Nonlinearity and wavelength control in ultrashort-pulse subsurface material processing

Roland A. Richter*
Department of Physics, NTNU, Norwegian University of Science and Technology, Trondheim, Norway

Vladimir Kalashnikov
Photonics Institute, Vienna University of Technology, Vienna, Austria and
Dipartimento di Ingegneria dell’Informazione, Sapienza University of Rome, Italy

Irina T. Sorokina
Department of Physics, NTNU, Norwegian University of Science and Technology, Trondheim, Norway and
ATLA Lasers AS, Trondheim, Norway

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The pronounced dependence of the nonlinear parameters of both dielectric and semiconductor materials on the wavelength, and the nonlinear interaction between the ultra-short laser pulse and the material requires precise control of the wavelength of the pulse, in addition to the precise control of the pulse energy, pulse duration and focusing optics. This becomes particularly important for fine sub-wavelength single pulse sub-surface processing. Based on two different numerical models and taking Si as example material, we investigate the spatio-temporal behavior of a pulse propagating through the material while covering a broad range of parameters. The wavelength-dependence of material processing depends on the different contributions of two- and three-photon absorption in combination with the Kerr effect which results in a particularly sharp nonlinear peak at $\approx 2100 \text{ nm}$. We could show that in silicon this makes processing preferable close to this wavelength. The impact of the nonlinear nonparaxial propagation effects on spatio-temporal beam structure is also investigated. It could be shown that with increasing wavelength and large focusing angles the aberrations at the focal spot can be reduced, and thereby cleaner and more precise processing can be achieved. Finally, we could show that the optimum energy transfer from the pulse to the material is within a narrow window of pulse durations between 600 to 900 fs.

I. INTRODUCTION & STATE OF THE ART

Fine material processing of semiconductors in general, and silicon in particular is a hot topic both in the scientific community as well as in industry. Up to now a lot of effort has been put into surface processing experiments using mainly nanosecond pulse durations and pulse energies above $1 \mu\text{J}$ [1–8]. Processing technology has been well understood both on the practical and theoretical level [7, 9]. In the last few years the focus has been shifted towards the next level fine material processing – in particular sub-surface modification of the crystalline silicon using ultra-short pulsed lasers (pico- and femtosecond) at the wavelengths above $1100 \text{ nm}$, where silicon becomes transparent. Several successful studies have been carried out since then [10–20]. Those sub-surface defects are targeted for such important industrial processes as thin wafer exfoliation [21], sub-surface structuring – [17, 21], data storage [22], waveguides [4, 22–24] and grating fabrication [6]. The common way to introduce sub-surface modifications in all these works is using multiple pulses and pulse trains [25]. Very recently it was shown that also single pulse processing of monocrystalline silicon is possible with ultra-short pulses at $2100 \text{ nm}$, producing particularly fine sub-wavelength modifications [26]. Besides optimization of the laser pulse parameters, the choice of the laser wavelength played a crucial role in this work, suggesting a much more complex nonlinear light-matter interaction with spatio-temporal dynamics that strongly depend on the wavelength. The latter is a subject of the present numerical investigation.

Besides nonlinear multi-photon absorption light propagating through the material experiences several different processes, such as self-focusing [16, 21] and plasma-induced de-focusing and reflection [11, 15], to name a few. Those processes strongly depend on the wavelength and intensity of the pulse propagating through the material [27, 28]. Depending on the wavelength the impact of each parameter will vary. Here, especially self-focusing effect due to the third-order nonlinearity has to be mentioned, as it becomes extremely strong within a rather narrow wavelength range at a certain wavelength, that depends on the forbidden band-gap of the material. An investigation of the wavelength-dependent interplay of several nonlinear optical effects and their impact on the spatio-temporal dynamics of ultra-short laser-pulses inside the material becomes particularly important for single-pulse sub-surface processing of any material, not only silicon. In the present work we for the for the first time perform such study using silicon as a model.

In silicon the third-order nonlinearity peaks between 2000 to $2200 \text{ nm}$ [27, 28], and therefore influences the pulse significantly stronger than at shorter or longer wavelengths. Within the same wavelength range the two- and three-photon absorption experiences a minimum at...
this wavelength, thus allowing to reduce the potential for surface damage (due to lower multi-photon absorption) while retaining the capability of delivering a large amount of energy to the focal spot [16]. This makes it possible to achieve pulse beam collapse and correspondingly high intensities at relatively low pulse energies. For certain parameters this might decrease the material processing threshold, while simultaneously increase the precision at which the material can be processed. In Ref. [21] we for the first time suggested to use the interplay of all these nonlinear optical effects to find an optimum wavelength for processing of silicon with fs- and ps-pulses. The modelling and experimental results were obtained at low NA values of 0.125–0.3, and it became clear that using a large focusing angle is necessary to avoid nonlinear absorption too close to the surface, as also mentioned by Zavedeev et al. [15].

In earlier theoretical works Zavedeev et al. [11, 15] it was shown that for a numerical aperture of 0.3 and a pulse duration of 250 fs the maximum deposited energy density reaches its maximum value at around 2000 nm and then, after dropping down, starts to grow with the wavelength again. This is in contrast with the maximum plasma density, which decreases with λ nearly monotonically. Moreover, Kononenko et al. [29] showed that an increase of the pulse energy not necessarily will increase the intensity at the focal spot due to multi-photon absorption, plasma defocusing and reflection. All this suggested the need of careful control of the laser parameters, including wavelength, to make use of the strong wavelength dependence of the material nonlinearity, to achieve a particularly precise single pulse shot processing.

Besides the wavelength, energy and pulse duration are equally important, as experimentally shown by Das et al. [18], Mareev et al. [19]. They demonstrate that a certain pulse duration is necessary to create sub-surface modifications within Si depending on the focusing angle. Das et al. [18] gives a minimum pulse duration of 5.4 ps for a focusing numerical aperture of 0.85 to induce modifications within Si when operating a laser with λ ≈ 1.55 μm. For pulses between 5.4 to 21 ps no increase of the modification threshold could be found [18], except when increasing the pulse duration to nanoseconds. For shorter pulse durations in the femtosecond range Mareev et al. [19] could show a decreasing amount of generated free carriers [19] when keeping the wavelength, pulse energy and focusing optics constant, indicating higher losses while propagating through Si for shorter pulse durations. This was experimentally confirmed by Chambonneau et al. [30].

To the best of our knowledge Zavedeev et al. [15] was the first to have investigated numerically the behavior of femto-second pulses propagating through Si at wavelengths ≫ 2000 nm. They stated that the pre-focal depletion of the pulse energy decreases significantly with wavelength, and that the maximum deposited energy density and the plasma defocusing decreases. The decrease in the free-carrier concentration does not cause a similar decrease in the density of the deposited energy, because the inverse bremsstrahlung absorption becomes stronger with longer wavelengths. In spite of all these advantages of going to longer wavelengths they did not expect a significant improvement in femto-second laser processing when going above 5 μm wavelength at an NA of 0.3.

