Development of nonperturbative nonlinear optics models including effects of high order nonlinearities and of free electron plasma. Part I: Maxwell-Schrödinger equations coupled with evolution equations for polarization effects. Part II: SFA-like nonlinear optics

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Abstract

This paper is dedicated to the exploration of non-conventional nonlinear optics models for intense and short electromagnetic fields propagating in a gas. When an intense field interacts with a gas, usual nonlinear optics models, such as cubic nonlinear Maxwell, wave and Schrödinger equations, derived by perturbation theory may become inaccurate or even irrelevant. As a consequence, and to include in particular the effect of free electrons generated by laser-molecule interaction, several heuristic models, such as UPPE, HOKE models, etc, coupled with Drude-like models [1], [2], were derived. The goal of this paper is to present alternative approaches based on non-heuristic principles. This work is in particular motivated by the on-going debate in the filamentation community, about the effect of high order nonlinearities versus plasma effects due to free electrons, in pulse defocusing occurring in laser filaments [3], [4], [5], [6], [7], [8], [9]. The motivation of our work goes beyond perturbative models, and is more generally related to models of interaction of any external intense and (short) pulse with a gas. In this paper, two different strategies are developed. The first one is based on the derivation of an evolution equation on the polarization, in order to determine the response of the medium (polarization) subject to a short and intense electromagnetic field. Then, we derive a combined semi-heuristic model, based on Lewenstein's SFA (Strong Field Approximation) model and the usual perturbative modeling in nonlinear optics. The proposed model allows for inclusion of high order nonlinearities as well as free electron plasma effects. We have developed alternative analytical techniques in order to properly include free electron and high order nonlinearity effects in polarization models.

Keywords: Nonlinear optics, laser, filamentation, Maxwell-Schrödinger

Contents

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1. Introduction

1.1. Introductory remarks

This paper is devoted to the derivation of non-standard nonperturbative nonlinear optics models for intense electromagnetic fields propagating in a gas. This work is strongly motivated by the recurrent debate in the nonlinear optics community, regarding appropriate choice of nonlinear optics models for modeling laser filaments, in particular appropriate models for high order nonlinearities and plasma of free electrons. These issues were already discussed in the celebrated papers [10], [11]. Over the years, several models were proposed and the interested reader can refer to several complete review papers [1], [2], [12]. Although, some ellaborated models (HOKE, UPPE [13], [14]) allow for accurate simulations and analysis of laser filamentation in several physical frameworks [13] , [15], [16], [9], [7], [17] at high intensity accurate modeling of the generation and evolution of plasma of free electron and nonperturbative nonlinearities, is still an open problem, which is in particular studied in [6] [18], [19], [3], [4], [8], [20].
The first approach which is proposed here, consists of an extension of a micro-macro model constituted by Maxwell's equations, ME's, and Schrödinger equations, TDSE's, modeling the nonlinear response of a gas to an electromagnetic field, see [21], [22] and [23]. From a practical point of view, this model confronts a major issue, which is the huge computational cost for computing the polarization using TDSE's. As a consequence realistic simulations are only possible on very short propagation distances, which makes this *ab-initio* model irrelevant for filamentation for instance. In order to reduce the overall computational complexity, a model is then proposed which is based on a transport-like equation, modeling the time evolution of polarization. This additional equation allows for a drastic reduction of the number of TDSE's to be solved, and then of the overall computational complexity of the numerical model. Although we still do not expect the model to be efficient enough for simulating filamentation, it may be a good candidate for a fundamental understanding of this phenomenon [1], [2]. This strategy is first developed in details for circularly polarized pulses and is valid for ultrashort pulses. Other strategies based on geometrical optics or Kirchoff's formula are also proposed for more general pulses.
In the second part of the paper, we propose to derive from the Strong Field Approximation (SFA) model [24], for intense laser-molecule interaction, a macroscopic nonperturbative nonlinear optics model. As first order nonlinearities play an important role in field propagation, Lewenstein's SFA model, which cannot be treated by 1-D models, is coupled with a traditional perturbative nonlinear optics model [25]. We will also show how far we can go in the explicit and rigorous derivation of the dipole moment including bound and continuous states, using the density matrix formalism.

1.2. Maxwell-Schrödinger model

We recall that the Maxwell-Schrödinger model developed in [21], [22] and [23] is based on the coupling of the 3D macroscopic ME's with many TDSE's under the dipole approximation (electric field is supposed to be constant in space at the molecular scale). This is valid when the smallest internal wavelengths $\lambda_{\min}$ of the electromagnetic field are much larger than the molecule size ℓ, that is $\ell \ll \lambda_{\min}$ (typically $\lambda_{\min} \approx 800nm$ where $\ell \approx 0.1nm$). We then define the ME's on a bounded space domain with a boundary Γ, and $\mathbf{x}' = (x', y', z')^T$ denotes the electromagnetic field space variable. At the molecular scale, and working under the Born-Oppenheimer approximation, we will denote by $\mathbf{x} = \Big(x, y, z\Big)^T$ the TDSE space variable (for electrons).

The molecular density is supposed to be constant in time, continuous in space, and is denoted by $\mathcal{N}(\mathbf{x}')$. The equations we consider are the following ones:

$$\left\{\begin{array}{lcl}\partial_t\mathbf{B}(\mathbf{x}',t) & = & -c\nabla\times\mathbf{E}(\mathbf{x}',t),\\ \partial_t\mathbf{E}(\mathbf{x}',t) & = & c\nabla\times\mathbf{B}(\mathbf{x}',t)-4\pi\big(\partial_t\mathbf{P}(\mathbf{x}',t)\big),\\ \nabla\cdot\mathbf{B}(\mathbf{x}',t) & = & 0,\\ \nabla\cdot\big(\mathbf{E}(\mathbf{x}',t)+4\pi\mathbf{P}(\mathbf{x}',t)\big) & = & e(\mathcal{N}_I-\mathcal{N}_e).\end{array}\right. \tag{1}$$

Polarization-TDSE is written as:

$$\left\{\begin{array}{lcl}\mathbf{P}(\mathbf{x}',t)=\mathcal{N}(\mathbf{x}')\sum_{i=1}^m\mathbf{P}_i(\mathbf{x}',t) & = & \mathcal{N}(\mathbf{x}')\sum_{i=1}^m\chi_{\Omega_i}(\mathbf{x}')\int_{\mathbb{R}^3}\psi_i(\mathbf{x},t)\mathbf{x}\psi_i^*(\mathbf{x},t)d\mathbf{x},\\ \mathtt{i}\partial_t\psi_i(\mathbf{x},t) & = & -\dfrac{\nabla_{\mathbf{x}}^2}{2}\psi_i(\mathbf{x},t)+\mathbf{x}\cdot\mathbf{E}_{\mathbf{x}_i'}(t)\psi_i(\mathbf{x},t)+V_c(\mathbf{x})\psi_i(\mathbf{x},t)\,\forall i\in\{1,..,m\}\end{array}\right. \tag{2}$$

where V_C denotes the "Coulomb" potential. Computation of the TDSE provides complete wavefunctions, including ionization, that is a continuum spectrum of free electrons propagating in a laser pulse. Such electrons can recombine with the parent ion with maximum energy $I_p+3.17U_p$, where I_p is the ionization potential and $U_p=E^2/4m\omega^2$, the ponderomotive energy acquired by a particle of mass m in a field E and frequency ω or with neighbours with energies exceeding $3U_p$, [26], [27]. In (1), Ω_i denotes the macroscopic spatial domain containing a molecule of reference associated to a wavefunction ψ_i, and $\mathbf{P}_i$ denotes the macroscopic polarization in this domain. In other words, Domain Ω_i contains $\mathcal{N}(\mathbf{x}')\mathrm{vol}(\Omega_i)$ molecules represented by one single eigenfunction ψ_i (under the assumption of a unique pure state). Naturally we have $\cup_{i=1}^m\Omega_i=\Omega$. We now assume that the spatial support of ψ_i is included in a domain $\omega_i\subset\mathbb{R}^3$, which is supposed to be *sufficiently large*. We allow free electrons to reach the boundary ω_i and we impose absorbing boundary conditions on $\partial\omega_i$. We refer to [23], [26] for a complete description of the geometry of this model. Functions χ_{Ω_i} are defined by $\chi\otimes\mathbf{1}_{\Omega_i}$ where χ is a plateau function and $\mathbf{1}_{\Omega_i}$ is the characteristic function of Ω_i. Finally $\mathbf{E}_{\mathbf{x}_i'}$ denotes the electric field (supposed constant in space) in Ω_i.

The overall complexity of this model is huge due to the very large number of TDSE which must be solved in order to get an accurate description of the medium response. Details can again be found in [26], [27]. In this paper we propose some models to reduce considerably the complexity of the ME-TDSE model. The principle is based on the derivation of a transport equation satisfied by the polarization vector, which will be coupled to ME's. Although the model still contains TDSE's, the evolution equation for the polarization, $\mathbf{P}$, allows to reduce drastically the number of TDSE's involved in the model. The most simple polarization evolution equation is an homogeneous transport equation. More accurate models are then proposed, including, in particular, the electric field variations. Models for circularly polarized electric pulses, Gaussian beams, as well as general electric fields are presented.

1.3. SFA model

The second strategy is based on the estimation of the medium polarization $\mathbf{P}(\mathbf{x}',t)$, modeling the response of a medium to an electromagnetic field, from molecular dipole moments, as a combination of contributions derived from Lewenstein's SFA model [24], and from usual perturbative modeling [25]. Classical nonlinear optics models are derived from ME's coupled with field-molecule TDSE's. The TDSE's are mathematically, solved using perturbation theory, allowing for explicit expressions of molecule dipole moments, then of the polarization. Although, this approach gives accurate nonlinear models for not too intense electromagnetic fields, when ionization occurs, these models can fail for precise description of all the complex nonperturbative nonlinear phenomena occurring during laser-molecule interactions (ATI, HHG, plasma of free electron generation, etc). Heuristic models are then derived [1], [2], in order to include, free electron contributions. The SFA model is derived from the transitions from free to ground states:

$$\psi_L(\mathbf{x},t)=e^{iI_pt}\Big(\phi_0(\mathbf{x})+\int d^3\mathbf{v}b(\mathbf{v},t)\mathbf{v}\Big)$$

for a ground state ϕ_0 and ionization potential I_p. It allows for an accurate modeling of laser-molecule interaction for intense pulses. The second part of this paper is then devoted to the derivation of a combined model, including the features of both the SFA model and the classical nonlinear optics perturbative models.

1.4. Organization of the paper

The paper is organized as follows. Section 2 is devoted to the modeling of the polarization, based on an evolution equation. More specifically, in Subsection 2.1 transport-like equations modeling the polarization vector evolution, are derived and are coupled to the ME's, for circularly ultrashort polarized pulses. The case of general 3-d electric fields is proposed in Subsection 2.2. In Appendix, other extensions are proposed, including an evolution equation of the polarization for Gaussian beams. Then, the methodology is extended for long pulses in Section 3, under the paraxial and slowly varying envelope approximations Subsection 3.1. Subsection 3.2 is dedicated to geometrical optics techniques for deriving a polarization equation. In Section 4, a combined SFA and perturbation approach is then presented. First, Subsection 4.1 is dedicated to the derivation of a Liouville-like equation for free and bound states. However, the complexity of the derived equations does not allow for an explicit computation of the matrix density, and of the dipole moment without using perturbation theory. Then Subsection 4.2 is devoted to SFA-like models including high order nonlinearities. Concluding remarks are proposed in Section 5.

2. Polarization evolution equation for very short pulses

2.1. Evolution equation on the dipole moment: circularly polarized field

We assume first that the external electromagnetic field is circularly polarized, which satisfies in vacuum, the following equations:

$$\left\{\begin{array}{rcl}
\partial_t E_{x'}(z',t) & = & -c^2\partial_{z'}B_{y'}(z',t)\\
\partial_t E_{y'}(z',t) & = & +c^2\partial_{z'}B_{x'}(z',t)\\
\partial_t B_{x'}(z',t) & = & +\partial_{z'}E_{y'}(z',t)\\
\partial_t B_{y'}(z',t) & = & -\partial_{z'}E_{x'}(z',t)
\end{array}\right. \tag{3}$$

A solution to these (Maxwell) equations is

$$\left\{\begin{array}{rcl}
E_{x'}(z',t) & = & E_0 f(kz'-\omega_0 t)\sin(kz'-\omega_0 t)\\
E_{y'}(z',t) & = & -E_0 f(kz'-\omega_0 t)\cos(kz'-\omega_0 t)\\
B_{x'}(z',t) & = & -B_0 f(kz'-\omega_0 t)\cos(kz'-\omega_0 t)\\
B_{y'}(z',t) & = & B_0 f(kz'-\omega_0 t)\sin(kz'-\omega_0 t)
\end{array}\right.$$

where f is the envelope of the initial electromagnetic field, $k=\omega_0/c$. In Fig. 1, we illustrate an example of a pulse we will consider, where we use atomic units: $E_0(a.u.) = 5\times 10^9 V\cdot cm^{-1}$ corresponding to $I = cE_0^2/8\pi = 3.5\times 10^{16} W\cdot cm^{-2}$, $T(a.u.) = 24\times 10^{-18}s = 24asec$, $a_0 = 0.0529nm$, $e=\hbar=m_e=1$. Within the ME-TDSE model, interaction with a molecule requires the solution to TDSE:

$$i\partial_t\psi = -\frac{1}{2}\triangle\psi + V_c(\mathbf{x})\psi + \mathbf{x}\cdot\mathbf{E}_{\mathbf{x}'}(t)\psi.$$

with $\mathbf{x}=(x,y)$, $\mathbf{E}_{\mathbf{x}'}=(E_{x'},E_{y'})$. In velocity gauge [28], this is written equivalently as:

$$i\partial_t\psi = -\frac{1}{2}\triangle\psi + V_c(\mathbf{x})\psi + i\mathbf{A}_{\mathbf{x}'}(t)\cdot\nabla\psi + \frac{\|\mathbf{A}_{\mathbf{x}'}(t)\|^2}{4}\psi.$$

with $\mathbf{A}_{\mathbf{x}'}=(A_{x'},A_{y'})$, $\mathbf{E}_{\mathbf{x}'}=-\partial_t\mathbf{A}_{\mathbf{x}'}$. The computation of the wavefunction ψ of a molecule "located in $\mathbf{x}'$", allows to deduce the dipole moment $\mathbf{d}$ as follows

$$\mathbf{d}(\mathbf{x}',t) = \int_{\mathbb{R}^2}|\psi(\mathbf{x},t)|^2\mathbf{x}dxdy$$

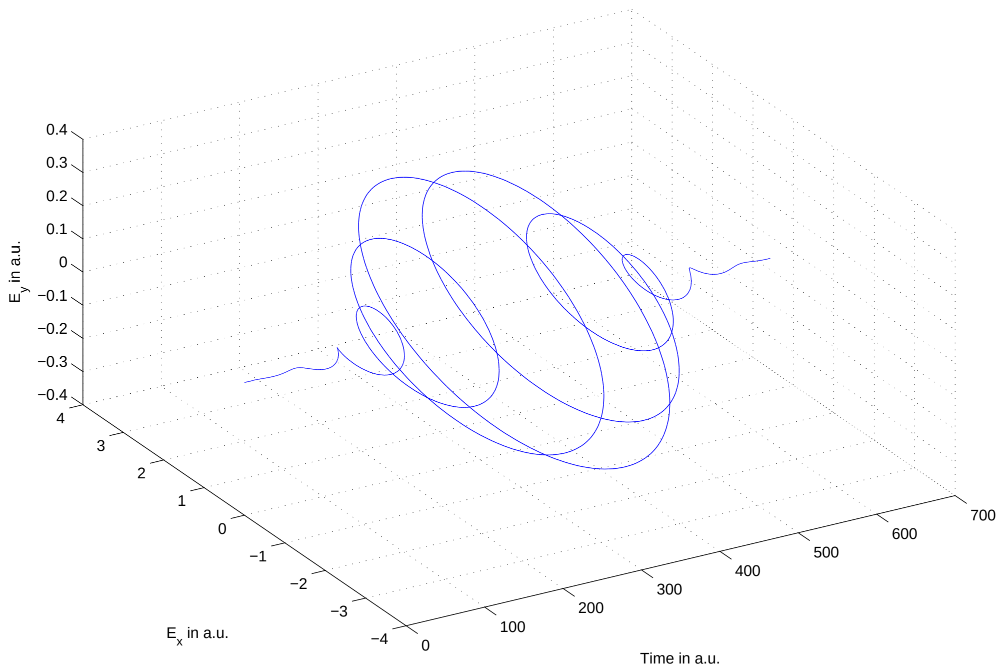

Figure 1: A circularly polarized pulse, $\mathbf{E}(x', y', t)$

Following [29] and [30], we now derive an evolution equation for $\mathbf{d}$. Assuming that the wavefunction of molecule m_1 "located" in $\mathbf{x}'_1 = (x'_1, y'_1, z'_1)$ is known, a sequence of evolution equations to estimate the dipole moment of molecule m_2 "located" in $\mathbf{x}'_2 = (x'_1, y'_1, z'_2)$ with $z'_2 > z'_1$, can be derived as follows.

Model 1. If $|\Delta z'| := |z'_2 - z'_1|$ is small enough, we may assume that a circularly polarized electromagnetic field interacting with molecule m_2 is almost identical (up to a time delay) to the one which molecule m_1 is subject to. Note that overall, we obviously do not assume that the electromagnetic field propagates as in a linear medium (or vacuum) in Maxwell's equations. In that case, let us define ψ_i the wavefunction of molecule m_i

$$i\partial_t \psi_i = -\frac{1}{2}\triangle\psi_i + V_c(\mathbf{x})\psi_i + \mathbf{x}\cdot\mathbf{E}_i(t)\psi_i.$$

and $\mathbf{d}(\mathbf{x}'_i, t)$ the corresponding dipole moment, with $i = 1, 2$. In addition $\mathbf{E}_i(t) = \mathbf{E}(\mathbf{x}'_i, t)$ denotes the electric field that molecule m_i is subject to. The above assumption mathematically implies: $\mathbf{E}_2(t) = \mathbf{E}_1\big(t - \Delta z'/v_g\big)$ and as a consequence

$$\mathbf{d}(\mathbf{x}'_2, t) = \mathbf{d}\Big(\mathbf{x}'_1, t - \frac{\Delta z'}{v_g}\Big) \tag{4}$$

where v_g is the group velocity (c in vacuum). Then the polarization $\mathbf{P}$ satisfies for $z' \in [z'_1, z'_2]$ the following transport equation

$$\partial_t \mathbf{P}(\mathbf{x}', t) + v_g \partial_{z'} \mathbf{P}(\mathbf{x}', t) = \mathbf{0} \tag{5}$$

with initial data $\mathbf{P}(\mathbf{x}'_1,) = \mathcal{N}_0 \mathbf{d}(\mathbf{x}'_1, \cdot)$. The model is then purely macroscopic (except for the computation of the initial data for $\mathbf{P}$), see Fig 2. *This model is applicable as long as $|\Delta z'|$ is small enough, or if the molecule* *density is small enough, that is as long as the effect of the medium on* $\mathbf{E}$ *during the pulse propagation from* $\mathbf{x}_1'$ *to* $\mathbf{x}_2'$ *is sufficiently negligible to not be included in the dipole moment calculation of molecule* m_2*.* In order to include medium effects on $\mathbf{E}$ in the propagation from $\mathbf{x}_1'$ to $\mathbf{x}_2'$ an improvement of the model is necessary. In particular, this approach will allow to consider larger propagation lengths.

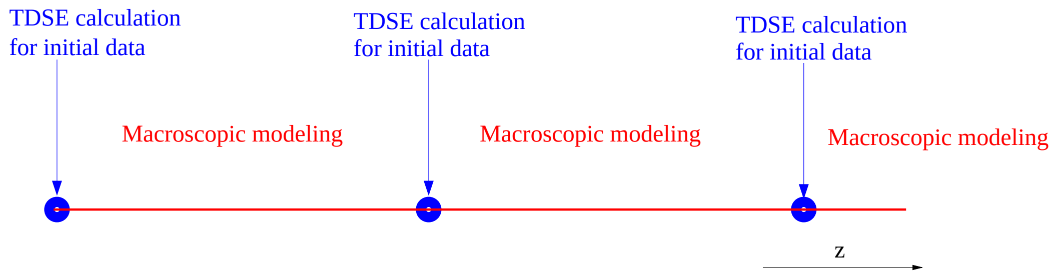

Figure 2: Macroscopic model with initial data from TDSE

Model 2. We now assume that the effect of the medium is sufficiently strong to make the assumption $\mathbf{E}_2(t) = \mathbf{E}_2\big(t - \Delta z'/v_g\big)$ inaccurate. In that case, we have to include $\mathbf{E}_2(t) - \mathbf{E}_1\big(t - \Delta z'/v_g\big)$ in the interaction of the field with molecule m_2. We define (see also the remark below about nonlinear modeling):

$$\Delta\boldsymbol{\mathcal{E}}(t) = \big(\Delta\boldsymbol{\mathcal{E}}^{(x)}(t), \Delta\boldsymbol{\mathcal{E}}^{(y)}(t)\big) := \mathbf{E}_2(t) - \mathbf{E}_1\Big(t - \frac{\Delta z'}{v_g}\Big)$$

Now assume that ψ_1 the solution to:

$$i\partial_t\psi_1 = -\frac{1}{2}\triangle\psi_1 + V_c(\mathbf{x})\psi_1 + \mathbf{x}\cdot\mathbf{E}_1(t)\psi_1$$

from which dipole moment $\mathbf{d}(\mathbf{x}_1', t)$, is obtained and that ii)

$$\left\{\begin{array}{l} \mathbf{c}^{(x)}(\mathbf{x}_1', t) = \int_{\mathbb{R}^2} x^2\mathbf{x}\big|\psi_1(\mathbf{x}, t)\big|^2 dxdy, \\ \\ \mathbf{c}^{(y)}(\mathbf{x}_1', t) = \int_{\mathbb{R}^2} y^2\mathbf{x}\big|\psi_1(\mathbf{x}, t)\big|^2 dxdy, \\ \\ \mathbf{c}^{(xy)}(\mathbf{x}_1', t) = \int_{\mathbb{R}^2} xy\mathbf{x}\big|\psi_1(\mathbf{x}, t)\big|^2 dxdy \end{array}\right. \tag{6}$$

Then, in order to solve:

$$i\partial_t\psi_2 = -\frac{1}{2}\triangle\psi_2 + V_c(\mathbf{x})\psi_2 + \mathbf{x}\cdot\mathbf{E}_2(t)\psi_2 \tag{7}$$

for $t \in [t_a, t_b]$, we solve, using an operator splitting method (from Trotter-Kato's formula)

$$\left\{\begin{array}{l} \left\{\begin{array}{ll} i\partial_t\psi_2 = -\frac{1}{2}\triangle\psi_2 + V_c(\mathbf{x})\psi_2 + \mathbf{x}\cdot\mathbf{E}_1\Big(t - \frac{\Delta z'}{v_g}\Big)\psi_2, & t \in [t_a, t_b^*], \\ \psi_2(\cdot, t_a) = \phi_0(\cdot) & \end{array}\right. \\ \\ \text{then} \\ \\ \left\{\begin{array}{ll} i\partial_t\psi_2 = \mathbf{x}\cdot\Delta\boldsymbol{\mathcal{E}}(t)\psi_2, & t \in [t_a, t_b], \\ \\ \psi_2(\cdot, t_a) = \psi_2(\cdot, t_b^*) & \end{array}\right. \end{array}\right.$$

where $t_b^* = t_b$. Using that

$$\psi_1\Big(\cdot, t_a - \frac{\Delta z'}{v_g}\Big) = \phi_0(\cdot)$$

The solution to this equation is approximated by:

$$\psi_2(\mathbf{x}, t_b) \approx \psi_1\Big(\mathbf{x}, t_b - \frac{\Delta z'}{v_g}\Big)\Big(1 - i\Delta T \mathbf{x}\cdot\Delta\boldsymbol{\mathcal{E}}(t_a)\Big)$$

where $\Delta T = t_b - t_a$. Note that the choice of approximating $\Delta\boldsymbol{\mathcal{E}}$ at t_a, is arbitrary (anytime time t in $[t_a, t_b]$ would be acceptable, in principle). That is

$$|\psi_2(\mathbf{x}, t_b)|^2 \approx \Big|\psi_1\Big(\mathbf{x}, t_b - \frac{\Delta z'}{v_g}\Big)\Big|^2\Big(1 + \Delta T^2\big(\mathbf{x}\cdot\Delta\boldsymbol{\mathcal{E}}(t_a)\big)^2\Big) \tag{8}$$

So that

$$\begin{aligned}\mathbf{d}(\mathbf{x}_2', t_b) \quad \approx \quad & \mathbf{d}\Big(\mathbf{x}_1', t_b - \frac{\Delta z'}{v_g}\Big) + \Delta T^2\Big(\big(\Delta\boldsymbol{\mathcal{E}}^{(x)}(t_a)\big)^2\mathbf{c}^{(x)}\Big(\mathbf{x}_1', t_b - \frac{\Delta z'}{v_g}\Big) \\ & + \big(\Delta\boldsymbol{\mathcal{E}}^{(y)}(t_a)\big)^2\mathbf{c}^{(y)}\Big(\mathbf{x}_1', t_b - \frac{\Delta z'}{v_g}\Big) + 2\Delta\boldsymbol{\mathcal{E}}^{(x)}(t_a)\Delta\boldsymbol{\mathcal{E}}^{(y)}(t_a)\mathbf{c}^{(xy)}\Big(\mathbf{x}_1', t_b - \frac{\Delta z'}{v_g}\Big)\Big)\end{aligned} \tag{9}$$

and from which we can evaluate $\mathbf{P}(\mathbf{x}_2', t_b) = \mathcal{N}_0(\mathbf{x}_2')\mathbf{d}(\mathbf{x}_2', t_b)$.

