Adiabatic Floquet model for the optical response in femtosecond filaments

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Abstract
The standard model of femtosecond filamentation is based on phenomenological assumptions which suggest that the ionization-induced carriers can be treated as a homogeneous, uncorrelated plasma according to the Drude model, while the nonlinear response of the bound carriers is responsible for the all-optical Kerr effect. Here, we demonstrate that the additional plasma generated at a multiphoton resonance dominates the saturation of the nonlinear refractive index. Since resonances are not captured by the standard model, we propose a modification of the latter in which ionization enhancements can be accounted for by an ionization rate obtained from non-Hermitian Floquet theory. In the adiabatic regime of long pulse envelopes, this augmented standard model is in excellent agreement with direct quantum mechanical simulations. Since our proposal maintains the structure of the standard model, it can be easily incorporated into existing codes for the numerical simulation of femtosecond filaments.

Keywords: light–matter interaction, \textit{ab initio} simulations, metastable states, strong-field ionization

(Some figures may appear in colour only in the online journal)

1. Introduction
Femtosecond filaments are a versatile tool of nonlinear optics. With the advent of the chirped pulse amplification technique which can deliver ultrashort, intense laser pulses \cite{1}, filament generation in gaseous media became possible for the first time \cite{2}. Since the optical power in femtosecond pulses can easily exceed the threshold for nonlinear self-focusing, they can be used to study the catastrophic nonlinear wave collapse \cite{3}. Under loose focusing conditions, however, the onset of plasma defocusing may avoid the collapse and lead to the formation of longitudinally extended filaments. Since filaments have highly directional beam characteristics, they are theoretically described by the forward Maxwell equation \cite{4} or the unidirectional pulse propagation equation \cite{5}. Usually, polarization and current densities are plugged into the propagation equation to account for the optical response of the medium. In the standard model of femtosecond filamentation these quantities are treated phenomenologically \cite{6}: it is assumed that the nonlinear part of the polarization density is governed by the $\chi^{(3)}$-nonlinearity, while the current density of the ionization-induced dilute plasma is described by the Drude model. Neglecting third harmonic generation, the standard model can be boiled down to a formula for the field-dependent refractive index change according to

$$\Delta n(t) = n_2 I(t) - \frac{\rho(t)}{2\rho_c}. \quad (1)$$

Here, $n_2$ is the nonlinear refractive index describing the strength of Kerr self-focusing, $I(t)$ is the temporal intensity envelope of the laser pulse, and $\rho$ is the electron density derived from Keldysh theory \cite{7}. Above the critical plasma density $\rho_c$, which is related to the plasma frequency according to $\omega_p = \omega_0 \sqrt{\rho/\rho_c}$, the electron plasma becomes opaque. The standard model has proven quite successful in many applications. However, its validity was questioned by an experimental observation of a saturating Kerr nonlinearity known as higher-order Kerr effect (HOKE) \cite{8}. This result
triggered conflicting studies either confirming [9–11] or rejecting [12–14] a possible new paradigm of filamentation.

Since there is a growing consensus that the measured deviations from the standard model cannot be described perturbatively via the HOKE model, one may ask if yet another physical mechanism is responsible? *Ab initio* simulations of light–matter interaction gave promising explanations which are not accounted for by the standard model. Among them are trapped population in Rydberg states [15], formation of Kramers–Henneberger atoms [16] or Freeman resonances [17]. These findings also shed some light on the conceptual problems of the standard model [18], e.g., the ambiguity in the definition of bound and free electrons in atoms dressed by strong laser fields, or the fact that resonance enhancements are neglected in the ionization rate. One striking argument is that the perturbative description of the polarization density fails in the regime of filamentation, rendering HOKE questionable [14, 19]. Therefore, instead of a phenomenological approach, the demand for an alternative model derived from quantum mechanical first principles is growing. For instance, one could calculate the polarization and current densities needed to propagate Maxwell’s equations by direct numerical simulations of the time-dependent Schrödinger equation (TDSE), as done in [20]. However, this coupled *ab initio* approach is numerically cumbersome and not feasible for the desired meter-scale propagation distance, the typical longitudinal extension of laboratory-generated filaments. Therefore, faster computational models for quantum-mechanical calculations of the atomic response have been recently proposed, like the non-local model of [21], or the approach proposed in [22] which describes the response in terms of the field-dressed, metastable ground state.

In the first part of this manuscript, we demonstrate the importance of multiphoton resonances which are not accounted for by the standard model. Our *ab initio* simulations show that resonances can significantly enhance the ionization yield which then dominates the nonlinear refractive index. In the second part, we offer a modified version of the standard model, in which the density of free electrons obtained from the direct numerical simulations replaces the electron density calculated according to the Keldysh theory.