In the last few years large progress has been achieved in development of ultrashort pulsed high energy lasers operating above 2000 nm. Three laser systems could be of potential interest in this wavelength region: a picosecond-fiber Ho:Yb:MOPA laser, operating at up to 10 mJ at 10 kHz repetition rates [21], a femtosecond Cr:ZnS [31] or Cr:ZnSe laser with up to 2 mJ pulse energy [32–34], and an Er:ZBLAN fiber laser operating around 3 μm, delivering pulse durations down to 100 to 200 fs [35–37]. The availability of commercial lasers in this wavelength range calls for a detailed numerical analysis that might be particularly important for practical applications of these lasers.

### A. Mechanisms of sub-surface modifications in silicon

Several mechanisms for the modification of the crystal structure of silicon under irradiation with short and ultra-short pulses have already been described. Depending on the pulse parameters and the chosen model each approach relies on different mechanisms, time scales and different thresholds. Below the most relevant approaches for the investigated pulse parameter range are given:

1. So-called “non-thermal melting” is proposed when irradiating silicon with ultra-short pulses [38–44]. By exciting more than ≈ 5 to 10 % of the valence electrons in the conduction band, corresponding to a plasma density of ≈ 5 × 10^{21} to 10 × 10^{21} cm^{-3}) the bonds between the atoms are weakened enough to enable a process which resembles melting, but without reaching the lattice temperatures associated with melting. This process is independent of the wavelength, after it only relies on the density of excited electrons.

2. Following Du et al. [45], B.C et al. [46], Du et al. [47], Tien et al. [48], Chimier et al. [49] reaching the critical plasma density in a material, defined as n_{c} = [(\varepsilon_{0} · m_{e}) / e^{2}] · ω^{2} is sufficient for reaching the modification/damage threshold in a material. This criterion is wavelength-dependent, and scales with λ^{-2}. Therefore, it is easier to reach the modification threshold with this definition using longer wavelengths compared to shorter wavelengths.

3. Following Chambonneau et al. [7], Chimier et al. [49], Zhokhov and Zheltikov [50], Werner et al. [51] reaching the threshold given in point 2 is necessary
for modifying the material, but not necessarily sufficient. Instead, they propose to relate to the deposited energy within the material when determining if the material will be modified. For ultra-short pulses it can be assumed that the energy initially is retained only within the electrons during the full pulse duration, and only afterwards will be transferred into the lattice. Therefore, as upper limit for the absorbed energy Equation (1) can be used.

\[ E_{\text{max}} = 2E_{\text{g}}\varrho_{\text{nt}}. \]  

Here, \( E_{\text{g}} \) gives the band gap, and \( \varrho_{\text{nt}} \) the generated free-carrier density. Other approaches for determining the threshold (such as the Gamaly-model [52]) are described by Werner et al. [51].

B. Applied methods and procedure

In this work we focus on the theoretical study of the spatio-temporal effects of the propagation of the ultra-short pulses inside silicon at different wavelengths above 1100 nm. These effects are a result of the interplay of nonlinear optical effects and have a strong wavelength dependence.

To numerically investigate numerical apertures above 0.2 a non-paraxial pulse propagation method has to be used, the paraxial approximation is not sufficient for numerical apertures larger than that [53]. For this purpose two different methods are used in this paper, the unidirectional pulse propagation [54] and the generalized Helmholtz equation [55]. While the former approach is a spectral technique using the Fourier split-step method for time stepping, the latter is a purely temporal technique, and is implemented and solved with a finite-element-method (FEM). This allows the generalized Helmholtz approach to better resolve spatial effects at the cost of neglecting spectral effects.

We use numerical simulations to investigate the influence of the wavelength dependence of nonlinear absorption, self-focusing and plasma-related effects for wavelengths between 1550 to 3250 nm for pulse durations between 100 to 5000 fs and compare the influence of the numerical aperture. Moreover, we also investigate the spatio-temporal behaviour of the pulse and demonstrate the significant influence of the generated plasma on the pulse shape while propagating through the material, and at the focal spot.

We apply the methods of the spatio-temporal structures and dynamics control in the nonlinear media under the condition of strong field localization. Our observations demonstrate a possibility of spatio-temporal control like the nonlinear mode-self-cleaning in the multimode fibers [56, 57] that bridges nonlinear fiber- and solid-state laser photonics. Manipulation with highly concentrated spatio-temporal patterns provides the tools for high-energy physics and technology in photonics, information processing, condensed matter modification and others.

II. NUMERICAL MODEL AND PHYSICAL PARAMETERS

A. Physical nonlinear parameters

A light pulse propagating through Silicon is influenced by several different wavelength-dependent nonlinear phenomena, such as the Kerr-effect, multi-photon absorption or plasma absorption and -defocusing.

Within the investigated wavelength range (between 1100 nm and 2500 nm) only two- and three-photon absorption can be seen as relevant, after single-photon absorption can be neglected for wavelengths above 1100 nm, and four-photon absorption can be neglected for wavelengths \( \leq 3300 \text{ nm} \). A graph displaying the values for the two-photon absorption (based on Bristow et al. [58] is shown in Figure 1 in the upper figure in blue, and the values for the three-photon absorption (based on Pearl et al. [59]) are shown in the same figure in orange.

The exact values for the wavelengths (as given in Section II) investigated in this paper are shown in Table I, together with the corresponding sources.

The strength of the Kerr-effect can vary depending on the doping and orientation of the silicon wafer, and therefore we chose to include two different values as an upper and lower boundary. The wavelength-dependent values are shown in the lower figure in Figure 1 for low \( n_2 \)-values (based on Lin et al. [27]) in red and in green for high \( n_2 \)-values (based on Wang et al. [28]). The main difference between the used materials in Lin et al. [27], Wang et al. [28] is the different doping of the material, which in turn results in a different nonlinear refractive index. Wang et al. [28] uses a p-doped material with a doping concentration of \( 1 \times 10^{15} \text{ cm}^{-3} \), which corresponds to a surface resistivity of \( \approx 13.5 \Omega \text{ cm} \). Lin et al. [27] uses a similar p-doped crystal, but with a surface resistivity of \( 20 \Omega \text{ cm} \), which corresponds to a dopant concentration of \( \approx 6.7 \times 10^{14} \text{ cm}^{-3} \). Therefore, a lower dopant concentration in a p-doped crystal corresponds to a lower nonlinear refractive index.

