Influence of an external electric field on the energy dissipation at the initial stage of laser ablation

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Abstract. A density-dependent two-temperature model is applied to describe laser excitation and the following relaxation processes of silicon in an external electric field. Two approaches on how to describe the effects of the external electric field are presented. The first approach avoids the buildup of internal electric fields due to charge separation by assuming ambipolar diffusion and adds an additional carrier-pair current. In the second approach, electrons and holes are treated separately to account for charge separation and the resulting shielding of the external electric field inside the material. The two approaches are compared to experimental results. Both the first approach and the experimental results show similar tendencies for optimization of laser ablation in the external electric field. © 2021 Society of Photo-Optical Instrumentation Engineers (SPIE) [DOI: 10.1117/1.OE.61.2.021003]

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1 Introduction

Engineering and control of surface and bulk properties of materials using ultrashort laser pulses are highly demanded for modern innovative technologies. Laser-assisted nanostructuring has already been employed for a number of applications such as optical waveguides, photonic crystals, solar fuels, functional colorization of surfaces, friction reduction, controlling of wetting properties, and many others. However, the quality of the micromachined areas still needs to be improved to fully exploit the potential of laser processing of solids. Several methods were proposed to optimize the ablation process, for instance, liquid confinement layers or laser treatment of materials in the presence of different gases. An alternative and promising way to control laser micromachining is via application of external magnetic or electric fields. An influence of the external magnetic field on the ablation process is still under intense investigations and debates, however, less attention was given so far to the role of an externally applied electric field to the energy deposition and dissipation. Moreover, the external E-field in a recent investigation was perpendicular to the laser beam wave vector. In contrast, in our experiments, we apply an external electric field parallel to the laser wave vector.

In this paper, we report on the influence of an external electric field on the ablation process of a silicon sample following a femtosecond laser pulse. Here, we focus on the theoretical description, including the effect of an external electrical field to the density-dependent two temperature model (nTTM). This model has been successfully applied to simulate laser-matter structuring for different numerical or experimental setups. The nTTM allows to take into account the transient free-carrier density in contrast to the conventional two-temperature model, which only tracks carrier and lattice temperature evolution. We also show results of accompanying experiments, revealing an influence of the external electric field on ablation depth.

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The paper is structured as follows: In Sec. 2, we summarize the main idea, equations, and result of the nTTM in its original form. In Sec. 3, we present two different theoretical approaches aiming to include the effect of an external electric field. Section 4 is devoted to the experimental part of the project. The results of theory and experiment are compared in Sec. 5. We close with a conclusion and summary.

2 Density-Dependent Two-Temperature Model (nTTM)

The excitation of silicon with an ultrashort laser pulse, as well as the following relaxation processes can be described by an nTTM as introduced by van Driel in Ref. 20 and presented in the form applied in this work by Rämer et al. 21 For completeness, a short description of the model is given here.

The nTTM tracks the spatially and temporarily resolved evolution of its three main parameters, namely free-carrier density $n$, electron temperature $T_e$ and phonon temperature $T_{ph}$. It is governed by three equations, denoting the energy and particle balance, respectively:

$$c_{ph} \frac{\partial T_{ph}}{\partial t} = \nabla \cdot (\kappa_{ph} \nabla T_{ph}) + g(T_e - T_{ph}),$$

$$c_e \frac{\partial T_e}{\partial t} = (\alpha_{SPA} + \alpha_{FCA})I + \beta_{TPA} I^2 - \nabla \cdot w - g(T_e - T_{ph}) - \frac{\partial u_e}{\partial n} \frac{\partial n}{\partial t} - \frac{\partial u_e}{\partial e_g} \frac{\partial e_g}{\partial t},$$

$$\frac{\partial n}{\partial t} = \frac{\alpha_{SPA} I}{\hbar \omega_L} + \frac{\beta_{TPA} I^2}{2 \hbar \omega_L} + \delta_H n - \gamma n^3 - \nabla \cdot j.$$  