The operator splitting used above induces an error between the approximate $\psi_2^{(a)}(\cdot, t_b)$ (computed in (8)), and exact wavefunction $\psi_2^{(e)}(\cdot, t_b)$ solution to

$$i\partial_t\psi_2 = -\frac{1}{2}\triangle\psi_2 + V_c(\mathbf{x})\psi_2 + \mathbf{x}\cdot\mathbf{E}_2(t)\psi_2$$

is of the form $\mathcal{O}\Big((t_b - t_a)^2\mathbf{x}\cdot\Delta\boldsymbol{\mathcal{E}}(t_a)\triangle\psi(\cdot, t_a)\Big)$. As a consequence, we can evaluate the error between the exact polarization $\mathbf{P}^{(e)}(\mathbf{x}_2', t_b)$

$$\mathbf{P}^{(e)}(\mathbf{x}_2', t_b) = \mathcal{N}_0(\mathbf{x}_2')\mathbf{d}_2^{(e)}(\mathbf{x}', t) = \mathcal{N}_0(\mathbf{x}_2')\int_{\mathbb{R}^2}|\psi_2^{(e)}(\mathbf{x}, t)|^2\mathbf{x}dxdy$$

and approximate polarization $\mathbf{P}^{(a)}(\mathbf{x}_2', t_b)$ computed from (9).

Note that from a practical point of view this approximation is accurate in principle, for $(t_b - t_a)$ small enough as the splitting error leads to

$$\Big|\mathbf{P}^{(e)}(\mathbf{x}_2', t_b) - \mathbf{P}^{(a)}(\mathbf{x}_2', t_b)\Big| = \mathcal{O}\bigg(\mathcal{N}_0(\mathbf{x}_2')(t_b - t_a)^4\Big|\Delta\boldsymbol{\mathcal{E}}\Big|_\infty^2\bigg)$$

In practice however, $t_b - t_a$ is large, corresponding to the overall computational time for TDSE. Typically for a N_c-cycle pulse of wavelength λ, as $(t_b - t_a) \approx N_c\lambda/v_g$

$$\Big|\mathbf{P}^{(e)}(\mathbf{x}_2', t_b) - \mathbf{P}^{(a)}(\mathbf{x}_2', t_b)\Big| = \mathcal{O}\bigg(\mathcal{N}_0(\mathbf{x}_2')\Big[\frac{N_c\lambda}{v_g}\Big]^4\Big|\Delta\boldsymbol{\mathcal{E}}\Big|_\infty^2\bigg) \tag{10}$$

This makes, in practice, this approximation only relevant for very short pulses, or equivalently for $\Big|\Delta\boldsymbol{\mathcal{E}}\Big|_\infty$ small enough.

We deduce from this estimate that the polarization computed above is accurate as long as $\mathcal{N}_0(\mathbf{x}_2')\big(\Delta z/v_g\big)^4\big|\Delta\boldsymbol{\mathcal{E}}\big|_\infty^2$ is small enough. This means that: for low density medium and/or short enough propagation length, and/or small electric field variation approximation $\mathbf{P}^{(a)}(\mathbf{x}_2', t_b)$ is accurate.

Under the above assumptions, a macroscopic model can then be derived from the above calculus. We set

$$\mathbf{Q}^{(x,y,xy)}(\cdot,t) := \mathcal{N}_0(\cdot)\mathbf{c}^{(x,y,xy)}(\cdot,t)$$

and

$$\left\{\begin{array}{rcl} \partial_t\mathbf{P}(\mathbf{x}',t) + v_g\partial_z\mathbf{P}(\mathbf{x}',t) & = & \int_0^t \Big[\Delta\boldsymbol{\mathcal{E}}(\mathbf{x}',s'), \Delta\boldsymbol{\mathcal{E}}(\mathbf{x}',s')\Big]_{\mathbf{Q}} ds' \\ \\ \partial_t\mathbf{Q}(\mathbf{x}',t) + v_g\partial_z\mathbf{Q}(\mathbf{x}',t) & = & \mathbf{0} \end{array}\right. \tag{11}$$

where for $\mathbf{\Omega} \in M_{42}(\mathbb{R})$ such that

$$\mathbf{\Omega} = \left(\begin{array}{cc} \mathbf{\Omega}^{(x)} & \mathbf{\Omega}^{(xy)} \\ \\ \mathbf{\Omega}^{(xy)} & \mathbf{\Omega}^{(y)} \end{array}\right)$$

with $\mathbf{\Omega}^{(x,y,xy)}$ in $M_{21}(\mathbb{R})$, $[\cdot,\cdot]_{\mathbf{\Omega}} : M_{21}(\mathbb{R}) \times M_{21}(\mathbb{R}) \to M_{21}(\mathbb{R})$ is defined as follows. For $\mathbf{\Gamma}, \mathbf{\Delta} \in M_{21}(\mathbb{R})$:

$$\big[\mathbf{\Gamma},\mathbf{\Delta}\big]_{\mathbf{\Omega}} = \Delta_x\Gamma_x\mathbf{\Omega}^{(x)} + \Delta_y\Gamma_y\mathbf{\Omega}^{(y)} + \big(\Delta_x\Gamma_y + \Delta_y\Gamma_x\big)\mathbf{\Omega}^{(xy)}$$

The initial data (at $t = t_a$) satisfies

$$\left\{\begin{array}{rcl} \mathbf{P}(\mathbf{x}',t_a) & = & \mathbf{P}\Big(\mathbf{x}_1', t_a - \dfrac{z'-z_1'}{v_g}\Big) \\ \mathbf{Q}(\mathbf{x}',t_a) & = & \mathbf{Q}\Big(\mathbf{x}_1', t_a - \dfrac{z'-z_1'}{v_g}\Big) \end{array}\right. \tag{12}$$

with $\mathbf{Q}$ matrix function with values in $M_{42}(\mathbb{R})$

$$\mathbf{Q}(\mathbf{x}',t) = \left(\begin{array}{cc} \mathbf{Q}^{(x)}(\mathbf{x}',t) & \mathbf{Q}^{(xy)}(\mathbf{x}',t) \\ \\ \mathbf{Q}^{(xy)}(\mathbf{x}',t) & \mathbf{Q}^{(y)}(\mathbf{x}',t) \end{array}\right)$$

and

$$\Delta\boldsymbol{\mathcal{E}}(\mathbf{x}',t) = \mathbf{E}(\mathbf{x}',t) - \mathbf{E}\big(\mathbf{x}', t - \frac{\Delta z'}{v_g}\big)$$

This set of equations is then coupled to ME's (only): (11), (12), (1). From $\mathbf{x}_1'$ to $\mathbf{x}_2'$, the model is then purely *macroscopic*. Schrödinger's equation is only solved to determine the initial data of $\mathbf{P}$ and $\mathbf{Q}$ at $\mathbf{x}_1'$. This approach allows to take into account variations (including linear, nonlinear medium effects as well as diffraction) of electromagnetic field which may occur during the propagation from $\mathbf{x}_1'$ to $\mathbf{x}_2'$. Note that the RHS of (11), can in particular be interpreted as the polarization change due to diffraction, and medium nonlinear effects.

The evolution equation for $\mathbf{P}$ is then coupled to ME's (1) for modeling the propagation of the laser pulse over (z_1', z_2'). Polarization at (x', y', z') for $z' > z_2'$, is then computed again using the TDSE from (2). More specifically, the domain is decomposed in subdomains in the z' direction, and only at certain locations of each sudomain, the TDSE's are computed to evaluate the dipole moment. At other points, the macroscopic evolution equations on $\mathbf{P}$ are used. This methodology is summarized in Fig. 3, where the dipole

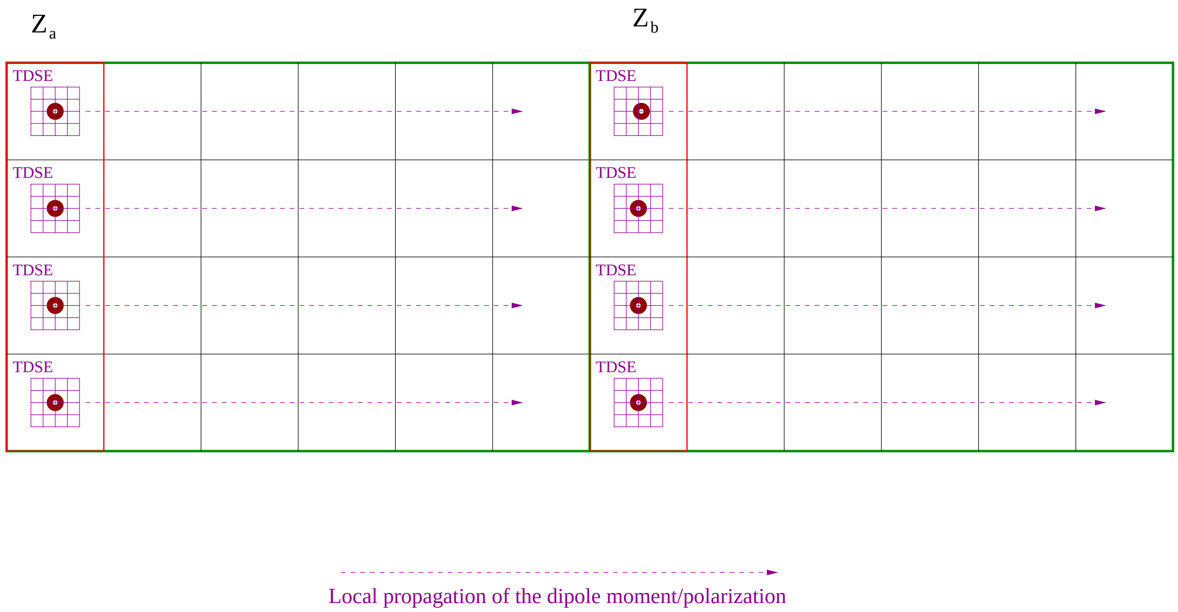

Figure 3: Spatial evolution of polarization

moment/polarization is computed from TDSE only for certain z' (Z_a, Z_b on the figure). Elsewhere, the evolution equation for $\mathbf{P}$, allows a "cheap" (as fully macroscopic) computational evaluation. Computational details can be found in [29], where this strategy is presented in 1-d dimension, and where it is shown that a reduction of almost two orders of magnitude in the computational complexity can be reached. From a practical point of view, the range of application of the presented approach is then limited only to very short pulses. We present in Appendix A, an extension of the method for Gaussian beams.

Remark 2.1 (Estimation of the group velocity). *In our models, to estimate the velocity group v_g in a given medium and for which n_0, n_2 are approximately known (from $\chi^{(1)}$, $\chi^{(3)}$), we use a standard approach. Start from $n \approx n_0 + n_2|\mathbf{E}|^2$, where n_0 (resp. n_2) is the linear (resp. second nonlinear) refractive index and n_g, the group velocity*

$$v_g = \frac{c}{n_g(\omega)} = \frac{c}{n(\omega) + \omega\dfrac{\partial n(\omega)}{\partial \omega}}$$

In first approximation, we can take $v_g \approx c/\sqrt{1+\chi^{(1)}}$, where $\chi^{(1)}$ is the instantaneous linear susceptibility. Note that a precise estimate of the group velocity is in fact not essential (except for Model 1), but allows for a reduction of $\left|\Delta\boldsymbol{\mathcal{E}}\right|_\infty$ and we can then expect a reduction of the splitting error, in particular when a low-order operator splitting is used (Model 2), and then a more accurate modeling of the polarization evolution.

We conclude this subsection by a remark regarding the backward propagation.

Remark 2.2. *As we are interested in multidimensional electromagnetic field propagation, it is important to mention that backward propagation should be included in the derivation of the polarization evolution equation. We should then derive a second PDE of the form*

$$\partial_{tt}\mathbf{P} - v_g^2 \triangle \mathbf{P} = \mathbf{S}(\mathbf{E})$$

with initial data to determine from quantum TDSE. This is possible extending the arguments developed above. This time, we will use: $\mathbf{P} \approx \chi^{(1)}\mathbf{E} + \chi^{(3)}\mathbf{E}^3$, where $\chi^{(1)}$ and $\chi^{(3)}$ are estimates of the first and third *instantaneous susceptibilities. That is*

$$\mathbf{E} \approx \frac{\mathbf{P}}{\chi^{(1)}} \frac{1}{1 + \dfrac{\chi^{(3)}}{\chi^{(1)}}|\mathbf{E}|^2} = \frac{\mathbf{P}}{\chi^{(1)}} \left(1 - \frac{\chi^{(3)}}{\chi^{(1)}}|\mathbf{E}|^2\right). \tag{13}$$

Then substituting $\mathbf{E}$ *from* (13)*, in the wave equation, and including the current density* $\mathbf{J}$*:*

$$\partial_t^2 \mathbf{E} - c^2 \triangle \mathbf{E} - \nabla(\nabla \cdot \mathbf{E}) \quad = \quad -4\pi(\partial_t^2 \mathbf{P} + \partial_t \mathbf{J})$$

Note that $\mathbf{J}$ *satisfies the following evolution equation [2], [21], where* ν_e *is the collision frequency:*

$$\partial_t \mathbf{J} + \nu_e \mathbf{J} \quad = \quad \frac{e^2 \rho}{m_e} \mathbf{E}$$

Thus, we get a general equation for $\mathbf{P}$*, which writes:*

$$\partial_t^2 \mathbf{P} - v_g^2 \triangle \mathbf{P} \quad = \quad -\frac{c^2 \chi^{(3)}}{\chi^{(1)}(1 + 4\pi\chi^{(1)})} \left[\triangle(\mathbf{P}|\mathbf{E}|^2) - \frac{1}{c^2}\partial_t^2(\mathbf{P}|\mathbf{E}|^2)\right] - \frac{4\pi\chi^{(1)}}{1 + 4\pi\chi^{(1)}}\partial_t \mathbf{J} \tag{14}$$

This equation is then coupled to the usual wave equation, and the initial data $\mathbf{P}(\cdot, 0)$*,* $\partial_t \mathbf{P}(\cdot, 0)$ *are computed from TDSE's, following a similar approach as above. Note that* (14) *is a general wave equation for* $\mathbf{P}$ *from which we can derive for instance* (5) *or* (11)*.*

To illustrate this model, its strengths and its limits, we propose a numerical example which consists of the comparison of harmonic spectra of an electric field solution to a non-homogeneous 1-d ME's, coupled with Schrödinger equations. Molecules are supposed to be aligned, and we compare the electric field spectra, when the response (polarization) of the medium is computed from, respectively, 1024, 256, 64, 16, 4 and 2 TDSE's. The physical data are as follows:

- $\mathcal{N}_0 = 1.2 \times 10^{-5}$ mol·(volume unit)$^{-1}$ in atomic unit.
- number of cycles ≈ 7, and wavelength $800nm$.
- the total propagation length is $\approx 30\mu m$, including $\approx 10\mu m$ in the gas.

As expected the spectra are quite close, even when the polarization is computed from 4 TDSE's versus 1024, see Fig. 4. In Fig. 5, we represent in logscale, the L^2-norm of the error with respect to the spectrum of reference (computed from 1024 TDSE's), as well as the estimation of the CPU/storage gain. Roughly, we can estimate that dividing by a factor N the number of the TDSE to solve (compared to a full Maxwell-Schrödinger model), allows to reduce by a factor N the overall computational complexity and data storage.

We now propose the same example (same data) except that the medium is 5 times denser that in the previous example. In that case, we expect that nonlinearities will deteriorate the numerical solution, see Fig. 6. In that case, a good representation of the medium requires more TDSE's, which increases the overall complexity of the simulation. More advanced models should then derived to include nonlinearities.

2.2. Generalization

This section is devoted to a generalization of the methodology which was developed above for circularly polarized and Gaussian fields (Appendix). We consider the full 3-D Maxwell equations coupled with TDSE's. We are first interested in computing the dipole moment of molecule m_α, located in $\mathbf{x}'_\alpha$ at time t. In this goal we have to evaluate the wavefunction ψ_α, solution to the following TDSE

$$i\partial_t \psi_\alpha(\mathbf{x}, t) = \Big(-\frac{1}{2}\triangle + V_c(\mathbf{x}) + \mathbf{x} \cdot \mathbf{E}_\alpha(t)\Big)\psi_\alpha(\mathbf{x}, t)$$

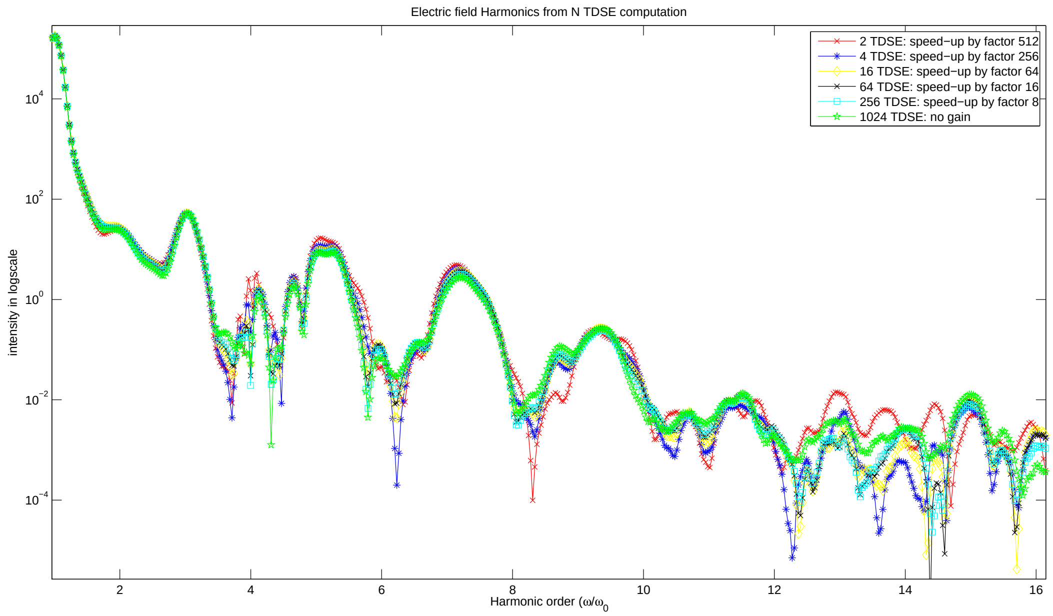

Figure 4: Electric field harmonic spectrum comparison for 4, 16, 64, 256 and 1024 TDSE's

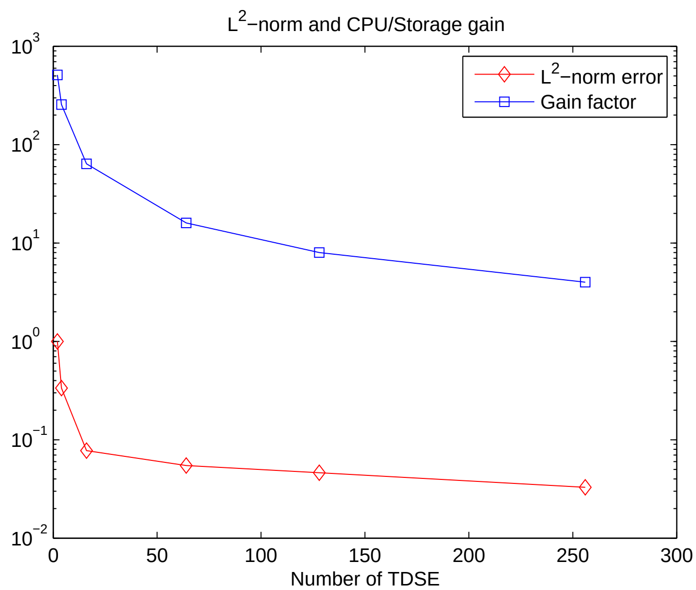

Figure 5: CPU/storage gain and L^2-norm error, as function of TDSE's

and deduce the corresponding dipole moment $\mathbf{d}(\mathbf{x}'_\alpha, t) = \mathbf{d}_\alpha(t)$. We assume that $\mathbf{E}_\alpha(t) \approx \sum_{i=1}^{L} \alpha_i \mathbf{E}_i(t-T)$ where T is some positive real number such that $t - T \geqslant 0$. This approximation comes typically from Kirchhoff's formula:

$$\mathbf{E}(\mathbf{x}'_\alpha, t) = \frac{1}{4\pi}\frac{\partial}{\partial t}\Big(t\int_{|\xi|=1}\mathbf{E}\big(\mathbf{x}'_\alpha + c(t-T)\xi\big)dS_\xi\Big) + \frac{t}{4\pi}\int_{|\xi|=1}\partial_t\mathbf{E}\big(\mathbf{x}'_\alpha + c(t-T)\xi\big)dS_\xi$$

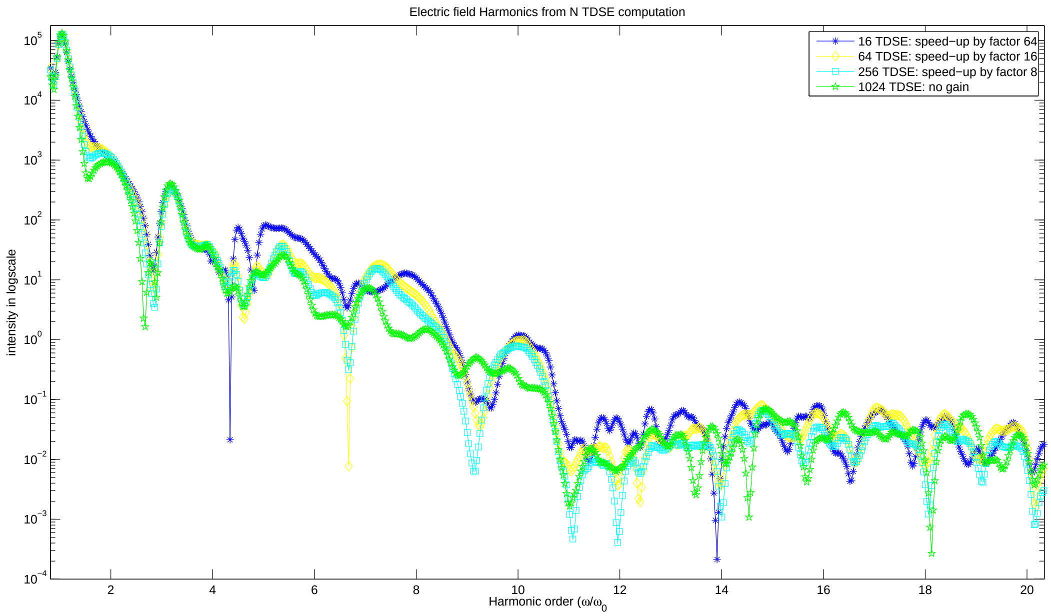

Figure 6: Electric field harmonic spectrum comparison, for 4, 16, 64, 256 and 1024 TDSE's.