Those parameters can be combined in the so-called Nonlinear Figure of Merit (NFOM). It usually is defined as the relative merit of the Kerr nonlinearity versus the two-photon nonlinearity by the ratio of both values, divided by the optical wavelength in vacuum, according to Mizrahi et al. [62]. When extending this ratio to multi-photon absorption, one obtains the NFoM in the following form [28]:

\[ \text{NFOM} = \begin{cases} \frac{n_2}{\lambda^2 \beta^2} & \forall \lambda \leq 2200 \text{ nm} \\ \frac{n_2}{\lambda^2 \beta^2} & \forall \lambda > 2200 \text{ nm} \end{cases} \]
with $\lambda$ the wavelength in vacuum, $n_2(\lambda)$ the wavelength-dependent nonlinear refractive index, $I$ the field intensity and $\beta^{(K)}$ the coefficient for $K$-photon absorption.

As shown in Figure 1 it has to be kept in mind that the three-photon absorption coefficient does not decrease to zero for wavelengths below 2200 nm. It still can influence the laser light, provided a certain intensity is reached. This intensity can be determined by comparing the contribution of both two- and three-photon absorption and setting

$$\beta^{(2)} \approx \beta^{(3)} I.$$  

(3)

For 1550 nm this threshold is reached at $I_{1550\text{ nm}} = 2.8\text{ PW m}^{-2}$, while for 2150 nm the threshold can be calculated to be $I_{2150\text{ nm}} = 10\text{ TW m}^{-2}$, and thus significantly lower. When no absorption is intended (such as in waveguides) the intensity typically stays below 10 TW m$^{-2}$ [63], and therefore three-photon absorption can be neglected for shorter wavelengths. For those intensities Equation (2) therefore stays valid.

For processing the absorption, on the contrary, the localized absorption of laser pulses is the main aim, and therefore the intensities will be much higher at least close to the focal spot. At those intensities three-photon absorption will still be an important factor at wavelengths shorter than 2200 nm, and therefore we propose a revised approach for the nonlinear figure of merit for large intensities:

$$NFOM_{rev}(\lambda, I_0) = \frac{n_2(\lambda)}{\lambda(\beta^{(2)} + \beta^{(3)} I_0)}.$$  

(4)

Similarly to three-photon absorption higher-order absorption processes will be present at shorter wavelengths, too. Therefore, it can be beneficial to verify if those processes have to be included in the revised nonlinear figure of merit, and potentially forming the following definition:

\begin{table}[h]
\centering
\begin{tabular}{|c|c|c|c|c|}
\hline
Wavelength & Two-photon & Three-photon & Four-photon & Nonlinear refractive & Refractive
\begin{tabular}{c}
absorption \ coefficient [58] \end{tabular} & \begin{tabular}{c}
absorption \ coefficient [59] \end{tabular} & \begin{tabular}{c}
absorption \ coefficient [60] \end{tabular} & \begin{tabular}{c}
index [28] \end{tabular} & \begin{tabular}{c}
index [61] \end{tabular} \\
\hline
1550 nm & $1.313 \times 10^{-11} \text{ m W}^{-1}$ & $4.711 \times 10^{-27} \text{ m}^3 \text{ W}^{-2}$ & $0 \text{ m}^2 / \text{ W}^3$ & $5.75 \times 10^{-18} \text{ m}^2 \text{ W}^{-1}$ & 3.4757 \\
1950 nm & $3.361 \times 10^{-12} \text{ m W}^{-1}$ & $1.403 \times 10^{-26} \text{ m}^3 \text{ W}^{-2}$ & $0 \text{ m}^2 / \text{ W}^3$ & $1.475 \times 10^{-17} \text{ m}^2 \text{ W}^{-1}$ & 3.453 \\
2150 nm & $2.030 \times 10^{-13} \text{ m W}^{-1}$ & $2.020 \times 10^{-26} \text{ m}^3 \text{ W}^{-2}$ & $0 \text{ m}^2 / \text{ W}^3$ & $1.375 \times 10^{-17} \text{ m}^2 \text{ W}^{-1}$ & 3.446 \\
2550 nm & $0 \text{ m W}^{-1}$ & $2.860 \times 10^{-26} \text{ m}^3 \text{ W}^{-2}$ & $0 \text{ m}^2 / \text{ W}^3$ & $2.25 \times 10^{-18} \text{ m}^2 \text{ W}^{-1}$ & 3.437 \\
3250 nm & $0 \text{ m W}^{-1}$ & $9.366 \times 10^{-28} \text{ m}^3 \text{ W}^{-2}$ & $1 \times 10^{-41} \text{ m}^5 / \text{ W}^3$ & $2 \times 10^{-18} \text{ m}^2 \text{ W}^{-1}$ & 3.428 \\
\hline
\end{tabular}
\caption{Physical parameter values for nonlinear interactions acting on light propagating through Si at the example wavelengths. The data is based on Figure 1 with the corresponding literature.}
\end{table}
NFOM_{revHigh}(\lambda, I_0) = \frac{n_2(\lambda)}{\lambda \sum_{K=2}^{\infty} (\beta(K)I_0^{K-2})}.

(5)

The calculation of the threshold intensity at which higher-order multi-photon coefficients will be of interest is complicated by sparse availability of measurement data for higher-order absorption coefficients in Si over a large range of wavelengths. Gai et al. [60] provides several measurement points for the four-, five- and six-photon absorption coefficients, but only within a limited wavelength range (unlike the data given for the three-photon absorption coefficient). The lowest wavelength where the four-photon absorption coefficient has been measured is 3250 nm [60], with a value of (1.00 ± 0.25) × 10^{-41} m^7/W^3. Compared to the three-photon absorption coefficients at 2150 nm and 2550 nm a threshold intensity can be calculated (by following Equation (3)) at \(I_0 \geq 2\) PW m\(^{-2}\). Together with the decrease of the four-photon absorption coefficient for shorter wavelengths (i.e. \(\lambda \leq 3250\) nm) it therefore can be safely assumed that for intensities below 2 PW m\(^{-2}\) and above 10 TW m\(^{-2}\) Equation (4) can be used, and replaces Equation (2) as the definition of the nonlinear figure of merit. Graphs based on Equations (2) and (4) showing the change of the nonlinear figure of merit for increasing intensities are given in Figure 2. Following the estimations made above the transition from Equation (2) to Equation (4) is at \(I_0 \geq \frac{1}{10} \times 10^{13}\) W m\(^{-2}\). The values for the nonlinear figure of merit depending on the intensity and wavelength are shown in Figure 2, with a cross-section at \(I_0 = \frac{1}{10} \times 10^{13}\) W m\(^{-2}\) shown in Figure 3. It has a clearly visible peak at \(\approx 2200\) nm due to the transition from two-photon absorption to three-photon absorption combined with the maximal Kerr-effect contribution.

![FIG. 2: Nonlinear figure of merit (NFOM) as defined by Equation (5), drawn against different intensity- and wavelength values](image)

In Figure 2 it can be seen that the three-photon absorption combined with the intensity gradually increases, which shifts the peak of the nonlinear figure of merit towards the shorter wavelengths (up to 1950 nm for 1 PW m\(^{-2}\)) while simultaneously lowering the whole nonlinear figure of merit. This indicates the growing importance of higher-order absorption compared to lower-order absorption due to the scaling with the intensity, a relation which also is displayed in Figure 4.

