Finite-Difference Time-Domain Formulation of Stochastic Noise in Macroscopic Atomic Systems

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Abstract—A numerical model based on the finite-difference time-domain method is developed to simulate fluctuations which accompany the dephasing of atomic polarization and the decay of excited state's population. This model is based on the Maxwell–Bloch equations with *c*-number stochastic noise terms. We successfully apply our method to a numerical simulation of the atomic superfluorescence process. This method opens the door to further studies of the effects of stochastic noise on light-matter interaction and transient processes in complex systems without prior knowledge of modes.

Index Terms—Finite-difference time-domain (FDTD) methods, Maxwell equations, noise, spontaneous emission, stochastic processes.

I. INTRODUCTION

T HE finite-difference time-domain (FDTD) method [1] has been extensively used in solving Maxwell's equations for dynamic electromagnetic (EM) fields. The incorporation of auxiliary differential equations, such as the rate equations for atomic populations [2], the Bloch equations for the density of states of atoms [3], and the multi-subband semiconductor Bloch equations [6], [7], has lead to comprehensive studies of light-matter interaction. Although the FDTD method has become a powerful tool in computational electrodynamics, it has been applied mostly to classical or semiclassical problems without noise.

Noise plays an important role in light-matter interaction. Marcuse solved the rate equations for light intensity and electron population including noise terms [6] to illustrate the effect of noise on lasing mode dynamics [7]. Gray and Roy extended the formulation by adding noise to the field equation in order to study the laser line shape [8]. Starting from a microscopic Hamiltonian, Kira *et al.* developed a semiconductor theory including spontaneous emission to describe semiconductor lasers [9]. While considerable progress has been made, these models

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remain in the modal picture. Knowledge of mode properties is required to characterize the noise, making it difficult to study complex systems in which the mode information is unknown a priori . Without invoking the modal picture, Hofmann and Hess obtained the quantum Maxwell-Bloch equations including spatiotemporal fluctuations [10]. Although it was useful to study spatial and temporal coherence in diode lasers, this formalism was based on the assumption that the temporal fluctuations of carrier density and photon density were statistically independent, which often breaks down above the lasing threshold. A FDTD simulation of microcavity lasers including quantum fluctuations was also done recently [11]. This simplified model added white Gaussian noise as a source to the electric field. The noise amplitude depended only on the excited state's lifetime. The dephasing process, which was much faster than the excited state's population decay, should have induced more noise but was neglected.

Our goal is to develop a FDTD-based numerical method to simulate fluctuations in macroscopic systems caused by interactions of atoms and photons with reservoirs (heatbaths). Such interactions induce temporal decay of photon number, atomic polarization, and excited state's population, which can be described phenomenologically by decay constants. The fluctuation-dissipation theorem demands temporal fluctuations or noise to accompany these decays. We intend to incorporate such noise in a way compatible with the FDTD method, that allows one to study the light-matter interaction in complex systems without prior knowledge of modes. In a previous work [12], we included noise caused by the interaction of the light field with an external reservoir in an open system. In this paper, we we develop a numerical model to simulate noise caused by the interaction of atoms with reservoirs such as lattice vibrations and atomic collisions. As an example, we apply the method to a numerical simulation of superfluorescence in a macroscopic system where the dominant noise is from the atoms rather than the light field.

We start with the Bloch equations for two-level atoms in one dimension where the direction of light propagation is along the x-axis

$$\begin{pmatrix} \dot{\rho}_1\\ \dot{\rho}_2\\ \dot{\rho}_3 \end{pmatrix} = \begin{pmatrix} 0 & \omega_0 & 0\\ -\omega_0 & 0 & 2\Omega_R\\ 0 & -2\Omega_R & 0 \end{pmatrix} \begin{pmatrix} \rho_1\\ \rho_2\\ \rho_3 \end{pmatrix} \\ - \begin{pmatrix} \frac{1}{T_2} & 0 & 0\\ 0 & \frac{1}{T_2} & 0\\ 0 & 0 & \frac{1}{T_1} + P_r \end{pmatrix} \begin{pmatrix} \rho_1\\ \rho_2\\ \rho_3 - \rho_3^{(s)} \end{pmatrix}$$
(1)

