Multi-level Maxwell-Bloch simulations.

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Abstract

We present the Maxwell-Bloch equations that are a model for the semi-classical description of laser-matter interactions. After having discussed both models and their numerical coupling in a general context regarding relaxation terms and the number of energy levels, we give examples of simulations that show the capacities of our approach.

1 Introduction

High intensity or short pulse lasers induce some strong non linear effects in matter where transient effects are important. Here a semi-classical model is used to describe laser-matter interaction. Such a model consists in a classical description for the electromagnetic wave (Maxwell equations) and a quantum description of matter leading to optical Bloch equations. The variables that describe matter are therefore the probability of electrons to be in certain quantum states and the coherence between these states.

The model we describe is not new in itself but we enable to choose more general coefficients in the equations. Besides the number of energy levels to describe matter may be any integer, but of course 2, 3 or 4 for practical applications.

Previous work on the subject include a 1D and 2D code by R. Ziolkowski et al. [7, 8, 9] that use a finite difference Yee scheme for Maxwell equations and two-level atoms. We may also cite a work by Martín et al. [3, 4] where the paraxial approximation is applied to Maxwell equations (and therefore the laser pulses may not be very short). Finally Nagra and York [5] couple Maxwell equations with rate equations involving up to four levels but not taking into account coherences and as we will see below very rapid transient phenomena are not modeled.

We design the numerical scheme in order to be able to model any number of levels and general relaxation terms. This induces extra problems in the time discretization of equations that are treated in [1] for Bloch equations with a forced electromagnetic field.

In section 2 we recall the Maxwell-Bloch model as well as the hypothesis which have to be imposed on the relaxation operator. In section 3 we describe the main features of the numerical model and finally section 4 is devoted to simulations that point out the advantages of being able to keep coherences and handling more than two level atoms.

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2 Maxwell-Bloch equations

The classical description for the propagation involve Maxwell equations:

$$\left\{ \begin{array}{ll} \partial_t \vec{B} = -\nabla \times \vec{E}, & \nabla \cdot \vec{B} = 0, \\ \partial_t \vec{D} = \nabla \times \vec{H}, & \nabla \cdot \vec{D} = 0. \end{array} \right. \tag{1}$$

Magnetic effects play no role in the applications we are interested in and we therefore take $\vec{B} = \mu_0 \vec{H}$. Our attention is focused on the other constitutive law: the displacement vector is defined by $\vec{D} = \varepsilon_0 \vec{E} + \vec{P}$ where $\vec{P}$ is the polarization due to the presence of a medium. In a classical description this polarization would be expressed in terms of $\vec{E}$, here we use a quantum mechanical model for the matter that is described by state vectors $|\psi\rangle$, the time evolution of which are given by the Schrödinger equation

$$i\hbar \partial_t |\psi\rangle = H|\psi\rangle.$$

The Hamiltonian H is the sum of the unperturbed Hamiltonian H_0 and the perturbation V induced by the electromagnetic field. The state vector $|\psi\rangle$ is decomposed on a basis of eigenstates $|j\rangle$ of the unperturbed Hamiltonian H_0: $|\psi\rangle = \sum a_j |j\rangle$. The quantum states $|j\rangle$ is associated to the eigenvalue $\mathcal{E}_j = \hbar\omega_j$. The observable variable we study is the density matrix which is defined by $\rho_{jk} = a_j a_k^*$ up to some statistical averaging. It is solution to the Bloch equation

$$i\hbar \partial_t \rho = [H, \rho], \tag{2}$$

where $[\cdot, \cdot]$ denotes the commutator of two operators. Keeping only N relevant levels, ρ is a $N \times N$ hermitian non negative matrix. Its diagonal elements ρ_{jj} represent the population of levels $|j\rangle$ and its off-diagonal elements ρ_{jk} the coherences between levels $|j\rangle$ and $|k\rangle$.

To express the perturbation V, we restrict ourselves to dipole moments because they induce the larger order in the perturbation series. Perturbation V reads $V = -e\vec{E} \cdot \vec{R}$ having taken into account the fact that $\vec{E}$ does not significantly vary over atomic distances. The dipole moment matrix is defined by its elements $\vec{p}_{jk} = \langle k | e\vec{R} | j \rangle$. With these notations equation (2) reads

$$\partial_t \rho_{jk} = -i\omega_{jk}\rho_{jk} - \frac{i}{\hbar}\vec{E} \cdot [\vec{p}, \rho]_{jk}, \tag{3}$$

where $\omega_{jk} = \omega_j - \omega_k$ is the frequency associated to the transition from level $|k\rangle$ to level $|j\rangle$.