**B. Numerical model and methods**

To model the propagation of the pulse through Silicon we use the scalar unidirectional pulse propagation equation (UPPE) [54, 64], written as

\[
\partial_z \hat{E}(\omega, z) = i k_z \hat{E}(\omega, z) + \frac{i \omega^2}{2 \sigma_0 c^2 k_z} \hat{P}(\omega, z) - \frac{\omega}{2 \sigma_0 c^2 k_z} \hat{J}(\omega, z),
\]

(6)

with \(\hat{E}\) the Fourier-transformed electric field of the pulse, \(P\) the (non-)linear polarizations acting on the pulse and \(J\) the (non-)linear current densities acting on the propagating pulse. The wave number is given by \(\omega\) and the propagation constant \(k_z\) in a cylindrical coordinate system is defined as (based on Fedorov et al. [53], Couairon et al. [65])

\[
k_z = k_0 - \frac{k_0^2}{2k_0},
\]

(7)

to allow the calculation of tightly focusing optics with an NA \(\geq 0.2\) while still using a scalar equation.

The main advantage of the UPPE compared to the nonlinear Schrödinger equation (NLSE) for simulating the nonlinear propagation of a light pulse in Silicon is the inclusion of non-paraxial effects. This allows the numerical simulation of optics with a very large numerical aperture.

One has to note, that Equations (6) and (7) correspond to the generalised Helmholtz equation written in a co-moving coordinate system (\(z = z, t = T - z/u\)) \((t\) is a local time, \(T\) is a time in the laboratory coordinate system, \(u\) is a group velocity of a pulse, and \(r\) is a radial coordinate within the axial symmetry approximation) [55]:
The transverse Laplacian in Equation (8) reads as
$$\nabla^2_\perp \rightarrow \frac{\partial^2}{\partial r^2} + (1/r) \frac{\partial}{\partial r},$$
where $r$ is a radial coordinate.

The contribution $\Xi(E(t,z))$ to the nonlinear polarization corresponds to represented by the terms in eq. (6). That are defined by the Kerr-effect, described in section II.A, while the nonlinear multi-photon absorption (two- and three-photon absorption) and the plasma absorption and -defocusing are part of the nonlinear currents.

The UPPE-equation (c.f. Equations (6) and (7)) is a very effective tool for simulations of field dynamics outside the paraxial approximation. Using the split-step Fourier approach could promise an acceleration of computation. However, the interweaving of spatial and space degrees of freedom requires the evaluation of big-size matrices, which is a memory-intensive task. The generalized Helmholtz-equation (Equation (8)) is evaluated using (from Couairon et al. [64])

$$\frac{\partial^2}{\partial r^2} + \left(\frac{1}{r}\right) \frac{\partial}{\partial r} E(t,z,r) = \Xi(E(t,z,r)). \quad (8)$$

The intensity $I$ is defined as $I = \frac{\omega_0^2}{2} |E|^2$, and $E_0$ is the band gap energy. $\tau_c$ in Equation (12) is the electron collision time. Finally, $\sigma_K$ in Equation (10) is given as

$$\sigma_K = \frac{\beta(K)}{\overline{\epsilon} \omega_0^2 \theta_{nt}} \quad \text{[64]}, \quad (13)$$

with $\theta_{nt}$ the neutral free carrier density.

III. NUMERICAL SIMULATIONS

Initial investigations of the wavelength dependence of the pulse propagation in silicon was already done before, as for example by Richter et al. [16, 21, 74], but those investigations were limited by the used method for numerical simulations. Richter et al. [16, 21] used the NLSE for simulating the pulse propagation, which limits the maximum focusing angle which can be used. On the other hand Das et al. [18] reported experimentally for $\lambda = 1550$ nm that an NA value of $\geq 0.85$ has to be used for pulse durations of $\approx 5$ ps, which limits the applicability of the NLSE for those pulse durations. Nevertheless, at least for lower focusing angles it could be shown that between 2000 to 2200 nm the efficiency of the energy delivery to the focal spot was increased compared to longer and shorter wavelengths.

To investigate and correlate the experimentally observed behavior with numerical simulations we chose to simulate a pulse at four different wavelengths, 1550 nm, 1950 nm, 2150 nm and 2350 nm with the pulse energies ranging from 0.001 $\mu$J to 1 $\mu$J. A full overview over all parameters is given in Table II. Here, the maximum pulse energy for an NA of 0.25 is 500 $\mu$J, while the maximum pulse energy for an NA of 0.85 is 5000 $\mu$J.

For all calculations with the UPPE the initial beam diameter was set to $d_0 = 2$ mm for a wavelength of 1550 nm. To achieve a similar fluence in the focal spot for linear propagation, the beam radius $r_\lambda$ for other wavelengths was adjusted using

$$r_\lambda = d_0 \frac{\lambda}{1550 \text{nm}} \quad \text{[16, 21, 74]}, \quad (14)$$

unless explicitly noted otherwise.

While this will result in a similar fluence level at the focal spot independently of the wavelength, it will result in changed focusing angles. This in turn will influence the impact of the nonlinear effects onto the propagation...
TABLE II: Overview over the used pulse- and lens parameters in the numerical simulations

| Parameter                  | Value      | Unit    |
|----------------------------|------------|---------|
| Pulse duration $t_0$       | 100, 500, 1000, 5000 fs |         |
| Pulse wavelength $\lambda_0$ | 1550, 1950, 2000, 2150, 2550, 3250 nm |         |
| Pulse energy $E_0$         | 1, 10, 62.5, 100, 500, 1000, 5000 nJ |         |
| Numerical aperture at 1550 nm | 0.1, 0.25, 0.85 |         |
| Beam radius at 1550 nm     | 1 mm       |         |
| Focus depth 100, 250, 500, 700 µm |   |         |

The numerical apertures used for the HHE are listed in Table IV, in comparison.

TABLE III: NA-values used for the wavelengths displayed in Table II when using Equation (6) as pulse propagation equation, following Equation (14), with an initial beam diameter of 2 mm

| Wavelength | Target NA: 0.25 | Target NA: 0.85 |
|------------|-----------------|-----------------|
| 1550 nm    | 0.25            | 0.85            |
| 1950 nm    | 0.31            | 1.07            |
| 2150 nm    | 0.35            | 1.18            |
| 2550 nm    | 0.41            | 1.40            |
| 3250 nm    | 0.52            | 1.78            |

As displayed in this figure the total absorption due to multi-photon absorption scales with the intensity. Depending on the order of the multi-photon absorption this scaling factor is either purely $I^2$, $I^3$ or a combination of both. A higher scaling factor can lead to a higher absorption current, even though the absorption coefficient itself is lower. This effect can be seen especially for 2550 nm. While the generated current for an intensity of $1 \times 10^9$ W m$^{-2}$ is five orders of magnitude lower than the current generated by 2150 nm, it is equal for an intensity of $\approx 3 \times 10^{13}$ W m$^{-2}$, and higher than that for increasing intensities. These intensities are roughly comparable to the intensities reached by a pulse with an energy of 1 nJ and a duration of 100 fs at a wavelength of 2150 nm, focused with a lens with $f = 4$ mm and a beam diameter of 1 mm. This gives a focal spot area of $\geq 90$ µm$^2$, and with a peak power (for a Gaussian pulse) of 8.8 kW the peak intensity is $\approx 9.4 \times 10^{13}$ W m$^{-2}$, i.e. an intensity which can be easily achieved.

