, same curve appears in Fig. 4 and 7) in comparison to experimental data published by Allenspacher et al. and Pronko et al.

While the predicted damage thresholds are both in good agreement with the experimental results for pulse durations up to about 300 fs, the non-degenerate calculation tends to overestimate the threshold for longer pulses.

This behavior is due to the fact that for long pulses, the carrier system tends to become more degenerate even during irradiation. FCA is more effective when the carrier system becomes degenerate which explains why the damage threshold is lower when a Fermi distribution is assumed. The reason why FCA is more effective for degenerate systems lies within the nature of the Fermi integrals. The ratio of Fermi integrals \( H^{\xi}_{\Omega}(\xi_c) \) increases with increasing degeneracy for \( \xi > \varsigma \). Consequently, the carrier-ion collision frequency and, hence, the FCA coefficient is larger in the degenerate case (27) than in the non-degenerate one (28).

We conclude that the classical assumption of a Maxwell-Boltzmann distribution oversimplifies the description. Though the choice of the distribution function does not influence the estimated damage threshold for pulses up to about 300 fs, it may, however, become important for longer pulses.
For all following calculations, we will therefore assume a Fermi distributed carrier system. Note, however, that all observations made on reflectivity and absorption for a non-degenerate system in Sec. IIIA and IIIB as well as all conclusions drawn there also apply to a degenerate system.

### D. Influence of Transport

In this section, we will investigate transport effects. To that end, we consider both particle (7) and energy (9) transport.

Figure 11 depicts carrier and lattice temperature as well as carrier density at the surface calculated either considering or neglecting transport. The maxima of carrier and lattice temperature are lower when transport is considered, because energy dissipates away from the surface. In addition, while lattice temperature still increases even on long timescales of tens of picoseconds in the case without transport, it reaches a maximum and starts to decrease on longer timescales when transport is considered.

For carrier density things are a bit different. While the maximum carrier density shows the same behavior as the temperatures, after about 5 ps the density at the surface becomes higher when transport is considered in comparison to the case without transport. This behavior of surface carrier density can be attributed to carrier confinement which has been investigated by Preston and van Driel\(^\text{11}\) for silicon irradiated with picosecond and nanosecond pulses.

Let us now briefly have a look at the influence of transport effects on the estimated damage thresholds.

Figure 12 depicts the damage thresholds calculated using the full model (red diamonds, same curve appears in Fig. 4, 7 and 10) and neglecting transport (blue triangles) in comparison with experimental data published by Allenspacher \textit{et al}.\(^\text{8}\) and Pronko \textit{et al}.\(^\text{66}\).

The damage thresholds estimated neglecting transport are lower than those when considering transport but show the same overall behavior with increasing pulse duration.
and are still in very good agreement with experimental data. The threshold being lower when neglecting transport can be explained by the fact that in this case no heat is carried away from the surface.

Thus, we conclude that as long as only the temperature evolution at the surface is of interest (e.g., for the estimation of damage thresholds) and no detailed investigation of temperature or density profiles is necessary, it is sufficient to implement an nTTM neglecting transport, which considerably reduces computational efforts.

IV. SUMMARY

In this work, we extended the nTTM first presented by van Driel\textsuperscript{11} to account for the changes in optical parameters, namely reflectivity and FCA coefficient, due to the highly transient free carrier density during the excitation with femtosecond laser pulses. To that end, we implemented a Drude model.

For the irradiation of silicon with a 100 fs-laser pulse at 800 nm, we analyzed the influence of the transient optical properties. We conclude, that it is of utter importance to consider changes in reflectivity and FCA coefficient due to the changing carrier density. This can not only be seen when looking at reflectivity, temperature and density evolution but is also clearly reflected in the calculated damage thresholds. Comparison with experimental data shows that the $T$-expression often used with the nTTM in earlier works strongly overestimates the threshold. Damage thresholds calculated with our improved approach using a Drude model considering both carrier-ion and carrier-phonon collisions are, however, in very good agreement with experimental data even over a wide range of pulse durations. Note that we did not fit any parameter of our model to reproduce experimental damage thresholds.