where $\Omega_R \equiv \gamma E_z / \hbar$ is the Rabi frequency, ω_0 the atomic transition frequency, E_z the electric field which is parallel to the *z*-axis, γ the dipole coupling term. Phenomenological decay times due to decoherence T_2 and the excited state's lifetime T_1 (which includes spontaneous emission and non-radiative recombination) are appended. In the absence of strong light confinement, which holds for macroscopic systems, T_1 and T_2 can be considered independent of the local density of states (LDOS). Hence, they do not have a dependence on spatial location nor frequency. We also include incoherent pumping of atoms from level 1 to level 2. The rate is proportional to the population in level 1, and can be written as $P_r \rho_{11}$. $\rho_3^{(s)}$ represents the steady-state value of ρ_3 when $E_z = 0$.

The relations between the Bloch vector and the density matrix are

$$\rho_1 = \rho_{12} + \rho_{21}$$

$$\rho_2 = i(\rho_{12} - \rho_{21})$$

$$\rho_3 = \rho_{22} - \rho_{11}.$$
(2)

The total polarization P_z of N atoms in a volume V is $P_z = -(N/V)|\gamma|\rho_1$ and inserted into the Maxwell's equations

$$\frac{dH_y}{dt} = -\frac{1}{\mu_0} \frac{dE_z}{dx}$$
$$\frac{dE_z}{dt} = \frac{1}{\epsilon} \frac{dH_y}{dx} - \frac{1}{\epsilon} \frac{dP_z}{dt}.$$
(3)

The atom-reservoir interactions not only cause decay of the Bloch vector, but also introduce noise according to the fluctuation-dissipation theorem. In Section II, we describe the model developed to include noise in the Maxwell–Bloch equations. The FDTD implementation of this model is presented in Section III. In Section IV, we simulate atomic superfluorescence and compare the results to previous experimental data and quantum-mechanical calculations.

II. NOISE MODEL

Starting from the quantum Langevin equation within the Markovian approximation, Drummond and Raymer derived a set of stochastic c-number differential equations describing light propagation and atom-light interaction in the many-atom limit [13]. The noise sources in these equations are from both the damping and the nonlinearity in the Hamiltonian. The latter represents the nonclassical component of noise, giving rise to nonclassical statistical behavior. Since our primary interests lie with classical behavior of macroscopic systems, such as superfluorescence and lasing, we neglect the nonclassical noise in this paper. The amplitude of classical noise accompanying the field decay is proportional to \sqrt{n} , where n is the thermal photon number. At room temperature the number of thermal photons at visible frequencies ($\hbar\omega \sim 1 \text{ eV}$) is on the order of 10^{-17} . This can be interpreted in a quantum mechanical picture as that most of the time there are no thermal photons at visible frequencies in the system. Thus, the noise related to field decay is neglected in this paper. At higher temperatures or longer wavelengths, this noise becomes significant and it can be incorporated into the FDTD algorithm following the approach we developed in our previous work [12].

The classical noise related to the pumping and decay of the atomic density matrix can be expressed as

$$\Gamma_{12} = (\xi_1 + i\xi_2)\sqrt{\gamma_p \rho_{22}}
\Gamma_{21} = (\xi_1 - i\xi_2)\sqrt{\gamma_p \rho_{22}}
\Gamma_{22} = \xi_3 \sqrt{\frac{\rho_{22}}{T_1} + P_r \rho_{11}}.$$
(4)