### 2. Time-dependent Schrödinger equation

We briefly describe the methods for simulating the optical response of a one-dimensional hydrogen atom under the influence of a strong laser field. One-dimensional model atoms are popular toy models and give good qualitative predictions of the dynamics of atoms in strong laser fields. In particular, they were successfully used for theoretical studies of high harmonic generation and atomic stabilization in strong fields [24–26]. Starting from first principles, we write down the TDSE in the dipole approximation using atomic units (au)

\[
\mathcal{i} \partial_t \psi(x, t) = \left[ -\frac{1}{2} \frac{\partial^2}{\partial x^2} - \frac{1}{\sqrt{x^2 + \alpha^2}} + E(t)x \right] \psi(x, t).
\]

To circumvent the singularity of the atomic potential at the origin, a soft-core potential is used which yields the correct ionization potential of hydrogen, \( I_0 = 13.6 \text{ eV} \equiv 0.5 \text{ au} \) if \( \alpha = \sqrt{2} \). For integrating the TDSE numerically we discretize time \( t \rightarrow t_n \) and space \( x \rightarrow x_j \) with equal spacings \( \tau = \Delta t = 0.01 \text{ au} \) and \( h = \Delta x = 0.1 \text{ au} \), respectively. For the second derivative in the Hamiltonian we can write the central difference

\[
\partial^2_t \psi \approx \frac{\psi_{j-1} - 2\psi_j + \psi_{j+1}}{h^2},
\]

where \( \psi_j \) is the wave function at time step \( t_n \) on the grid point \( x_j \). Thus, the matrix representing \( H \) on the grid has a tri-diagonal structure. The implicit Crank–Nicolson propagator is commonly chosen to compute the wave function for the next time step

\[
U(t_{n+1}, t_n) = \left( 1 + \frac{\mathcal{i}}{2} H(t_{n+\frac{1}{2}}) \right)^{-1} \left( 1 - \frac{\mathcal{i}}{2} H(t_{n+\frac{1}{2}}) \right),
\]

which preserves unitarity and is second order accurate in space and time [27]. When the propagator is applied to the wave function, i.e., \( \psi^{n+1} = U(t_{n+1}, t_n)\psi^n \), we obtain a set of linear equations

\[
M_{n+\frac{1}{2}}^+ \psi^{n+1} = M_{n+\frac{1}{2}}^- \psi^n
\]

with \( M_{n+\frac{1}{2}}^\pm \equiv \left( 1 \pm \frac{\tau}{2} \frac{H(t_n)}{H(t_n)} \right) \).

These equations can be efficiently solved using the tridiagonal matrix algorithm [28]. As an initial condition we provide that \( \psi^0 \) is the atom’s ground state. Reflections at the spatial ends
of the integration domain are suppressed by a 40 au wide absorbing boundary layer [29].

### 2.2. Nonlinear refractive index and resonantly enhanced ionization

We consider a laser pulse with a central wavelength of 800 nm, which corresponds to an angular frequency of \( \omega_0 = 0.057 \text{ au} \) and an optical cycle length of \( T = 2.67 \text{ fs} \equiv 110.3 \text{ au} \). To ensure that the electric field \( E(t) \) has a vanishing DC component, we introduce the vector potential \( A(t) \), from which we reconstruct the electric field according to \( E(t) = -\partial_t A(t) \). The vector potential has a cosine square envelope

\[
A(t) = A_0 \cos^2(\pi t/T_p) \cos(\omega_0 t)
\]

for \( |t| \leq T_p/2 \), where \( T_p \) is the total pulse duration, and the peak amplitudes of \( E \) and \( A \) are related via \( E_0 = \omega_0 A_0 \). We already know from our previous work that Freeman resonances between laser-dressed states lead to drops in the refractive index, and that those resonances are more pronounced when a long flat-top pulse is used. We therefore additionally employ a flat-top pulse with a four-cycle ramp up and down, respectively, according to (7) but with an 80-cycle long central region of constant amplitude in between.

Having solved the TDSE for a single electron, the polarization density for a gas of hydrogen atoms at standard condition, with number density \( \sigma = 4 \times 10^{-8} \text{ au} \), can be obtained from the atomic dipole moment

\[
P(t_n) = -\sigma \langle \psi^n| x |\psi^n\rangle.
\]

Denoting \( \tilde{X} (\omega) \) as the Fourier transform of \( X(t) \), we can define a time-averaged susceptibility and thus a refractive index of the hydrogen gas interacting with the laser pulse

\[
\chi = \frac{\tilde{P} (\omega_0)}{\epsilon_0 \tilde{E} (\omega_0)}, \quad n = \text{Re} (\sqrt{1 + \chi}).
\]

By repeating the simulation with variable peak intensity \( I \) up to \( 40 \text{ TW cm}^{-2} \), we obtain an intensity-dependent, nonlinear refractive index

\[
\Delta n (I) = n (I) - \lim_{I \to 0} n (I).
\]

To ensure that \( n (I) \) correctly captures the response from ionized electrons, the simulation volume is constantly enlarged with increasing \( I \). Thus, less than 0.1% of the total ionization is absorbed at the boundaries.