We set $\chi(\cdot,t) := \sum_j^L \alpha_j\phi_j(\cdot,t-T)$ for $t \geqslant T$, where ϕ_j is solution to

$$i\partial_t\phi_j(\mathbf{x},t) = \Big(-\frac{1}{2}\triangle + V_c(\mathbf{x}) + \mathbf{x}\cdot\mathbf{E}_j(t)\Big)\phi_j(\mathbf{x},t)$$

which are assumed known at time $t-T$, as well as $\mathbf{d}_j(t-T)$. These are wavefunctions of molecules "located" on the sphere of radius $c(t-T)$ and center $\mathbf{x}'$ according to Kirchhoff's formula. Thus

$$i\partial_t\chi(\mathbf{x},t) = \Big(-\frac{1}{2}\triangle + V_c(\mathbf{x})\Big)\chi(\mathbf{x},t) + \sum_{j=1}^{L}\alpha_j\mathbf{x}\cdot\mathbf{E}_j(t-T)\phi_j(\mathbf{x},t-T)$$

that we rewrite

$$i\partial_t\chi(\mathbf{x},t) = \Big(-\frac{1}{2}\triangle + V_c(\mathbf{x}) + \mathbf{x}\cdot\mathbf{E}_\alpha(t)\Big)\chi(\mathbf{x},t) + F(\mathbf{x},t)$$

where

$$F(\mathbf{x},t) = \mathbf{x}\cdot\sum_{j=1}^{L}\alpha_j\big(\mathbf{E}_j(t-T) - \mathbf{E}_\alpha(t)\big)\phi_j(\mathbf{x},t-T)$$

We denote

$$i\partial_t\psi_\alpha(\mathbf{x},t) = \Big(-\frac{1}{2}\triangle + V_c(\mathbf{x}) + \mathbf{x}\cdot\mathbf{E}_\alpha(t)\Big)\psi_\alpha(\mathbf{x},t)$$

We denote $\mathbf{d}_\beta(t)$ the dipole moment associated to a virtual molecule of Wavefunction χ, that is:

$$\mathbf{d}_\beta(t) \quad = \quad \int_{\mathbb{R}^3}|\chi(\mathbf{x},t)|^2\mathbf{x}dxdydz = \sum_{i,j=1}^{L}\alpha_i\alpha_j\int_{\mathbb{R}^3}\phi_i(\mathbf{x},t-T)\bar{\phi}_j(\mathbf{x},t-T)\mathbf{x}dxdydz$$

Now we introduce $\mathbf{d}_\beta^{(c)}(t)$, $\mathbf{d}_\beta^{(u)}(t)$ such that

$$\mathbf{d}_\beta(t) = \mathbf{d}_\beta^{(c)}(t) + \mathbf{d}_\beta^{(u)}(t)$$

with

$$\begin{aligned}
\mathbf{d}_\beta^{(c)}(t) &= \textstyle\sum_{j=1}^L \alpha_j^2 \mathbf{d}_j(t-T) \\
\mathbf{d}_\beta^{(u)}(t) &= \textstyle\sum_{i\neq j; i,j=1}^L \alpha_i\alpha_j \int_{\mathbb{R}^3} \phi_i(\mathbf{x}, t-T)\bar{\phi}_j(\mathbf{x}, t-T)\mathbf{x}dxdydz
\end{aligned}$$

We also have

$$\psi_\alpha(\mathbf{x}, t) = \chi(\mathbf{x}, t) + \varepsilon_\alpha(\mathbf{x}, t)$$

where ε_α satisfies the equation

$$i\partial_t \varepsilon_\alpha(\mathbf{x}, t) = \big(-\frac{1}{2}\triangle + V_c(\mathbf{x}) + \mathbf{x}\cdot\mathbf{E}_\alpha(t)\big)\varepsilon_\alpha(\mathbf{x}, t) - F(\mathbf{x}, t)$$

with null initial condition.

$$\varepsilon_\alpha(\mathbf{x}, t) = -i\int_T^t \Big[\big(-\frac{1}{2}\triangle + V_c(\mathbf{x}) + \mathbf{x}\cdot\mathbf{E}_\alpha(s)\big)\varepsilon_\alpha(\mathbf{x}, s)\Big]ds + i\int_T^t F(\mathbf{x}, s)ds$$

In the following, we denote

$$\eta_j(\mathbf{x}, t) := i\int_T^t \big(\mathbf{E}_j(s-T) - \mathbf{E}_\alpha(s)\big)\phi_j(\mathbf{x}, s-T)ds$$

so that

$$\int_T^t F(\mathbf{x}, s)ds = \sum_{j=1}^L \alpha_j \mathbf{x}\cdot\eta_j(\mathbf{x}, t)$$

We now use the approximation

$$\psi_\alpha(\mathbf{x}, t) \approx \chi(\mathbf{x}, t) + \sum_{j=1}^L \alpha_j \mathbf{x}\cdot\eta_j(\mathbf{x}, t)$$

and

$$\begin{aligned}
\mathbf{x}(t) &= \textstyle\sum_{i,j=1}^L \alpha_i\alpha_j \int_{\mathbb{R}^3} \big(\mathbf{x}\cdot\eta_j(\mathbf{x}, t)\big)\big(\mathbf{x}\cdot\bar{\eta}_i(\mathbf{x}, t)\big)\mathbf{x}dxdydz \\
&\quad -\textstyle\sum_{j=1}^L \alpha_j \int_{\mathbb{R}^3} \mathbf{x}\cdot\big(\eta_j(t)\bar{\chi}(t) + \bar{\eta}_j(t)\chi(t)\big)\mathbf{x}dxdydz
\end{aligned}$$

which is again justified by the assumption of non-interaction of electrons attached to distinct molecules.

$$\mathbf{d}_\alpha(t) = \mathbf{d}_\beta^{(c)}(t) + \mathbf{d}_\beta^{(u)}(t) + \mathbf{x}(t)$$

Now we argue that in first approximation $\mathbf{x}$ and $\mathbf{d}_\beta^{(u)}$ can be neglected. Indeed, the Maxwell-Schrödinger model is derived assuming that molecules m_i and m_j for $i \neq j$, do not interact, that is $\langle\phi_i, \phi_j\rangle = \langle\eta_i, \eta_j\rangle = 0$. That is

$$\mathbf{d}_\alpha(t) \approx \mathbf{d}_\beta^{(c)}(t) = \sum_{j=1}^L \alpha_j^2 \mathbf{d}_i(t-T)$$

This equation leads to an expression of the polarization of the form

$$\mathbf{P}(\mathbf{x}'_\alpha, t) \approx \sum_{j=1}^L \alpha_j^2 \mathbf{d}(\mathbf{x}'_i, t-T)$$

where the $\mathbf{x}_i$ lie on a sphere of radius cT and center $\mathbf{x}'_\alpha$. From a practical point of view, this relation is of little interest, as the dipole moment computation in one single location requires computations in several loci. However, from a more theoretical view point, it gives interesting information about the global picture.

3. Polarization evolution equation for longer pulses

The next two subsections are dedicated to the derivation of an evolution equation for the polarization under the paraxial and slowly varying envelope approximations. We then no more assume that the pulse duration is ultrashort.

3.1. General model under the paraxial approximation

The interaction of the laser field with the medium breaks the possible symmetry of the incoming pulse. We here propose an extension of Subsection 2.1, when paraxial approximation is assumed [31]. We write the electric field propagation in direction $\mathbf{e}_{z'}$ as

$$\mathbf{E}(\mathbf{x}'_{\perp}, z', t) = A(\mathbf{x}'_{\perp}, z', t)e^{i(k_{z'}z'-\omega t)}\mathbf{e}_{z'}$$

Say at time t_a and in $(\mathbf{x}'_{\perp,1}, z'_1)$, the initial polarization $\mathbf{P}(\mathbf{x}'_{\perp,1}, t_a)$ and its derivative $\partial_t\mathbf{P}(\mathbf{x}'_{\perp,1}, t_a)$, will be computed from a TDSE

$$i\partial_t\psi_1 = -\frac{1}{2}\triangle\psi_1 + V_c(\mathbf{x})\psi_1 + \mathbf{x}\cdot\mathbf{E}(t)\psi_1$$

with initial data $\psi_1(t = t_a) = \phi$. Now we seach for an evolution equation for $\mathbf{P}$ starting from (14) but under the paraxial approximation. In this goal we search for $\mathbf{P}$ and $\mathbf{J}$ in the form

$$\mathbf{P}(\mathbf{x}'_{\perp}, z', t) = \Pi(\mathbf{x}'_{\perp}, z', t)e^{i(k_{z'}z'-\omega t)}\mathbf{e}_{z'} \qquad (15)$$

$$\mathbf{J}(\mathbf{x}'_{\perp}, z', t) = \Lambda(\mathbf{x}'_{\perp}, z', t)e^{i(k_{z'}z'-\omega t)}\mathbf{e}_{z'} \qquad (16)$$

where A, Π, Λ are the slowly varying complex amplitudes. We intend to rewrite the wave equation coupled with (14) under the SVAE. We first get

$$\left\{\begin{array}{lcl} \triangle\mathbf{E} & = & \big(\triangle_{\perp}A + \partial^2_{z'}A + 2ik_{z'}\partial_{z'}A - k^2_{z'}A\big)e^{i(k_{z'}z'-\omega t)}\mathbf{e}_{z'} \\ \partial^2_t\mathbf{E} & = & \big(\partial^2_t A - 2i\omega\partial_t A - \omega^2 A\big)e^{i(k_{z'}z'-\omega t)}\mathbf{e}_{z'} \end{array}\right.$$

Now as

$$\partial_t\mathbf{J} = \big(\partial_t\Lambda - i\omega\Lambda\big)e^{i(k_{z'}z'-\omega t)}\mathbf{e}_{z'}$$

the continuity equation can be rewritten

$$\partial_t\Lambda = (i\omega - \nu_e)\Lambda + \frac{e^2\rho}{m_e}A$$

Thus we also have

$$\partial_t\mathbf{J} = \Big(-\nu_e\Lambda + \frac{e^2\rho}{m_e}A\Big)e^{i(k_{z'}z'-\omega t)}\mathbf{e}_{z'}$$

We now rewrite the nonlinearity in (14) (RHS), as follows

$$\begin{array}{rcl} \big(\partial^2_t - c^2\triangle\big)\big(\mathbf{P}|\mathbf{E}|^2\big) & = & \Big(\partial^2_t\big(e^{i(k_{z'}z'-\omega t)}\Pi|A|^2\big) - c^2\triangle_{\perp}(\Pi|A|^2) \\ & & -c^2\partial^2_{z'}\big(e^{i(k_{z'}z'-\omega t)}\Pi|A|^2\big)\Big)e^{i(k_{z'}z'-\omega t)}\mathbf{e}_{z'} \end{array}$$

Now we rewrite

$$\begin{cases} \triangle_\perp\big(\Pi|A|^2\big) & = & |A|^2\triangle_\perp\Pi+\Pi\triangle_\perp|A|^2+2\nabla_\perp|A|^2\cdot\nabla_\perp\Pi \\ \partial^2_{z'}\big(e^{i(k_{z'}z'-\omega t)}\Pi|A|^2\big) & = & e^{i(k_{z'}z'-\omega t)}\Big(-k^2\Pi|A|^2+2ik|A|^2\partial_{z'}\Pi+2ik\Pi\partial_{z'}|A|^2 \\ & & +2\big(\partial_{z'}|A|^2\big)\big(\partial_{z'}\Pi\big)+\Pi\partial^2_{z'}|A|^2+|A|^2\partial^2_{z'}\Pi\Big) \\ \partial^2_t\big(e^{i(k_{z'}z'-\omega t)}\Pi|A|^2\big) & = & e^{i(k_{z'}z'-\omega t)}\Big(-\omega^2\Pi|A|^2-2i\omega|A|^2\partial_t\Pi-2i\omega\Pi\partial_t|A|^2 \\ & & +2\big(\partial_t|A|^2\big)\big(\partial_t\Pi\big)+\Pi\partial^2_t|A|^2+|A|^2\partial^2_t\Pi\Big) \end{cases}$$

We now apply the SVEA that is

$$\begin{cases} \big|\partial^2_{z'}A\big| & \ll & k_{z'}|\partial_{z'}A| & \ll & k^2_{z'}|A| \\ \big|\partial^2_{z'}\Pi\big| & \ll & k_{z'}|\partial_{z'}\Pi| & \ll & k^2_{z'}|\Pi| \\ \big|\partial^2_t A\big| & \ll & \omega|\partial_t A| & \ll & \omega^2|A| \\ \big|\partial^2_t\Pi\big| & \ll & \omega|\partial_t\Pi| & \ll & \omega^2|\Pi| \end{cases}$$

Then, we get a full model.

$$\begin{cases} \partial_t A+c\partial_{z'}A & = & \dfrac{ic}{2k_{z'}}\triangle_\perp A+4\pi\left(-\partial_t\Pi+\dfrac{ik_{z'}c}{2}\Pi+\dfrac{i\nu_e}{2\omega}\Lambda-\dfrac{i\rho}{\omega}A\right) \\ \partial_t\Pi+\dfrac{c}{1+4\pi\chi^{(1)}}\partial_{z'}\Pi & = & \dfrac{ic}{2k_{z'}\big(1+4\pi\chi^{(1)}\big)}\triangle_\perp A-4\pi\chi^{(1)}\Pi+\dfrac{2i\pi\chi^{(1)}\nu_e}{\big(1+4\pi\chi^{(1)}\big)ck_{z'}}\Lambda-\dfrac{2i\pi\chi^{(1)}\rho}{\big(1+4\pi\chi^{(1)}\big)ck_{z'}}A \\ & & -\dfrac{2ic}{k_{z'}\big(1+4\pi\chi^{(1)}\big)}\dfrac{\chi^{(3)}}{\chi^{(1)}}\Big[|A|^2\triangle_\perp\Pi+\Pi\triangle_\perp|A|^2+2(\nabla_\perp|A|^2)\cdot(\nabla_\perp\Pi)\Big] \\ & & -\dfrac{\chi^{(3)}}{\chi^{(1)}\big(1+4\pi\chi^{(1)}\big)}\big[|A|^2(\partial_t\Pi+c\partial_{z'}\Pi)+\Pi(\partial_t|A|^2+c\partial_{z'}|A|^2)\big] \\ \partial_t\Lambda & = & (i\omega-\nu_e)\Lambda+\rho A \end{cases} \tag{17}$$

The interest of this model is that it gives an accurate description of the polarization envelope (then of the electric field envelope). Again, the evolution equation on Π is used, only in a localized spatial region, and from initial data $\mathbf{P}(\cdot,t_a)$, $\partial_t\mathbf{P}(\cdot,t_a)$, computed from TDSEs following the same technique as the one described in Section 2, except that we can now consider much larger propagation distances. Naturally from (17) it is possible to derive more simple models neglecting certains terms of the RHS.

3.2. Geometrical optics approach (two-dimensional case)

We here discuss an approach based on the geometrical optics techniques which allows to derive a more simple model than (17). We will follow strategy and notation from [32], in order to reduce TDSE computations. Starting first from $\mathbf{E}(\mathbf{x}',t)=\mathrm{Re}\big[A(\mathbf{x}',t)e^{i(k_0S_E(\mathbf{x}')-\omega t)}\big]\mathbf{e}_{z'}$ then working in the moving frame $z'\leftarrow z'$ and $t'\leftarrow t'-z'/v_g$, allows to get ride of the time dependence in the envelope calculation, with $\mathbf{x}'=(x',z')$

$$\mathbf{E}(\mathbf{x}')=A(\mathbf{x}')e^{ik_0S_E(\mathbf{x}')}\mathbf{e}_{z'}$$

which satisfies the following Helmholtz equation

$$\nabla^2\mathbf{E}(\mathbf{x}')+k^2(\mathbf{x}')\mathbf{E}(\mathbf{x}')=\mathbf{0}$$

with $k^2(\mathbf{x}') = k_0^2\big(1 + F(|U(\mathbf{x}')|^2)\big)$, and F a function, modeling to the medium response to the electric field. Note that F null corresponds to a pulse propagation in vacuum. The eikonal and transport equations which follow, by identifying terms in k_0 and k_0^2, are

$$\left\{\begin{array}{lcl} \nabla S_{\mathbf{E}}(\mathbf{x}')\cdot\nabla S_{\mathbf{E}}(\mathbf{x}') & = & 1 + F\big(A^2(\mathbf{x}')\big), \\ \\ 2\nabla S_{\mathbf{E}}(\mathbf{x}')\cdot\nabla A(\mathbf{x}') + A(\mathbf{x}')\nabla^2 S_{\mathbf{E}}(\mathbf{x}') & = & 0 \end{array}\right.$$

Then, under the paraxial approximation, the envelope can be rewritten $U(\mathbf{x}') = A(\mathbf{x}')e^{ik_0 S_U(\mathbf{x}')}$, assuming $k_0 \gg 1$ and $S_U = S_{\mathbf{E}} - z'$ where $S_U(\mathbf{x}') = F\big(|U_0|^2\big)z'$. Ray trajectories are given by $\Big\{\big(\mathbf{x}'(\sigma),\sigma\big), \sigma \in \mathbb{R}\Big\}$, where

$$\frac{d\mathbf{x}'}{d\sigma}(\sigma) = \nabla S_U\big(\mathbf{x}'(\sigma)\big)$$

According again to [32], we can assume $z' \approx \sigma$ (high power beam) so that we can parameterize the rays in z' (direction of propagation of the pulse).

The main idea is now to use rays in order to reduce the TDSE computations.

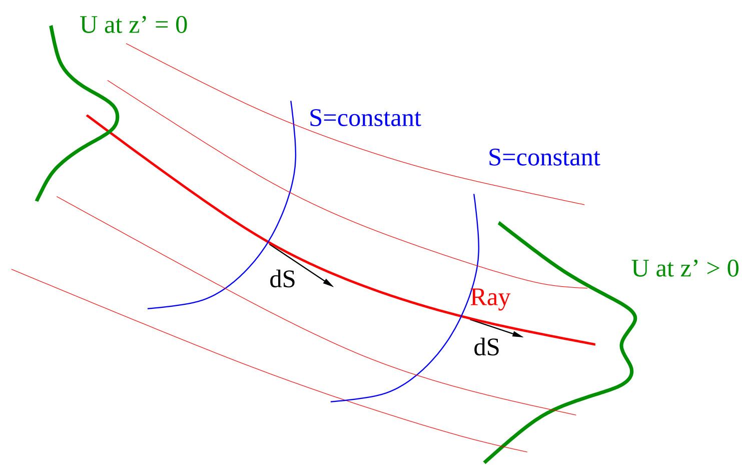

Figure 7: Ray path

1. The starting point is to model $S_U(\mathbf{x}') = z' + F\big(|U(\mathbf{x}')|^2\big)$, where F is a medium dependent function, modeling the Kerr effect (self-focusing), such that:

$$F(|U|^2) = \chi^{(3)}|U|^2$$

The susceptibility $\chi^{(3)}$ is medium, as well as time and space dependent. In first approximation $\chi^{(3)}$ can be taken constant. However, it is possible to more precisely determine its value *via* TDSE computation [3]. We can rewrite F_α, such that

$$F_\alpha(|U|^2) = \chi_\alpha^{(3)}|U|^2$$

where $\chi_\alpha^{(3)}$ is computed from a TDSE for a molecule "located" at $(x'_\alpha, 0)$. In vacuum, F is set to zero.

2. Determine the ray trajectories assuming that $z' \approx \sigma$:

$$\frac{dx'}{dz'}(z') = -\frac{1}{2}\partial_{x'} S_U\big(x'(z'), z'\big) \qquad (18)$$

with $x'(0) = x'_0$ given.

