Modeling of ultrafast laser pulse propagation

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ABSTRACT

Computer simulations of ultrafast optical pulses face multiple challenges. This requires one to construct a propagation model to reduce the Maxwell system so that it can be efficiently simulated at the temporal and spatial scales relevant to experiments. The second problem concerns the light-matter interactions, demanding novel approaches for gaseous and condensed media alike. As the nonlinear optics pushes into new regimes, the need to honor the first principles is ever greater, and requires striking a balance between computational complexity and physical fidelity of the model. With the emphasis on the dynamics in intense optical pulses, this paper discusses some recent developments and promising directions in the field of ultrashort pulse modeling.

Keywords: Nonlinear optics, pulse propagation, light-matter interaction

1. INTRODUCTION

Modern nonlinear optics, and in particular the field of ultra-short, high-intensity optical pulses continues to explore new regimes.[1] One of the recent trends concerns the push towards longer wavelengths. A lot of research over the last two decades in the nonlinear optics was made possible by the availability of high-power pulsed laser sources in the near infrared, this is the wavelength region where the efforts have been concentrated so far. The fields of optical filamentation,[2] strong-field and attosecond physics,[3] and high-harmonic generation[4] are just a few examples of disciplines that have been driven by and benefited tremendously from accurately controlled, femtosecond-duration optical pulses. Naturally, the nonlinear optics community generated significant volume of knowledge about the processes that govern this specific wavelength regime. However, as a longer-wavelength sources gradually become more available, the interests start to shift toward the mid-infared regime. This regime offers new opportunities and poses new challenges, not only from the point of view of sources, detection, applications etc., but also from the standpoint of basic physics. A question immediately emerges about the applicability of our knowledge relevant to the near infrared. It turns out that as we move between the different laser wavelength regions, the physics changes in a qualitative way.[5,6] This fact will have deep consequences on both, nonlinear optics applications and on the theory and simulation. We will start this contribution with a brief review of the dynamics of light-matter interactions in order to contrast different regimes and their mathematical models...

2. MODELING NONLINEAR OPTICAL PULSE PROPAGATION

2.1 Nonlinear pulse propagation in near infrared

Physical effects that govern the propagation of high-intensity optical pulses in various media are relatively well understood.[2] A great deal of this understanding was attained with the help of modeling and numerical simulations.[7] This was especially the case in the area of optical filamentation[8] which in fact motivated a lot of the effort on the theoretical side.

Without going into unnecessary detail, let us simply say that the essential mathematical model that captures the most important physical effects in the near infrared and also in the visible region is that of the Nonlinear Schrödinger Equation[9] (NLS),

$$\partial_z A = \frac{i}{2k_0}\Delta A - \frac{k''}{2}\partial_{tt} A + \frac{i\omega_0 n_2}{c}|A|^2 A \ .$$

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Needless to say that NLS as shown here is rather inadequate to describe the dynamics of light pulses on a *quantitative* level. It was recognized very quickly in the early nineties that one has to go beyond this celebrated equation in order to make connections to experimental observations.

Nevertheless, from the standpoint of capturing the basics physics, NLS does have the right ingredients, at least for the near-infrared nonlinear optics. In general, it describes the wave propagation (first term on RHS) in dispersive (second term), weakly nonlinear (third term) media. Here, the important distinction is the relative strength of the two medium properties: For NLS to apply, the chromatic dispersion of linearly propagating (small-amplitude) waves has to be the stronger effect, with the nonlinearity being relatively weaker. This is indeed the case in the nonlinear optics in the visible and near infrared regions.

Another important characteristics of NLS is that it does not describe the optical field directly, but represents it in terms of its envelope A. This is assumed to evolve or vary slowly with the propagation distance and it allows, at least in principle, to decrease the computational complexity in the computer-aided modeling. While this kind of approximation has long been recognized as degrading the fidelity of the numerical model, the envelope description was and still is frequently applied. While resorting to the envelope language is not something we would recommend under any circumstances, it is fair to say that in the near-infrared the optical pulse description in terms of envelopes does not necessarily imply a fundamentally flawed model. However, this changes fundamentally as soon as one transitions to longer wavelengths...