Our next objective is to seek for correlations between the nonlinear refractive index and the probability of ionization, the population of excited states and the ground state depletion. Thus, we compute the probability that the electron occupies a specific field-free bound state after the pulse is gone. This can be extracted directly from the wave function at the end of the pulse by projecting onto those eigenvectors of the discrete Hamiltonian which correspond to bound atomic states, denoted as \( \phi_k \),

\[
\beta_k = |\langle \phi_k | \psi (T_p/2) \rangle|^2 \quad \text{and} \quad \beta_{\text{bound}} = \sum_k \beta_k.
\]

In practice, the sum in (11) includes the first 100 bound states which is enough to ensure convergence of \( \beta_{\text{bound}} \). Obviously, the ground state depletion is \( 1 - \beta_0 \), and the probability for the electron to be in an excited state is \( \beta_{\text{bound}} - \beta_0 \). The ionization probability is defined as the non-bound part of the wave function, \( \beta_{\text{free}} = 1 - \beta_{\text{bound}} \).

Results for the 80-cycle flat-top pulse with \( T_{\text{FWHM}} = 215.8 \text{ fs} \) are shown in figure 1. The nonlinear refractive index (purple) exhibits local drops around peak intensities of 15, 22 and 30 TW cm\(^{-2} \) as well as above 31 TW cm\(^{-2} \). Apparently, these drops coincide with an increased ground state depletion (red). The depletion is mainly caused by enhanced transitions into the continuum (green), since the probability to end up in an excited bound state (orange) is 1–2 orders of magnitude smaller. The origin of the enhanced ionization probability are Freeman resonances, as discussed in [17]. To consolidate the existence of

![Figure 1](image-url)
Freeman resonances in the optical response, we briefly sketch how they occur: since the ground state of an atom is strongly bound, its energy remains almost constant when an external field is applied. With increasing intensity, however, the AC Stark effect shifts the energy levels of the dressed Rydberg states by the ponderomotive potential, \( U_p = E^2/(4\omega_0^2) \). Eventually, a Freeman resonance arises when the energy difference between a Rydberg state and the ground state is a multiple of the photon energy. These resonances are usually preceded by a channel closure, which occurs when the ground state is in multiphoton resonance with the ponderomotively up-shifted continuum threshold. In consequence, we expect an enhanced population of the participating Rydberg state at a Freeman resonance. Indeed, above the 9-photon channel closure at 5.5 TW cm\(^{-2}\) in figure 1, we observe an increase and successive peaks in the occupation probability of the excited states. Interestingly, this seems to be anticorrelated to the ionization probability, which is decreased at the same intensities, most likely due to interference stabilization [30]. This stabilization vanishes for peak intensities \( \geq 15 \) TW cm\(^{-2}\), where transitions into both bound and continuum states are resonantly enhanced.

We now look closer at individual Rydberg states which should be dominantly populated at a Freeman resonance. As shown in figure 2(a), we resolve transitions up to the 31st excited state just after the 9-photon channel closure. Note that parity conservation prohibits a 9-photon transition from the even ground state to even excited states, thus only transitions to odd-parity states are allowed. After the 10-photon channel closure at 30.2 TW cm\(^{-2}\) in figure 2(d), we again observe Freeman resonances, now with 10-photon transitions to even-parity excited states. Somehow different are the resonances near 15, 22, and 30 TW cm\(^{-2}\) in figures 2(b) and (c): increasing the peak intensity beyond the range of figure 2(a), we expect transitions to the 7th, 5th, and 3rd excited state, since with increasing intensity lower lying excited states come into multiphoton resonance with the ground state. However, these transitions are not dominant since the population of other excited states is of same magnitude. Obviously, multiple excited states contribute to these resonances. This may be related to the fact that the energy shift of lower lying excited states shows a more complicated behavior than that of Rydberg states. Resulting interactions between dressed excited states may lead to a parity change, which explains that also even-numbered excited states are occupied. Nevertheless, these ‘mixed resonances’ facilitate enhanced ionization as seen in figure 1.

2.3. Modification of the standard model

The observations of the last paragraph clearly indicated that resonances in the refractive index are correlated to the ground state depletion. Our results also revealed that the ground state depletion is dominated by transitions into the continuum. They therefore confirm the standard model assumption that the refractive index saturation stems from free electrons. A significant difference to the standard model, however, lies in the physical mechanism underlying the ionization enhancement which we identified as resonances between laser-dressed states. Thus, the standard model should be able to reproduce our simulation results if the resonantly enhanced ionization is taken into account. To test this assumption, we write down a modified, time-independent version of the nonlinear refractive index (1)

\[
\Delta n(I) = n_2 I - \frac{\rho_{\text{lin}}(I)}{2\rho_0}, \tag{12}
\]

where \( I \) is the peak intensity of the laser pulse. Furthermore, we have to take into account that the density of free carriers is unambiguously defined only for a vanishing external electric field [31]. We therefore replaced in (1) the Keldysh-like ionization density \( \rho(I) \) with the electron density at the end of the pulse \( \rho_{\text{lin}}(I) \) calculated from the direct numerical simulations, multiplied by a correction factor \( \eta = 0.5 \) as justified in the appendix. We then compare three different cases:

\[
\rho_{\text{lin}}(I) = \sigma \begin{cases} 
\beta_{\text{free}}(I) & \text{case A} \\
\beta_{\text{free}}(I) + \beta_{\text{Ryd}}(I) & \text{case B} \\
1 - \beta_0(I) & \text{case C.}
\end{cases} \tag{13}
\]