3. Determine A along the rays, from the transport equation

$$\frac{dA}{dz'}\big(x'(z')\big) = -\frac{1}{2}A\big(x'(z')\big)\partial_{x'}S_U\big(x'(z'),z'\big) \tag{19}$$

   From there, U is deduced along the rays: $U\big(x'(z'),z'\big) = A\big(x'(z'),z'\big)e^{ik_0S_U\big(x'(z'),z'\big)}$. In practice, the electric field will be computed from ME. However, this information is relevant from a practical point of view, in order to estimate the polarization.
4. We assume that for a molecule "located" at $\mathbf{x}'_\alpha$, there exists a trajectory $\big\{\mathbf{x}'_\alpha(\sigma),\, \sigma \geqslant 0\big\}$, passing through that point. Except in vacuum, $U(\mathbf{x}') = U\big(\mathbf{x}'_\alpha(\sigma)\big) \neq U\big(x'_\alpha,0)\big)$. In the moving frame, the electric field the molecule is subject to, is identical to the one applied to a molecule "located" at $(0,z'_\beta)$, with $z'_\beta \leqslant z'_\alpha$. More specifically, there exists a level set denoted by $\mathcal{C}_\alpha(\sigma)$, of normal vector $\nabla S_U\big(\mathcal{C}(\sigma)\big)$ and passing though $\mathbf{x}'_\alpha$, which intersects the line $x'=0$, at say z'_β. Along this curve, the electric field is constant, and in particular: $\mathbf{E}\big(\mathbf{x}'_\alpha(\sigma)\big) = \mathbf{E}\big(0,z'_\beta\big)$ as well as $\mathbf{P}\big(\mathbf{x}'_\alpha(\sigma)\big) = \mathbf{P}\big(0,z'_\beta\big)$. We may assume that $\mathbf{P}\big(0,z'_\beta\big)$ was evaluated from a direct TDSE computation. From this remark, we can construct a continuous equation, modeling the time evolution of the polarization. As along $\mathcal{C}_\alpha(\sigma)$, the polarization is constant, we obviously get:

$$\partial_\sigma\Big\{\mathbf{P}\big(\mathcal{C}(\sigma)\big)\Big\} = \mathbf{0}$$

   By assumption $z' \approx \sigma$, so that

$$\partial_{z'}\Big\{\mathbf{P}\big(\mathcal{C}(z')\big)\Big\} \;=\; \frac{d\mathcal{C}(z')}{dz'}\cdot\nabla\mathbf{P}\big(\mathcal{C}(z')\big) \;=\; \mathbf{0}$$

   or equivalently in the moving frame

$$\partial_{z'}\mathbf{P}\big(\mathcal{C}(z')\big) + \frac{d\mathcal{C}_{x'}(z')}{dz'}\partial_{x'}\mathbf{P}\big(\mathcal{C}(z')\big) \;=\; \mathbf{0}$$

   In the fixed frame we get

$$\partial_t\mathbf{P}\big(\mathcal{C}(z'),t\big) + v_g\partial_{z'}\mathbf{P}\big(\mathcal{C}(z'),t\big) + v_g\frac{d\mathcal{C}_{x'}(z')}{dz'}\partial_{x'}\mathbf{P}\big(\mathcal{C}(z'),t\big) \;=\; \mathbf{0} \tag{20}$$

   The equation models, along the level sets, the evolution of the polarization, taken into account, the propagation and nonlinear effects. Diffraction effects are here assumed negligible.

From the above analysis we can then determine the rays, $\mathbf{x}'(\sigma)$, as well as $U\big(\mathbf{x}'(\sigma)\big)$ and $S_U\big(\mathbf{x}'(\sigma)\big)$ (U and S_U along the rays), starting from any $(\mathbf{x}^{'}_\perp,0)$. We denote by $\mathbf{x}'_\alpha(\sigma)$ the ray:

$$\frac{d\mathbf{x}'_\alpha}{d\sigma}(\sigma) = \nabla S_U\big(\mathbf{x}'(\sigma)\big), \qquad \mathbf{x}'_\alpha(0) = (\mathbf{x}'_{\perp,\alpha},0)$$

At for any $\mathbf{x}'$ such that $\mathbf{x}' = \mathbf{x}'_\alpha(\sigma)$, a molecule located at $\mathbf{x}'$, will be subject to the field

$$\mathbf{E}(\mathbf{x}',t) = U\big(\mathbf{x}'_\alpha(\sigma),t\big)\cos\Big(\omega t - k_0S_U\big(\mathbf{x}'_\alpha(\sigma)\big)\Big)\mathbf{e}_{z'}$$

For $(0,z'_\beta)$, such that $U\big(\mathbf{x}'_\alpha(\sigma),t\big) = U\big((0,z'_\beta),t\big)$, then

$$\mathbf{E}(\mathbf{x}',t) = U\big((z^{'}_\beta,0)\big)\cos\big(\omega t - k_0S_U\big((0,z'_\beta)\big)\big)\mathbf{e}_{z'}$$

From a practical view point, it is then necessary to evaluate the level sets $\mathcal{C}(\sigma)$. in order to solve (20). In conclusion, geometrical optics is not used here to directly update the electric field, but (only) to determine the nonlinear response of the medium in the wave equation, without a direct TDSE computation. Note that in vacuum, $F=0$, $dA\big(\mathbf{x}'(\sigma)\big)/d\sigma \approx 0$, that is A is constant along the ray ($= A\big(\mathbf{x}'(0)\big)$) and only the phase $S_U\big(\mathbf{x}'(\sigma)\big)$ evolves.

3.3. Polarization reconstruction

Polarization is often deduced from TDSE in the hypothesis of a unique pure state, that is at $(\mathbf{x}', t)$, polarization is given by

$$\mathbf{P}(\mathbf{x}', t) = \mathcal{N}_0(\mathbf{x}')\mathbf{d}(\mathbf{x}', t)$$

where $\mathbf{d}(\mathbf{x}', t)$ is the dipole moment of a molecule "located" in $\mathbf{x}'$. A natural extension to p pure states consists of setting

$$\mathbf{P}(\mathbf{x}', t) = \mathcal{N}_0(\mathbf{x}')\sum_{l=1}^{p}\mathbf{d}^{(l)}(\mathbf{x}', t)$$

where $\mathbf{d}^{(l)}(\mathbf{x}', t) = \langle\psi^{(l)}|\mathbf{x}|\psi^{(l)}\rangle$ and $\psi^{(l)}$ is solution to

$$i\partial_t\psi(\mathbf{x}, t) = \Big(H_0 + \mathbf{x}\cdot\mathbf{E}_\alpha(t)\Big)\psi(\mathbf{x}, t), \qquad \psi(\mathbf{x}, 0) = \phi_l(\mathbf{x}).$$

with $H_0\phi_l = \varepsilon_l\phi_l$. From a practical point of view, solving this p TDSE's can easily be implemented in parallel (multi-threading for instance) as each TDSE computation is done independently.

4. SFA nonlinear optics models

The method which is developed in this section consists of coupling bound and free states in the solving of TDSE, in order to determine molecule dipole moments. More specifically, the overall strategy is to determine the bound state contribution combining the usual perturbative approach for weak fields, and a Lewenstein's SFA approach for the free state contribution, as well as the bound-continuous state interactions; however we *do not* limit the interaction to free states of the continuum with the ground state like in [24]. In fine, we determine an explicit approximate solution to the TDSE, and of the dipole moment allowing to accurately model the polarization in Maxwell's equations. We refer mainly to [25] for the notation and derivation of perturbative nonlinear optics modeling and [24] for the SFA model. Before presenting this model, we first derive a general Liouville equation, Subsection 4.1, including bound and continuous states, from which, in principle we could derived macroscopic polarization. However, due to the complexity of the derived model, it is expected of poor interest from a practical point of view. 1

4.1. General non-perturbative approach. How far can we go ?

In this subsection, we consider the general situation:

$$i\partial_t\psi = \big(H_0 + \mathbf{x}\cdot\mathbf{E}(t)\Big)\psi, \; \psi(\mathbf{x}, 0) = \phi_0$$

We consider the case a unique pure state. We search for a wavefunction in the general form:

$$\psi(\mathbf{x}, t) = \sum_n c_n(t)\phi_n(\mathbf{x}) + \int_{\sigma_c} c(t, \eta)\phi_c(\mathbf{x}, \eta)\rho_c(\eta)d\eta \tag{21}$$

where i) σ_c denotes the continuous spectrum of H_0, ii) ρ_c denotes the spectral density in σ_c, iii) ϕ_c satisfies $H_0\phi_c(\mathbf{x}, \lambda) = \lambda\phi_c(\mathbf{x}, \lambda)$ (where "$\|\phi_c(\cdot, \lambda)\|_{L^2} = +\infty$", and finally iv) $c(t, \lambda) := \langle\phi(\cdot, \lambda)|\psi(\cdot, t)\rangle$. Assuming that the full spectrum (including eigenelements) is known, we have to determine $\big(c_n(t)\big)_n$, $\big(c(t, \lambda)\big)_\lambda$ as well as $\rho(\lambda)$. In the following, the discrete spectrum elements of H_0 are denoted ε_n. For any $n \in \mathbb{N}$:

$$i\langle\partial_t\psi|\phi_n\rangle = \langle\big(H_0 + \mathbf{x}\cdot\mathbf{E}(t)\big)\psi|\phi_n\rangle$$

and for any $\lambda \in \sigma_c$

$$i\langle\partial_t\psi|\phi_c(\cdot, \lambda)\rangle = \langle\big(H_0 + \mathbf{x}\cdot\mathbf{E}(t)\big)\psi|\phi_c(\cdot, \lambda)\rangle$$

We set

$$\begin{cases} H_{nm} & = & \mathbf{E}(t)\cdot\langle \mathbf{x}\phi_n|\phi_m\rangle \\ H_n(\lambda) & = & \mathbf{E}(t)\cdot\langle \mathbf{x}\phi_n|\phi_c(\cdot,\lambda)\rangle \\ K_n(\lambda) & = & \langle \mathbf{E}(t)\cdot\langle \mathbf{x}\phi_c(\cdot,\lambda)|\phi_n\rangle \\ H(\lambda,\mu) & = & \mathbf{E}(t)\cdot\langle \mathbf{x}\phi_c(\cdot,\lambda)|\phi_c(\cdot,\mu)\rangle \\ D(\lambda,\mu) & = & \langle \phi_c(\cdot,\lambda)|\phi_c(\cdot,\mu)\rangle \end{cases}$$

and

$$\rho_{mn}(t) = c_m^*(t)c_n(t),\ \rho_{m\lambda}(t) = c_m^*(t)c(t,\lambda),\ \rho_{\lambda m}(t) = c^*(t,\lambda)c_m, \rho_{\lambda\mu}(t) = c^*(t,\lambda)c(t,\mu)$$

We prove in Appendix B, that for all m, n, λ, μ:

$$\begin{cases} \dot{\rho}_{mn}(t) & = & i\sum_\nu H_{m\nu}\rho_{\nu n}(t) - \rho_{m\nu}(t)H_{\nu n} + i\int_{\sigma_c}\Big(\rho_\eta(t)K_m^*(\eta)\rho_c^*(\eta) - \rho_{m\eta}(t)K_n(\eta)\rho(\eta)\Big)d\eta \\ \dot{\rho}_{m\lambda}(t) & = & i\sum_n\Big[\rho_{n\lambda}(t)H_{nm}^* - \rho_{mn}(t)H_n(\lambda)\Big] \\ & & +i\int_{\sigma_c}\Big[K_n^*(\eta)\rho_c^*(\eta)\rho_{\eta\lambda}(t) - H(\eta,\lambda)\rho_c(\eta)\rho_{m\eta}(t)\Big]d\eta \\ \dot{\rho}_{\lambda\mu}(t) & = & i\sum_n\Big[\rho_{n\mu}(t)H_n^*(\lambda) - \rho_{\lambda n}(t)H_n(\mu)\Big] \\ & & +i\int_{\sigma_c}\Big[H^*(\eta,\lambda)\rho_c^*(\eta)\rho_{\eta\mu}(t) - H(\eta,\mu)\rho_c(\eta)\rho_{\lambda\eta}(t)\Big]d\eta \end{cases}$$

Remark 4.1. *This system is a Liouville-like equation which is comparable to the usual one, which does not include the continuum of the Hamiltonian spectrum and which writes*

$$\dot{\rho}_{mn} = -\frac{i}{\hbar}\big[\widehat{H},\widehat{\rho}\big]_{mn}$$

from which is deduced $Tr(\widehat{\rho}\widehat{\mathbf{x}}) = \sum_{nm}\rho_{nm}\mathbf{x}_{mn}$ *in the classical theory.*

The above equation contains, in principle, all the state transitions. However, from a practical point of view, it is of moderate interest, due to its complexity. The following section is devoted to an analogue approach, except that the integral over the continuum in (21), is approximated by SFA.

4.2. SFA-like nonlinear optics model: unique continuous state

The principle of the following model is to include perturbative and nonperturbative contribution, using in particular on Lewenstein's SFA model, which has the ability to accurately capture phenomena such as ionization and high order harmonic generation. We search for a wavefunction ψ solution to

$$i\partial_t\psi = \Big(H_0 + V(\mathbf{x},t)\Big)\psi,\ \psi(\mathbf{x},0) = \phi_0(\mathbf{x})$$

where H_0 denotes the laser-free Hamiltonian and V the electric potential, as the sum of bound and bound-free state contributions:

$$\psi(\mathbf{x},t) = \psi_B(\mathbf{x},t) + \psi_L(\mathbf{x},t)$$

and where the purely bound part is of the form

$$\psi_B(\mathbf{x},t) = \sum_{l\in\mathbb{N}}\lambda^l\psi_B^{(l)}(\mathbf{x},t)$$

with

$$\psi_B^{(l)}(\mathbf{x},t) = \sum_{k\in\mathbb{N}} a_k^{(l)}(t)\phi_k(\mathbf{x})e^{-i\omega_k t}$$

where we have denoted ϕ_k the eigenvectors of the field-free Hamiltonian: $H_0\phi_k = \varepsilon_k\phi_k$ and $\omega_k = \hbar\varepsilon_k$. We assume that

$$\psi_B^{(0)}(\mathbf{x},t) = \big(1-\alpha(t)e^{iI_p t}\big)\phi_0(\mathbf{x})e^{-i\omega_0 t}$$

where $\alpha(t)$ is a time-dependent complex. The bound-continuous part is of the form of a SFA:

$$\psi_L(\mathbf{x},t) = e^{iI_p t}\Big(\alpha(t)\phi_0(\mathbf{x}) + \int d^3\mathbf{p}\, b(\mathbf{p},t)e^{i\mathbf{p}\cdot\mathbf{x}}\Big)$$

with I_p the ionization potential and where $\psi_L(\mathbf{x},0) = \alpha_0\phi_0(\mathbf{x})$ and $b(\mathbf{p},0) = 0$. A natural choice for α is then the depletion coefficient. According to [24], uncoupling the equation in α and the TDSE, α can be modeled by the following equation

$$\dot{\alpha}(t) = -\gamma(t)\alpha(t),\ \alpha(0) = \alpha_0 \in (0,1)$$

where γ is defined by (52) in [24] and is naturally maximal at the electric field peaks. In practice, and as proposed in that paper γ will be approximated by its average $\bar{\gamma} \in \mathbb{C}$ with positive real part, so that

$$\alpha(t) \approx \alpha_0 e^{-\bar{\gamma}t}$$

Now, we set

$$\delta := \bar{\gamma} - iI_p$$

so that

$$e^{iI_p t}\alpha(t) \approx \alpha_0 e^{-\delta t} \tag{22}$$

The unknowns of the problem are $(a_l)_{l\geqslant 1}$ and b. Now by linearity, we have

$$i\partial_t\psi_{B,L} = \Big(H_0 + V(\mathbf{x},t)\Big)\psi_{B,L}$$

Parameter α is chosen such that initially

$$\psi(\mathbf{x},0) = \psi_B(\mathbf{x},0) + \psi_L(\mathbf{x},0) = (1-\alpha_0)\phi_0(\mathbf{x}) + \alpha_0\phi_0(\mathbf{x}) = \phi_0(\mathbf{x})$$

Naturally, we first have:

$$\psi_B^{(0)}(\mathbf{x},t) \approx \left(1-\alpha_0 e^{-\delta t}\right)\phi_0(\mathbf{x})e^{-i\omega_0 t} \tag{23}$$

Coefficients $(a_l)_{l\geqslant 1}$ will be searched following the usual perturbative method as presented in [25]. Denoting $\omega_l = \hbar\varepsilon_l$, for all l

$$a_m^{(N)}(t) = \frac{1}{i\hbar}\sum_{l\in\mathbb{N}}\int_{-\infty}^{t} V_{ml}(t')a_l^{(N-1)}(t')e^{i\omega_{ml}t'}dt'$$

where

$$V_{ml}(t) = \langle\phi_m|\mathbf{x}\cdot\mathbf{E}(t)|\phi_l\rangle = \int \mathbf{x}\cdot\mathbf{E}(t)\phi_m^*\phi_l d^3\mathbf{x}$$

and the transition frequencies are denoted $\omega_{mg} = \omega_m - \omega_g$. We deduce that for $\mathbf{E}(t) = \sum_{p\in\mathbb{N}}\mathbf{E}(\omega_p)e^{-i\omega_p t}$, and using (22)

$$a_m^{(1)}(t) \approx \frac{1}{\hbar}\sum_{p\in\mathbb{N}}\boldsymbol{\mu}_{mg}\cdot\mathbf{E}(\omega_p)\Big\{\frac{1}{\omega_{mg}-\omega_p} - \frac{\alpha(t)e^{iI_p t}}{\omega_{mg}-\omega_p+i\delta}\Big\}e^{i(\omega_{mg}-\omega_p)t}$$

then $a_m^{(2)}$, etc, see [25] for details.

Remark 4.2 (Important remark on the contribution of $\chi^{(l)}$ in SFA's Lewenstein.). *As it is well-known, SFA's Lewenstein model contributes to $\chi^{(l)}$. Then, it is important to separate the contribution of the susceptibility tensors from the bound states and from SFA's Lewenstein function. In this goal, we will sum $(a_n)_n$ only over the bound states, omitting the contribution from "bound-continuum" from the usual perturbative approach, as this contribution is already present in SFA. We denote the corresponding indices by $\mathbb{B}$ (sum over the bound states only).*

For instance

$$
\begin{aligned}
a_n^{(2)}(t) \quad \approx \quad & \frac{1}{\hbar^2}\sum_{(p,q)\in\mathbb{N}^2}\sum_{m\in\mathbb{B}}\big(\boldsymbol{\mu}_{mg}\cdot\mathbf{E}(\omega_p)\big)\big(\boldsymbol{\mu}_{nm}\cdot\mathbf{E}(\omega_q)\big) \\
& \times\Big\{\frac{1}{(\omega_{ng}-\omega_p-\omega_q)(\omega_{mg}-\omega_p)} - \frac{\alpha(t)e^{iI_pt}}{(\omega_{mg}-\omega_p+i\delta)(\omega_{ng}-\omega_p-\omega_q+i\delta)}\Big\}e^{i(\omega_{ng}-\omega_p-\omega_q)t}
\end{aligned}
$$

and similarly

$$
\begin{aligned}
a_\nu^{(3)}(t) \quad \approx \quad & \frac{1}{\hbar^3}\sum_{(p,q,r)\in\mathbb{N}^3}\sum_{(m,n)\in\mathbb{B}^2}\big(\boldsymbol{\mu}_{\nu n}\cdot\mathbf{E}(\omega_r)\big)\big(\boldsymbol{\mu}_{nm}\cdot\mathbf{E}(\omega_q)\big)\big(\boldsymbol{\mu}_{mg}\cdot\mathbf{E}(\omega_p)\big) \\
& \times\Big\{\frac{1}{(\omega_{\nu g}-\omega_p-\omega_q-\omega_r)(\omega_{ng}-\omega_p-\omega_q)(\omega_{mg}-\omega_p)} \\
& - \frac{\alpha(t)e^{iI_pt}}{(\omega_{\nu g}-\omega_p-\omega_q-\omega_r+i\delta)(\omega_{ng}-\omega_p-\omega_q+i\delta)(\omega_{mg}-\omega_p+i\delta)}\Big\}e^{i(\omega_{ng}-\omega_p-\omega_q-\omega_r)t}
\end{aligned}
$$

In the above formula, if $m \geqslant 1$ (recall that in atomic unit the electronic charge is $e = 1$):

$$
\boldsymbol{\mu}_{mg} = -\int \bar{\phi}_m\widehat{\mathbf{x}}\phi_0 d^3\mathbf{x}
$$

We deduce:

$$
\left\{
\begin{aligned}
\psi_B^{(1)}(\mathbf{x},t) \quad = \quad & \sum_{m\in\mathbb{B}} a_m^{(1)}\phi_m(\mathbf{x})e^{-i\omega_m t} \\
= \quad & \sum_{m\in\mathbb{B}}\sum_{p\in\mathbb{N}}\big(\boldsymbol{\mu}_{mg}\cdot\mathbf{E}(\omega_p)\big)\Big[\frac{1}{\omega_{mg}-\omega_p} - \frac{\alpha(t)e^{iI_pt}}{\omega_{mg}-\omega_p+i\delta}\Big]\phi_m(\mathbf{x})e^{-i(\omega_0+\omega_p)t} \\
\psi_B^{(2)}(\mathbf{x},t) \quad = \quad & \sum_{(m,n)\in\mathbb{B}^2}\sum_{(p,q)\in\mathbb{N}^2}\big(\boldsymbol{\mu}_{nm}\cdot\mathbf{E}(\omega_q)\big)\big(\boldsymbol{\mu}_{mg}\cdot\mathbf{E}(\omega_p)\big)\Big[\frac{1}{(\omega_{mg}-\omega_p)(\omega_{ng}-\omega_p-\omega_q)} \\
& - \frac{\alpha(t)e^{iI_pt}}{(\omega_{mg}-\omega_p+i\delta)(\omega_{ng}-\omega_p-\omega_q+i\delta)}\Big]\phi_n(\mathbf{x})e^{-i(\omega_0+\omega_p+\omega_q)t} \\
\psi_B^{(3)}(\mathbf{x},t) \quad = \quad & \sum_{(m,n,\nu)\in\mathbb{B}^3}\sum_{(p,q,r)\in\mathbb{N}^3}\big(\boldsymbol{\mu}_{\nu n}\cdot\mathbf{E}(\omega_r)\big)\big(\boldsymbol{\mu}_{nm}\cdot\mathbf{E}(\omega_q)\big)\big(\boldsymbol{\mu}_{mg}\cdot\mathbf{E}(\omega_p)\big) \\
& \times\Big[\frac{1}{(\omega_{mg}-\omega_p)(\omega_{ng}-\omega_p-\omega_q)(\omega_{\nu g}-\omega_p-\omega_q-\omega_r)} \\
& - \frac{\alpha(t)e^{iI_pt}}{(\omega_{mg}-\omega_p+i\delta)(\omega_{ng}-\omega_p-\omega_q+i\delta)(\omega_{\nu g}-\omega_p-\omega_q-\omega_r+i\delta)}\Big]\phi_\nu(\mathbf{x})e^{-i(\omega_0+\omega_p+\omega_q+\omega_r)t}
\end{aligned}
\right.
$$