2.2 Nonlinear pulse propagation in mid infrared

In media with wide transparency windows, the chromatic dispersion often decreases with the increasing wavelength. On the other hand, the strength of the nonlinear interactions may not change that drastically. For example, recent measurements of the nonlinear index in a number of gases[10] showed that the nonlinear dispersion is rather weak. In the crudest approximation one can say that the Kerr effect, at least its electronic component, is as strong in the mid infrared as it is in the near infrared. As a consequence of these trends, the relative role of the dispersion and nonlinearity switches, which in turn means that the essential mathematical model also needs to change. Instead of NLS, the generic description of nonlinear wave propagation relevant to the mid-infrared is the so-called modified Kadomtsev Petviashvili (MKP) equation.[5,6] It describes strongly nonlinear, weakly dispersive wave propagation regimes:

$$\partial_\tau \left(\partial_z E - a \partial_\tau^3 E + \frac{4n_2}{3c} \partial_\tau E^3 \right) = \frac{c}{2n_0} \Delta_\perp E \ .$$

Like NLS, the above MKP equation captures the effects of chromatic dispersion (parameter a) coupled to the medium nonlinearity (nonlinear index n_2), but there is a qualitative difference between the two equations. The MKP equation describes the optical field E directly, without approximations involving envelopes which are characteristic of NLS. In other words, the MKP paradigm is a carrier-resolved description,[11] and this becomes crucial for modeling in the mid-infrared.

The qualitative difference in physics is perhaps best appreciated in the mechanism that arrests the self-focusing collapse initiated and driven by the Kerr nonlinearity in pulses with sufficiently high peak powers. In the NLS-based description one needs, in general, to invoke processes "outside of the NLS model," such as nonlinear losses, ionization in strong fields, and subsequent de-focusing of the light field by freed electrons. As the collapsing pulse reaches sufficient peak intensity, its energy will be diverted in the outward spatial direction by the action of the defocusing "plasma." The result is a dynamic balance, termed nonlinear spatial replenishment,[12] in which recurring collapses are arrested, and are followed by a decrease of the peak intensity before the energy of the pulse is replenished in the on-axis spatial region again, thus starting a new cycle in the collapse sequence. One can say that both the "self-focusing forces" and "collapse-arrest forces" act in the spatial dimensions transverse to the pulse propagation direction.

The corresponding mechanism is different in the mid-infrared region,[6] and this is faithfully described by the MKP equation. Unlike in the previous case, the MKP actually contains all necessary ingredients responsible for the arrest of the self-focusing collapse. This is because the equation in fact describes yet another type of singularity, and that is the carrier-wave shock formation.[5] This occurs before a fully developed collapse can occur, and has the consequence of adding higher-harmonic (3rd, 5th, 7th, ...) spectral components to the optical field. These propagate with velocities different from that of fundamental and that is why they walk-off in the propagation direction from the main pulse. The energy focused by the Kerr effect is thus redirected in the longitudinal direction in the form of high-frequency waves that are left behind in the wake of the main pulse. The mechanism is illustrated in the following Fig. 1, depicting a snapshot of the temporal profile of the electric field in a mid-infrared pulse:

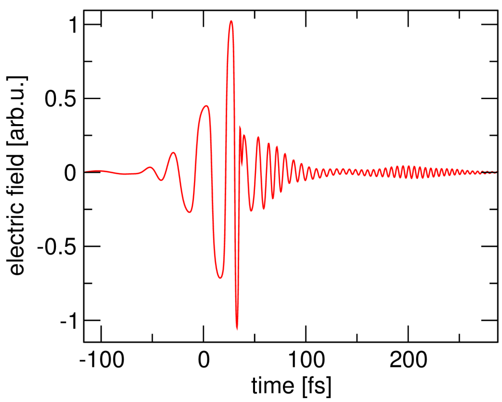

Figure 1. Self-focusing collapse arrest in a mid-infrared optical pulse. The initial pulse had the central wavelength of 6μm and propagated through dry air (with no absorption included in the simulation). The leading edge of this waveform exhibits non-sinusoidal oscillations — note the blunt peaks that appear due to carrier-wave shocking. The leading portion of the pulse propagates in a nearly z-invariant form: While it receives energy from off-axis directions due to self-focusing, it "emits" it in the longitudinal direction, thus preventing the run-away collapse. The high-frequency components evident in the trailing edge are the harmonics generated in the optical shock, now seen lagging behind due to their lower (as compared to fundamental) propagation velocity.