Case A considers only ionized electrons, whereas case B includes the population in Rydberg states. This is motivated by the fact that their contribution to the refractive index is similar to that of free electrons [32]. Additionally, electrons which are freed during the pulse may be captured back into Rydberg states when the field is gone. Here, we consider all states above the ninth excited state as Rydberg states since their corresponding Floquet energies shift with the ponderomotive potential (cf figure 1 in [17]). The probability to find the atom in a Rydberg state is then given by \( \beta_{\text{Ryd}} = \sum_{k>9} \beta_k \). The overall population of excited states, however, is small compared to the ionization probability as seen in figure 1. Therefore, case C considers the full ground state depletion and may serve as a simple, but accurate approximation.

Results are presented in figure 3 together with the exact TDSE calculation (10) for different pulse shapes and lengths. First, we employed the 80-cycle flat-top pulse. For low peak intensities shown in panel (a), the refractive index for both model (A: green, B: orange, C: red) and simulation (purple, dashed) follows the Kerr-nonlinearity \( n_2 I \) (gray, dashed–dotted) until ionization becomes non-negligible. However, beyond 8 TW cm\(^{-2}\), the TDSE result is obviously superlinear, indicating that the next coefficient in a perturbative description \( n_4 \) would be positive. Actually, a super-linear correction proportional to \( I^2 \) renders the agreement between model and simulation astounding close. For the 15 TW cm\(^{-2}\) resonance, the drop in the refractive index is significantly larger than predicted by model case A and B (which overlap in this panel). Case C, on the other hand, is closer to the simulation result. This observation also holds for resonances at 22 TW cm\(^{-2}\) and 29.8 TW cm\(^{-2}\) in panel (b) which shows the full intensity range on a larger scale for the same flat-top pulse. All in all the deviation...
remains small. After the 10-photon channel closure, the increased population of Rydberg states (cf figure 2) becomes noticeable in the refractive index. As expected, case A (ionization only) deviates from the simulation while case B and C are in good agreement. Model and simulation astonishingly coincide even for a purely cos²-pulse as shown in panel (c), where the total pulse duration is 96 cycles ($T_{\text{FWHM}} = 93.3$ fs). Again, we notice deviations in case A for peak intensities larger than $30 \text{ TW cm}^{-2}$, but case B and C are close to the numerical result. For the 24-cycle pulse ($T_{\text{FWHM}} = 23.3$ fs) in panel (d), we observe increasing deviations from our model as the Kerr nonlinearity and the plasma response assume comparable magnitudes, i.e., for intensities beyond the zero-crossing of the nonlinear refractive index. These deviations, however, remain small and the qualitative behavior is reproduced.

Overall, our modification of the standard model is in remarkable agreement with the nonlinear refractive index from pure TDSE calculations, manifesting the conclusion that resonantly enhanced ionization is the driving component in its saturation and sign inversion.

### 3. Adiabatic Floquet resonance model for the nonlinear optical response

In this part of the manuscript, the results of our numerical studies will be cast into an augmented standard model of the optical response in filaments which can be easily built into existing numerical simulation codes for femtosecond filamentation. Our proposed adiabatic Floquet resonance (AFR) model is based on the following observations: first, we have
convinced ourselves that the optical response is mainly determined by resonantly enhanced transitions from the ground state and the resulting ground state depletion. Second, we have shown that the Kerr part of the standard model is nearly unaffected by the considered non-perturbative effects. In consequence, our model for the time-dependent nonlinear refractive index is

$$\Delta n_{AFR}(t) = n_2 I(t) - \frac{\rho_0(t)}{2\rho_c},$$

where $$\rho_0(t) = \sigma (1 - \beta_0(t))$$ is the macroscopic ground state depletion. In principle, $$\rho_0(t)$$ has to be extracted from direct numerical simulations of the TDSE, and nothing would be gained. Nevertheless, in the adiabatic regime of a slowly varying pulse envelope, we can make use of non-Hermitian Floquet theory to calculate the decay rate $$w[I]$$ of the ground state. The ground state depletion $$\rho_0(t)$$ is then governed by the rate equation

$$\partial_t \rho_0(t) = w[I(t)](\sigma - \rho_0(t)).$$

Since in the adiabatic case, no transitions to excited states occur, the decay rate $$w[I]$$ is identical with the ionization rate, and $$\rho_0$$ is the density of free electrons, in complete correspondence to the usual standard model. For our AFR model it is therefore sufficient to replace the Keldysh-type ionization rate in the standard model by an ionization rate obtained from the non-Hermitian Floquet theory.