Here, Eq. (1) tracks the evolution of the phonon temperature $T_{ph}$ with the corresponding phonon heat capacity $c_{ph}$. Heat transport in the phonon system is described by the first term on the right-hand side of Eq. (1), where $\kappa_{ph}$ is the phonon heat conductivity. The second term considers electron–phonon coupling, being proportional to the temperature difference between electrons and phonons. An important parameter is the electron–phonon coupling parameter $g$. Here, it is assumed as $g = c_e / \tau_{rel}$, where $c_e$ is the electron heat capacity and $\tau_{rel}$ is a constant relaxation time. Equation (2) describes the evolution of the electron temperature $T_e$. The first two terms on the right-hand side consider laser absorption with the absorption coefficients for single-photon absorption (SPA) $\alpha_{SPA}$, free-carrier absorption (FCA) $\alpha_{FCA}$, and two-photon absorption (TPA) $\beta_{TPA}$. The third term takes into account heat transport in the electron system by the heat current density $w$. The fourth term is the electron–phonon coupling, similar as in Eq. (1) and here with the opposite sign to ensure energy conservation. The last two terms in Eq. (2) are analogous to the left-hand side of the equation. All three terms together build the total time derivative of the free-carrier energy density $u_e$ depending on the electron temperature, free-carrier density and also the band-gap energy $e_p$. The band-gap energy is a transient parameter due to its dependence on the free-carrier density and phonon temperature. The evolution of the free-carrier density $n$ is described by Eq. (3). The first two terms take into account free-carrier generation by SPA and TPA. Here, $\hbar \omega_L$ is the photon energy. The third and fourth term describe impact ionization with the impact ionization coefficient $\delta_H$ and Auger recombination with the Auger recombination coefficient $\gamma$, respectively. Both processes also change the kinetic energy and thus the temperature of the free carriers: in impact ionization, kinetic energy of a free electron is used for a bound one to overcome the band gap. In the opposite process of Auger recombination, the potential energy released by the recombination of an electron-hole pair is transferred to kinetic energy of a third free carrier. The change of temperature of the electron system is reflected by the term $-(\partial u_e / \partial n)(\partial n / \partial t)$ in Eq. (2). Particle transport with the particle density current $j$ is considered by the last term of Eq. (3).

For transport in the electron system, ambipolar diffusion is assumed, which means that electrons and holes move together as pairs due to their Coulomb attraction. 20 The resulting electron-hole pair density current $j$ and electron heat density current $w$ are described as

$$j = \frac{1}{e^2} \frac{\sigma_e \sigma_h}{\sigma_e + \sigma_h} [\nabla (\mu_h - \mu_e) + e (S_e - S_h) \nabla T_e],$$  

$$w = \frac{\partial}{\partial t} (n \epsilon_{tot} (T_e, T_{ph})).$$
\[ w = \Pi j - (\kappa_e + \kappa_h) \nabla T_e, \] (5)

with the electron charge \( e \), electric conductivity \( \sigma \), chemical potential \( \mu \), Seebeck coefficient \( S \), Peltier coefficient \( \Pi \), and the heat conductivity \( \kappa \). The subscripts \( e \) and \( h \) stand for electrons and holes, respectively.

The temporal evolution of the three main parameters at the laser-irradiated surface obtained by the nTTM\textsuperscript{21} is shown in Fig. 1 for a laser pulse duration of \( \tau = 100 \text{ fs} \), a laser wavelength of \( \lambda = 800 \text{ nm} \) and a sub-threshold laser fluence of \( F = 130 \text{ mJ/cm}^2 \). The material parameters used in this work are taken from Ref. 21.