Similarly, the transition intensity for the shorter wavelengths can be taken from Figure 4, and can be approxi-
mated to be $\approx 1 \times 10^{15}$ W m$^{-2}$, corresponding to a pulse with $\approx 10$ nJ and the parameters used above. Nevertheless, one has to keep in mind that the intensity at the focal spot was taken into account here. When focusing deep into the material the affected area increases, which in turn decreases the intensity. Setting the focal depth to 500 µm in Si, corresponding to $\approx 144$ µm in air, will increase the affected area by a factor of $\approx 11$, and reduce the intensity accordingly. Decreasing the focal length of the lens (and thereby increasing the numerical aperture) will affect the intensity. Going from an NA of 0.125 as used above to an NA of 0.85 (which is typically used for processing experiments [18, 21]) by decreasing $f$ from 4 mm to 0.58 mm will increase the affected area by $\approx 130$ and lower the intensity accordingly. This is especially important for longer wavelengths, after the total absorption for higher-order multi-photon absorption processes scales faster with the intensity.

B. Influence of the non-paraxiality on the pulse shape

To demonstrate the importance of using methods which do not rely on the paraxial approximation for propagating the laser pulse a pulse was simulated using Equation (8), once while including the paraxial approximation, and once without, while keeping the pulse parameters constant. The pulse structure at the same position within silicon is shown in Figure 5(a) for the case of included paraxial approximation, and in Figure 5(b) for the case without approximation.

In comparison a clear difference is visible. While the paraxial approach results in an elongated structure, the non-paraxial approach shows a clear intensity dip in the center of the pulse. Despite a relatively small NA Figure 5, the non-paraxial approximation addresses more precisely the nonlinear factors causing the spatial-time reshaping of a beam.

C. Spatio-temporal structure and impact of spherical aberrations

While propagating through the material the pulse changes its shape both in space and in time. This is visualized in Figures 7(a) to 7(d). These figures demonstrate the spatio-temporal evolution of a light pulse with $t_0 = 100$ fs, $E_0 = 100$ nJ and a wavelength of $\lambda = 1550$ nm, focused into Si with a lens with NA = 0.25. The focal spot was placed at 500 µm in air, corresponding to 1736 µm in Si due to the increased refractive index.

Simulations based on both Equations (6) and (8) demonstrate the appearance of spherical aberrations (c.f. Figure 6(a)). A spatial substructure appears in the form of “light rings”, which evolves to $r = 0$ µm and squeezes with a distance due to self-focusing (c.f. Figure 6(b)).

Several points can be noted about the pulse behavior

FIG. 5: Contour plots of intensity demonstrating the time profile asymmetry due to plasma generation. The pulse parameters are (following Equation (A5)) $P_0 = 0.5$ MW, $f = 700$ µm, NA = 0.1, $w_0 = 6.4$ µm, $\lambda = 2000$ nm, and $t_0 = 250$ fs. The displayed field profiles are taken in a depth of 670 µm below the surface.

FIG. 6: Contour plots of “light rings” in the intensity (spherical aberrations) in dependence on the propagation distance inside the medium. The pulse parameters are (following Equation (A5)) $P_0 = 10$ MW, $f = 500$ µm, NA = 0.25, $w_0 = 2$ µm, $\lambda = 1550$ nm, and $t_0 = 100$ fs.
Figure 7: Spatio-temporal intensity field distribution of a pulse with $\lambda = 1550\,\text{nm}$, $E_0 = 100\,\text{nJ}$ and a duration of 100 fs. The pulse is focused using a lens with $f = 500\,\text{µm}$ and a numerical aperture of 0.25.

Figure 7(b) shows the pulse structure shortly before the focal spot at $z = 500\,\text{µm}$, Figure 7(c) shortly after the focal spot, and Figure 7(d) far away from the focal spot. The effect of self-focusing is clearly visible from Figure 7(a) to Figure 7(b), followed by nonlinear absorption and plasma-induced defocusing after the focal spot.

Already before the focal spot. Going from $423\,\text{µm}$ (Figure 7(a)) to $454\,\text{µm}$ (Figure 7(b)) not only the pulse in total is focused towards the focal spot, but also the effect of self-focusing is visible. The innermost part of the pulse with the highest intensity is focused more compared to the rest of the pulse, creating an asymmetric shape along the spatial axis. Nevertheless, the pulse stays symmetric along the time axis.

This effect can be considered as analogue to the Kerr-beam self-cleaning in multi-mode fibers, as demonstrated by Krupa et al. [57]. The calculations demonstrate that the NA growth enhances the tendency for generating multiple rings. Still, this effect degrades quickly at the wavelength close to the maximum of NFOM and longer wavelengths.

Shortly before the focusing point, though, the self-focusing effect together with the generated plasma leads to a deformation of the beam not only in space, but also in time. As also shown below in Figures 14(a) to 14(d), the leading part of the pulse generates a large amount of free carriers, which lead to diffraction and multi-photon absorption. Due to the multi-photon absorption scaling nonlinearly with the intensity this affects the central part of the pulse dis-proportionally more compared to the leading and trailing edge. Generated free carriers also lead to absorption, but only linearly, and therefore can be neglected for high intensities. Still, for a suffi-
cient density the absorption due to free carriers is visible, and can be seen for the trailing part of the pulse in Figures 7(c) and 7(d). For this part the intensity is reduced by \( \approx 50\% \) compared to the leading edge.

Figure 5, based on Equation (8), demonstrates the effect of time asymmetry in the vicinity of the first maximum of the NFOM (c.f. Figures 2 and 3) at \( \lambda = 2000 \text{ nm} \). This effect is connected with the plasma generated by the pulse front that initiates the pulse “explosion” (c.f. Figure 8).

(a) Temporal field distribution of the pulse at different positions inside the material.

(b) Contour plots and time dependence of plasma density at the beam axis, corresponding to Figure 8(a).

D. Impact of the numerical aperture (NA) and focusing depth on the nonlinear pulse propagation

Changing the physical position of the lens itself (i.e. shifting the depth of the focal spot inside the material) and the parameters of the lens itself (i.e. changing the numerical aperture) influences how the beam propagates inside the material, and how the different nonlinear ma-

(c) Intensity evolution.