3.1. Calculation of Floquet multipliers and eigenstates

Non-Hermitian Floquet theory describes the atom dynamics in terms of Floquet resonances [33]. These are metastable electronic states in a finite volume which are subject to outgoing wave boundary conditions and thus take ionization effects into account. In this approach, the ionization rate is identical to the inverse lifetime of the Floquet ground state resonance. To ensure $$L^2$$-integrability of the resonance wave functions, we employed complex rotation $$x \rightarrow xe^{i\theta}$$ to the Hamiltonian

$$H(x) = -\frac{1}{2} \partial_x^2 - \frac{1}{\sqrt{x^2 + \alpha^2}} - iA(t) \frac{\partial_x}{\partial x}.$$  

Here, we opted for the velocity gauge, since in the length gauge, complex rotation would result in a blow-up of the wave function during time propagation. The ionization rate and Floquet resonances can be obtained from the Floquet eigenvalue equation [34–36]

$$(H(xe^{i\omega t}) - i\epsilon_\alpha) \phi(x, t) = \epsilon_\alpha \phi(x, t),$$

where $$\epsilon_\alpha$$ is the complex quasi-energy and $$\Gamma_\alpha = -2 \text{Im} \epsilon_\alpha$$ is the inverse lifetime of a resonance. The Floquet wave function $$\Psi = e^{-i\omega t'\phi}$$ [37] is quasi-periodic with

$$\Psi(x, t + T) = e^{-i\omega T} \Psi(x, t),$$

where $$T = 2\pi / \omega_0$$ is period of the laser pulse. $$\Psi$$ decays exponentially due to the negative imaginary part of the quasi-
energy \( e_\alpha \) (with \( \Gamma_\alpha > 0 \)). Typically, the ground state resonance \( \phi_0 \) is the most stable eigensolution with the smallest decay rate \( \Gamma_\alpha \), which can be interpreted as the rate of ionization from the ground state. This is exactly the quantity we are interested in to build our augmented standard model. Instead of solving (17) directly, we note that by substituting \( \Psi(x, t + T) = U(T) \Psi(x, t) \) into (18), it can be rewritten as an eigenvalue equation for the one-cycle propagator \( \mathcal{U} \equiv U(T) \). Consequently, the Floquet wave functions are eigenfunctions of \( \mathcal{U} \), whereas the Floquet multipliers \( e^{-i\epsilon_i T} \) are the corresponding eigenvalues [38]. For diagonalization of \( \mathcal{U} \) we employ the eigenvectors \( \chi_n \) of the discrete counterpart of the free particle Hamiltonian \( \hat{H}_0 = -\partial_x^2 / 2 \). By propagating \( \chi_n \) along one optical cycle we calculate the matrix elements

\[
\mathcal{U}_{mn} = \langle \chi_m(0) | \mathcal{U} | \chi_n(0) \rangle = \langle \chi_m(0) | \chi_n(T) \rangle. \tag{19}
\]

Since the results of this section require an accurate determination of the complex quasienergies, for the numerical propagation of the \( | \chi_n \rangle \), we employed a fourth-order scheme for the spatial derivatives based on the Numerov approximation employed in [39]. In fact, this procedure does not affect the tridiagonal property of the short-distance propagator, such that the Crank-Nicolson scheme of section 2.1 can be maintained here. Diagonalization of \( \mathcal{U} \) then yields the complex Floquet multipliers \( e^{-i\epsilon_i T} \) and the corresponding Floquet states \( \phi_i \). Note that in the analytic theory, according to the Balslev–Combes theorem [40, 41], the quasi-energies of the quasi-bound resonances do not depend on the chosen rotation angle \( \theta \). However, this does not necessarily hold true for the numerical treatment, since one usually diagonalizes \( \mathcal{U} \) using either a truncated set of basis functions or a finite space grid. In these cases, the rotation angle has to be chosen such that the quasi-energies become stationary with respect to a variation of \( \theta \). In our case, we obtain reasonable results for \( \theta = 0.3 \).

In figure 4, we plot the Floquet multipliers \( e^{-i\epsilon_i T} \) in the complex plane for two different values of \( \theta \). The multipliers corresponding to the discrete quasi-continuum can be observed to spiral into the origin. In accordance with the Balslev–Combes theorem, the continuum energies are transformed as \( e_\epsilon \rightarrow e_\epsilon e^{-2i\theta} \), and the positions of the corresponding Floquet multipliers consequently depend on the rotation angle \( \theta \). In contrast, the positions of the multipliers corresponding to Floquet resonances are stationary w.r.t. \( \theta \)-variation. Therefore, a small change in \( \theta \) allows for a straightforward identification of the Floquet resonances. Furthermore, the distance of the Floquet multipliers to the unit circle is a measure of the lifetime \( \Gamma' \). While the Floquet multipliers of stable bound states (\( \Gamma' > 0 \)) are located on the contour \( \mathcal{C} \) of the unit circle, the lifetime \( \tau = 1 / \Gamma' \) of a resonance decreases with increasing distance from \( \mathcal{C} \). Going from \( I = 5 \) TW cm\(^{-2} \) in figure 4(a) to 22 TW cm\(^{-2} \) in (b), resonances move closer to the origin for higher intensities, i.e., they are less stable and ionization is increased. Additionally, in panel (b), we observe resonances with almost the same complex angle in the polar plot. Since a full rotation corresponds to an energy difference of one photon energy, these resonances facilitate multiphoton transitions.