The gray shadowed area shows the temporal laser intensity profile centered around a time of \( t = 300 \text{ fs} \). At first, the electron system is heated by the far-left flank of the Gaussian laser intensity profile and reaches an electron temperature plateau. At this initial stage, SPA is the dominating absorption process in which valence electrons absorb one photon and consequently are excited across the band gap. The excess energy is transferred into kinetic energy of the free electrons. Since at these initially very low intensities each excited electron carries the same amount of energy, a transient temperature plateau builds up.\textsuperscript{21} However, it is worth noting that the energy distribution is not thermalized at these low densities. As the laser pulse maximum is reached, the carrier-pair density increases rapidly and the electron temperature increases furthermore by FCA, TPA, and Auger heating. The latter is caused by an interplay of Auger recombination and the term \(- (\partial u_e / \partial n) (\partial n / \partial t)\) in Eq. (2). Shortly after the laser intensity peak, the electron temperature and density reach a maximum and decrease again. Both quantities are affected by transport processes due to gradients in the chemical potential and temperature. Additionally, the decrease in electron temperature is caused by electron–phonon coupling, where the electron and phonon system exchange energy and thereby level their temperatures, and the decrease of carrier density is influenced by Auger recombination.

### 3 Models for the Effect of an External Electrical Field

When we apply an electrical field perpendicular to the surface of the irradiated material, electrons and holes will screen the field towards the bulk of the material. Such conditions can be usually described with help of the dielectric function. However, the dielectric function of laser-excited silicon upon and directly after irradiation is changing rapidly, due to the rapidly changing carrier densities and temperatures.

Since a full inclusion of transient optical properties is numerically challenging, we study as a first step, two extreme cases of the microscopic response of the charges within the bulk material. The first case maintains the assumption of ambipolar diffusion within the material: An external electrical field is capable to accelerate the mobile electrons within the skin layer. Further inside the material, the external electrical field is screened and the accelerated electrons move freely through the bulk. With the concept of ambipolar diffusion, which essentially avoids the buildup
of further internal electrical fields, electrons and holes will then move together through the bulk. Within this approach, we do not calculate the details of the charge-acceleration within the skin layer, but assume a constant drift velocity for electron-hole pairs. We describe the extension of our model and the obtained results in Sec. 3.1. The second case studies the other extreme, which is the full charge separation of electrons and holes. Depending on the mobility of the charge carriers, they are able to annihilate the electrical field within the material. We describe this approach and the results in Sec. 3.2.

3.1 Additional Carrier-Pair Current

Here, we study the influence of a constant drift velocity of the electron-hole pairs on the temporal and spatial free-carrier density as well as on the electron and phonon temperature.

A current of

\[ j_{\text{add}} = v \cdot n, \]  

is added to the electron–hole pair density current \( j \) in Eq. (4). Here, \( v \) is a constant drift velocity and \( n \) is the density of electron-hole pairs as above. The direction of the drift velocity is perpendicular to the laser-irradiated surface, with the sign given according to Fig. 2.

Figure 2 shows a schematic view of the laser-irradiated material. The arrows indicate the direction of the additional carrier-pair current \( j_{\text{add}} \) for different signs of the drift velocity \( v \). For a negative sign of \( v \), the current is headed from the back side of the material towards the laser-irradiated surface, for a positive sign in the opposite direction.

Figure 3 shows the temporal evolution of the three main parameters at the laser-irradiated surface for different directions of the additional current \( j_{\text{add}} \). A laser wavelength of \( \lambda = 800 \text{ nm} \), pulse duration of \( \tau = 100 \text{ fs} \) and a fluence of \( P = 130 \text{ mJ/cm}^2 \) is used. For the negative (positive) direction, the free-carrier density \( n \) and carrier temperature \( T_e \) are increased (decreased) by...
the additional current. As a consequence, the phonon temperature $T_{\text{ph}}$ is also increased (decreased).

Figure 4 shows the spatial phonon temperature profile within the sample of 800-nm thickness. At the moment of time $t = 40$ ps, the phonon temperature near the surface has roughly reached its maximum. For the negative direction of the additional current, i.e., pointing against the laser wave vector (see Fig. 2), a higher phonon temperature and a steeper gradient in the temperature profile is obtained near the laser-irradiated surface in comparison to the case without an additional current. In contrast to that, the phonon surface temperature is lowered and the gradient in the temperature profile flattened for the positive direction of the additional current, when it is pointing along the laser wave vector. For this direction, a considerable increase of phonon temperature can be observed at the back side of the material.