Additionally, we found that the choice of carrier collision frequency used in the Drude model strongly influences the lattice temperature and, consequently, the calculated damage threshold. Thus, we conclude that it does not suffice to assume a constant carrier collision frequency. Both carrier-ion collisions and carrier-phonon collisions have to be considered especially when dealing with pulses longer than about 300 fs.

We analyzed the influence of the distribution function assumed for the carriers and found that degeneracy effects might become important for pulses in the picosecond range.

Furthermore, we investigated transport of carriers and energy and found that the influence of the transient band gap during the excitation can lead to carrier confinement in a region below the incident surface. Moreover, we found that damage thresholds calculated neglecting transport are a bit lower than those when considering transport but show the same overall behavior with increasing pulse duration and are still in very good agreement with experimental data. We therefore conclude, that it suffices to use an nTTM neglecting transport but considering transient optical parameters, which will finally be used to describe laser-excited semiconductors.

ACKNOWLEDGMENTS

The authors thank K. Sokolowski-Tinten and V. P. Lipp for helpful suggestions. Financial support by the Deutsche Forschungsgemeinschaft through the Emmy Noether (grant no. RE 1141/11) and the Heisenberg program (grant no. RE 1141/15) is gratefully acknowledged.

\begin{thebibliography}{9}
\bibitem{1} J. S. Preston and H. M. van Driel, \textit{Phys. Rev. B} \textbf{30}, 1950 (1984).
\bibitem{2} X. Y. Wang, D. M. Riffe, Y.-S. Lee, and M. C. Downer, \textit{Phys. Rev. B} \textbf{50}, 8016 (1994).
\bibitem{3} J. R. Goldman and J. A. Prybyla, \textit{Phys. Rev. Lett.} \textbf{72}, 1364 (1994).
\bibitem{4} B. N. Chichkov, C. Momma, S. Nolte, F. Alvensleben, and A. Tümermann, \textit{Applied Physics A} \textbf{63}, 109 (1996).
\bibitem{5} T. Sjodin, H. Petek, and H.-L. Dai, \textit{Phys. Rev. Lett.} \textbf{81}, 5664 (1998).
\bibitem{6} K. Sokolowski-Tinten and D. von der Linde, \textit{Phys. Rev. B} \textbf{61}, 2643 (2000).
\bibitem{7} A. J. Sabbah and D. M. Riffe, \textit{Phys. Rev. B} \textbf{66}, 165217 (2002).
\bibitem{8} P. Allenspacher, B. Hüttner, and W. Riede, \textit{Proc. SPIE} \textbf{4932}, 358 (2003).
\bibitem{9} L. Englert, M. Wollenhaupt, L. Haag, C. Sarpe-Tudoran, B. Rethfeld, and T. Baumert, \textit{Applied Physics A} \textbf{92}, 749 (2008).
\bibitem{10} S. I. Anisimov, B. L. Kapeliovich, and T. L. Perel’man, Zh. Eksp. Teor. Fiz. \textbf{66}, 776 (1974), [Sov. Phys. JETP \textbf{39}, 375 (1974)].
\bibitem{11} H. M. van Driel, \textit{Phys. Rev. B} \textbf{35}, 8166 (1987).
\bibitem{12} B. Rethfeld, A. Kaiser, M. Vicanek, and G. Simon, \textit{Applied Physics A} \textbf{69}, S109 (1999).
\bibitem{13} B. Rethfeld, \textit{Phys. Rev. Lett.} \textbf{92}, 187401 (2004).
\bibitem{14} N. M. Bulgakova, R. Stoian, A. Rosenfeld, I. V. Hertel, and E. E. B. Campbell, \textit{Phys. Rev. B} \textbf{69}, 054102 (2004).
\bibitem{15} N. M. Bulgakova, R. Stoian, A. Rosenfeld, I. V. Hertel, W. Marine, and E. E. B. Campbell, \textit{Applied Physics A} \textbf{81}, 345 (2005).
\end{thebibliography}