To calculate an intrapulse ionization rate \( \Gamma_0 \) for the ground state resonance, we solve the eigenvalue equation (18) for a set of intensities up to 40 TW cm\(^{-2} \). When the intensity increases above a channel closure, \( \Gamma_0 \) exhibits pronounced peaks stemming from multiphoton or Freeman resonances, as shown in figure 6(a). In principle, the obtained ionization rate is valid only in the adiabatic regime of a slowly varying pulse envelope. In this case, resonantly enhanced ionization transfers an electron in the ground state resonance directly into the continuum, without populating the intermediate resonant state, as pointed out in [42]. However, as the pulse approaches the few-cycle regime, population transfer to excited Floquet resonances has to be taken into account, and the pronounced
resonance peaks in $I_0(I)$ are expected to smear out for shorter pulses.

3.2. Augmented standard model with ionization rate from non-Hermitian Floquet theory

Having calculated the ionization rate $I_0$, we can compare the predictions from our AFR model (14) and (15) with the results of direct numerical simulations. First, we solve the TDSE to obtain the polarization density $P(t)$ for the given electric field $E(t)$. From these quantities, a time dependent susceptibility $\chi(t)$ is computed by forming the ratio of the corresponding complex analytic signals [43]

$$\chi(t) = \frac{P(t)}{\epsilon_0 E(t)}$$

with $P = P + i \mathcal{H}(P)$ (and likewise $E$), and $\mathcal{H}$ is the Hilbert transform. From (20), the (ab initio) nonlinear refractive index is obtained as $\Delta n(t) = \sqrt{1 + \chi(t)} - n_0$, where $n_0$ is the field-free background refractive index. In order to obtain the corresponding predictions of the AFR model, denoted as $\Delta n_{AFR}(t)$, we have to determine the nonlinear index $n_2$ for intensities below 1 TW cm$^{-2}$, i.e., in the perturbative Kerr regime, the refractive index reproduces the intensity profile of the pulse, $\Delta n(t) = n_2 I(t)$. After evaluating $\Delta n(t)$ from TDSE at the local maxima of $I(t)$ for a series of low peak intensities, a linear fit yields $n_2 = 6.5 \times 10^{-7}$ cm$^2$ TW$^{-1}$. This allows the evaluation of the Kerr part of the nonlinear refractive index $\Delta n_{AFR}(t)$. The plasma part in (14) can be calculated by plugging the ionization rate $I_0$ into the rate equation (15) and by integrating the latter for temporal intensity profiles $I(t)$.

Figure 5 shows temporally resolved refractive indices for peak intensities 6.2, 22.2 and 38.2 TW cm$^{-2}$ using a cos$^2$-pulse with 96 cycles. If we compare the AFR results (orange) to that of the direct simulations (purple, dashed), we find that our model gives excellent predictions for the nonlinear refractive index: we clearly see that in the leading edge the pulse profile is recovered in accordance with the Kerr nonlinearity $\Delta n(t) = n_2 I(t)$, panel (a) and (b). However, as plasma accumulates during the laser-atom interaction, the refractive index correction in the trailing edge of the pulse becomes more negative and turns the nonlinearity into a defocusing one, panel (b) and (c). This behavior is also well-known from the standard model and responsible for the dynamic spatial replenishment scenario described in [44].

To conclude our comparison with TDSE results, we derive the intensity dependence of the refractive index as predicted by the AFR model. In particular, if we know the polarization density $P_{AFR}(t)$, we can employ (9) to calculate the time-averaged susceptibility $\chi_{AFR}(\omega_0)$ at the center frequency of the pulse. In order to construct $P_{AFR}(t)$, we use the general relation

$$P(t) = \epsilon_0 \chi(t) E(t)$$

and

$$\chi(t) = n^2(t) - 1,$$

where, in this case, $n(t) = n_0 + \Delta n_{AFR}(t)$. The nonlinear refractive index $\Delta n_{AFR}(I)$ then directly follows from (9) upon Fourier transform of $P_{AFR}(t)$ and $E(t)$ with various peak intensities $I$.

In figures 6(b)–(d), we show the intensity-dependent refractive indices obtained from both model and simulation versus peak intensity $I$: for a flat-top comprising of 80 optical cycles (b) as well as for cos$^2$-envelopes with $T_p = 96$ T (c) and $T_p = 24$ T (d). For the long flat-top pulse, the agreement between the direct numerical calculation and the augmented standard model are indeed excellent. In fact, for long pulses we can employ the adiabatic approximation with respect to the pulse envelope: here, the wave function remains in the ground state resonance which adiabatically adapts to the instantaneous intensity. In this case, the ionization rate $I_0$ accurately describes the actual ionization from the ground state resonance. Moving to shorter pulses in figures 6(c) and (d), however, we see deviations between the two calculations. Especially, above 30 TW cm$^{-2}$, the AFR refractive index exhibits strong resonances, which appear smeared out in the $\Delta n(I)$ from the direct simulations. Obviously, for shorter pulses, the evolution of the envelope can no longer be considered adiabatic. In consequence, transitions occur from the ground state resonance to higher lying resonances, and the ionization rate $I_0$ becomes less accurate.