In the long-wavelength regime the interplay between the self-focusing nonlinearity and chromatic dispersion thus brings a new paradigm into optical filamentation. The high-frequency walk-off mechanism is capable of subtracting the focused pulsed beam energy at a rate that prevents the collapse altogether. Expressed in another way, the MKP equation predicts that the shock-forming singularity is approached before the spatial collapse occurs. The singularity avoidance happens thanks to the chromatic dispersion. The latter must be sufficiently small (otherwise we would be in the classical NLS-like regime) in order to allow shock formation in accompaniment to the generation of new spectral components, but it must not be too small in order to provide for sufficiently fast walk-off of higher frequencies. In this regime, the plasma generation is of significantly lesser importance than in the optical filaments created by 800nm laser pulses.[6]

2.3 Pulse propagation model for mid-infrared

It is obvious from the above that the mechanism relevant for the mid-infrared nonlinear optics is genuinely carrier-wave based. It is not the question of accuracy or of the model fidelity that MKP should be preferred over NLS in the mid-infrared. The crucial fact is that NLS, and in fact any envelope-based description, simply fails to capture the most basic mechanism that keeps the self-focusing in check. Only carrier resolving pulse propagation equations[13] can be utilized to model nonlinear processes at longer wavelengths.

The above described MKP equation is similarly robust as the NLS. In fact it can capture the qualitative behavior shown in Fig. 1 to such a degree that it may be difficult to decide if the MKP or a more realistic model was used in the simulation. Nevertheless, for a realistic experiment modeling only pulse evolution equations with minimal approximation must be used.[13] The generalized Unidirectional Pulse Propagation Equation (gUPPE) is one such model. In a symbolic form it can be written as follows:[14]

$$\partial_z E(\omega, x, y, z) = +\frac{i}{2\sqrt{L}} e^{-i\sqrt{L}z} \mathcal{F}\left\{ N\left(\mathcal{F}^{-1}\left\{ e^{+i\sqrt{L}z} E \right\}\right)\right\} \ .$$

This propagation system represents a large set of coupled differential equations for the spectral amplitudes $E(\omega, x, y, z)$ of the propagating optical waveform. It can be derived from the Maxwell equations with the minimal possible approximations, namely solely under assumption of the unidirectional propagation. This assumes that the nonlinear interaction, here described by the "operator" N returning the nonlinear polarization given the electric field history, is such that the new waves generated to propagate in the opposite direction than the incident pulse can be neglected. Then only the forward propagating wave needs to be simulated in the above form which explicitly separates the linear propagator from the nonlinear evolution. The former is symbolically written here in terms of the Helmholtz operator L. While in the homogeneous medium this reduces to the well known spectral-method propagation utilizing suitable spectral transforms in the space transverse to the direction of propagation,[13] in wave-guide structures $\sqrt{L}$ can be treated by a number of available beam-propagation techniques.[14] In such a case, L is linear and therefore diagonal in the frequency space, and the underlying BPM problems are decoupled between different frequencies. With sufficient computational resources, this technique allows extremely nonlinear pulse propagation simulation in waveguides with full spatial resolution.[15] Simulations in homogeneous media are usually less complex and require more modest efforts.

The important property of the gUPPE is that the linear and nonlinear properties of the simulated system are treated in a complete separation. In other words, the above equation remains valid for an arbitrary medium model represented in $N(E)$. This is why one can say that the pulse propagation side of the pulse-medium interaction problem has been solved, despite the fact that fully satisfactory models for the light-matter interactions are not yet available. This is the issue that we address in the following Section 3.

2.4 Highly nonlinear propagation and mid-infrared optical filaments

With the gUPPE model, we are in position to explore the novel regimes that appear in the mid-infrared wavelength region, and we can answer the question of what exactly are the consequences when the mathematical paradigm for pulse propagation switches from that defined by the Nonlinear Schödinger Equation to that underlined by the modified Kadomtsev-Petviashvili equation.

However, the first consideration is the simple argument concerning wavelength. Taking the scaling of the critical power for self-focusing and and the fact that longer wavelength diffracts faster, one can reasonably expect that for example optical filaments would be thicker in comparison with those created with the 800 nm lasers. Arguing along similar lines, the modulation instability driven by the Kerr nonlinearity becomes effectively weaker at long wavelengths. While these properties all point in the direction of mid-infrared filaments attaining coarser scales, the most important issue is that the collapse mechanism no longer needs to depend on the free electron generation and subsequent de-focusing action of the dilute plasma.