Now regarding the continuum, we have

$$
\partial_t b(\mathbf{p},t) = -i\Big(\frac{\mathbf{p}^2}{2}+I_p\Big)b(\mathbf{p},t) - \mathbf{E}(t)\cdot\nabla_{\mathbf{p}}b(\mathbf{p},t) - i\alpha(t)\mathbf{E}(t)\mathbf{d}_g(\mathbf{p})
$$

where $\mathbf{d}_g(\mathbf{p}) = \langle\phi_0|\mathbf{x}|\mathbf{p}\rangle$. We can then show (see [24], and method of characteristics) that

$$b(\mathbf{p},t) = -i\int_0^t dt'\alpha(t')\mathbf{E}(t)\cdot\mathbf{d}_g\big(\mathbf{p}-\mathbf{A}(t)-\mathbf{A}(t')\big)\exp\Big(-i\int_{t'}^t dt''\big(\mathbf{p}-\mathbf{A}(t)-\mathbf{A}(t'')\big)^2/2+I_p\Big)$$

From there, we can give an approximate expansion of the overall dipole moment defined as

$$\mathbf{d}(t) = \langle\psi|\widehat{\boldsymbol{\mu}}|\psi\rangle$$

using that

$$\begin{aligned}\mathbf{d}(t) &= \langle\psi_B|\widehat{\boldsymbol{\mu}}|\psi_B\rangle + \langle\psi_L|\widehat{\boldsymbol{\mu}}|\psi_L\rangle + \langle\psi_B|\widehat{\boldsymbol{\mu}}|\psi_L\rangle + \langle\psi_L|\widehat{\boldsymbol{\mu}}|\psi_B\rangle \\ &= \mathbf{d}_{BB}(t) + \mathbf{d}_{LL}(t) + \mathbf{d}_{BL}(t)\end{aligned}$$

with

$$\mathbf{d}_{BB}(t) = \sum_{l\in\mathbb{N}}\lambda^l\mathbf{d}_{BB}^{(l)}(t) \tag{24}$$

In order to simplify the presentation, we will assume that damping phenomena are not incorporated in the model, which allows to deal with real transition frequencies. Extension to complex transitions is possible following [25]. We then get

$$\mathbf{d}_{BB}^{(0)}(t) = 0 \tag{25}$$

then

$$\mathbf{d}_{BB}^{(1)}(t) = \langle\psi_B^{(0)}|\widehat{\boldsymbol{\mu}}|\psi_B^{(1)}\rangle + \langle\psi_B^{(1)}|\widehat{\boldsymbol{\mu}}|\psi_B^{(0)}\rangle \tag{26}$$

leading, from $\mathbf{E}^*(\omega_p) = -\mathbf{E}(\omega_p)$ to

$$\begin{aligned}\mathbf{d}_{BB}^{(1)}(t) &= \frac{1}{\hbar}\textstyle\sum_{m\in\mathbb{B}}\sum_{p\in\mathbb{N}}\Bigg\{\boldsymbol{\mu}_{gm}\big(\boldsymbol{\mu}_{mg}\cdot\mathbf{E}(\omega_p)\big)\Big(\dfrac{1-\alpha^*(t)e^{-iI_pt}}{\omega_{mg}-\omega_p} - \dfrac{\alpha(t)e^{iI_pt}\big(1-\alpha^*(t)e^{-iI_pt}\big)}{\omega_{mg}-\omega_p+i\delta}\Big)e^{-i\omega_pt} \\ &\quad + \big(\boldsymbol{\mu}_{mg}\mathbf{E}(\omega_p)\big)^*\boldsymbol{\mu}_{mg}\Big(\dfrac{1-\alpha(t)e^{iI_pt}}{\omega_{mg}-\omega_p} - \dfrac{\alpha^*(t)e^{-iI_pt}\big(1-\alpha(t)e^{iI_pt}\big)}{\omega_{mg}-\omega_p-i\delta^*}\Big)\Bigg\}e^{i\omega_pt}\end{aligned} \tag{27}$$

This can be rewritten

$$\begin{aligned}\mathbf{d}_{BB}^{(1)}(t) &= \frac{1}{\hbar}\textstyle\sum_{m\in\mathbb{B}}\sum_{p\in\mathbb{N}}\Bigg\{\boldsymbol{\mu}_{gm}\big(\boldsymbol{\mu}_{mg}\cdot\mathbf{E}(\omega_p)\big)\Big(\dfrac{1-\alpha^*(t)e^{-iI_pt}}{\omega_{mg}-\omega_p} - \dfrac{\alpha(t)e^{iI_pt}\big(1-\alpha^*(t)e^{-iI_pt}\big)}{\omega_{mg}-\omega_p+i\delta}\Big) \\ &\quad + \big(\boldsymbol{\mu}_{gm}\cdot\mathbf{E}(\omega_p)\big)\boldsymbol{\mu}_{mg}\Big(\dfrac{1-\alpha(t)e^{iI_pt}}{\omega_{mg}+\omega_p} - \dfrac{\alpha^*(t)e^{-iI_pt}\big(1-\alpha(t)e^{iI_pt}\big)}{\omega_{mg}+\omega_p-i\delta^*}\Big)\Bigg\}e^{-i\omega_pt}\end{aligned} \tag{28}$$

Similarly

$$\mathbf{d}_{BB}^{(2)}(t) = \langle\psi_B^{(0)}|\widehat{\boldsymbol{\mu}}|\psi_B^{(2)}\rangle + \langle\psi_B^{(2)}|\widehat{\boldsymbol{\mu}}|\psi_B^{(0)}\rangle + \langle\psi_B^{(1)}|\widehat{\boldsymbol{\mu}}|\psi_B^{(1)}\rangle \tag{29}$$

From above, we deduce that

$$\begin{aligned}\langle\psi_B^{(0)}|\widehat{\boldsymbol{\mu}}|\psi_B^{(2)}\rangle &= \frac{1}{\hbar^2}\textstyle\sum_{(p,q)\in\mathbb{N}^2}\sum_{(m,n)\in\mathbb{B}^2}\boldsymbol{\mu}_{gn}(\boldsymbol{\mu}_{nm}\cdot\mathbf{E}(\omega_q))(\boldsymbol{\mu}_{mg}\cdot\mathbf{E}(\omega_p)) \\ &\quad\times\Bigg\{\dfrac{1-\alpha^*(t)e^{-iI_pt}}{(\omega_{ng}-\omega_p-\omega_q)(\omega_{mg}-\omega_p)} - \dfrac{\big(1-\alpha^*(t)e^{-iI_pt}\big)\alpha(t)e^{iI_pt}}{(\omega_{mg}-\omega_p+i\delta)(\omega_{ng}-\omega_p-\omega_q+i\delta)}\Bigg\}e^{-i(\omega_p+\omega_q)t}\end{aligned}$$

and

$$
\begin{aligned}
\langle\psi_B^{(1)}|\widehat{\boldsymbol{\mu}}|\psi_B^{(1)}\rangle &= \frac{1}{\hbar^2}\sum_{(p,q)\in\mathbb{N}^2}\sum_{(m,n)\in\mathbb{B}^2}\big(\boldsymbol{\mu}_{ng}\cdot\mathbf{E}(\omega_q)\big)^*\boldsymbol{\mu}_{nm}\big(\boldsymbol{\mu}_{mg}\cdot\mathbf{E}(\omega_q)\big)\\
&\times\Big(\frac{1}{\omega_{mg}-\omega_p}-\frac{\alpha(t)e^{iI_pt}}{\omega_{mg}-\omega_p+i\delta}\Big)\Big(\frac{1}{\omega_{ng}-\omega_q}-\frac{\alpha^*(t)e^{-iI_pt}}{\omega_{ng}-\omega_q-i\delta^*}\Big)e^{-i(\omega_p-\omega_q)t}
\end{aligned}
$$

So that:

$$
\begin{aligned}
\mathbf{d}_{BB}^{(2)}(t) &= \frac{2}{\hbar^2}\sum_{(p,q)\in\mathbb{N}^2}\sum_{(m,n)\in\mathbb{B}^2}\mathrm{Re}\Bigg\{\boldsymbol{\mu}_{gn}\big(\boldsymbol{\mu}_{nm}\cdot\mathbf{E}(\omega_q)\big)\big(\boldsymbol{\mu}_{mg}\cdot\mathbf{E}(\omega_p)\big)\\
&\times\Big(\frac{1-\alpha^*(t)}{(\omega_{ng}-\omega_p-\omega_q)(\omega_{mg}-\omega_p)}-\frac{\big(1-\alpha^*(t)e^{-iI_pt}\big)\alpha(t)e^{iI_pt}}{(\omega_{mg}-\omega_p+i\delta)(\omega_{ng}-\omega_p-\omega_q+i\delta)}\Big)e^{-i(\omega_p+\omega_q)t}\Bigg\}\\
&+\frac{1}{\hbar^2}\sum_{(p,q)\in\mathbb{N}^2}\sum_{(m,n)\mathbb{B}^2}\big(\boldsymbol{\mu}_{ng}\cdot\mathbf{E}(\omega_q)\big)^*\boldsymbol{\mu}_{nm}\big(\boldsymbol{\mu}_{mg}\cdot\mathbf{E}(\omega_q)\big)\\
&\times\Big(\frac{1}{\omega_{mg}-\omega_p}-\frac{\alpha(t)e^{iI_pt}}{\omega_{mg}-\omega_p+i\delta}\Big)\Big(\frac{1}{\omega_{ng}-\omega_q}-\frac{\alpha^*(t)e^{-iI_pt}}{\omega_{ng}-\omega_q-i\delta^*}\Big)e^{-i(\omega_p-\omega_q)t}
\end{aligned}
\tag{30}
$$

or written similarly

$$
\begin{aligned}
\mathbf{d}_{BB}^{(2)}(t) &= \frac{1}{\hbar^2}\sum_{(m,n)\in\mathbb{B}^2}\sum_{(p,q)\in\mathbb{N}^2}\Bigg\{\boldsymbol{\mu}_{gn}\big(1-\alpha^*(t)e^{-iI_pt}\big)\big(\boldsymbol{\mu}_{nm}\cdot\mathbf{E}(\omega_q)\big)\big(\boldsymbol{\mu}_{mg}\cdot\mathbf{E}(\omega_p)\big)\\
&\times\frac{1}{(\omega_{mg}-\omega_p)(\omega_{ng}-\omega_p-\omega_q)}-\frac{\alpha(t)e^{iI_pt}}{(\omega_{mg}-\omega_p+i\delta)(\omega_{ng}-\omega_p-\omega_q+i\delta)}\\
&+\boldsymbol{\mu}_{nm}\big(\boldsymbol{\mu}_{gn}\cdot\mathbf{E}(\omega_q)\big)\big(\boldsymbol{\mu}_{mg}\cdot\mathbf{E}(\omega_p)\big)\left(\frac{1}{\omega_{ng}+\omega_q}-\frac{\alpha^*(t)e^{-iI_pt}}{\omega_{ng}+\omega_q-i\delta^*}\right)\left(\frac{1}{\omega_{mg}-\omega_p}-\frac{\alpha(t)e^{iI_pt}}{\omega_{mg}-\omega_p+i\delta}\right)\\
&+\boldsymbol{\mu}_{ng}\big(1-\alpha(t)e^{iI_pt}\big)\big(\boldsymbol{\mu}_{mn}\cdot\mathbf{E}(\omega_q)\big)\big(\boldsymbol{\mu}_{gm}\cdot\mathbf{E}(\omega_p)\big)\\
&\times\frac{1}{(\omega_{mg}+\omega_p)(\omega_{ng}+\omega_p+\omega_q)}-\frac{\alpha^*(t)e^{-iI_pt}}{(\omega_{mg}+\omega_p-i\delta^*)(\omega_{ng}+\omega_p+\omega_q-i\delta^*)}\Bigg\}e^{-i(\omega_p+\omega_q)t}
\end{aligned}
$$

Similarly, one can deduce $\mathbf{d}_{BB}^{(3)}$, from

$$
\mathbf{d}_{BB}^{(3)}(t)=\langle\psi_B^{(0)}|\widehat{\boldsymbol{\mu}}|\psi_B^{(3)}\rangle+\langle\psi_B^{(3)}|\widehat{\boldsymbol{\mu}}|\psi_B^{(0)}\rangle+\langle\psi_B^{(1)}|\widehat{\boldsymbol{\mu}}|\psi_B^{(2)}\rangle+\langle\psi_B^{(2)}|\widehat{\boldsymbol{\mu}}|\psi_B^{(1)}\rangle \tag{31}
$$

where

$$
\begin{aligned}
\langle\psi^{(0)}|\widehat{\boldsymbol{\mu}}|\psi^{(3)}\rangle &= \sum_{(m,n,\nu)\in\mathbb{B}^3}\sum_{(p,q,r)\in\mathbb{N}^3}\boldsymbol{\mu}_{g\nu}\big(1-\alpha^*(t)e^{-iI_pt}\big)\\
&\times\big(\boldsymbol{\mu}_{\nu n}.\mathbf{E}(\omega_r)\big)\big(\boldsymbol{\mu}_{nm}\cdot\mathbf{E}(\omega_q)\big)\big(\boldsymbol{\mu}_{mg}\cdot\mathbf{E}(\omega_p)\big)\\
&\times\Big(\frac{1}{(\omega_{mg}-\omega_p)(\omega_{ng}-\omega_p-\omega_q)(\omega_{\nu g}-\omega_p-\omega_q-\omega_r)}\\
&-\frac{\alpha(t)e^{iI_pt}}{(\omega_{mg}-\omega_p+i\delta)(\omega_{ng}-\omega_p-\omega_q+i\delta)(\omega_{\nu g}-\omega_p-\omega_q-\omega_r+i\delta)}\Big)e^{-i(\omega_q+\omega_p+\omega_r)t}
\end{aligned}
$$

and

$$\langle\psi^{(1)}|\widehat{\boldsymbol{\mu}}|\psi^{(2)}\rangle = \sum_{(m,n,\nu)\in\mathbb{B}^3}\sum_{(p,q,r)\in\mathbb{N}^3}\boldsymbol{\mu}_{mn}\big(\boldsymbol{\mu}_{g\nu}\cdot\mathbf{E}(\omega_r)\big)\big(\boldsymbol{\mu}_{nm}\cdot\mathbf{E}(\omega_q)\big)\big(\boldsymbol{\mu}_{mg}\cdot\mathbf{E}(\omega_p)\big)$$
$$\times\Big(\frac{1}{\omega_{\nu g}+\omega_r}-\frac{\alpha^*(t)e^{-iI_pt}}{\omega_{\nu g}+\omega_r-i\delta^*}\Big)\Big(\frac{1}{(\omega_{mg}-\omega_p)(\omega_{ng}-\omega_p-\omega_q)}$$
$$-\frac{\alpha(t)e^{iI_pt}}{(\omega_{mg}-\omega_p+i\delta)(\omega_{ng}-\omega_p-\omega_q+i\delta)}\Big)e^{-i(\omega_p+\omega_q+\omega_r)t}$$

and

$$\langle\psi^{(2)}|\widehat{\boldsymbol{\mu}}|\psi^{(1)}\rangle = \sum_{(m,n,\nu)\in\mathbb{B}^3}\sum_{(p,q,r)\in\mathbb{N}^3}\boldsymbol{\mu}_{nm}\big(\boldsymbol{\mu}_{\nu g}\cdot\mathbf{E}(\omega_r)\big)\big(\boldsymbol{\mu}_{mn}\cdot\mathbf{E}(\omega_q)\big)\big(\boldsymbol{\mu}_{gm}\cdot\mathbf{E}(\omega_p)\big)$$
$$\times\Big(\frac{1}{\omega_{\nu g}-\omega_r}-\frac{\alpha(t)e^{iI_pt}}{\omega_{\nu g}-\omega_r+i\delta}\Big)\Big(\frac{1}{(\omega_{mg}+\omega_p)(\omega_{ng}+\omega_p+\omega_q)}$$
$$-\frac{\alpha^*(t)e^{-iI_pt}}{(\omega_{mg}+\omega_p-i\delta^*)(\omega_{ng}+\omega_p+\omega_q-i\delta^*)}\Big)e^{-i(\omega_p+\omega_q+\omega_r)t}$$

and

$$\langle\psi^{(3)}|\widehat{\boldsymbol{\mu}}|\psi^{(0)}\rangle = \sum_{(m,n,\nu)\in\mathbb{B}^3}\sum_{(p,q,r)\in\mathbb{N}^3}\boldsymbol{\mu}_{\nu g}\big(1-\alpha(t)e^{iI_pt}\big)\big(\boldsymbol{\mu}_{n\nu}.\mathbf{E}(\omega_r)\big)\big(\boldsymbol{\mu}_{mn}\cdot\mathbf{E}(\omega_q)\big)\big(\boldsymbol{\mu}_{gm}\cdot\mathbf{E}(\omega_p)\big)$$
$$\times\Big\{\frac{1}{(\omega_{mg}+\omega_p)(\omega_{ng}+\omega_p+\omega_q)(\omega_{\nu g}+\omega_p+\omega_q+\omega_r)}$$
$$-\frac{\alpha^*(t)e^{-iI_pt}}{(\omega_{mg}+\omega_p-i\delta^*)(\omega_{ng}+\omega_p+\omega_q-i\delta^*)(\omega_{\nu g}+\omega_p+\omega_q+\omega_r-i\delta^*)}\Big\}e^{-i(\omega_q+\omega_p+\omega_r)t}$$

Now from [24] and denoting $\mathbf{d}_g(t)=\langle\mathbf{p}|\mathbf{x}|\phi_0\rangle$, we have

$$\mathbf{d}_{LL}(t)=i\int_0^t\int d\tau d^3\mathbf{v}\mathbf{E}(t-\tau)\cdot\mathbf{d}_g\big(\mathbf{v}-\mathbf{A}(t-\tau)\big)\boldsymbol{\mu}_g^*\big((\mathbf{v}-\mathbf{A}(t)\big)e^{-iS(\mathbf{v},t,\tau)-\delta^*t-\delta(t-\tau)} \tag{32}$$

where

$$S(\mathbf{v},t,\tau):=\int_\tau^t ds\Big[\frac{(\mathbf{v}-\mathbf{A}(s))^2}{2}+I_p\Big]$$

Naturally (29) and (32) are trivially deduced from the usual theory. We have now to evaluate

$$\mathbf{d}_{BL}(t)=\langle\psi_B|\widehat{\boldsymbol{\mu}}|\psi_L\rangle+\langle\psi_L|\widehat{\boldsymbol{\mu}}|\psi_B\rangle$$

In this goal, we will follow a strategy close the one developed by [24]. That is:

$$\mathbf{d}_{BL}(t)=\sum_{l\in\mathbb{N}}\lambda^l\mathbf{d}_{BL}^{(l)}(t) \tag{33}$$

with

$$\mathbf{d}_{BL}^{(l)}(t)=2\mathrm{Re}\Big\{\langle\psi_B^{(l)}(\mathbf{x},t)|\widehat{\boldsymbol{\mu}}|\psi_L(\mathbf{x},t)\rangle\Big\} \tag{34}$$

where

$$\langle\psi_B^{(l)}(\mathbf{x},t)|\widehat{\boldsymbol{\mu}}|\psi_L(\mathbf{x},t)\rangle = \sum_{m\in\mathbb{B}}\int_{\mathbb{R}^3}d^3\mathbf{x}\big(a_m^{(l)}(t)\big)^*\phi_m(\mathbf{x})e^{i\omega_mt}\boldsymbol{\mu}e^{iI_pt}\big(\alpha(t)\phi_0(\mathbf{x})+\int_{\mathbb{R}^3}d^3\mathbf{p}b(\mathbf{p},t)e^{i\mathbf{p}.\mathbf{x}}\big)$$

and

$$\mathbf{d}_{BL}^{(l)}(t) \;=\; 2\mathrm{Re}\Big\{\sum_{m\in\mathbb{B}}\Big(\big(a_m^{(l)}(t)\big)^* e^{i(\omega_m+I_p)t}\boldsymbol{\mu}_{mg}\alpha(t)+\int_{\mathbb{R}^3} d^3\mathbf{p}\big(a_m^{(l)}(t)\big)^* b(\mathbf{p},t)e^{i(\omega_m+I_p)t}\mathbf{d}_m(\mathbf{p})\Big)\Big\} \quad (35)$$

where for all $l \geqslant 1$

$$\mathbf{d}_l(\mathbf{p}) = \langle\phi_l|\widehat{\boldsymbol{\mu}}|\mathbf{p}\rangle$$

and

$$b(\mathbf{p},t) \;=\; -i\int_0^t \mathbf{E}\cos(\tau)\cdot\mathbf{d}\big(\mathbf{p}(t)+\mathbf{A}(t)-\mathbf{A}(\tau)\big)\exp\Big(-\int_\tau^t i\big(\frac{(\mathbf{p}(t)+\mathbf{A}(t)-\mathbf{A}(s))^2}{2}+I_p\big)ds\Big)d\tau \quad (36)$$

with for $l \geqslant 1$, $\mathbf{d}_l(\mathbf{p})$ which is negligible. Indeed, as mentioned before in SFA model an implicit assumption is that the matrix elements of the Hamiltonian between bound states (except for the ground state) and free states are negligible. It is however possible to estimate these contributions within our model. Moreover, a second model Subsection 4.3, will be developed to go beyond this assumption. Now for $l = 0$

$$\mathbf{d}_{BL}^{(0)}(t) = -i\int_{\mathbf{R}^3} d^3\mathbf{v}\int_0^t d\tau\big(1-\alpha_0 e^{-\delta^* t}\big)\mathbf{E}(\tau)\cdot\mathbf{d}_g^*\big(\mathbf{v}-A(t)\big)\mathbf{d}_g\big(\mathbf{v}-A(\tau)\big)e^{iS(\mathbf{v},t,\tau)} \quad (37)$$

In addition, for the $l \geqslant 1$ we have

$$\mathbf{d}_{BL}^{(l)}(t) = \sum_{m\in\mathbb{B}}\big(a_m^{(l)}(t)\big)^* e^{i(\omega_m+I_p)t}\boldsymbol{d}_{mg}\alpha(t) \quad (38)$$