(d) Density evolution.

FIG. 8: Contour plots of the intensity evolution, demonstrating the time profile asymmetry and pulse explosion (Figure 8(a)) due to plasma generation (corresponding contour plots and time dependence of plasma density at the beam axis are shown in Figure 8(b)). The intensity and the plasma density evolution are illustrated in Figures 8(c) and 8(d), respectively. The parameters are the same as in Figure 6.

When increasing the numerical aperture from 0.25 to 0.85 the initial intensity along the propagation path is lower, and therefore nonlinear effects will start closer to the focal spot. Therefore, almost no difference is visible material parameters can act on the propagating beam. For a varying depth and a fixed numerical aperture the difference is visible for a numerical aperture of 0.25, as shown in Figure 9.

Moving the focal spot deeper into the material will change the intensity distribution along the propagation path, such that nonlinear effects, especially self-focusing, can act on the beam for a longer period, and therefore change the resulting intensity field distribution at the focal spot. This behavior is visible especially for \( \lambda = 1950 \text{ nm}, 2150 \text{ nm}, \) after those wavelengths experience a stronger self-focusing compared to shorter and longer wavelengths (as shown in Figure 1). This effect is visible especially along the beam propagation axis, as shown in Figure 14.
in the structure of the intensity field distribution for an NA of 0.85 is visible (c.f. Figures 14(c) and 14(d)), while the pulse focused with a lower NA value changes its structure when changing the focal depth. This demonstrates that larger focusing angles are necessary for processing closer to the surface, where the interaction length with nonlinear effects will be reduced around the focal spot compared to lower focusing angles.

E. Spatio-temporal pulse behavior depending on pulse wavelength and -energy

The spatio-temporal splitting described in Section III C and shown in Figures 7(a) to 7(d) does depend on several different factors such as pulse wavelength, pulse energy, pulse duration and the focusing optics. While the first parameter influences the value of the material parameters (as seen in Table I), the latter three influence how the intensity of the pulse will behave while travelling through the material. A higher intensity will result in both earlier self-focusing and a faster onset of multi-photon absorption, which in turn are both influenced by the value of the material parameters set by the wavelength. As a first comparison the intensity of a pulse with \( t_0 = 100 \text{ fs} \) and \( E_0 = 100 \text{ nJ} \) directly at the focal spot \( z = 100 \mu\text{m} \) for Figures 9(a) to 9(d) and \( z = 250 \mu\text{m} \) for Figures 9(e) to 9(l)). Small variations in the field distribution are visible for 1950 nm (c.f. Figures 9(b) and 9(f)) and 2150 nm (c.f. Figures 9(c) and 9(g)). The intensity field distribution for 1550 nm and 2550 nm does not change, in comparison. In Figures 9(i) to 9(l) the intensity field for an NA of 0.85 is displayed in comparison, with similar parameters as Figures 9(e) to 9(h). Unlike as for a numerical aperture of 0.25 the field distribution for a numerical aperture of 0.85 does not change significantly when decreasing the focal depth from 250 \( \mu\text{m} \) to 100 \( \mu\text{m} \), and therefore was omitted.

FIG. 9: Electric field (Intensity) for \( t_0 = 100 \text{ fs}, f_{\text{inside}} = 100 \mu\text{m}, E_0 = 100 \text{ nJ} \) directly at the focal spot \( z = 100 \mu\text{m} \) for Figures 9(a) to 9(d) and \( z = 250 \mu\text{m} \) for Figures 9(e) to 9(l)). Small variations in the field distribution are visible for 1950 nm (c.f. Figures 9(b) and 9(f)) and 2150 nm (c.f. Figures 9(c) and 9(g)). The intensity field distribution for 1550 nm and 2550 nm does not change, in comparison. In Figures 9(i) to 9(l) the intensity field for an NA of 0.85 is displayed in comparison, with similar parameters as Figures 9(e) to 9(h). Unlike as for a numerical aperture of 0.25 the field distribution for a numerical aperture of 0.85 does not change significantly when decreasing the focal depth from 250 \( \mu\text{m} \) to 100 \( \mu\text{m} \), and therefore was omitted.
of 250 µm.

The effect of multi-photon absorption and plasma diffraction can clearly be seen for all pulse wavelengths. The trailing part of the pulse has been absorbed almost completely, resulting in a shift of the peak intensity towards the leading edge. In addition, the self-focusing effect introduces oscillations, especially for 1950 nm and 2150 nm.

Here, it is also worth to mention the intensity distribution over the wavelengths at the focal spot. For the linear propagation without absorption or diffraction, the intensities should be equal at the focal spot, but even though the self-focusing effect is stronger at 1950 nm and 2150 nm compared to the shorter and longer wavelengths, the peak intensity follows the absorption curve shown in Figure 4 at \( I_0 \geq 1 \times 10^{15} \text{W m}^{-2} \), with the shortest wavelength having the highest peak intensity. Nevertheless, the difference between the peak intensities is negligible, with the peak intensity at 2550 nm only \( \approx 1.7 \) times smaller than the peak intensity at 1550 nm. In comparison to this difference in intensity, the density of the generated free carriers is roughly comparable for all wavelengths independently of the depth of the focus point (as shown in Figures 15(b) and 15(d)), with the generated density being slightly higher for 2550 nm by \( \approx 8\% \). This indicates that the absorbed energy going into the generation of free carriers is roughly the same, independently of the wavelength.

In comparison to a lens with an NA of 0.85 one also notices that the field is longer in time (especially visible in the slices shown in Figures 14(a) to 14(d)). For a numerical aperture of 0.25 the pulse is stretched to over \( \approx 800 \) fs, while for a numerical aperture of 0.85 the pulse is only stretched to 100 to 200 fs, depending on the observed wavelength, and thereby much closer to the original pulse duration.

For long wavelengths, the spherical aberrations decay so that a field concentrates in a comparatively symmetrical spot at some propagation distance inside a medium (see Figure 10). As one can see from Figures 1 and 2, the two-photon absorption process decreases towards this wavelength range so that a relative contribution of the Kerr-effect increases. Thus, a plasma influence on a beam focusing destabilization and a damage threshold decays. As a result, a non-destructive modification of material becomes possible. One must note that the weakening of plasma contribution breaks the spatial-time relation mentioned above. Namely, a beam squeezes mainly in spatial dimension than in the time one for the large NA (compare Figure 10). Our analysis demonstrates that such an effect could cause a spatial-time splitting at some propagation distances, which requires control of initial beam parameters.

F. Correlation between the pulse duration, the pulse wavelength, the pulse energy and the peak intensity/peak free carrier density/absorbed energy

As shown in Figure 4 the absorption of the pulse largely depends on the pulse intensity. To reduce the intensity of the pulse there are several options: Increasing the focal spot size, decreasing the pulse energy or increasing the pulse duration. With the size of the modifications in Si as the main goal the first option is not ideal, and therefore either the pulse energy has to be decreased, or the pulse duration increased. The effectiveness of both approaches is compared in Figure 11.