As a result of these trends, the predicted filament properties are strikingly different when one compares the well known near-infrared regime. First of all we should note that achieving the self-focusing collapse requires a significantly higher peak power, and pulses with sufficient energies are not yet widely available in the mid-infrared. Nevertheless, it it is interesting to look at what possibilities could be offered by the long-wavelength pulse propagation regime once truly high-intensity pulses become more common. Besides the obvious appeal of the delivery of high-intensity light on a distant target, theoretical exploration of the pulse propagation serves as a window into the physics, some aspects of which may turn out to be quite different from that we are familiar with in 800 nm wavelength pulses.

One specific aspect which we would like to emphasize in this contribution is that the pulse interactions at longer wavelength are in a well defined sense more nonlinear than those in the visible or near infrared regions. In the latter, the self-focusing Kerr effect is usually used to achieve very high light intensity, which in turn initiates the nonlinear dynamics resulting in harmonic generation, ionization, white-light generation etc. Interestingly, in the regimes that fall into the category described by the modified Kadomtsev Petviashvili equation, the self-focusing collapse is not a necessary condition to induce highly nonlinear behavior. To illustrate this, let us look at an example of a $\lambda = 4\mu$m pulse which can generate super-continuum spectrum, and deliver high-intensity, broad-band radiation at a target twice as far as the initial beam Rayleigh range:

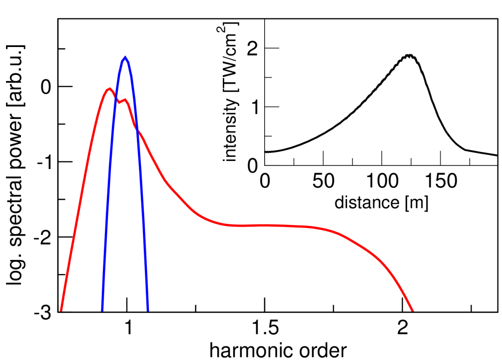

Figure 2. Super-continuum generation without self-focusing. Initially collimated, 100 fs duration, 40mJ energy, 4μm wavelength pule propagating in dry air. While the pulse energy and the corresponding peak power is insufficient to induce a true self-focusing collapse, the propagation can be still characterized as highly nonlinear: With the Rayleigh range of the initial beam around 75 m, high-intensity light is delivered to distances twice as far. At the same time, an extremely broad spectrum (super-continuum) develops as a result of the carrier-wave shock process described in Sec. 2.2.

Thus, even with the pulse energies that are not too high above what is currently available in a couple of research groups at the time of this writing, one can achieve highly nonlinear interactions even without creation of a classical (i.e. similar to those that occur in the near infra-red) filament.

However, should significantly higher pulsed power become available for ultra-short duration pulses, many interesting possibilities would open. As an example, Fig. 3 shows a long-distance optical filament held together by the dynamic balance between the self-focusing and high-frequency wave walk-off:

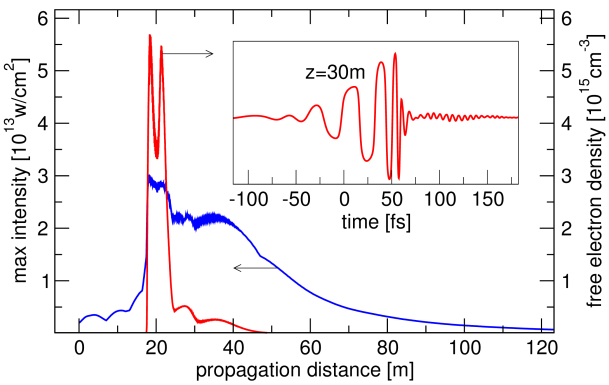

Figure 3. Optical filament formation in a very high energy optical pulse at $\lambda = 8\mu$m. Initially collimated, 350mJ pulse generates a high-intensity (left vertical axis) filament several tens of meters long, but without high free electron density (right vertical axis). Instead of plasma-induced de-focusing, the self-focusing collapse is kept in check by the generation of the high-frequency waves "ejected" through the trailing edge of the pulse (insert).