3.2 Charge Separation

In this approach, free electrons and holes are treated separately to account for the charge separation in an external electric field. Thus, the free-carrier density $n$ is replaced by the free electron density $n_e$ and the hole density $n_h$. However, free electrons and holes are still assumed to possess a common temperature $T_e$.

Therefore, the evolution of the free-carrier density in Eq. (3) is modified to account for separate densities

$$\frac{\partial n_{e,h}}{\partial t} = \frac{a_{\text{SPA}} I}{\hbar \omega} + \beta_{\text{TPA}} I^2 + \delta_{\text{eh}} n_e n_h + \delta_{\text{ehh}} n_e n_h n_h - \gamma_{\text{eh}} n_e^2 n_h - \gamma_{\text{ehh}} n_e n_h^2 - \nabla \cdot j_{e,h},$$

where the subscripts $e$ and $h$ represent electrons and holes, respectively. The first two terms in Eq. (7) which describe laser excitation are the same for both carrier types since electrons and holes are created as pairs. The next four terms account for impact ionization and Auger recombination. Both events which involve two electrons and one hole (eeh) and events which involve one electron and two holes (ehh) are considered. The last term describes particle transport with two separate particle density currents which read

$$j_{e,h} = \mp \frac{\sigma_{e,h}}{e} \left[ E + \frac{\nabla \mu_{e,h}}{e} + S_{e,h} \nabla T_e \right].$$

Here, $E$ is the internal electric field which is found by solving $\nabla \cdot (\varepsilon E) = e(n_h - n_e)$ with the boundary condition, that the electric field at the laser-irradiated surface is $E(z = 0) = E_{\text{ext}} / \varepsilon$. Here, $E_{\text{ext}}$ is the applied external electric field and $\varepsilon$ the static dielectric constant.

Fig. 4 Spatial phonon temperature profile for different drift velocities $v$, 40 ps after laser irradiation.
Furthermore, the heat current of electrons and holes, respectively, read

\[ w_{e,h} = \Pi_{e,h} J_{e,h} - \kappa_{e,h} \nabla T_{e,h} \]  

(9)

with the total carrier heat current given by \( w = w_e + w_h \). The material properties, newly used in this extension of the model, are listed in Table 1. The mobilities of electrons and holes, respectively, are both assumed to be constant.

Figure 5 shows the spatial profile of the lattice temperature after 20 ps obtained with this model for different fluences and different external electric fields. As in Sec. 3.1, a laser wavelength of \( \lambda = 800 \) nm and a pulse duration of \( \tau = 100 \) fs have been applied. In all investigated cases, the effect of the electric field on the phonon temperature profile is small, despite of large electric field strengths.

Figure 6 shows the temporal evolution of the electric field between the first and second discretization cell at a depth of 1 nm [Fig. 6(a)], as well as the change in electron density by transport in the first cell \( \frac{dn_e}{dt} |_{J} \) [Fig. 6(b)] for a laser fluence of \( F = 190 \) mJ/cm\(^2\). The electric field