These noise terms are associated with ρ_{12} , ρ_{21} , and ρ_{22} , respectively. $\gamma_p = 1/T_2 - 1/2T_1$. The ξ_j terms are real, Gaussian, random variables with zero mean and the following correlation relation:

$$\langle \xi_j(t)\xi_k(t')\rangle = \delta_{jk}\delta(t-t') \tag{5}$$

where j, k = 1, 2, 3. The noise terms Γ_{12} and Γ_{21} represent fluctuations corresponding to decoherence by dephasing, while Γ_{22} is the fluctuation corresponding to relaxation of and pumping to the excited state's population. Only the linear term for pump noise is included here, a common first order approximation [14]. Furthermore, because we assume $T_2 \ll T_1$, pump fluctuations are neglected in Γ_{12} and Γ_{21} since they are orders of magnitude smaller than noise due to dephasing. According to (2), the noise terms for the Bloch vector are reduced to real variables as

$$\Gamma_{1} = 2\xi_{1}\sqrt{\gamma_{p}\rho_{22}}
\Gamma_{2} = -2\xi_{2}\sqrt{\gamma_{p}\rho_{22}}
\Gamma_{3} = 2\xi_{3}\sqrt{\frac{\rho_{22}}{T_{1}} + P_{r}\rho_{11}}.$$
(6)

They can be added directly to (1).

In a 1-D system, the total number of atoms N are split equally among M grid cells, giving the number of atoms per cell $N_s = N/M$. All quantities are defined at each individual grid cell, e.g., the term $\rho_3(x)$ is the *number* of inverted atoms in one cell at position x. The number of atoms in each cell is assumed to be constant assuring $\dot{\rho}_{11} + \dot{\rho}_{22} = 0$. We forcibly keep N_s constant via the relation $\rho_{11} = N_s - \rho_{22}$ and only calculate the excited state's population $\rho_{22}(t)$. The final stochastic equations to be solved are

$$\frac{d\rho_1(x,t)}{dt} = \omega_0 \rho_2(x,t) - \frac{1}{T_2} \rho_1(x,t) + \Gamma_1(x,t)
\frac{d\rho_2(x,t)}{dt} = -\omega_0 \rho_1(x,t) + \frac{2|\gamma|}{\hbar} E_z(x,t) \left(2\rho_{22}(x,t) - N_s\right)
- \frac{1}{T_2} \rho_2(x,t) + \Gamma_2(x,t)
\frac{d\rho_{22}(x,t)}{dt} = -\frac{|\gamma|}{\hbar} E_z(x,t) \rho_2(x,t) - \frac{1}{T_1} \rho_{22}(x,t)
+ P_r(N_s - \rho_{22}(x,t)) + \Gamma_{22}(x,t).$$
(7)

In the above equation, the steady-state value of ρ_3 in (1) is substituted by $\rho_3^{(s)} = N_s(T_1P_r - 1)/(T_1P_r + 1)$, an expression obtained by setting the time derivatives in (1) to zero. ρ_{11} in the expression of Γ_{22} in (4) can be replaced by $N_s - \rho_{22}$.

III. NUMERICAL IMPLEMENTATION

The most commonly used method of solving the Maxwell–Bloch equations is the "strongly coupled method."

With Δt being the time step, E, and ρ are both computed at $n\Delta t$, $(n + 1)\Delta t$, etc., while H is computed at $(n - 1/2)\Delta t$, $(n + 1/2)\Delta t$, etc. This produces equations with coupled terms such as $E^{n+1}\rho^{n+1}$ that must be solved by a predictor-corrector scheme (as used in [3]) or a fixed-point procedure, both of which are computationally inefficient. Therefore, we use a weakly coupled method that is easily implemented and efficient for 1-D systems.

The weakly coupled method was put forth by Bidégaray [15]. The electric field E_z is computed at times $n\Delta t$, $(n+1)\Delta t$, but ρ is calculated at $(n-1/2)\Delta t$, $(n+1/2)\Delta t$, thereby decoupling those discretized equations and creating a simple leap-frog type propagation system for 1-D. The noise terms in (7) are present throughout the entirety of the simulation and thus, should be incorporated efficiently. After discretization, the ξ_i terms are correlated according to $\langle \xi_j(x_u, t_m) \xi_k(x_v, t_n) \rangle = (1/\Delta t) \delta_{jk} \delta_{uv} \delta_{mn}$, and can be generated quickly with the Marsaglia and Bray modification of the Box–Müller Transformation [16]. Because the noise terms contain $\sqrt{\rho_{22}}$, as seen in (4) and (6), we are not able to use the weakly coupled scheme to solve for ρ_1, ρ_2 and ρ_{22} as precisely as possible. Instead, the approximation of using the previous time step value $\sqrt{\rho_{22}^{n-1/2}}$ is employed. It is valid as long as the atomic population is varying slowly. For the simulation of superfluorescence in Section IV, the maximum change of ρ_{22} over one time step Δt is only 0.0007%.