We can now estimate the susceptibility tensors from bound-free state contribution. More specifically, for $l = 1$, we have

$$\begin{aligned}
\mathbf{d}_{BL}^{(1)}(t) \;=\;& \textstyle\sum_{m\in\mathbb{B}}\sum_{p\in\mathbb{N}}\Bigg\{\Bigg[\big(\boldsymbol{\mu}_{gm}\cdot\mathbf{E}(\omega_p)\big)\Big(\frac{1}{(\omega_{mg}+\omega_p)}-\frac{\alpha^*(t)e^{-iI_pt}}{(\omega_{mg}+\omega_p-i\delta^*)}\Big)\boldsymbol{\mu}_{mg}\alpha(t)\\
&+\big(\boldsymbol{\mu}_{mg}\cdot\mathbf{E}(\omega_p)\big)\Big(\frac{1}{(\omega_{mg}-\omega_p)}-\frac{\alpha(t)e^{iI_pt}}{(\omega_{mg}-\omega_p+i\delta)}\Big)\boldsymbol{\mu}_{gm}\alpha(t)\\
&+\textstyle\int_{\mathbb{R}^3} d^3\mathbf{p}\,\big(\boldsymbol{\mu}_{gm}\cdot\mathbf{E}(\omega_p)\big)\Big(\frac{1}{(\omega_{mg}+\omega_p)}-\frac{\alpha^*(t)e^{-iI_pt}}{(\omega_{mg}+\omega_p-i\delta^*)}\Big)b(\mathbf{p},t)\mathbf{d}_m(\mathbf{p})\\
&+\textstyle\int_{\mathbb{R}^3} d^3\mathbf{p}\,\big(\boldsymbol{\mu}_{mg}\cdot\mathbf{E}(\omega_p)\big)\Big(\frac{1}{(\omega_{mg}-\omega_p)}-\frac{\alpha(t)e^{iI_pt}}{(\omega_{mg}-\omega_p+i\delta)}\Big)b^*(\mathbf{p},t)\mathbf{d}_m^*(\mathbf{p})\Bigg]e^{-i\omega_pt}\Bigg\}
\end{aligned} \quad (39)$$

For the 2^{nd} order dipole moment we have

$$\begin{aligned}
\mathbf{d}_{BL}^{(2)}(t) \;=\;& \textstyle\sum_{(m,n)\in\mathbb{B}^2}\sum_{(p,q)\in\mathbb{N}^2}\Bigg\{\Bigg[\big(\boldsymbol{\mu}_{mn}\cdot\mathbf{E}(\omega_q)\big)\big(\boldsymbol{\mu}_{gm}\cdot\mathbf{E}(\omega_p)\big)\Big[\frac{1}{(\omega_{mg}+\omega_p)(\omega_{ng}+\omega_p+\omega_q)}\\
&-\frac{\alpha^*(t)e^{-iI_pt}}{(\omega_{mg}+\omega_p-i\delta^*)(\omega_{ng}+\omega_p+\omega_q-i\delta^*)}\Big]\boldsymbol{\mu}_{ng}\alpha(t)\\
&+\big(\boldsymbol{\mu}_{nm}\cdot\mathbf{E}(\omega_q)\big)\big(\boldsymbol{\mu}_{mg}\cdot\mathbf{E}(\omega_p)\big)\Big[\frac{1}{(\omega_{mg}-\omega_p)(\omega_{ng}-\omega_p-\omega_q)}\\
&-\frac{\alpha(t)e^{iI_pt}}{(\omega_{mg}-\omega_p+i\delta)(\omega_{ng}-\omega_p-\omega_q+i\delta)}\Big]\boldsymbol{\mu}_{0n}\alpha^*(t)\\
&+\textstyle\int_{\mathbb{R}^3} d^3\mathbf{p}\,\big(\boldsymbol{\mu}_{mn}\cdot\mathbf{E}(\omega_q)\big)\big(\boldsymbol{\mu}_{gm}\cdot\mathbf{E}(\omega_p)\big)\Big[\frac{1}{(\omega_{mg}+\omega_p)(\omega_{ng}+\omega_p+\omega_q)}\\
&-\frac{\alpha^*(t)e^{-iI_pt}}{(\omega_{mg}+\omega_p-i\delta^*)(\omega_{ng}+\omega_p+\omega_q-i\delta^*)}\Big]b(\mathbf{p},t)\mathbf{d}_n(\mathbf{p})\\
&+\textstyle\int_{\mathbb{R}^3} d^3\mathbf{p}\,\big(\boldsymbol{\mu}_{nm}\cdot\mathbf{E}(\omega_q)\big)\big(\boldsymbol{\mu}_{mg}\cdot\mathbf{E}(\omega_p)\big)\Big[\frac{1}{(\omega_{mg}-\omega_p)(\omega_{ng}-\omega_p-\omega_q)}\\
&-\frac{\alpha(t)e^{iI_pt}}{(\omega_{mg}-\omega_p+i\delta)(\omega_{ng}-\omega_p-\omega_q+i\delta)}\Big]b^*(\mathbf{p},t)\mathbf{d}_n^*(\mathbf{p})\Bigg]e^{-i(\omega_p+\omega_q)t}\Bigg\}
\end{aligned} \quad (40)$$

Finally for the 3^{rd} order bound-free dipole moment we have

$$
\begin{aligned}
\mathbf{d}_{BL}^{(3)}(t) \quad = \quad & \sum_{(m,n,\nu)\in\mathbb{B}^3}\sum_{(p,q,r)\in\mathbb{N}^3}\Bigg\{\Bigg[\big(\boldsymbol{\mu}_{n\sigma}.\mathbf{E}(\omega_r)\big)\big(\boldsymbol{\mu}_{mn}\cdot\mathbf{E}(\omega_q)\big)\big(\boldsymbol{\mu}_{gm}\cdot\mathbf{E}(\omega_p)\big) \\
& \times\Bigg(\frac{1}{(\omega_{mg}+\omega_p)(\omega_{ng}+\omega_p+\omega_q)(\omega_{\nu g}+\omega_p+\omega_q+\omega_r)} \\
& -\frac{\alpha^*(t)e^{-iI_pt}}{(\omega_{mg}+\omega_p-i\delta^*)(\omega_{ng}+\omega_p+\omega_q-i\delta^*)(\omega_{\nu g}+\omega_p+\omega_q+\omega_r-i\delta^*)}\Bigg)\boldsymbol{\mu}_{\nu g}\alpha(t) \\
& +\big(\boldsymbol{\mu}_{\sigma n}\cdot\mathbf{E}(\omega_r)\big)\big(\boldsymbol{\mu}_{nm}\cdot\mathbf{E}(\omega_q)\big)\big(\boldsymbol{\mu}_{mg}\cdot\mathbf{E}(\omega_p)\big) \\
& \times\Bigg(\frac{1}{(\omega_{mg}-\omega_p)(\omega_{ng}-\omega_p-\omega_q)(\omega_{\nu g}-\omega_p-\omega_q-\omega_r)} \\
& -\frac{\alpha(t)e^{iI_pt}}{(\omega_{mg}-\omega_p+i\delta)(\omega_{ng}-\omega_p-\omega_q+i\delta)(\omega_{\nu g}-\omega_p-\omega_q-\omega_r+i\delta)}\Bigg)\boldsymbol{\mu}_{g\nu}\alpha(t) \\
& +\int_{\mathbb{R}^3}d^3\mathbf{p}\,\big(\boldsymbol{\mu}_{n\sigma}.\mathbf{E}(\omega_r)\big)\big(\boldsymbol{\mu}_{mn}\cdot\mathbf{E}(\omega_q)\big)\big(\boldsymbol{\mu}_{gm}\cdot\mathbf{E}(\omega_p)\big) \\
& \times\Bigg(\frac{1}{(\omega_{mg}+\omega_p)(\omega_{ng}+\omega_p+\omega_q)(\omega_{\nu g}+\omega_p+\omega_q+\omega_r)} \\
& -\frac{\alpha^*(t)e^{-iI_pt}}{(\omega_{mg}+\omega_p-i\delta^*)(\omega_{ng}+\omega_p+\omega_q-i\delta^*)(\omega_{\nu g}+\omega_p+\omega_q+\omega_r-i\delta^*)}\Bigg)b(\mathbf{p},t)\mathbf{d}_\nu(\mathbf{p}) \\
& +\int_{\mathbb{R}^3}d^3\mathbf{p}\,\big(\boldsymbol{\mu}_{\sigma n}\cdot\mathbf{E}(\omega_r)\big)\big(\boldsymbol{\mu}_{nm}\cdot\mathbf{E}(\omega_q)\big)\big(\boldsymbol{\mu}_{mg}\cdot\mathbf{E}(\omega_p)\big) \\
& \times\Bigg(\frac{1}{(\omega_{mg}-\omega_p)(\omega_{ng}-\omega_p-\omega_q)(\omega_{\nu g}-\omega_p-\omega_q-\omega_r)} \\
& -\frac{\alpha(t)e^{iI_pt}}{(\omega_{mg}-\omega_p+i\delta)(\omega_{ng}-\omega_p-\omega_q+i\delta)(\omega_{\nu g}-\omega_p-\omega_q-\omega_r+i\delta)}\Bigg)b^*(\mathbf{p},t)\mathbf{d}^*_\nu(\mathbf{p}) \\
& \Bigg]e^{-i(\omega_p+\omega_q+\omega_r)t}\Bigg\}
\end{aligned} \tag{41}
$$

We then get

$$\mathbf{P}(\mathbf{x}',t)=\mathbf{P}_{BB}(\mathbf{x}',t)+\mathbf{P}_{BL}(\mathbf{x}',t)+\mathbf{P}_{LL}(\mathbf{x}',t) \tag{42}$$

with

$$\mathbf{P}_{BB}(\mathbf{x}',t)=\mathcal{N}\mathbf{d}_{BB}(t),\qquad \mathbf{P}_{BL}(\mathbf{x}',t)=\mathcal{N}\mathbf{d}_{BL}(t),\qquad \mathbf{P}_{LL}(\mathbf{x}',t)=\mathcal{N}\mathbf{d}_{LL}(t)$$

where $\mathbf{d}_{BB}(t)$, $\mathbf{d}_{BL}(t)$ and $\mathbf{d}_{LL}(t)$ are approximated from (24), (27), (44) then (32) and finally (34), (40), (41). To conclude we take $\lambda=1$. In fine, $\mathbf{P}$ has computed in (42) is used in (1).

Remark 4.3. *The contribution $\mathbf{d}_{BL}$ is estimated using [24], where transitions from "continuum-continuum" and "continuum-excited states" are neglected. As a consequence, the contribution $\mathbf{d}_{BL}^{(l)}$, for $l\geqslant 1$ may be, in principle, neglected as by construction of SFA model, these transitions were neglected. In the next section, we will show how to improve the modeling, including contribution more "continuum-bound state" transitions.*

That is

$$
\begin{cases}
\mathbf{P}_{BB}(\mathbf{x}',t) & \approx & \frac{\mathcal{N}}{\hbar}\sum_{m\in\mathbb{B}}\sum_{p\in\mathbb{N}}\Big\{\boldsymbol{\mu}_{mg}\cdot\big(\boldsymbol{\mu}_{mg}\cdot\mathbf{E}(\omega_p)\big)\Big(\frac{1-\alpha^*(t)e^{-iI_pt}}{\omega_{mg}-\omega_p}-\frac{\alpha(t)\big(1-\alpha^*(t)e^{-iI_pt}\big)}{\omega_{mg}-\omega_p+i\delta}\Big) \\
& & +\big(\boldsymbol{\mu}_{gm}\cdot\mathbf{E}(\omega_p)\big)\boldsymbol{\mu}_{mg}\Big(\frac{1-\alpha(t)}{\omega_{mg}+\omega_p}-\frac{\alpha^*(t)e^{-iI_pt}\big(1-\alpha(t)e^{iI_pt}\big)}{\omega_{mg}+\omega_p-i\delta^*}\Big)\Big\}e^{-i\omega_pt} \\
& & +\frac{2\mathcal{N}}{\hbar^2}\sum_{(p,q)\in\mathbb{N}^2}\sum_{(m,n)\in\mathbb{B}^2}\mathrm{Re}\Big\{\boldsymbol{\mu}_{gn}\big(\boldsymbol{\mu}_{nm}\cdot\mathbf{E}(\omega_q)\big)\big(\boldsymbol{\mu}_{mg}\cdot\mathbf{E}(\omega_p)\big) \\
& & \times\Big(\frac{1-\alpha^*(t)e^{-iI_pt}}{(\omega_{ng}-\omega_p-\omega_q)(\omega_{mg}-\omega_p)}-\frac{\big(1-\alpha^*(t)\big)\alpha(t)e^{iI_pt}}{(\omega_{mg}-\omega_p+i\delta)(\omega_{ng}-\omega_p-\omega_q+i\delta)}\Big)e^{-i(\omega_p+\omega_q)t}\Big\} \\
& & +\frac{\mathcal{N}}{\hbar}\sum_{(p,m)\in\mathbb{N}\times\mathbb{B}}\Big\{\boldsymbol{\mu}_{mg}\big(\boldsymbol{\mu}_{mg}\cdot\mathbf{E}(\omega_p)\big)\Big(\frac{1-\alpha^*(t)}{\omega_{mg}-\omega_p}-\frac{\alpha(t)e^{iI_pt}\big(1-\alpha^*(t)e^{-iI_pt}\big)}{\omega_{mg}-\omega_p+i\delta}\Big) \\
& & +\big(\boldsymbol{\mu}_{gm}\cdot\mathbf{E}(\omega_p)\big)\cdot\boldsymbol{\mu}_{mg}\Big(\frac{1-\alpha(t)}{\omega_{mg}+\omega_p}-\frac{\alpha^*(t)e^{-iI_pt}\big(1-\alpha(t)e^{iI_pt}\big)}{\omega_{mg}+\omega_p-i\delta^*}\Big)\Big\}e^{-i\omega_pt} \\
\mathbf{P}_{LL}(\mathbf{x}',t) & \approx & i\mathcal{N}\int_0^t\int d\tau d^3\mathbf{p}\mathbf{E}(t-\tau)\cdot\mathbf{d}_g\big(\mathbf{p}-\mathbf{A}(t-\tau)\big)\boldsymbol{d}_g^*\big((\mathbf{p}-\mathbf{A}(t)\big)e^{-iS(\mathbf{p},t,\tau)-\delta^*t-\delta(t-\tau)} \\
\mathbf{P}_{BL}(\mathbf{x}',t) & \approx & -2\mathcal{N}\mathrm{Re}\Big\{i\int_{\mathbf{R}^3}d^3\mathbf{v}\int_0^t d\tau\big(1-\alpha_0e^{-\delta^*t}\big)\mathbf{E})(\tau)\cdot\mathbf{d}_g^*\big(\mathbf{v}-A(t)\big)\mathbf{d}_g\big(\mathbf{v}-A(\tau)\big)e^{iS(\mathbf{v},t,\tau)}\Big\}
\end{cases}
$$

Now a bit more detailed analysis is necessary to estimate the susceptibility tensors.
First, the linear susceptibility $\chi^{(1)}_{BB}$, is given through $P^{(1)}_{BB,i}=\sum_j\chi^{(1)}_{ij,BB}E_j(\omega_p)$ by

$$\chi^{(1)}_{BB,ij}(t)=\frac{\mathcal{N}}{\hbar}\sum_{m\in\mathbb{B}}\Big\{\mu^i_{mg}\mu^j_{mg}\Big(\frac{1-\alpha^*(t)}{\omega_{mg}-\omega_p}-\frac{\alpha(t)\big(1-\alpha^*(t)\big)}{\omega_{mg}-\omega_p+i\delta}\Big)+\mu^j_{gm}\mu^i_{mg}\Big(\frac{1-\alpha(t)}{\omega_{mg}+\omega_p}-\frac{\alpha^*(t)\big(1-\alpha(t)\big)}{\omega_{mg}+\omega_p-i\delta^*}\Big)\Big\}$$

Assuming that $\alpha=0$, the linear susceptibility is the same usual (from perturbation theory).
Next, using again [25]'s notations, and

$$P^{(2)}_{BB,i}=\sum_{jk}\sum_{pq}\chi^{(2)}_{BB,ijk}\big(\omega_p+\omega_q,\omega_q,\omega_p\big)E_j(\omega_q)E_k(\omega_p)$$

the second-order susceptibility is given by

$$
\begin{aligned}
\chi^{(2)}_{BB,ijk}(\omega_p+\omega_q,\omega_q,\omega_p) \quad = \quad & \frac{\mathcal{N}}{\hbar^2}\mathcal{P}_I\sum_{(m,n)\in\mathbb{B}^2}\Bigg(\mu^i_{gn}\mu^j_{nm}\mu^k_{mg} \\
& \times\Big(\frac{\big(1-\alpha^*(t)e^{-iI_pt}\big)}{(\omega_{mg}-\omega_p)(\omega_{ng}-\omega_p-\omega_q)}-\frac{\alpha(t)e^{iI_pt}\big(1-\alpha^*(t)e^{-iI_pt}\big)}{(\omega_{mg}-\omega_p+i\delta)(\omega_{ng}-\omega_p-\omega_q+i\delta)}) \\
& +\mu^j_{gn}\mu^i_{nm}\mu^k_{mg}\Big(\frac{1}{\omega_{ng}+\omega_q}-\frac{\alpha^*(t)e^{-iI_pt}}{\omega_{ng}+\omega_q-i\delta^*}\Big)\Big(\frac{1}{\omega_{mg}-\omega_p}-\frac{\alpha(t)e^{iI_pt}}{\omega_{mg}-\omega_p+i\delta}\Big) \\
& +\mu^j_{gn}\mu^k_{nm}\mu^i_{mg} \\
& \times\Big(\frac{\big(1-\alpha(t)e^{iI_pt}\big)}{(\omega_{mg}+\omega_p)(\omega_{ng}+\omega_p+\omega_q)}-\frac{\alpha^*(t)e^{-iI_pt}\big(1-\alpha(t)e^{iI_pt}\big)}{(\omega_{mg}+\omega_p-i\delta^*)(\omega_{ng}+\omega_p+\omega_q-i\delta^*)}\Big)\Bigg)
\end{aligned}
$$

where $\mathcal{P}_I$ the intrindic permutation operator.

Finally the expression of the third-order susceptibility $\chi^{(3)}_{BB,kjih}$ can be deduced from above calculation through:

$$P^{(3)}_{BB,k}(\omega_p+\omega_q+\omega_r) = \sum_{hij}\sum_{pqr}\chi^{(3)}_{BB,kjih}\big(\omega_p+\omega_q+\omega_r,\omega_r,\omega_q,\omega_p\big)E_j(\omega_r)E_i(\omega_q)E_h(\omega_p)$$

by

$$\begin{aligned}
\chi^{(3)}_{BB,kjih}(\omega_p+\omega_q+\omega_r,\omega_r,\omega_q,\omega_p) \quad = \quad & \frac{\mathcal{N}}{\hbar^3}\mathcal{P}_I\sum\nolimits_{(m,n,\nu)\in\mathbb{B}^3}\Bigg(\mu^k_{g\nu}\mu^j_{\nu n}\mu^i_{nm}\mu^h_{mg}\\
&\times\Bigg(\frac{\big(1-\alpha^*(t)e^{-iI_pt}\big)}{(\omega_{mg}-\omega_p)(\omega_{ng}-\omega_p-\omega_q)(\omega_{\nu g}-\omega_p-\omega_q-\omega_r)}\\
&-\frac{\big(1-\alpha^*(t)e^{-iI_pt}\big)\alpha(t)e^{iI_pt}}{(\omega_{mg}-\omega_p+i\delta)(\omega_{ng}-\omega_p-\omega_q+i\delta)(\omega_{\nu g}-\omega_p-\omega_q-\omega_r+i\delta)}\Bigg)\\
&+\mu^j_{g\nu}\mu^k_{\nu n}\mu^i_{nm}\mu^h_{mg}\left(\frac{1}{\omega_{\nu g}+\omega_r}-\frac{\alpha^*(t)e^{-iI_pt}}{\omega_{\nu g}+\omega_r-i\delta^*}\right)\\
&\times\left(\frac{1}{(\omega_{mg}-\omega_p)(\omega_{ng}-\omega_p-\omega_q)}-\frac{\alpha(t)e^{iI_pt}}{(\omega_{mg}-\omega_p+i\delta)(\omega_{ng}-\omega_p-\omega_q+i\delta)}\right)\\
&+\mu^j_{g\nu}\mu^i_{\nu n}\mu^k_{nm}\mu^h_{mg}\left(\frac{1}{\omega_{\nu g}+\omega_r}-\frac{\alpha(t)e^{iI_pt}}{\omega_{\nu g}-\omega_r+i\delta}\right)\\
&\times\left(\frac{1}{(\omega_{mg}+\omega_p)(\omega_{ng}+\omega_p+\omega_q)}-\frac{\alpha^*(t)e^{-iI_pt}}{(\omega_{mg}+\omega_p-i\delta^*)(\omega_{ng}+\omega_p+\omega_q-i\delta^*)}\right)\\
&+\mu^k_{g\nu}\mu^j_{\nu n}\mu^i_{nm}\mu^h_{mg}\\
&\times\Bigg(\frac{\big(1-\alpha(t)e^{iI_pt}\big)}{(\omega_{mg}+\omega_p)(\omega_{ng}+\omega_p+\omega_q)(\omega_{\nu g}+\omega_p+\omega_q+\omega_r)}\\
&-\frac{\alpha^*(t)e^{-iI_pt}\big(1-\alpha(t)e^{iI_pt}\big)}{(\omega_{mg}+\omega_p-i\delta^*)(\omega_{ng}+\omega_p+\omega_q-i\delta^*)(\omega_{\nu g}+\omega_p+\omega_q+\omega_r-i\delta^*)}\Bigg)\Bigg)
\end{aligned}$$