To conclude this section, we point out that we cross checked our results for the one-dimensional model atom with results from non-Hermitian Floquet calculations for a full three-dimensional (3D) model of atomic hydrogen,
employing the STRFLO eigensolver of [45], which is available for online download [46]. For the 3D case, we found qualitatively comparable resonance structures in the decay rate of the Floquet ground state resonance. These resonances were also present in full 3D TDSE simulations [39] for finite envelope pulses, giving rise to resonantly enhanced ionization yields at the end of the pulses. However, we observed that finite envelope effects, i.e., non-adiabaticity in the pulse envelope, have a more pronounced impact in 3D: the resonances seen by a 90 fs pulse are slightly more smeared out than in the discussed one-dimensional case. We therefore repeated our 3D simulations for slightly longer 120 fs pulses, in case of which the clear visibility of the resonances in the ionization fraction was restored. This leads us to conclude that the qualitative predictions of our model are equally valid for the three-dimensional model atom.

4. Conclusion

In our manuscript, we propose an augmented standard model for the phenomenological description of the optical response in femtosecond filaments. Our AFR model maintains the structure of the standard model, which is written as a sum of the Kerr-induced refractive index change and a plasma-induced one, where the time-dependence of the plasma generation is governed by an ionization rate $w[I]$. However, instead of an ionization rate derived from Keldysh theory, we employ an ionization rate obtained from non-Hermitian Floquet theory.

The development of our model was led by numerical experiments which clearly showed that resonantly enhanced ionization has a strong impact on the nonlinear refractive index modifications. Of course, this raises the question whether these resonances actually influence the dynamics of femtosecond filaments. Clearly, for pulse durations of several picoseconds and field strength much larger than $10^{18} \text{ W m}^{-2}$, this is not the case. Instead, for these pulse parameters, Freeman resonances are washed out since the energy spectrum of free electrons resulting from above-threshold, multiphoton ionization is averaged over a focal volume. The latter effect can be attributed to the ponderomotive force which pushes the electrons out of the laser focal volume [23]. However, as shown in [6], for the typical pulse durations and field-strengths in femtosecond filaments, the resulting electron displacement due to the ponderomotive force is negligible compared to the optical beam waist. We thus may expect that resonances have a noticeable impact on filamentary propagation for the pulse durations of some 100 femtoseconds considered in the present work. The possible impact of these resonances on the filamentary propagation on femtosecond pulses was theoretically explored in our previous work [17]. Moreover, multiphoton resonances in nitrogen N$_2$ may enhance molecular dissociation processes, an important prerequisite for experimentally inducing bidirectional air lasing [47]. Interestingly, resonance-related drops in the nonlinear susceptibility or polarizability were identified in [16, 48, 49]. However, the main mechanism leading to the local decrease of the refractive index was not discussed, namely the resonantly enhanced ionization yield. Our numerical results showed that the optical response is dominated by the ordinary Kerr effect and the ground-state depletion, i.e., the plasma-like response of Rydberg and free electrons. We were thus led to the conclusion that an augmented standard model may be obtained by replacing the Keldysh-like ionization rate in the plasma term by an ionization rate derived from non-Hermitian Floquet theory-since in the adiabatic regime Floquet theory accurately captures ionization enhancements due to multiphoton resonances.

Figure 6. Ionization rate $\Gamma_0$ of the ground state Floquet resonance versus intensity (a). Intensity-dependent refractive index for a 80-cycle flat-top pulse (b) or $\cos^2$-pulses with 96 (c) and 24 optical cycles (d).
The AFR model yields excellent results in agreement with the *ab initio* TDSE calculation of the nonlinear refractive index. Our results also seem to rule out the necessity to introduce higher-order Kerr terms, since our modified standard model only contains the lowest order Kerr term \( n_2 J \). We therefore suggest that it is mainly resonance-enhanced ionization which led to the discrepancies between the experimental results of [8] and the standard model predictions. For a conclusive proof of our suggestion, though, it is essential to numerically reenact the experimental protocol of [8], which utilized a non-collinear pump-probe setup to detect the Kerr-induced birefringence. This requires full three-dimensional simulations of the TDSE and offers the chance for a detailed understanding of what was actually measured in the experiment.