The discretized equations with noise are

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$$E_z^{n+1} = E_z^n + \frac{\Delta t}{\epsilon} \frac{dH_y}{dx} - \Delta t A \rho_1^{n+1/2}$$

$$+ \Delta t B \rho_2^{n+1/2}$$
(8a)

$$H_y^{n+1/2} = H_y^{n-1/2} - \frac{\Delta t}{\mu_0} \frac{dE_z}{dx}$$
(8b)

$$\rho_1^{n+1/2} = \rho_1^{n-1/2} + \frac{1}{2} \Delta t \omega_0 \left(\rho_2^{n+1/2} + \rho_2^{n-1/2} \right) - \frac{1}{2} \frac{\Delta t}{T_2} \left(\rho_1^{n+1/2} + \rho_1^{n-1/2} \right) + \Delta t \Gamma_1 \qquad (8c)$$

$$p_{2}^{n+1/2} = \rho_{2}^{n-1/2} - \frac{1}{2} \Delta t \omega_{0} \left(\rho_{1}^{n+1/2} + \rho_{1}^{n-1/2} \right) \\ + \frac{2\Delta t |\gamma|}{\hbar} E_{z}^{n} \left(\rho_{22}^{n+1/2} + \rho_{22}^{n-1/2} - N_{s} \right) \\ - \frac{1}{2} \frac{\Delta t}{T_{2}} \left(\rho_{2}^{n+1/2} + \rho_{2}^{n-1/2} \right) + \Delta t \Gamma_{2}$$
(8d)

$$\rho_{22}^{n+1/2} = \rho_{22}^{n-1/2} - \frac{1}{2} \frac{\Delta t |\gamma|}{\hbar} E_z^n \left(\rho_2^{n+1/2} + \rho_2^{n-1/2} \right) - \frac{1}{2} \Delta t \left(\frac{1}{T_1} + P_r \right) \left(\rho_{22}^{n+1/2} + \rho_{22}^{n-1/2} \right) + \Delta t P_r N_s + \Delta t \Gamma_{22}$$
(8e)

where we have defined $A \equiv |\gamma|/V_s \epsilon T_2$ and $B \equiv |\gamma|\omega_0/V_s \epsilon$. These equations are solved to obtain the final FDTD equations for E_z , H_y , ρ_1 , ρ_2 , and ρ_{22} .

IV. RESULTS AND DISCUSSION

We apply the Maxwell–Bloch equations with noise to a FDTD simulation of superfluorescence (SF) and compare the results to previous data obtained experimentally [17] and theoretically [18]. SF is the cooperative radiation of an initially

inverted but incoherent two-level medium resulting from spontaneous buildup of a macroscopic coherent dipole. This is an interesting and suitable case to study with our method because both spatial propagation of light and noise are important. Noise caused by collisional dephasing can seriously disturb SF and change the emission character to amplified spontaneous emission (ASE). We simulate the transition from SF to ASE with increasing dephasing rate, corresponding to the experiment by Malcuit *et al.* on super-oxide ions in potassium chloride (KCl:O₂) [17].

Experimentally, the ions inside a cylinder of diameter $d = 80 \ \mu m$ and length $L = 7 \ mm$ were excited by a short pulse. The total number of excited ions is $N = 3 \times 10^9$. The emission wavelength is $\lambda = 629 \ mm$. The Fresnel number for the excitation cylinder is $F = A/\lambda L \sim 1$, where A is the area of the cylinder cross-section. $T_1 = 76 \ mm$, and T_2 was varied via temperature change. The "cooperative lifetime" or the duration of SF pulse $\tau_r = 8\pi A T_1/3\lambda^2 N$ is 2.7 ps. The estimated delay time for the SF peak after the excitation pulse

$$\tau_d = \tau_r \left[\frac{1}{4} \ln(2\pi N) \right]^2 \tag{9}$$

is 94 ps.