The corresponding fields (for 1550 nm) are shown in Figure 11. Several effects can be seen here, and are worth mentioning:

- A longer pulse looses a larger amount of energy, leading to the larger shift of the intensity peak towards the leading edge (c.f. Figures 11(a) and 11(b))

- While the pulse with \( \lambda = 1550 \) nm generates the highest intensity at the focal spot for the pulses with \( t_0 = 100 \) fs (c.f. Figures 11(a), 14(c) and 14(d)). For longer pulse durations (\( \geq 1000 \) fs) higher intensities will be generated at longer wavelengths.

- Even though the pulse energy is one order of magnitude lower in Figure 11(a) than in Figure 14(c)
the peak intensity value is approximately equal, indicating intensity clamping due to high absorption and diffraction.

- The generated plasma density for a pulse with \( t_0 = 1000 \text{ fs} \) is 50 % larger compared to a pulse with \( t_0 = 100 \text{ fs} \), as shown in Figures 11(e) and 15(b), if both pulses have an energy of \( E_0 = 100 \text{ nJ} \). This indicates a better energy deposition of the pulse energy into the material for longer pulses.

- While the pulse with \( \lambda = 2550 \text{ nm} \) at \( t_0 = 100 \text{ fs} \) could generate the highest free-carrier density, this role is taken over by the pulse with \( \lambda = 1950 \text{ nm} \) for \( t_0 = 1000 \text{ fs} \), with the pulses at \( \lambda = 2150 \text{ nm} \) and \( \lambda = 2550 \text{ nm} \) on second position.

This indicates that pulses longer than 100 fs are necessary to efficiently transfer energy from the pulse into the material. For a pulse duration of 1000 fs exemplary results are displayed in Figures 11(b) and 11(e), and for a pulse duration of 5000 fs those results are displayed in Figures 11(c) and 11(f). Several major differences can be noted for a pulse duration of 5000 fs in comparison with the data in Figure 11(b) (for \( t_0 = 1000 \text{ fs} \)):

- The pulses at \( \lambda = 1950 \text{ nm} \) and \( \lambda = 2150 \text{ nm} \) now share the peak intensity value, compared with the pulses with a shorter and longer wavelength

- The peak intensity for \( t_0 = 5000 \text{ fs} \) is decreased by \( \approx 65 \% \) compared to \( t_0 = 1000 \text{ fs} \)

- The peak density decreased by \( \approx 20 \% \) compared to \( t_0 = 1000 \text{ fs} \), but is still higher by \( \approx 25 \% \) compared to a pulse duration of 100 fs.

As it can be seen in Figures 11(a) to 11(f) and 14 the peak intensity and the carrier density vary depending on pulse duration, pulse energy and pulse wavelength if the focal depth and focusing angle is kept at a constant value. Following those graphs for a focusing depth of 100 \( \mu \text{m} \) and a numerical aperture of 0.85/1.07/1.18/1.40 and a focus depth of 100 \( \mu \text{m} \),

The results shown in the figures above (Figures 11(d) and 11(f)) indicate that there is an optimum of the pulse...
and ranging between $2 \times 10^5$ to $6 \times 10^6 \text{ m}^{-3}$, one obtains absorbed energies between $\approx 72$ to $215 \text{ J cm}^{-3}$. Absorbing these energies will lead to a temperature increase of $\approx 44$ to $130 \text{ K}$. This is not sufficient for increasing the temperature of Si from room temperature to the melting temperature, but sufficient to expand the heated spot slightly (with $\Delta V/V = 9 \times 10^{-6} \text{ K}^{-1}$ [75]). This local expansion can lead to a permanent local change of the refractive index (even without melting), which then in turn can be detected with optical methods.

This direct correlation between the generated free carrier density and the resulting change in temperature also confirms that the modification threshold does not only depend on the wavelength, but rather on the interplay between several different pulse parameters including the pulse wavelength. Especially for longer pulse durations and large focusing angles several different wavelengths can achieve approximately the same energy deposit (c.f. Figures 11(d) to 11(f) and 15(a) to 15(d)). In that case pulse parameters other than the wavelength have a larger influence, such as initial pulse energy, pulse duration and focusing angle.

IV. CONCLUSION AND OUTLOOK

We investigated the behavior of a single pulse propagating through a material on example of Si, with specific focus on achieving particularly fine processing at low pulse energies. The influence of the wavelength, the nonlinear multi-photon absorption, the third-order non-linearity, the focusing angle, the focal depth, the pulse energy and the pulse duration were considered, with several important conclusions:

- We confirmed that using wavelengths between 2000 to 2200 nm is beneficial for maximizing the absorption of light and generation of free carriers in Si even at high focusing angles compared to shorter and longer wavelengths at the optimum pulse duration. This follows from the predicted curve in the nonlinear figure of merit, which has two distinct maxima at $\approx 2100 \text{ nm}$ and $\approx 3200 \text{ nm}$ (as shown in Figure 2).

- The importance of nonlinear effects inside Si for the propagation is larger for smaller NA values. The larger the focusing angle, the less influence self-focusing and multi-photon absorption has on the propagating pulse before the focal spot. This increases the repeatability of modification generation.

- The effect of nonlinear spherical aberrations or “light ring generation” was investigated. As it was shown, this effect degrades close to the maximum of the NFOM. The longer the wavelength, the better the quality of the processed structures.

- The wavelength-dependent effects related to the Kerr effect and the multi-photon absorption de-
FIG. 13: Influence of the pulse parameters energy and duration at a constant wavelength of 1550 nm, numerical aperture of 0.85 and focus depth of 100 µm below the surface. A shorter pulse duration is directly correlated with a higher peak intensity (c.f. Figure 13(a)), while a pulse with a duration between 500 to 1000 fs results in an increased free carrier density compared to longer and shorter pulse durations (c.f. Figure 13(b)). In Figure 13(c) the correlation between pulse duration, pulse energy and the relative energy loss within ±5% around the focal spot (i.e. within 95 to 105 µm in the material) are shown. Here the longest pulse durations together with the highest pulse energy give the maximum relative energy loss at the focal spot. The colorbar in Figure 13(a) gives the intensity in [W m\(^{-2}\)], in Figure 13(b) the density of the free carriers in [m\(^{-3}\)] and in Figure 13(c) the relative energy loss in [%].

fined by NFOM illustrate the different mechanisms of material processing. Namely, the plasma generation due to two- and three-photon absorption enhanced at \(\approx 2100\) nm by the Kerr-effect would decrease the modification threshold.

- We could show that the field distribution at the focal spot only varies slightly when changing the focal depth. Increasing the focusing angle decreases changes, i.e. for larger numerical apertures the distribution of the electric field changes less when changing the target depth compared to smaller numerical apertures.