An important issue which has to be deepened in further research is the transition from adiabatic to non-adiabatic pulse envelopes. As our calculations have shown, the ionization rate \( I_\text{ion} \) fails to accurately describe the ionization dynamics in the latter case. For, in fact, for cycle-pulse envelopes whose envelope evolves on time-scales comparable to the optical carrier, the quantum adiabatic theorem is not applicable, and the dynamics are no longer governed by the Floquet ground state resonance alone. Instead, transitions to excited resonance states have to be considered. In principle, this can be taken into account by solving the time-dependent Schrödinger equation in a basis of resonant states, by employing, e.g., non-Hermitian generalizations of the short-pulse Floquet method suggested in [50] or the adiabatic perturbation theory of [51]. We also note that a closely related approach has recently been proposed in [22]. However, while our approach is based on Floquet theory and assumes that the time scale on which the envelope evolves is much larger than the optical cycle, the aforementioned authors employ the adiabatic approximation on the level of the optical carrier wave. Their approach is thus especially suited to pulses in the quasistatic, long wavelength regime, and non-adiabatic corrections have to be considered for shorter wavelength pulses, which is further discussed in [52]. In contrast, the domain of validity of our approach extends to shorter wavelengths, but requires non-adiabatic corrections for few-cycle pulses, as discussed above. Nevertheless, we expect that our adiabatic Floquet model and future non-adiabatic corrections thereof are especially suited for capturing the essential physics when multiphoton resonances exist. Then, the time-dependent electron density satisfies a rate equation \( \partial_t \rho(t) = w[I(t)](\sigma - \rho(t)) \), whose solution for small ionization ratios may be approximated as

\[
\rho(t) \approx \sigma \int_{-\infty}^{t} w[I(\tau)] \mathrm{d} \tau. \tag{22}
\]

Under the assumption that the intensity profile \( I(t) \) is an even function, the electron density exhibits the symmetry \( \rho(-t) = -\rho(t) + \rho_{\text{fin}} \). The contribution of the free electrons to the \( \omega_0 \)-component of the polarization density may be written as

\[
\tilde{P}_{\text{ion}}(\omega_0) = - \frac{e_0}{\rho_i} \int_{-\infty}^{\infty} \mathrm{d} t \, \rho(t) E(t) e^{i\omega_0 t} = - \frac{e_0}{2\rho_i} \int_{-\infty}^{\infty} \mathrm{d} t \, \rho(t) E_{\text{env}}(t), \tag{23}
\]

where \( E_{\text{env}}(t) = E(t) e^{i\omega_0 t} \) denotes the complex envelope of the real electric field \( E(t) = (E(t) + E^*(t))/2 \). Here, we neglected the integral over the fast oscillating term \( E_{\text{env}} e^{i\omega_0 t} \). We may now use the symmetry of \( \rho(t) \) to recast this equation into

\[
\tilde{P}_{\text{ion}}(\omega_0) = - \frac{e_0}{4\rho_i} \int_{-\infty}^{\infty} \mathrm{d} t \, (\rho(t) - \rho(-t) + \rho_{\text{fin}}) E_{\text{env}}(t). \tag{24}
\]

Noting that our employed pulse envelopes \( E_{\text{env}} \) are real-valued and symmetric, the integral over the odd function \( (\rho(t) - \rho(-t)) E_{\text{env}}(t) \) vanishes, and we are left with

\[
\tilde{P}_{\text{ion}}(\omega_0) = - \frac{e_0}{4\rho_i} \rho_{\text{fin}} \int_{-\infty}^{\infty} \mathrm{d} t \, E(t) e^{i\omega_0 t} = - \frac{e_0 \rho_{\text{fin}}}{2\rho_i} \tilde{E}(\omega_0), \tag{25}
\]

where we used \( \tilde{E}(\omega_0) = 2\tilde{E}(\omega_0) \). Finally, using \( \chi(\omega_0) = \tilde{P}^{*}(\omega_0)/(e_0 \tilde{E}(\omega_0)) \) and \( n(\omega_0) = \sqrt{1 + \chi(\omega_0)} \approx 1 + \chi(\omega_0)/2 \), we find that the plasma correction \( \Delta n_{\text{ion}} \) to the nonlinear refractive index is given by

\[
\Delta n_{\text{ion}}(\omega_0) = -\frac{\eta \rho_{\text{fin}}}{2\rho_i}, \tag{26}
\]

with a correction factor \( \eta = 0.5 \) which is absent in the time-dependent formulation.

Deviations from this factor can result from memory effects in the ionization rate, i.e., an explicit time dependence of the ionization rate \( w[I(t), t] \). Such an explicit time-dependence can occur when a significant fraction of the ground-state population is transferred to higher excited states.

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**Appendix**

In the following, we discuss the introduction of the correction factor \( \eta \) employed in section 2.3, which, for the employed pulse profiles, is found to be close to a numerical value of 0.5. First of all, albeit an exact definition of an instantaneous, intrapulse electron density suffers from the gauge ambiguity, we assume for the derivation that an instantaneous ionization rate \( w[I(t)] \) exists. Then, the time-dependent electron density satisfies a rate equation \( \partial_t \rho(t) = w[I(t)](\sigma - \rho(t)) \), whose solution for small ionization ratios may be approximated as

\[
\rho(t) \approx \sigma \int_{-\infty}^{t} w[I(\tau)] \mathrm{d} \tau. \tag{22}
\]
The latter effect decreases with the adiabaticity of the laser pulse envelope.

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