Since $F \sim 1$, the EM modes propagating nonparallel to the cylinder axis are not supported [19]. Those modes propagating along the cylinder axis do not have a strong radial dependence, nor are there significant diffraction losses. Thus, the system can be considered as 1-D in our FDTD simulation. The grid resolution is $\Delta x = 70$ nm and the total running time is $\tau_{\rm sim} = 3$ ns. The Courant number S is set to 0.999999. The magic time step, S = 1, was seen to cause an instability in some cases. The value S = 1/2, however, does not propagate the large sudden impulses of the noise accurately. Setting $S = 1 - 10^{-6}$ preserves the accuracy to an acceptable degree while eliminating the instability at S = 1. There is some numerical dispersion and reflection from the absorbing boundary layer, but the error is of the order 10^{-6} . Ignoring non-radiative recombination, the atomic dipole coupling term $|\gamma| = \sqrt{3\lambda^3 \hbar \epsilon_0 / 8\pi^2 T_1} = 1.1 \times 10^{-29} \,\mathrm{C} \cdot \mathrm{m}.$

The simulation is started with the initial condition of all the atoms being excited ($\rho_{22} = N_s$). However, because the atomic population and polarization operators do not commute, the uncertainty principle demands a nonvanishing variance in the initial values of the Bloch vector [19]. This results in a tipping angle θ of the initial Bloch vector away from the top of Bloch sphere ($\rho_1 = 0, \rho_2 = 0, \rho_3 = N_s$). The value of θ is given by a Gaussian random variable centered at zero with a standard deviation $\theta_T = 2/\sqrt{N_s}$. Since there is no incoherent pumping at $t > 0, P_r$ is set to 0.

Fig. 1 shows the output EM energy at a spatial grid point outside the system for four different values of the dephasing time T_2 . When $T_2 = 100 \text{ ps} > \tau_d$, the cooperative emission characteristic of SF is clearly seen in Fig. 1(a). The number of atoms that emit cooperatively is estimated to be $N_c = 8\pi c T_1 A/3\lambda^2 L = 3.5 \times 10^8$ and is known as the Arecchi–Courtens cooperation number. Since $N_c < N = 3 \times 10^9$, the SF oscillates in time, with the maximal



Fig. 1. Numerical results of the output EM energy from initially-inverted two-level atoms, obtained by FDTD solution of the Maxwell–Bloch equations with noise. The left three columns show the output energy for three random realizations. The last column on the right shows the output energy averaged over 30 random realizations. All insets in the last column magnify the temporal range 0 < t < 1 ns. Dephasing time $T_2 = 100$ ps (first row), 33.3 ps (second row), 25.0 ps (third row), and 14.3 ps (fourth row).

emission intensity at $t \sim 170$ ps. This behavior agrees well with the previous result in [18]. For $T_2 = 33.3$ ps $< \tau_d$, there is enough dephasing to disturb the cooperative emission. The emitted pulse broadens and the time delay increases, as shown in Fig. 1(b). For $T_2 = 25$ ps, a further damping of superfluorescence is seen in Fig. 1(c). As T_2 decreases more, the pulse continues to broaden but the time delay begins to decrease. When T_2 reaches the critical value $\sqrt{\tau_r \tau_d} = 15.9$ ps, the amount of dephasing is sufficient to prevent the occurrence of cooperative emission. No macroscopic dipole moment can build up and the atoms simply respond to the instantaneous value of the radiation field. Hence, SF is replaced by ASE. Fig. 1(d) plots the ASE pulse for $T_2 = 14.3$ ps. The time delay is almost immeasurably small and the emission intensity is very noisy. Fig. 2 compares the delay times taken from our FDTD simulations to previous results obtained experimentally [17] and by full quantum-mechanical theory of SF [18]. The excellent agreement validates our FDTD-based numerical method. We emphasize that inclusion of the noise terms in (7)is essential to obtain the correct variation of τ_d with T_2 . As found in [17], the previous approach of modeling the initial fluctuations as random tipping angles of the Bloch vector and ignoring the noise at later times brings about good agreement with experiment only when T_2 is large making the amplitude of the noise terms in (7) small. As the dephasing rate increases, fluctuations can no longer be modeled simply as an initial noise.