To take into account the lack of information about the statistical averaging and of some phenomena that are not included in the present model as spontaneous emission of light, but also collisions, vibrations in crystal lattices or thermal perturbations in fluids, we add phenomenological relaxation terms to equation (3) that becomes

$$\partial_t \rho_{jk} = -i\omega_{jk}\rho_{jk} - \frac{i}{\hbar}[V, \rho]_{jk} + Q(\rho)_{jk}. \tag{4}$$

The off-diagonal relaxation term reads $Q(\rho)_{jk} = -\gamma_{jk}\rho_{jk}$ and diagonal terms follow "Pauli's master equation"

$$Q(\rho)_{jj} = \sum_{l \neq j} W_{jl}\rho_{ll} - \sum_{l \neq j} W_{lj}\rho_{jj} = \sum_{l \neq j} W_{jl}\rho_{ll} - \Gamma_j \rho_{jj}. \tag{5}$$

The relaxation to equilibrium states is obtained by imposing

$$W_{jl} = W_{lj} e^{\beta(\mathcal{E}_j - \mathcal{E}_l)} \tag{6}$$

(see e.g. Bloembergen [2]). Here $\beta = 1/\kappa T$, where κ is Boltzmann's constant and T is the temperature. Extra conditions have to be imposed on relaxation rates to conserve some of the main properties of the density matrix, mainly positiveness properties (see [1]). Thus we will suppose in the sequel that W has non negative coefficients and that

$$\gamma_{jk} = \frac{1}{2}(\Gamma_j + \Gamma_k) + \gamma_j^{coll} + \gamma_k^{coll} - A_j \cdot A_k. \tag{7}$$

where $\gamma_j^{coll} \in \mathbb{R}$, $A_j \in \mathbb{R}^N$ and $\gamma_j^{coll} \geq \frac{1}{2}\|A_j\|^2$ for all j. This last condition implies in particular that $\gamma_{jk} = \gamma_{kj}$.

The system is closed by computing the polarization as $\vec{P} = N_a \mathrm{Tr}(\vec{p}\rho)$ where N_a is the density of atoms.

3 Numerical issues

Our aim was not to find out a brand new method only designed for this model but to use well known methods for Maxwell equations and couple them with Bloch equations. Bloch equations only involve time derivatives and are therefore local in space. A discussion about their discretization may be found in [1] comparing the most commonly used Crank-Nicolson scheme and a method that conserve positiveness properties. Both a one-dimensional code and a two dimensional code have been written. The one dimensional code uses Yee's scheme to discretize Maxwell equations following [9]. To allow more complex geometries than in [7] (who uses a FDTD approach) our two-dimensional code is based on a finite volume approach using Delaunay-Voronoï meshes.

4 Simulations

In [9] we may find numerical simulation dealing with self-induced transparency where the number of transitions is monitored by the intensity of the incident field and ultra fast single-cycle pump pulses to invert populations. Here we choose some tests that take advantage of 3-level atoms and different relaxation times. We also show simulation that could not be obtained without the modeling of coherence as the transfer of coherence. Only 1D simulations are presented here.

4.1 Second harmonic generation

This test shows the advantage of taking into account more than two levels. We use here a three-level atom with equally spaced energy level and pump with the exact transition energy between two neighboring levels. Inversion of population may be observed as well as transient coherence evolution. Besides harmonics – and mainly the second one – of the incident frequency may be observed after a certain distance through matter. This is due to the fact that the wave interacts directly with the three possible transitions.