- It could be shown that there is an optimum pulse duration, located between 600 to 900 fs, explained by the necessity to optimize the spectral overlap between the pulse and the nonlinear figure of merit.

- Based on the two independent numerical approaches, we show that a nonparaxial beam structure under a plasma contribution could cause a spatio-temporal asymmetry and pulse splitting even for a comparatively small NA. Advantages of these numerical approaches are compared.

This work has thus demonstrated the importance of the careful control of laser parameters and in particular wavelength, pulse duration and energy within a large window for optimization of the energy transfer from the light pulse to the material at the focal spot. It sheds light on the different physical phenomena acting on the pulse, including the Kerr-effect, and how they can be used for optimizing the energy transfer. Further optimization and analysis can involve several directions. For example, wavelengths larger than 3000 nm can be investigated. As shown in Figures 2 and 3 there is a transition at \(\approx 3300\) nm from the three-photon absorption to the four-photon absorption. Placing a pulse within those two absorption regions (for example at 3250 nm) would have the advantage of a decreased absorption up close to the focal spot (c.f. Figure 4), but would lose the additional benefit of increased self-focusing, as the pulses at 1950 to 2150 nm can use.

Finally, the wavelength dependence and the impact of tunnel ionization versus multi-photon ionization and avalanche ionization can be studied and compared in a future work.

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DISCLOSURES

The authors declare no conflicts of interest.

Appendix A: Initial conditions for the electric field

To model the pulse as close as possible to existing experimental conditions, a Gaussian shape in time and space was chosen for the incoming pulse.

\[
E(r, t) = \sqrt{\frac{\mathcal{P}_0}{\pi w_0^2}} \cdot \exp \left( - \left( \frac{r}{w} \right)^2 - \frac{i k_0 r^2}{2 f} - i \phi - \left( \frac{\Delta t}{T} \right)^2 - i \omega_0 t \right).
\] (A1)

Here, \( r \) gives the position in space perpendicular to the propagation axis, \( w \) the beam radius, \( k_0 \) the wave number in vacuum, \( f \) the focal length and \( c \) the speed of light in vacuum. \( z_d = t_0^2/(2 \cdot \text{GVD}) \) is the dispersion length, with GVD the group velocity dispersion (typically expressed in fs\(^2\) m\(^{-1}\)), and labelled as \( \beta_2 \). This value is defined as \( \beta_2 = 1/c \left( 2 d^2/d\omega n(\omega) + \omega d^2/d\omega^2 n(\omega) \right) \), based on the pulse frequency \( \omega \) and the refractive index \( n(\omega) \). The pulse broadening due to dispersion is given by \( T = t_0 \sqrt{1 + (z/z_d)^2} \), and the phase shift \( \phi \) is defined as \( \phi = \arctan \left( -\frac{c t}{z} \right) \). Finally, the phase shift introduced by the focusing lens (\( \Delta t \) in Equation (A1)) is defined as \( \Delta t = t + \frac{r^2}{2c(f - z)} \).

In Equation (8) we use somewhat more general initial conditions for generality. These conditions correspond to the dynamics of the Gaussian beam in the paraxial approximation:

\[
\frac{\partial E(z = 0, t)}{\partial z} = 0 \quad \text{(A2)}
\]

\[
\Re(E(z = 0, t)) = \sqrt{\frac{\mathcal{P}_0}{\pi w_0^2}} \cdot \exp \left( -\frac{r^2}{w^2} \right) \cdot \text{sech} \left( \frac{t}{t_0} \right) \cdot \cos \left( \psi - \frac{k_0 r^2}{2R} \right)
\] (A3)

\[
\Im(E(z = 0, t)) = \sqrt{\frac{\mathcal{P}_0}{\pi w_0^2}} \cdot \exp \left( -\frac{r^2}{w^2} \right) \cdot \text{sech} \left( \frac{t}{t_0} \right) \cdot \cos \left( \psi - \frac{k_0 r^2}{2R} \right)
\] (A4)

with the parameters

\[
R = f \cdot \left( 1 + \left( \frac{2R}{f} \right)^2 \right), \quad z_R = \frac{w_0}{\pi NA}, \quad w_0 = \frac{\lambda}{\pi NA}
\] (A5)

Of course, real dynamics will differ from the Gaussian one due to nonlinear effects and non-paraxiality. Here, \( \mathcal{P}_0 \) is the initial power (as in Equation (A1)), \( w \) is the initial beam size, \( w_0 \) is the waist size, \( R \) is the wave front curvature, \( z_R \) is the Rayleigh length, and \( \psi \) is the Gouy phase. The value of \( \Re(E(z = 0, t))^2 + \Im(E(z = 0, t))^2 \) has a sense of intensity.

Appendix B: Distribution of the temporal field and density along the propagation axis

The temporal electric field and the distribution of the generated free carriers can not only be displayed as contour plot (as shown in Figure 9), but also observed along the propagation axis. This has the advantage that slight variations in field intensity or carrier density between different focal depths or different numerical apertures are clearer, while only covering a single line along the propagation axis. Still, this also allows a direct overlap of the distribution of field or carrier density, which makes a comparison easier, and which is not possible for contour plots. The slices for the temporal field for a numerical aperture of 0.25 and a focal depth of 100 µm (corresponding to Figures 9(a) to 9(d)) are shown in Figure 14(a), and for a focal depth of 250 µm at the same numerical aperture (corresponding to Figures 9(e) to 9(h)) are shown in Figure 14(b). For a numerical aperture of 0.85 and a focal depth of 100 µm (corresponding to Figures 9(i) to 9(l)) the temporal intensity is displayed in Figure 14(c), and for a focal depth of 250 µm (without corresponding figures in Figure 9) the intensity is displayed in Figure 14(d). The density of the excited free carriers corresponding to Figures 14(a) to 14(d) is displayed in Figures 15(a) to 15(d).
FIG. 14: Slice of the temporal field intensity for $t_0 = 100$ fs, $f_{\text{inside}} = 100 \mu m/ f_{\text{inside}} = 250 \mu m$, $E_0 = 100 \text{nJ}$ focused with a lens with an NA = 0.25/0.31/0.35/0.41 in Figures 14(a) and 14(b) and an NA = 0.85/1.07/1.18/1.40 in Figures 14(c) and 14(d) directly at the focal spot ($z = 100 \mu m/z = 250 \mu m$). The intensity is given in [W m$^{-2}$].

FIG. 15: Slice of the generated free carrier density for $t_0 = 100$ fs, $f_{\text{inside}} = 100 \mu m/ f_{\text{inside}} = 250 \mu m$, $E_0 = 100 \text{nJ}$ focused with a lens with an NA = 0.25/0.31/0.35/0.41 in Figures 14(a) and 14(b) and an NA = 0.85/1.07/1.18/1.40 in Figures 14(c) and 14(d) directly at the focal spot ($z = 100 \mu m/z = 250 \mu m$).

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