We have also studied the decoherence process. The amplitude of the Bloch vector $\rho_B \equiv \sqrt{\rho_1^2 + \rho_2^2 + \rho_3^2} = \sqrt{N_s^2 + 4\rho_{12}\rho_{21} - 4\rho_{22}\rho_{11}}$. In the absence of decoherence, $\rho_{12}\rho_{21} = \rho_{22}\rho_{11}$, and $\rho_B = N_s$. The presence of decoherence



Fig. 2. Comparison of delay times of emission pulse obtained by our numerical simulation (black solid circles) with previous experimental data (red crosses) and quantum-mechanical calculation results (blue open diamonds) taken from [18]. Numerical delay times are obtained from the emission pulses averaged over 30 realizations.

decreases the off-diagonal terms of the density matrix, thus $\rho_{12}\rho_{21} < \rho_{22}\rho_{11}$ and $\rho_B < N_s$ [20]. We estimate the degree of decoherence through the ratio ρ_3/ρ_B , which is plotted in Fig. 3 for four different values of T_2 . Each curve is obtained by spatial average of ρ_3 and ρ_B over the entire excitation region and then ensemble-average over 30 realizations.

When the dephasing time is large $(T_2 > \tau_d)$, a macroscopic dipole moment is spontaneously formed. The enhanced radiative decay rate results in quick depletion of the population inversion ρ_3 . Despite $T_2 \ll T_1$, the decay of ρ_1 and ρ_2 by dephasing is overshadowed by the decay of ρ_3 by SF, leading to a rapid drop of ρ_3/ρ_B in time. This behavior is shown by the red dotted



Fig. 3. Ratio ρ_3/ρ_B as a function of time for $T_2 = 100$ ps (red dotted line), 33.3 ps (blue dashed line), 25.0 ps (green solid line), and 14.3 ps (black dash-dotted line).

line in Fig. 3. The non-monotonic decay is caused by SF oscillations as can be seen in Fig. 1(a). The oscillatory SF is a result of the number of atoms being greater than the Arecchi–Courtens cooperation number $(N > N_c)$. The intensity oscillation leads to an oscillation of population inversion which is 90° out of phase. The local maximum of ρ_3 at t = 320 ps (red dotted curve in Fig. 3) occurs just before the second peak of intensity at t = 370 ps [Fig. 1(a)]. As T_2 is reduced, the increased amount of decoherence frustrates the buildup of a macroscopic dipole moment and reduces the radiative decay rate. Consequently, the depletion of population inversion is slowed down. It leads to a slower decay of ρ_3/ρ_B and the disappearance of damped oscillations. Finally when the dephasing time is small enough $(T_2 < \sqrt{\tau_r \tau_d})$, the system stays in a decoherent state, and ρ_3/ρ_B remains close to one for a very long time.

V. CONCLUSION

We have developed a FDTD algorithm to incorporate stochastic noise in macroscopic systems into the Maxwell-Bloch equations. Such noise, resulting from atom-reservoir interactions, accompanies the dephasing of atomic polarization and decay of and pumping to the excited state population. We applied our algorithm to a numerical simulation of superfluorescence in a 1-D system. The results are in good agreement with previous experimental and theoretical studies. Although our simulations only include classical noise, nonclassical noise may be incorporated as well. Since they consist of nonlinear terms [13], the incorporation of nonclassical fluctuations to the FDTD algorithm may be numerically challenging. Given the rapid progress in development of various numerical methods of including nonlinearity in the Maxwell-Bloch equations [21], [22], we are optimistic that the quantum noise terms may be successfully integrated into our method. Therefore, our FDTD-based model can be used for numerical studies of light-matter interaction and transient processes in complex systems without prior knowledge of modes.

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