3. MODELING LIGHT-MATTER INTERACTIONS

The conventional approach in the modeling[7] of a medium exposed to strong femtosecond pulses was originally designed for pulse propagation equations written in terms of the optical field envelope.[8] This in turn was adopted from computational models in use since the typical pulse duration was in the pico- and even nano-second region. It has been recognized for some time that such models need improvement, especially in situations in which the carrier wave description becomes necessary.

Ideally, one would like to integrate into a single model system the Maxwell equations with the quantum-theory based description of the medium. Unfortunately such an approach is extremely demanding numerically. This is because for each and every spatial point resolved in the simulation of the optical field, one must evaluate the whole history of the quantum system and extract from it the medium polarization and/or induced current. This would of course imply that at a minimum a single-electron wavefunction in three spatial dimensions is calculated for all necessary times. In most situations, solution of only a single such system is by far more expensive than the whole of the optical pulse simulation. It is therefore very impractical; There are several proof of principle examples in the literature which demonstrate that a simultaneous simulation of a Maxwell-Schrödinger system[16–19] is at least conceptually possible and that it reproduces the basic features observed in experiments with optical filaments. However, to enable simulations on realistic scales corresponding to laboratory-scale NLO experiments, different methods must be devised, and a number of research groups pursue efforts in this direction (see e.g.[20–26]). Here we want to describe a method proposed recently, that aims to bypass the need for repetitive solution of quantum-system evolution. It is based on a one-time characterization of a given quantum model with the help of TDSE simulations. As the method utilizes Stark resonances for the description of both ionization and of the nonlinear polarization, it is termed the Metastable Electronic State Approach.[27]

The method starts with a TDSE calculation of Stark resonances for a chosen model system. For example a single-active-electron[28] model of an atom, exposed to a homogeneous external field is simulated for a range of field intensities. For each value of the latter, the least unstable resonance is found and its complex-valued energy and a generalized dipole moment[29] are evaluated and tabulated. The outcome of this stage is illustrated in Fig. 4. It shows the results for the nonlinear portion of the field-induced dipole moment in several noble gas atoms.

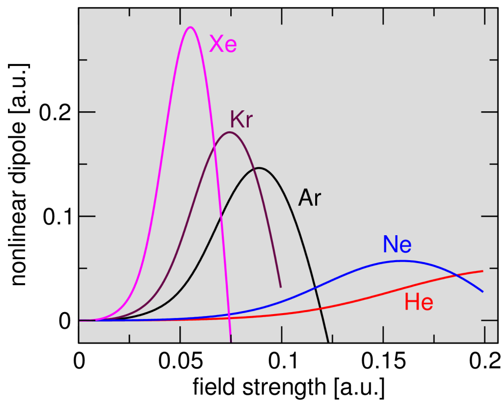

Figure 4. Nonlinear components of the dipole moment induced in a model atom by an external field. Single-active-electron approximation was utilized to obtain these functions. Having tabulated the nonlinear polarization over a relevant range of the field strengths, they are used to obtain the response in the optical pulses. While not directly used in MESA, nonlinear index can be estimated form these data sets — without adjustable parameters, the nonlinearity agrees with the experimental values to within 60-90 percent, depending on the species.[30] This is a clear indication that metastable states indeed carry a lot of relevant physical information.

It has been recognized for a long time that the complex energy of a Stark resonance can be used to approximate the ionization rate in an optical field, especially for longer wavelengths. Here we find that this is actually also true for the nonlinear polarization. This is illustrated in the following figure:

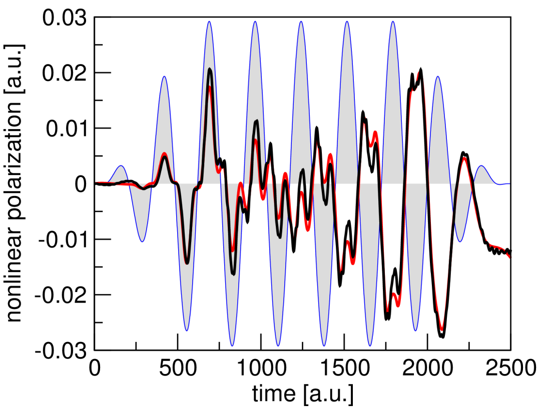

Figure 5. Nonlinear polarization induced in a Hydrogen-like quantum system by an excitation pulse depicted as a scaled shaded curve. The initial few cycles correspond to a situation with a lower light intensity, when the nonlinear response is initially dominated by self-focusing, and the response oscillates in phase with the driving field. At later times, the probability of ionization increases and the response is out of phase with the driving field, and is therefore de-focusing.