Now regarding the bound-free susceptibility tensors we have:

$$\begin{aligned}
\chi^{(1)}_{BL,ij}(\omega_p) \quad = \quad & \frac{\mathcal{N}}{\varepsilon_0}\sum_{m\in\mathbb{B}}\sum_{p\in\mathbb{N}}\Bigg[\mu^j_{gm}\mu^i_{mg}\left(\frac{1}{(\omega_{mg}+\omega_p)}-\frac{\alpha^*(t)e^{-iI_pt}}{(\omega_{mg}+\omega_p-i\delta^*)}\right)\alpha(t)\\
&+\mu^j_{mg}\mu^i_{gm}\left(\frac{1}{(\omega_{mg}-\omega_p)}-\frac{\alpha(t)e^{iI_pt}}{(\omega_{mg}-\omega_p+i\delta)}\right)\alpha^*(t)\\
&+\int_{\mathbb{R}^3}d^3\mathbf{p}\,\mu^j_{gm}d^i_m(\mathbf{p})\left(\frac{1}{(\omega_{mg}+\omega_p)}-\frac{\alpha^*(t)e^{-iI_pt}}{(\omega_{mg}+\omega_p-i\delta^*)}\right)b(\mathbf{p},t)\\
&+\int_{\mathbb{R}^3}d^3\mathbf{p}\,\mu^j_{mg}(d^i_m(\mathbf{p}))^*\left(\frac{1}{(\omega_{mg}-\omega_p)}-\frac{\alpha(t)e^{iI_pt}}{(\omega_{mg}-\omega_p+i\delta)}\right)b^*(\mathbf{p},t)\Bigg]
\end{aligned}$$

then

$$\chi^{(2)}_{BL,ijk}(\omega_r,\omega_q,\omega_p) = \sum_{(m,n)\in\mathbb{B}^2}\Bigg[\mu^j_{mn}\mu^k_{gm}\mu^i_{ng}\Big[\frac{1}{(\omega_{mg}+\omega_p)}\frac{1}{(\omega_{ng}+\omega_p+\omega_q)}-\frac{1}{(\omega_{mg}+\omega_p-i\delta^*)}\frac{\alpha^*(t)e^{-iI_pt}}{(\omega_{ng}+\omega_p+\omega_q-i\delta^*)}\Big]\alpha(t)$$
$$+\mu^j_{nm}\mu^k_{mg}\mu^i_{0n}\Big[\frac{1}{(\omega_{mg}-\omega_p)}\frac{1}{(\omega_{ng}-\omega_p-\omega_q)}-\frac{\alpha(t)e^{iI_pt}}{(\omega_{mg}-\omega_p+i\delta)(\omega_{ng}-\omega_p-\omega_q+i\delta)}\Big]\alpha^*(t)$$
$$+\int_{\mathbb{R}^3}d^3\mathbf{p}\,\mu^j_{mn}\mu^k_{gm}d^i_n(\mathbf{p})\Big[\frac{1}{(\omega_{mg}+\omega_p)(\omega_{ng}+\omega_p+\omega_q)}$$
$$-\frac{\alpha^*(t)e^{-iI_pt}}{(\omega_{mg}+\omega_p-i\delta^*)(\omega_{ng}+\omega_p+\omega_q-i\delta^*)}\Big]b(\mathbf{p},t)$$
$$+\int_{\mathbb{R}^3}d^3\mathbf{p}\,\mu^j_{nm}\mu^k_{mg}(\mathbf{d}^i_n)^*(\mathbf{p})\Big[\frac{1}{(\omega_{mg}-\omega_p)(\omega_{ng}-\omega_p-\omega_q)}$$
$$-\frac{\alpha(t)e^{iI_pt}}{(\omega_{mg}-\omega_p+i\delta)(\omega_{ng}-\omega_p-\omega_q+i\delta)}\Big]b^*(\mathbf{p},t)\Bigg]$$

and

$$\chi^{(3)}_{BL,kjih}(\omega_\sigma,\omega_r,\omega_q,\omega_p) = \sum_{(m,n,\nu)\in\mathbb{B}^3}\sum_{(p,q,r)\in\mathbb{N}^3}\Bigg[\mu^j_{n\sigma}\mu^i_{mn}\mu^h_{gm}\mu^k_{\nu g}\Big(\frac{1}{(\omega_{mg}+\omega_p)(\omega_{ng}+\omega_p+\omega_q)(\omega_{\nu g}+\omega_p+\omega_q+\omega_r)}$$
$$-\frac{\alpha^*(t)e^{-iI_pt}}{(\omega_{mg}+\omega_p-i\delta^*)(\omega_{ng}+\omega_p+\omega_q-i\delta^*)(\omega_{\nu g}+\omega_p+\omega_q+\omega_r-i\delta^*)}\Big)\alpha(t)$$
$$+\mu^j_{\sigma n}\mu^i_{nm}\mu^h_{mg}\mu^k_{g\nu}\Big(\frac{1}{(\omega_{mg}-\omega_p)(\omega_{ng}-\omega_p-\omega_q)(\omega_{\nu g}-\omega_p-\omega_q-\omega_r)}$$
$$-\frac{\alpha(t)e^{iI_pt}}{(\omega_{mg}-\omega_p+i\delta)(\omega_{ng}-\omega_p-\omega_q+i\delta)(\omega_{\nu g}-\omega_p-\omega_q-\omega_r+i\delta)}\Big)\alpha(t)$$
$$+\int_{\mathbb{R}^3}d^3\mathbf{p}\,\mu^j_{n\sigma}\mu^i_{mn}\mu^h_{gm}d^k_\nu(\mathbf{p})\Big(\frac{1}{(\omega_{mg}+\omega_p)(\omega_{ng}+\omega_p+\omega_q)(\omega_{\nu g}+\omega_p+\omega_q+\omega_r)}$$
$$-\frac{1}{(\omega_{mg}+\omega_p-i\delta^*)}\frac{1}{(\omega_{ng}+\omega_p+\omega_q-i\delta^*)}\frac{\alpha^*(t)e^{-iI_pt}}{(\omega_{\nu g}+\omega_p+\omega_q+\omega_r-i\delta^*)}\Big)b(\mathbf{p},t)$$
$$+\int_{\mathbb{R}^3}d^3\mathbf{p}\,\mu^j_{\sigma n}\mu^i_{nm}\mu^h_{mg}(\mathbf{d}^k_\nu)^*(\mathbf{p})\times\Big(\frac{1}{(\omega_{mg}-\omega_p)(\omega_{ng}-\omega_p-\omega_q)(\omega_{\nu g}-\omega_p-\omega_q-\omega_r)}$$
$$-\frac{\alpha(t)e^{iI_pt}}{(\omega_{mg}-\omega_p+i\delta)(\omega_{ng}-\omega_p-\omega_q+i\delta)(\omega_{\nu g}-\omega_p-\omega_q-\omega_r+i\delta)}\Big)b^*(\mathbf{p},t)\Bigg]$$

and where $\omega_\sigma=\omega_p+\omega_q+\omega_r$.

Interpretation and simplification of these tensors, such as the third harmonic generation from $\chi^{(3)}(3\omega)$, will be analyzed is a forthcoming paper.

4.3. SFA-like nonlinear optics model: possible extension to multiple continuous states

The use of the SFA modeling implies (implicitly) that some transitions are (by construction) neglected: "continuum-continuum" and "continuum-excited states" transitions [24]. In the following section, we propose to formally include more properly and accurately some of these transitions, which will allow us to model more accurately $\mathbf{d}_{BL}$ and $\mathbf{d}_{LL}$. In this goal, we will search for ψ in the form:

$$\psi(\mathbf{x},t)=\psi_B(\mathbf{x},t)+\psi_L(\mathbf{x},t)$$

where the purely bound part is of the form

$$\psi_B(\mathbf{x},t) = \sum_{l\in\mathbb{N}} \lambda^l \psi_B^{(l)}(\mathbf{x},t)$$

with

$$\psi_B^{(l)}(\mathbf{x},t) = \sum_{k\in\mathbb{N}} a_k^{(l)}(t)\phi_k(\mathbf{x})e^{-i\omega_k t}$$

where we have denoted ϕ_k the eigenvectors of the field-free Hamiltonian: $H_0\phi_k = \varepsilon_k\phi_k$. The bound-continuous part is of the form of SFA:

$$\psi_L(\mathbf{x},t) = \sum_{l\in\mathbb{N}} \lambda^l \psi_L^{(l)}(\mathbf{x},t)$$

where for $l \geqslant 0$

$$\psi_L^{(l)}(\mathbf{x},t) = e^{iI_p^{(l)}t}\Big(\alpha_l(t)\phi_l(\mathbf{x}) + \int d^3\mathbf{p}\, b^{(l)}(\mathbf{p},t)e^{i\mathbf{p}\cdot\mathbf{x}}\Big)$$

and with $\alpha_l(t)$ a time-dependent parameter and $I_P^{(l)}$ is defined as a relative ionization potential, from Level l energy

$$I_p^{(l)} = I_p - (\omega_l - \omega_0) = -\omega_l \tag{43}$$

In addition, we impose:

- for all $l \geqslant 0$, $b^{(l)}(\mathbf{p},0) = 0$,
- for $l = 0$, $\alpha_0(0) \in (0,1)$ and for $l \geqslant 1$, $\alpha_l(0) \ll 1$.

$$\begin{aligned}
\mathbf{d}(t) &= \langle\psi_B|\widehat{\boldsymbol{\mu}}|\psi_B\rangle + \langle\psi_L|\widehat{\boldsymbol{\mu}}|\psi_L\rangle + \langle\psi_B|\widehat{\boldsymbol{\mu}}|\psi_L\rangle + \langle\psi_L|\widehat{\boldsymbol{\mu}}|\psi_B\rangle \\
&= \mathbf{d}_{BB}(t) + \mathbf{d}_{BL}(t) + \mathbf{d}_{LL}(t)
\end{aligned}$$

with

$$\mathbf{d}_{BB}(t) = \sum_{l\in\mathbb{N}} \lambda^l \mathbf{d}_{BB}^{(l)}(t) \tag{44}$$

which takes the same value as above (28), (30), (31). Then

$$\mathbf{d}_{BL}(t) = \sum_{(k,l)\in\mathbb{N}^2} \lambda^{k+l}\mathbf{d}_{BL}^{(kl)}(t), \qquad \mathbf{d}_{LL}(t) = \sum_{(k,l)\in\mathbb{N}^2} \lambda^{k+l}\mathbf{d}_{LL}^{(kl)}(t)$$

where

$$\mathbf{d}_{BL}^{(kl)}(t) = \sum_{(k,l)\in\mathbb{N}^2} \Big(\langle\psi_B^{(k)}|\widehat{\boldsymbol{\mu}}|\psi_L^{(l)}\rangle + \langle\psi_L^{(k)}|\widehat{\boldsymbol{\mu}}|\psi_B^{(l)}\rangle\Big) \tag{45}$$

and

$$\mathbf{d}_{LL}^{(kl)}(t) = \sum_{(k,l)\in\mathbb{N}^2} \Big(\langle\psi_L^{(k)}|\widehat{\boldsymbol{\mu}}|\psi_L^{(l)}\rangle + \langle\psi_L^{(k)}|\widehat{\boldsymbol{\mu}}|\psi_L^{(l)}\rangle\Big) \tag{46}$$

As $\mathbf{d}_{BL,LL}$ is computed according to mathematical assumptions from [24], the following terms are neglected: $\langle\psi_B^{(k)}|\widehat{\boldsymbol{\mu}}|\psi_L^{(l)}\rangle \approx 0$ and $\langle\psi_L^{(k)}|\widehat{\boldsymbol{\mu}}|\psi_L^{(l)}\rangle \approx 0$ for $k \neq l$. Then for all l:

$$\mathbf{d}_{BL}(t) \approx \sum_l \lambda^{2l}\mathbf{d}_{BL}^{(ll)}(t) = 2\sum_l \lambda^{2l}\mathrm{Re}\langle\psi_B^{(l)}|\widehat{\boldsymbol{\mu}}|\psi_L^{(l)}\rangle$$

where

$$\mathbf{d}_{BL}^{(ll)}(t) = 2\mathrm{Re}\Big\{ \sum_{m\in\mathbb{B}} \Big(\big(a_m^{(l)}(t)\big)^* e^{i(\omega_m + I_p^{(l)})t}\mathbf{d}_{mg}\alpha_l(t) + \int_{\mathbb{R}^3} d^3\mathbf{p}\big(a_m^{(l)}(t)\big)^* b^{(l)}(\mathbf{p},t)e^{i(\omega_m+I_p^{(l)})t}\mathbf{d}_m(\mathbf{p})\Big)\Big\} \tag{47}$$

where $\mathbf{d}_l(\mathbf{p}) = \langle \mathbf{p}|\widehat{\mu}|\phi_l\rangle$. Regarding the continuum-continuum contribution

$$\mathbf{d}_{LL}(t) \approx \sum_l \lambda^{2l}\mathbf{d}_{LL}^{(ll)}(t) = 2\sum_l \lambda^{2l}\mathrm{Re}\langle \psi_L^{(l)}|\widehat{\boldsymbol{\mu}}|\psi_L^{(l)}\rangle$$

According to (32), we get

$$\mathbf{d}_{LL}^{(ll)}(t) = i\int_0^t \int d\tau d^3\mathbf{p}\mathbf{E}(t-\tau)\cdot \mathbf{d}_l\big(\mathbf{p}-\mathbf{A}(t-\tau)\big)\mathbf{d}_{Ll}^*\big((\mathbf{p}-\mathbf{A}(t)\big)e^{-iS(\mathbf{p},t,\tau)-\delta_l^* t-\delta_l(t-\tau)}$$

We get a full description of macroscopic polarization.

$$\left\{\begin{array}{lcl}
\mathbf{P}_{BB}^l(\mathbf{x}',t) & \approx & \dfrac{\mathcal{N}}{\hbar}\sum_{(p,m)\in\mathbb{N}\times\mathbb{B}}\Big\{\boldsymbol{\mu}_{mg}\cdot\big(\boldsymbol{\mu}_{mg}\cdot\mathbf{E}(\omega_p)\big)\Big(\dfrac{1-\alpha_l^*(t)}{\omega_{mg}-\omega_p} - \dfrac{\alpha_l(t)\big(1-\alpha_l^*(t)\big)}{\omega_{mg}-\omega_p+i\delta}\Big\} \\
& & +\big(\boldsymbol{\mu}_{gm}\cdot\mathbf{E}(\omega_p)\big)\cdot\boldsymbol{\mu}_{mg}\Big(\dfrac{1-\alpha_l(t)}{\omega_{mg}+\omega_p} - \dfrac{\alpha_l^*(t)\big(1-\alpha_l(t)\big)}{\omega_{mg}+\omega_p-i\delta^*}\Big)\Big\}e^{-i\omega_p t} \\
& & +\dfrac{2\mathcal{N}}{\hbar^2}\sum_{(p,q)\in\mathbb{N}^2}\sum_{(m,n)\in\mathbb{B}^2}\mathrm{Re}\Big\{\mu_{gn}(\mu_{nm}\cdot\mathbf{E}(\omega_q))(\mu_{mg}\cdot\mathbf{E}(\omega_p)) \\
& & \times\Big(\dfrac{1-\alpha_l^*(t)}{(\omega_{ng}-\omega_p-\omega_q)(\omega_{mg}-\omega_p))} - \dfrac{\big(1-\alpha_l^*(t)\big)\alpha_l(t)e^{iI_p t}}{(\omega_{mg}-\omega_p+i\delta)(\omega_{ng}-\omega_p-\omega_q+i\delta)}\Big)e^{-i(\omega_p+\omega_q)t}\Big\} \\
& & +\dfrac{\mathcal{N}}{\hbar}\sum_{(p,m)\in\mathbb{N}\times\mathbb{B}}\Big\{\boldsymbol{\mu}_{mg}\cdot\big(\boldsymbol{\mu}_{mg}\cdot\mathbf{E}(\omega_p)\big)\Big(\dfrac{1-\alpha_l^*(t)}{\omega_{mg}-\omega_p} - \dfrac{\alpha_l(t)\big(1-\alpha_l^*(t)\big)}{\omega_{mg}-\omega_p+i\delta}\Big) \\
& & +\big(\boldsymbol{\mu}_{gm}\cdot\mathbf{E}(\omega_p)\big)\cdot\boldsymbol{\mu}_{mg}\Big(\dfrac{1-\alpha_l(t)}{\omega_{mg}+\omega_p} - \dfrac{\alpha_l^*(t)\big(1-\alpha_l(t)\big)}{\omega_{mg}+\omega_p-i\delta^*}\Big)\Big\}e^{-i\omega_p t} \\
 & & \\
\mathbf{P}_{LL}^l(\mathbf{x}',t) & \approx & i\mathcal{N}\sum_{l\in\mathbb{N}}\int_0^t\int d\tau d^3\mathbf{p}\mathbf{E}(t-\tau)\cdot\mathbf{d}_l\big(\mathbf{p}-\mathbf{A}(t-\tau)\big)\boldsymbol{d}_{Ll}^*\big((\mathbf{p}-\mathbf{A}(t)\big)e^{-iS(\mathbf{p},t,\tau)-\delta_l^* t-\delta_l(t-\tau)} \\
 & & \\
\mathbf{P}_{BL}^l(\mathbf{x}',t) & \approx & 2\mathcal{N}\sum_{l\in\mathbb{N}}\mathrm{Re}\Big\{\sum_{m\in\mathbb{B}}\Big(\big(a_m^{(l)}(t)\big)^* e^{i(\omega_m+I_p^{(l)})t}\mathbf{d}_{mg}\alpha_l(t) \\
& & +\int_{\mathbb{R}^3} d^3\mathbf{p}\big(a_m^{(l)}(t)\big)^* b^{(l)}(\mathbf{p},t)e^{i(\omega_m+I_p^{(l)})t}\mathbf{d}_m(\mathbf{p})\Big)\Big\}
\end{array}\right.$$

Remark 4.4. *One can also consider wavefunction of the type*

$$\psi_L(\boldsymbol{r},t) = \alpha_m(t)\phi_m(\boldsymbol{r}) + \alpha_n(t)\phi_n(\boldsymbol{r}) + \int_{\mathbb{R}^3} d^3\boldsymbol{p}\, b^{(mn)}(\boldsymbol{p},t)e^{i\boldsymbol{p}\cdot\boldsymbol{r}} \tag{48}$$

By plugging this expression into the Schrödinger equation and taking the inner product with $|\phi_m(\boldsymbol{r})\rangle$, $|\phi_n(\boldsymbol{r})\rangle$

and $|\boldsymbol{p}\rangle$ respectively, we get:

$$\begin{aligned}
i\dot{\alpha}_m(t) &= E_m\alpha_m(t) + \alpha_n(t)\mathbf{E}(t)\cdot\boldsymbol{\mu}_{mn} \\
&\quad + \int_{\mathbb{R}^3} d^3\boldsymbol{p}\, b^{(mn)}(\boldsymbol{p},t)\langle\phi_m|x|\boldsymbol{p}\rangle\cdot\mathbf{E}(t) \qquad (49)\\
i\dot{\alpha}_n(t) &= E_n\alpha_n(t) + \alpha_m(t)\mathbf{E}(t)\cdot\boldsymbol{\mu}_{nm} \\
&\quad + \int_{\mathbb{R}^3} d^3\boldsymbol{p}\, b^{(mn)}(\boldsymbol{p},t)\langle\phi_n|x|\boldsymbol{p}\rangle\cdot\mathbf{E}(t) \qquad (50)\\
i\dot{b}(\boldsymbol{p},t) &= \big(a_m(t)\mathbf{E}(t)\cdot\boldsymbol{d}_x^{(m)}(\boldsymbol{p}) + a_n(t)\boldsymbol{d}_x^{(n)}(\boldsymbol{p})\big) \\
&\quad -\frac{\boldsymbol{p}^2}{2}b^{(mn)}(\boldsymbol{p},t)\mathbf{E}(t)\partial_{p_x}b^{(mn)}(\boldsymbol{p},t) \qquad (51)
\end{aligned}$$

assuming $a_m(t)$ and $a_n(t)$ are constant in time, can be solved using the method of characteristics as shown above. Doing so implicitly means that the rate of change of $a_m(t)$ and $a_n(t)$ are much less than that of $b^{(mn)}(\boldsymbol{p},t)$. One acquires a coupled ODE with time dependent coefficients.

Alternatively, we can propose a close approach, which may be cheaper numerically, as follows: we search for ψ_L in the form of a discrete sum (which is then an approximation of the integral over the continuum). Let us denote by $\mathbb{W}$, the corresponding indices and including as well, the ground state. That is

$$\psi_L(\mathbf{x},t) = c_g(t)\phi_g(\mathbf{x}) + \sum_{m\in\mathbb{W}} c_m(t)\phi_m(\mathbf{x})$$

Plugging in the system in the TDSE gives:

$$\left\{\begin{array}{lcl}
i\dot{c}_g(t) &=& \varepsilon_g c_g(t) + \sum_{m\in\mathbb{W}}\mathbf{E}(t)\cdot\langle\phi_m(\mathbf{x})|\mathbf{x}|\phi_g\rangle c_m(t) \\
i\dot{c}_m(t) &=& \varepsilon_m c_m(t) + \mathbf{E}(t)\cdot\langle\phi_m(\mathbf{x})|\mathbf{x}|\phi_g(\mathbf{x})\rangle c_g(t)
\end{array}\right.$$

neglecting again the continuum-continuum transitions. Then ψ_L is combined with ψ_B are proposed in the above sections.

5. Conclusion

This paper was devoted to the derivation of two non-classical nonlinear optics models. In both cases, contribution of bound-bound, bound-continuous and to a certain extent continuous-continuous state transitions are evaluated in order to determine the macroscopic polarization in Maxwell's equations. Although the first model, fully non-pertrubative, is an important improvement of the Maxwell-Schrödinger model in term of computational complexity, it however still contains microscopic components: TDSE computations have to be performed to determined initial polarizations. In addition, from a practical point of view in this configuration, it is valid only for very short pulses. The second model, although fully macroscopic is however semi-empirical and partially perturbative.
In order to validate and to analyze these models, we plan, in a close future, to test them on realistic situations. In particular simple filamentation simulations will be performed. The inclusion of nonlinearities coming from bound states as well as free electrons, is essential in order to describe accurately filament dynamics [1], [2].