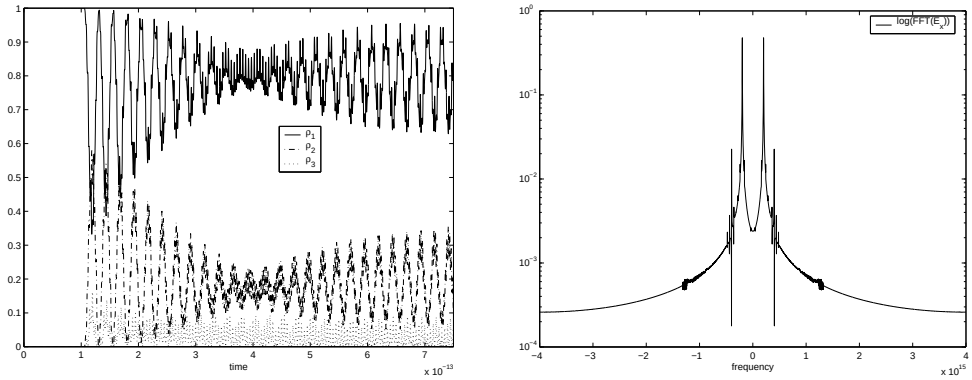

FIG. 1. *Second harmonic generation: Populations and Fourier Transform of E.*

4.2 Transfer of coherence

This test is inspired by an example by Suter [6]. At the initial time levels one and two are occupied and only coherence between level one and two is non zero. The dipole moment matrix is taken such that $\vec{p}_{12} = 0$ and $\vec{p}_{13} = 10\vec{p}_{23}$. This leads after a certain time to the transfer of coherence between level one and three. Such a test need of course a precise modeling of coherences and is impossible with a rate equations model.

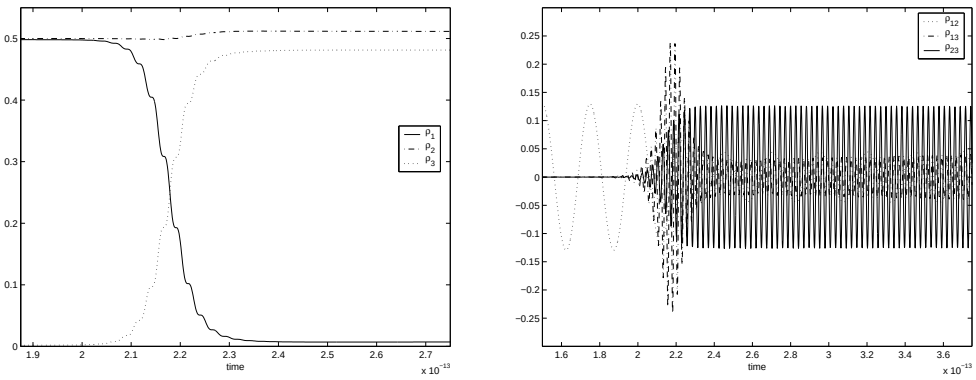

FIG. 2. *Transfer of coherence: Populations and Coherences.*

4.3 Different relaxation rates

The test we present here shows that we may take into account different relaxations rates. However we took here relatively close values and no collisions in order to see everything on the same figure. We may notice that relaxation induces some coherence between energy levels. Th shape of the envelope of coherences is directly connected to their relaxation rates.

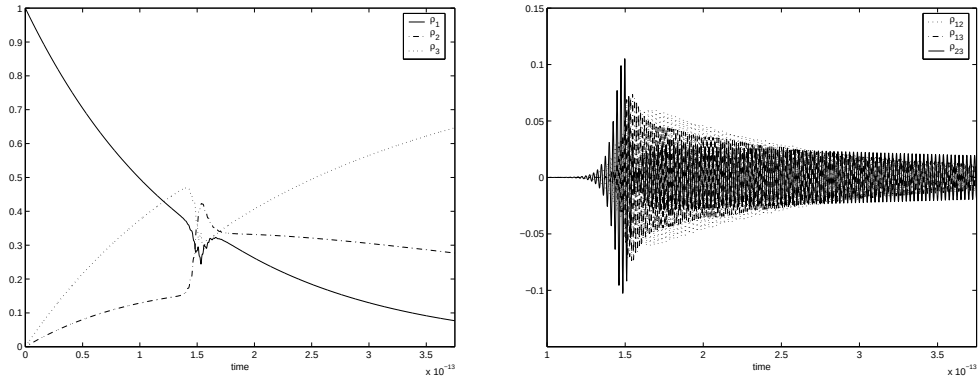

FIG. 3. *Different relaxation rates: Populations and Coherences.*

5 Conclusion

The above example show that we are able to model various physical phenomena. Other tests have been performed as 1D laser cavities and 2D results are in study too. The effort to model Bloch equations correctly may be used to couple with any existing Maxwell code in any dimension since the evolution of the density matrix is local in space.

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