Without corrections, the MESA approach yields a quasi-static, or adiabatic description of both the nonlinear response and strong-field ionization. The resulting computational model has a structure similar to that in the conventional model and has therefore only slightly higher computational complexity.[27] Specifically, the ionization rate is obtained from the field-dependent imaginary part of the Stark-resonance energy, and the polarization response is evaluated from the tabulated nonlinear dipole moment. More importantly, the two quantities can be show to be connected by an exact relation.[31] As a consequence, the appropriate proportions between the ionization and self-focusing nonlinearity are naturally enforced. Comparisons with the time-dependent Schrödinger equations shows that this becomes accurate already for pulse wavelength longer than about two microns. Shorter-wavelength excitation modeling can be improved with post-adiabatic corrections which will be described in detail elsewhere.

To complete the description of the light-matter interaction for the purposes of simulation, the linear (w.r.t. the field strength) portion of the total dipole moment induced in the resonant states must be removed. This is to avoid double counting of a portion of the linear chromatic dispersion; In pulse propagation simulations, the latter is usually described in terms of Sellmeier-like function or tabulation. This naturally encompasses influence from a large number of transitions — it would be unrealistic to expect that our simple quantum model utilizing a single-electron approximation could capture the linear susceptibility (and its frequency dependence) with accuracy sufficient for realistic modeling. This is why only the nonlinear response is calculated from the metastable states, and the linear properties of the medium are "encoded" in the propagator L in the generalized unidirectional pulse propagation equation shown above.

Here we would like to show a simple illustration to demonstrate that the method naturally captures the dynamics of optical filamentation:

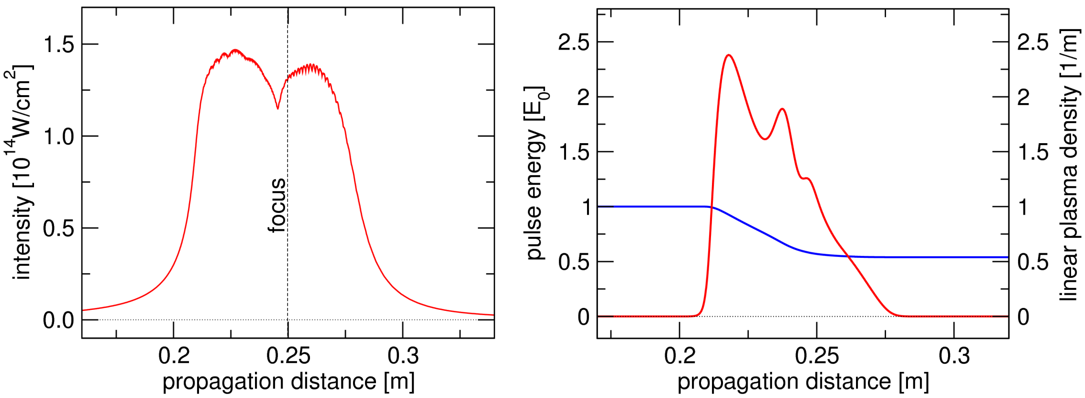

Figure 6. Femtosecond filament simulated with the MESA model based description of the light-medium interaction. The left panel shows the peak intensity versus the propagation distance in the vicinity of the linear beam focus. The right hand panel depicts the free electron density per unit length (right vertical axis, red line) and the pulse energy decreasing due to nonlinear losses. A Hydrogen-like atom model was used in this simulation. The description was completed by the linear frequency-dependent susceptibility taken as that of Argon.[32]

To conclude, the above example thus illustrates that our approach utilizing metastable electronic states can capture the filamentation dynamics with the similar computation effort as the conventional model. However, the crucial difference and advantage of the present method lies in the fact that all aspects of the nonlinear response are described in a unified way. In particular, we can by-pass the weakest aspect of the conventional model which is the uncertainty of the relative contribution of focusing and de-focusing effects.[33] MESA thus represents an important step toward the goal of fully self-consistent, first-principle based description of light-matter interactions in large-scale numerical simulations.

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