| Table 1 | Applied material properties for the Charge Separation model. All other parameters are identical to the ones used in the nTTM.\(^{21}\) |
|-----------------|-----------------|
| **Quantity** | **Value** |
| See Ref. 20 for the calculation of the transport coefficients \( \sigma_{e,h}, S_{e,h} \) and \( \Pi_{e,h} \). |
| **Carrier properties** | |
| Electron mobility \( \sigma_e \) | 0.0085 m\(^2\)/Vs\(^{22}\) |
| Hole mobility \( \sigma_h \) | 0.0019 m\(^2\)/Vs\(^{22}\) |
| Auger coefficient (eeh) \( \gamma_{eeh} \) | \( 2.8 \times 10^{-31} \) cm\(^6\)/s\(^{27}\) |
| Auger coefficient (ehh) \( \gamma_{ehh} \) | \( 0.99 \times 10^{-31} \) cm\(^6\)/s\(^{27}\) |
| Impact ionization rate \( \delta_{eeh} = \delta_{ehh} = (\delta_{II}/2) \) | \( 1/2 \times 3.6 \times 10^{10} \exp[-1.5 E_{gap}/(k_B T_e)] \) s\(^{-10}\) |
| **Optical properties** | |
| Static dielectric constant \( \epsilon \) | 11.97\(^{28}\) |

**Fig. 5** Spatial phonon temperature profile after 20 ps for different external electric fields and fluences.
strength decreases significantly before the laser pulse maximum is reached. This is caused by the rapid change in electron density, which corresponds to the charge separation of the electron–hole pairs which are generated by the left flank of the Gaussian laser pulse. After the laser pulse maximum, the electric field strength increases again due to particle currents caused by the relaxation of gradients in the material. The increase in electric field strength has no significant effect on the change in electron density after the laser pulse maximum, compared to the case of no applied external electric field.

3.3 Comparison of the Models

For the two presented models, very different assumptions were made which also yield very different results. For the model presented in Sec. 3.1, electrons and holes are assumed to move as pairs such that quasi neutrality is not broken and a significant effect on the phonon temperature is observed for a constant drift velocity. However, the drift velocity cannot be directly attributed to a electric field strength since electron–hole pairs do not have a net charge. Contrary to that, electrons and holes are separated by the external electric field in the model presented in Sec. 3.2. This charge separation leads to rapid shielding inside the material and thus the effect of the external electric field on the phonon temperature is marginal. It is worthy to note that further studies are planned, particularly considering the microscopic development of a free particle drift on one hand and a transient dielectric function on the other hand. To date, we cannot compare the results quantitatively. However, we have performed experimental investigations to estimate the quantity of the effect of an external electric field. They are presented in the next section.

4 Experiment

Figure 7 shows the schematic diagram of the experimental setup. For the experiments, a femtosecond fiber laser system (BlueCut, Menlo Systems GmbH), with a pulse duration of about 400 fs, a central wavelength of 1030 nm, and maximum pulse energy of 10 μJ, was used to irradiate the surface. The laser polarization was linear. To focus the laser beam onto the sample surface a telecentric F-Theta lens with \( f_j = 100 \) mm focal length, which provides a beam waist of \( w_0 = 9.5 \) μm, was applied.

The electric field was generated parallel to the laser wave vector. To achieve this, the sample is positioned between a metallic foil and a metallic sample, which were attached to a voltage generator. The top electrode is a 100-μm thick metal foil and to avoid breaking through in silicon when a high electric field is applied it is placed 1.5 mm above the sample surface. Additionally, to guarantee a nearly homogenous electric field within the processing area a through-hole with 1 mm in diameter was processed in the middle of the top electrode. The electric field strength decreases significantly before the laser pulse maximum is reached. This is caused by the rapid change in electron density, which corresponds to the charge separation of the electron–hole pairs which are generated by the left flank of the Gaussian laser pulse. After the laser pulse maximum, the electric field strength increases again due to particle currents caused by the relaxation of gradients in the material. The increase in electric field strength has no significant effect on the change in electron density after the laser pulse maximum, compared to the case of no applied external electric field.
was set to be \((400 \pm 15) \text{ V/mm}\) and we investigated the influence in both directions. The electric field direction is regarded as plus (+) direction when it points from the top to the bottom and vice versa for the negative (−) direction. In this study, single-pulse laser ablation on the polished side of the silicon pieces with the dimensions of \(10 \times 10 \times 0.5 \text{ mm}^3\) was performed across a range of laser pulse energies. The ablated profile was measured with a confocal microscope (Zeiss smart proof 5, Carl Zeiss AG).