Appendix A. (Gaussian beam). We consider the extension to Gaussian beams of the method developed above. Using [25]'s notations (see also [33]), we denote by w_0 the beam waist, λ its wavelength, and $\mathcal{A}$ its maximal amplitude, see Fig. 8, and [22] for details about this Gaussian pulse. We assume that

$$\mathbf{E}(\mathbf{x}', t) = A(\mathbf{x}')e^{i(kz'-\omega t)}\mathbf{e}_{z'}$$

where $\mathbf{x}' = (r', z')$ and

$$A(r', z') = \frac{\mathcal{A}}{1+i\xi}\exp\Big(-\frac{r'^2}{w_0^2(1+i\xi)}\Big)$$

with $\xi = 2z'/b$ and $b = kw_0^2$. The beam power is given by

$$\mathcal{P} = \int \frac{nc}{2\pi}|A|^2 = \frac{1}{4}n\omega_0|\mathcal{A}|^2$$

Assuming that the dipole moment $\mathbf{d}(\mathbf{x}'_1, t)$ at $\mathbf{x}'_1 := (r'_1, z'_1)$ for any r'_1 is known, from the solution to

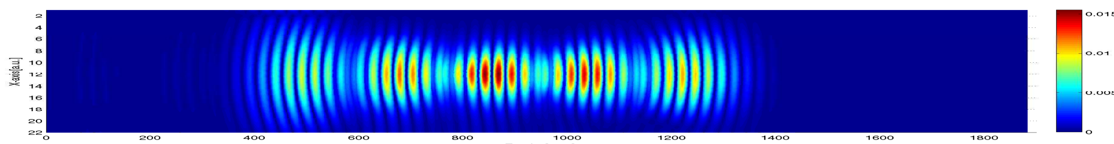

Figure 8: Gaussian beam intensity at different propagation times, in vacuum, in the (x, z)-plan, $I = 10^{13}\mathrm{W{\cdot}cm}^{-2}$

$$i\partial_t\psi = -\frac{1}{2}\triangle\psi + V_c\psi + \mathbf{x}\cdot\mathbf{E}_{\mathbf{x}'_1}\psi$$

we construct $\mathbf{d}(\mathbf{x}'_2, t)$ with $\mathbf{x}'_2 := (r'_1, z'_2)$ as follows. For $|\Delta z'|/b = |z'_2 - z'_1|/b$ small enough, we next rewrite $A(r'_1, z'_2)$ as a function of $A(r'_1, z'_1)$

$$A(r'_1, z'_2) = \frac{\mathcal{A}}{1+i\xi_2}\exp\Big(-\frac{r'^2_1}{w_0^2(1+i\xi_2)}\Big),\ \xi_2 = \frac{2z'_2}{b}$$

which can be rewritten, denoting $\Delta\xi = 2\Delta z'/b$

$$A(r'_1, z'_2) = \frac{\mathcal{A}}{1+i\xi_1+i\Delta\xi}\exp\Big(-\frac{r'^2_1}{w_0^2(1+i\xi_1+i\Delta\xi)}\Big)$$

and

$$\begin{aligned} A(r'_1, z'_2) &= A(r, z'_1)\frac{1+i\xi_1}{1+i\xi_1+i\Delta\xi}\exp\Big(-\frac{r'^2_1}{\omega_0^2}\big(\frac{1}{1+i\xi_1+i\Delta\xi} - \frac{1}{1+i\xi_1}\big)\Big) \\ &= A(r, z'_1)\frac{1+i\xi_1}{1+i\xi_1+i\Delta\xi}\exp\Big(-\frac{r'^2_1}{\omega_0^2(1+i\xi_1)}\Big)\exp\Big(\frac{1+i\xi_1}{1+i\xi_1+i\Delta\xi} - 1\Big) \end{aligned}$$

Using $|\Delta\xi| \ll 1$, we then have

$$\frac{1+i\xi_1}{1+i\xi_1+i\Delta\xi} = 1 - \frac{i\Delta\xi}{1+i\xi_1} - \frac{\Delta\xi^2}{(1+i\xi_1)^2} + \cdots$$

and as a consequence

$$A(r'_1, z'_2) \approx A(r, z'_1)\Big(1 - \frac{i\Delta\xi}{1+i\xi_1} - \frac{\Delta\xi^2}{(1+i\xi_1)^2}\Big)\Big(1 + \frac{r'^2_1}{\omega_0^2(1+i\xi_1)}\big(\frac{i\Delta\xi}{1+i\xi_1} + \frac{\Delta\xi^2}{(1+i\xi_1)^2}\big)\Big)$$

This gives a useful relation between $A(r_1', z_1')$ and $A(r_1', z_2')$. Denoting, now

$$\varepsilon(\xi_1, r_1', \Delta z') = -\frac{i\Delta\xi}{1+i\xi_1} + \frac{ir_1'^2\Delta\xi}{(1+i\xi_1)^2\omega_0^2} - \frac{\Delta\xi^2}{(1+i\xi_1)^2} + \frac{2r_1'^2\Delta\xi^2}{(1+i\xi_1)^3\omega_0^2}$$

we obtain

$$A(r_1', z_2') \approx A(r_1', z_1')\big(1+\varepsilon(\xi_1, r_1', \Delta z')\big)$$

In general $|A(r_1', z_2')| \not\approx |A(r_1', z_1')|$, so that $\mathbf{d}(\mathbf{x}_2', t) \not\approx \mathbf{d}\Big(\mathbf{x}_1', t - \Delta z'/v_g\Big)$. However, we can define $\Delta\rho_1'$ from

$$|A(r_1', z_2')| \approx |A(r_1' + \Delta\rho_1', z_1')| \tag{52}$$

so that $\mathbf{d}(\mathbf{x}_2', t) \approx \mathbf{d}\Big(\widetilde{\mathbf{x}_1'}, t - \dfrac{\Delta z'}{v_g}\Big)$, where $\widetilde{\mathbf{x}_1'} = (r_1' + \Delta\rho_1', z_1')$. In order to determine $\Delta\rho_1'$, we expand

$$\begin{aligned} A(r_1' + \Delta\rho_1', z_1') &= A(r_1', z_1')\exp\Big(-\frac{2r_1'\Delta\rho_1' + \Delta\rho_1'^2}{w_0^2(1+i\xi_1)}\Big) \\ &\approx A(r_1', z_1')\Big(1 - \frac{2r_1'\Delta\rho_1' + \Delta\rho_1'^2}{w_0^2(1+i\xi_1)} + \frac{(2r_1'\Delta\rho_1' + \Delta\rho_1'^2)^2}{2w_0^4(1+i\xi_1)^2}\Big) \end{aligned}$$

We set

$$\delta(\xi_1, \Delta\rho_1') := -\frac{2r_1'\Delta\rho_1' + \Delta\rho_1'^2}{w_0^2(1+i\xi_1)} + \frac{(2r_1'\Delta\rho_1' + \Delta\rho_1'^2)^2}{2w_0^4(1+i\xi_1)^2}$$

so that

$$A(r_1' + \Delta\rho_1', z_1') \approx A(r_1', z_1')\big(1+\delta(\xi_1, r_1', \Delta\rho_1')\big)$$

We now determine $\Delta\rho_1'$ such that (52) holds, that is $\Delta\rho_1'$ is such that

$$\delta(\xi_1, r_1', \Delta\rho_1') = \varepsilon(\xi_1, r_1', \Delta z')$$

As a first approximation

$$\Delta\rho_1' \approx \Delta z'\Gamma(r_1', z_1', \omega_0, b)$$

where we have set, recalling that $\xi_1 = 2z_1'/b$

$$\Gamma(r_1', z_1', \omega_0, b) := \pm\frac{\omega_0^2}{r_1' b}\Big|1 - \frac{r_1'^2}{(1+2iz_1'/b)\omega_0^2}\Big|$$

In Fig. 9, we represent the dipole moment propagating from $\widetilde{\mathbf{x}_1'}$ to $\mathbf{x}_2'$. We then deduce that

$$\mathbf{P}(\mathbf{x}_2', t) \approx \mathbf{P}\Big(\widetilde{\mathbf{x}_1'}, t - \frac{\Delta z'}{v_g}\Big)$$

A corresponding quasilinear model is, for $(r', z') \in \mathbb{R}_+^* \times (z_1', \infty)$

$$\partial_t\mathbf{P}(r', z', t) + \mathbf{V}(\mathbf{E}, \omega_0, b)\cdot\nabla\mathbf{P}(r', z', t) = \mathbf{0}$$

with initial data $\mathbf{P}(\widetilde{\mathbf{x}_1'}, 0)$ assumed to be known and where $\mathbf{V} = \big(\Gamma(r', z', \omega_0, b), v_g\big)$.

We conclude this section by a remark on a more accurate approximation for $\mathbf{P}(\mathbf{x}_2', t)$.

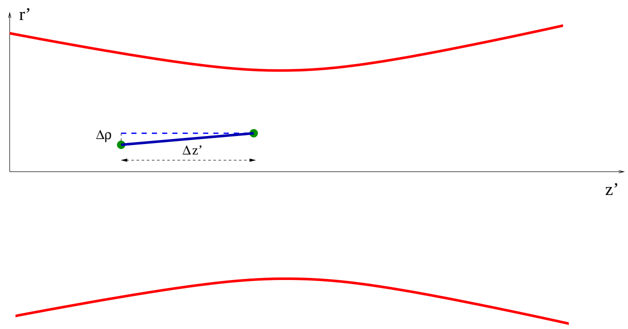

Figure 9: Dipole moment evolution

Remark 5.1. *In order to include electromagnetic field variation between* $\mathbf{x}'_1$ *and* $\widetilde{\mathbf{x}'_2}$*, it is possible to use an approach similar as the one proposed in Model 2 of Section 2.1. For the same reason as above, we get for* $t \in (t_a, t_b]$.

$$\begin{aligned}\mathbf{P}(\mathbf{x}'_2, t) \quad &\approx \quad \mathbf{P}\Big(\widetilde{\mathbf{x}'_1}, t - \frac{\Delta z'}{v_g}\Big) + \Delta T^2 \Big(\big(\Delta\boldsymbol{\mathcal{E}}^{(x)}(t_a)\big)^2 \mathbf{c}^{(x)}\Big(\widetilde{\mathbf{x}'_1}, t - \frac{\Delta z'}{v_g}\Big) \\ &+ \big(\Delta\boldsymbol{\mathcal{E}}^{(x)}(t_a)\big)^2 \mathbf{Q}^{(y)}\Big(\widetilde{\mathbf{x}'_1}, t_b - \frac{\Delta z'}{v_g}\Big) + 2\Delta\boldsymbol{\mathcal{E}}^{(x)}(t_a)\Delta\boldsymbol{\mathcal{E}}^{(y)}(t_a)\mathbf{Q}^{(xy)}\Big(\widetilde{\mathbf{x}'_1}, t_b - \frac{\Delta z'}{v_g}\Big)\Big)\end{aligned}$$

where

$$\Delta\boldsymbol{\mathcal{E}}(t_a) = \big(\Delta\boldsymbol{\mathcal{E}}^{(x)}(t_a), \Delta\boldsymbol{\mathcal{E}}^{(y)}(t_a)\big) := \mathbf{E}_{\mathbf{x}'_2}(t_a) - \mathbf{E}_{\widetilde{\mathbf{x}'_1}}\big(t - \Delta z'/v_g\big)$$

and $\mathbf{Q}^{(x,y,xy)}$ *are defined in* (6)*. Again, for* $z' > z'_1$*, we can model the evolution by*

$$\begin{cases} \partial_t \mathbf{P}(\mathbf{x}', t) + v_g \partial_z \mathbf{P}(\mathbf{x}', t) + \Gamma(r', z', \omega_0, b)\partial_{r'}\mathbf{P}(r', z', t) & = \quad \int_0^t \langle \mathbf{Q}(\mathbf{x}', s') \cdot \Delta\boldsymbol{\mathcal{E}}(\mathbf{x}', s'), \Delta\boldsymbol{\mathcal{E}}(\mathbf{x}', s')\rangle ds' \\ \partial_t \mathbf{Q}^{(x)}(\mathbf{x}', t) + v_g \partial_z \mathbf{Q}^{(x)}(\mathbf{x}', t) + \Gamma(r', z', \omega_0, b)\partial_{r'}\mathbf{Q}^{(x)}(r', z', t) & = \quad 0 \\ \partial_t \mathbf{Q}^{(y)}(\mathbf{x}', t) + v_g \partial_z \mathbf{Q}^{(y)}(\mathbf{x}', t) + \Gamma(r', z', \omega_0, b)\partial_{r'}\mathbf{Q}^{(y)}(r', z', t) & = \quad 0 \\ \partial_t \mathbf{Q}^{(xy)}(\mathbf{x}', t) + v_g \partial_z \mathbf{Q}^{(xy)}(\mathbf{x}', t) + \Gamma(r', z', \omega_0, b)\partial_{r'}\mathbf{Q}^{(xy)}(r', z', t) & = \quad 0 \end{cases}$$

with initial data $\mathbf{P}(\widetilde{\mathbf{x}_1}, 0)$.

An alternative approach can be derived from simple expansions as follows.

Remark 5.2. *Starting again from* $A(r' + \Delta\rho', z'_1)$ *but without using the Gaussian shape of the pulse, we first get* $A(r' + \Delta\rho', z'_1) \approx A(r', z'_1) + \Delta\rho'\partial_r A(r', z'_1)$*. Similarly in* $A(r', z'_2) \approx A(r', z'_1) + \Delta z'\partial_{z'} A(r', z'_1)$*. We deduce in first approximation that for a given* $\Delta z'$, $|\Delta\rho'| \approx \Big|\Delta z'\partial_{z'} A(r', z'_1)/\partial_{r'} A(r', z'_1)\Big|$.

Appendix B. (Generalized Liouville equation). For any $n \in \mathbb{N}$:

$$i\langle \partial_t \psi | \phi_n \rangle = \langle \big(H_0 + V(\mathbf{x}, t)\big)\psi | \phi_n \rangle$$

and for any $\lambda \in \sigma_c$

$$i\langle \partial_t \psi | \phi_c(\cdot, \lambda) \rangle = \langle \big(H_0 + V(\mathbf{x}, t)\big)\psi | \phi_c(\cdot, \lambda) \rangle$$

First, we get

$$i\partial_t\psi(\mathbf{x},t) = \sum_n \dot{c}_n(t)\phi_n(\mathbf{x}) + \int_{\sigma_c} \partial_t c(t,\eta)\phi_c(\mathbf{x},\eta)\rho_c(\eta)d\eta$$

and

$$\big(H_0 + V(\mathbf{x},t)\big)\psi(\mathbf{x},t) = \sum_n \big(\varepsilon_n + V(\mathbf{x},t)\big)c_n(t)\phi_n(\mathbf{x}) + \int_{\sigma_c} \big(\eta + V(\mathbf{x},t)\big)c(t,\eta)\phi_c(\mathbf{x},\eta)\rho_c(\eta)d\eta$$

As $\langle\phi_n|\phi_m\rangle = \delta_{nm}$,

$$i\langle\partial_t\psi|\phi_m\rangle = i\dot{c}_m(t) + i\int_{\sigma_c} \langle\phi_c(\cdot,\eta)|\phi_m\rangle\partial_t c(t,\eta)\rho_c(\eta)d\eta$$

and

$$i\langle\partial_t\psi|\phi_c(\cdot,\mu)\rangle = i\sum_n \dot{c}_n(t)\langle\phi_n|\phi_c(\cdot,\mu)\rangle + i\int_{\sigma_c} \langle\phi_c(\cdot,\eta)|\phi_c(\cdot,\mu)\rangle\partial_t c(t,\eta)\rho_c(\eta)d\eta$$

Similarly

$$\langle\big(H_0 + V(\mathbf{x},t)\big)\psi|\phi_m\rangle = \sum_n c_n(t)\langle\big(\varepsilon_n + V(\mathbf{x},t)\big)\phi_n|\phi_m\rangle + \int_{\sigma_c} c(t,\eta)\langle\big(\eta + V(\mathbf{x},t)\big)\phi_c(\cdot,\lambda)|\phi_m\rangle\rho_c(\lambda)d\lambda$$

and

$$\langle\big(H_0 + V(\mathbf{x},t)\big)\psi|\phi_c(\cdot,\mu)\rangle = \sum_n c_n(t)\langle\big(\varepsilon_n + V(\mathbf{x},t)\big)\phi_n|\phi_c(\cdot,\mu)\rangle + \int_{\sigma_c} \big(\eta + V(\mathbf{x},t)\big)c(t,\eta)\langle\phi_c(\cdot,\eta)|\phi_c(\cdot,\mu)\rangle\rho_c(\eta)d\eta$$

We set

$$\begin{cases} H_{nm} & := & \langle\big(\varepsilon_n + V(\mathbf{x},t)\big)\phi_n|\phi_m\rangle = \varepsilon_n\delta_{nm} + \langle V(\mathbf{x},t)\phi_n|\phi_m\rangle \\ H_n(\lambda) & := & \langle\big(\varepsilon_n + V(\mathbf{x},t)\big)\phi_n|\phi_c(\cdot,\lambda)\rangle = \langle V(\mathbf{x},t)\phi_n|\phi_c(\cdot,\lambda)\rangle \\ K_n(\lambda) & := & \langle\big(\lambda + V(\mathbf{x},t)\big)\phi_c(\cdot,\lambda)|\phi_n\rangle = \langle V(\mathbf{x},t)\phi_c(\cdot,\lambda)|\phi_n\rangle \\ H(\lambda,\mu) & := & \langle\big(\lambda + V(\mathbf{x},t)\big)\phi_c(\cdot,\lambda)|\phi_c(\cdot,\mu)\rangle = \langle V(\mathbf{x},t)\phi_c(\cdot,\lambda)|\phi_c(\cdot,\mu)\rangle \\ D(\lambda,\mu) & := & \langle\phi_c(\cdot,\lambda)|\phi_c(\cdot,\mu)\rangle \\ D_n(\mu) & := & \langle\phi_n|\phi_c(\cdot,\mu)\rangle \end{cases}$$

So that

$$\begin{cases} i\dot{c}_m(t) + i\int_{\sigma_c} D_m^*(\eta)\rho_c(\eta)\partial_t c(t,\eta)d\eta & = & \sum_n c_n(t)H_{nm} + \int_{\sigma_c} c(t,\eta)K_n(\eta)\rho_c(\eta)d\eta \\ i\sum_n \dot{c}_n(t)D_n(\mu) + i\int_{\sigma_c} D(\eta,\mu)\rho_c(\eta)\partial_t c(t,\eta)d\eta & = & \sum_n c_n(t)H_n(\mu) + \int_{\sigma_c} H(\eta,\mu)\rho_c(\eta)c(t,\eta)d\eta \end{cases}$$

As the operator $H_0 + V(\mathbf{x},t)$ is self-adjoint, we deduce that: $D(\lambda,\mu) = 0$ if $\lambda \neq \mu$ and $D_n(\lambda) = 0$ then

$$i\sum_n \dot{c}_n(t)D_n(\mu) + i\int_{\sigma_c} D(\eta,\mu)\rho_c(\eta)\partial_t c(t,\eta)d\eta = i\partial_t c(t,\mu)$$

This gives us a new set of equations:

$$\begin{cases} i\dot{c}_m(t) & = & \sum_n c_n(t)H_{nm} + \int_{\sigma_c} c(t,\eta)K_n(\eta)\rho_c(\eta)d\eta \\ i\partial_t c(t,\mu) & = & \sum_n c_n(t)H_n(\mu) + \int_{\sigma_c} H(\eta,\mu)\rho_c(\eta)c(t,\eta)d\eta \end{cases}$$

As $V(\mathbf{x},t)=\mathbf{x}\cdot\mathbf{E}(t)$

$$\begin{cases} H_{nm} &= \mathbf{E}(t)\cdot\langle\mathbf{x}\phi_n|\phi_m\rangle \\ H_n(\lambda) &= \mathbf{E}(t)\cdot\langle\mathbf{x}\phi_n|\phi_c(\cdot,\lambda)\rangle \\ K_n(\lambda) &= \langle\mathbf{E}(t)\cdot\langle\mathbf{x}\phi_c(\cdot,\lambda)|\phi_n\rangle \\ H(\lambda,\mu) &= \mathbf{E}(t)\cdot\langle\mathbf{x}\phi_c(\cdot,\lambda)|\phi_c(\cdot,\mu)\rangle \\ D(\lambda,\mu) &= \langle\phi_c(\cdot,\lambda)|\phi_c(\cdot,\mu)\rangle \end{cases}$$

We are interested in $\mathbf{d}(t)=\langle\mathbf{x}\rangle=\int|\psi|^2\mathbf{x}d^3x$. In this goal, we set

$\mathbf{d}_{mn}(t)=\langle\phi_m|\widehat{\mathbf{x}}|\phi_n\rangle$, $\rho_{mn}(t)=c_m^*(t)c_n(t)$, $\rho_{m\lambda}(t)=c_m^*(t)c(t,\lambda)$, $\rho_{\lambda m}(t)=c^*(t,\lambda)c_m, \rho_{\lambda\mu}(t)=c^*(t,\lambda)c(t,\mu)$

In order to compute $\mathbf{d}$ or equivalently $\mathrm{Tr}(\widehat{\rho}\widehat{\mathbf{x}})$, we derive a system of differential equiations. First, as

$$\dot\rho_{mn}(t)=\dot c_m^*(t)c_n(t)+c_m^*(t)\dot c_n(t)$$

then

$$c_n^*(t)\dot c_m(t)=-i\sum_\nu c_n^*(t)c_\nu(t)H_{\nu m}-i\int_{\sigma_c}c_n^*(t)c(t,\eta)K_m(\eta)\rho_c(\eta)d\eta$$

and

$$\dot c_n^*(t)c_m(t)=i\sum_\nu c_m(t)c_\nu^*(t)H_{\nu n}+i\int_{\sigma_c}c_m(t)c^*(t,\eta)K_n^*(\eta)\rho^*(\eta)d\eta$$

so that

$$\dot\rho_{mn}(t)=i\sum_\nu H_{m\nu}\rho_{\nu n}(t)-\rho_{m\nu}(t)H_{\nu n}+i\int_{\sigma_c}\Big(\rho_{n\eta n}(t)K_m^*(\eta)\rho_c^*(\eta)-\rho_{m\eta}(t)K_n(\eta)\rho_c(\eta)\Big)d\eta$$

Similarly, we have to evaluate $\partial_t\rho_{\lambda,\mu}(t)$. From

$$\dot\rho_{\lambda\mu}(t)=\partial_t c^*(t,\lambda)c(t,\mu)+c^*(t,\lambda)\partial_t c(t,\mu)$$

We then get

$$\begin{aligned}\dot\rho_{\lambda\mu}(t) &= i\textstyle\sum_n\Big[\rho_{n\mu}(t)H_n^*(\lambda)-\rho_{\lambda n}(t)H_n(\mu)\Big] \\ &\quad +i\textstyle\int_{\sigma_c}\Big[H^*(\eta,\lambda)\rho_c^*(\eta)\rho_{\eta\mu}(t)-H(\eta,\mu)\rho_c(\eta)\rho_{\lambda\eta}(t)\Big]d\eta\end{aligned}$$

Similarly

$$\dot\rho_{m\lambda}(t)=\dot c_m^*(t)c(t,\lambda)+c_m^*(t)\partial_t c(t,\lambda)$$

so that

$$\begin{aligned}\dot\rho_{m\lambda}(t) &= i\textstyle\sum_n\Big[\rho_{n\lambda}(t)H_{nm}^*-\rho_{mn}(t)H_n(\lambda)\Big] \\ &\quad +i\textstyle\int_{\sigma_c}\Big[K_n^*(\eta)\rho_c^*(\eta)\rho_{\eta\lambda}(t)-H(\eta,\lambda)\rho_c(\eta)\rho_{m\eta}(t)\Big]d\eta\end{aligned}$$

That is, we get the following system, for all m, n, λ, μ:

$$\begin{cases} \dot{\rho}_{mn}(t) & = & i\sum_{\nu} H_{m\nu}\rho_{\nu n}(t) - \rho_{m\nu}(t)H_{\nu n} + i\int_{\sigma_c}\Big(\rho_{\eta}(t)K_m^*(\eta)\rho_c^*(\eta) - \rho_{m\eta}(t)K_n(\eta)\rho(\eta)\Big)d\eta \\ \dot{\rho}_{m\lambda}(t) & = & i\sum_n\Big[\rho_{n\lambda}(t)H_{nm}^* - \rho_{mn}(t)H_n(\lambda)\Big] \\ & & +i\int_{\sigma_c}\Big[K_n^*(\eta)\rho_c^*(\eta)\rho_{\eta\lambda}(t) - H(\eta,\lambda)\rho_c(\eta)\rho_{m\eta}(t)\Big]d\eta \\ \dot{\rho}_{\lambda\mu}(t) & = & i\sum_n\Big[\rho_{n\mu}(t)H_n^*(\lambda) - \rho_{\lambda n}(t)H_n(\mu)\Big] \\ & & +i\int_{\sigma_c}\Big[H^*(\eta,\lambda)\rho_c^*(\eta)\rho_{\eta\mu}(t) - H(\eta,\mu)\rho_c(\eta)\rho_{\lambda\eta}(t)\Big]d\eta \end{cases}$$

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