Figure 8 shows how the ablation depth changes in the presence of an electric field in dependence on the applied pulse energy \(E_p\). The pulse energy of \(0.81 \mu\text{J}\) is highlighted by the dashed line. Hence, the ablation threshold peak fluence is \(0.57 \text{ J/cm}^2\) and is comparable to already presented results from other groups.\(^{29-32}\) The change of ablation depth will be considered by the ratio of the ablation depth with an electric field \((d_{E})\) to without electric field \((d_{E=0})\). With increasing pulse energy the change in removal depth increases up to a pulse energy of \(\sim 6 \mu\text{J}\). Above this pulse energy a drop occurs and the change in removal depth decreases.
significantly and remains almost constant for pulse energies >7 \( \mu \)J. This behavior can be attributed to the two ablation regimes—the gentle ablation and strong ablation regime. In the gentle ablation regime, close to the ablation threshold energy, the applied pulse energy is absorbed in the shallow region defined by the optical penetration depth. In the strong ablation regime thermal processes dominate the ablation process and the heat affected zone is defined by the thermal penetration depth. In this case the laser excited region of silicon will be melted and after cooling the material re-solidified within the crater. Due to this re-solidification of the molten material the relative change in ablation depth remains constant in the strong ablation regime and the effect of an applied electric field is smaller compared to the gentle ablation regime. Moreover, by an applied positive electric field the ablation crater becomes even smaller in depth compared to the ablated structures without an applied electric field. In general, the investigations show that the most pronounced effect is present by applying a negative electric field during the laser ablation. The created craters are always \( \sim 10\% \) to 20\% larger in depth compared to an applied positive electric field.

5 Comparison

The experiments reveal a significant effect of the external electric field on ablation depth, while the charge separation model described in Sec. 3.2 only shows marginal effects on phonon temperature for larger electric field strengths. However, a qualitative comparison can be made between the experiments and the additional carrier-pair current model described in Sec. 3.1.

Figure 9 shows the spatial phonon temperature profile near the laser-irradiated surface obtained by the “additional carrier-pair current” model. A laser wavelength of \( \lambda = 800 \) nm, pulse duration of \( \tau = 100 \) fs and two different fluences were used. The lower fluence of \( F = 173 \) mJ/cm\(^2\) lies slightly above the melting threshold and the higher fluence of \( F = 250 \) mJ/cm\(^2\) is chosen, such that phonon temperatures near the laser-irradiated surface above the boiling temperature are reached. The results show a reversal of the effect of the additional current on melting depth for higher fluences. For the lower fluence, the melting depth is increased for the negative direction and decreased for the positive direction. The opposite effect is observed for the higher fluence.

Since the electron mobility is higher than the hole mobility, we attribute the positive drift direction in this model to the negative field direction in the experiment. The higher melting depth for the positive drift direction at the higher fluence corresponds to the observation of higher ablation depths for negative field direction in the experiment, where laser fluences far above the ablation threshold were used. From these results we conclude that a flattened phonon temperature profile as observed for the higher fluence and \( v = +10^4 \) m/s in Fig. 9 lowers heat loss near the laser-irradiated surface and thus is advantageous to achieve higher ablation depths.

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**Fig. 9** Spatial phonon temperature profile obtained by the “additional carrier-pair current” model described in Sec. 3.1, 40 ps after laser irradiation for different laser fluences.
6 Summary and Conclusion

We presented two approaches on how to describe the effects of an external electric field on the initial stage of laser ablation in silicon. Also, experimental measurements have been performed, which show an effect of the external electric field on ablation depth. Similar tendencies are observed for the first approach, in which ambipolar diffusion is assumed and a carrier-pair current with constant drift velocity is added. The second approach that accounts for charge separation and shielding of the external electric field inside the material shows only marginal effects on the spatial lattice temperature profile. Further studies are needed to refine both approaches and to allow for quantitative comparisons between theory and experiment.

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