Nonlinearity in the medium for electromagnetic wave propagation induced by quantum mechanical interaction of the field with finite-state atoms

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1 Abstract

We consider the interaction between a classical time-varying electric field and the atomic electrical dipole moment operator of a finite state atom and calculate approximately the mixed state evolution of the atom with such an interaction Hamiltonian up to quadratic orders in the external classical electric field using Schrodinger's equation for mixed state evolution. From this approximate expression for the state evolution, we evaluate the average atomic electric dipole moment as a linear-quadratic function of the electric field. Later on, we also take into account Brownian motion bath noise that couples to the atomic dynamics, so that the Schrodinger equation dynamics gets modified to the Lindblad dynamics of an open quantum system, i.e., the master equation. From the average atomic dipole moment, we obtain an expression for the average polarization field of atoms constituting the medium through which the classical electromagnetic wave propagates. This average polarization is a linear-quadratic causal functional of the electric field. The resulting Maxwell equations for wave propagation, taking into account this polarization current density, thus acquire a quadratic nonlinearity which can be used to explain higher harmonic frequencies at the output of an optical fiber when the input is excited by the electric field coming from a monochromatic laser. We are then also able to calculate the mean square fluctuations in the polarization field using the evolving atomic state, and hence the electric field propagating through the medium of atoms acquires quantum fluctuations which can be used to explain time-varying random shifts in the spectral lines. As mentioned above, we include in this article an extension of this problem to the case when there is white Gaussian noise corrupting the state evolution dynamics of the atom and also the case when there is a pairwise interaction between the spin dipoles of atoms at two different

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locations. In the former case, we derive an approximate formula for the polarization field as well as its temporal statistical correlations, and we show how this computation can be extended to the latter case too. When there is a pairwise interaction between the atomic spins, we use a version of the approximate quantum Boltzmann equation to calculate the first and second order marginals of the atomic states, which is then used to compute the average polarization as well as its spatio-temporal correlations. Temporal correlations using second marginals of the atomic states cannot be computed using classical probabilistic methods owing to Heisenberg uncertainty, so we propose a sequential measurement-based strategy involving computation of the joint probabilities of the atomic spin components at two different times by first measuring one component of the spin, allowing for state collapse, and then following it up with nonlinear time evolution from the collapsed two-particle state using the quantum Boltzmann equation, followed further by measurement of another component of the spin at the second time.

Keywords: spin 1/2 atoms, spin dipole interaction, classical and quantum fluctuations in polarization, quantum Boltzmann equation, polarization current in Maxwell equations, nonlinear wave propagation for the electric field, spin j particles interacting with electric field, Brownian motion, stochastic differential equation, Ito correction term, unitary evolution.

2 Problem formulation and approximate solution

2.1 The case of spin 1/2 atoms

At each point $r \in \mathbb{R}^3$, there is a two-state atom with upspin state $|+>$ and downspin state $|->$. These are respectively eigenvectors of σ_z with eigenvalue ± 1. The electric dipole moment vector operator of the atom is given by $\mu.\sigma = \mu(\sigma_x, \sigma_y, \sigma_z)$. Let us fix a point r and let at time t, $\rho_A(t) = \rho_A(t|r)$ denote the mixed state of the atom at time t. Then, its average electric dipole moment at time t is given by

$$p(t) = p(t,r) = \mu.Tr(\sigma.\rho_A(t))] \in \mathbb{R}^3$$

The atom state evolves according to its interaction Hamiltonian with the electric field given by

$$H(t) = -\mu(\sigma, E(t,r))$$

Thus $\rho_A(t)$ satisfies

$$\rho_A'(t) = -i[H(t), \rho_A(t)] = i\mu[(\sigma, E(t,r)), \rho_A(t)]$$

Upto quadratic orders in the electric field, the solution is

$$\rho_A(t) = \rho_A(0) - i\int_0^t [H(s), \rho_A(0)]ds - \int_{0<s_2<s_1<t} [H(s_1), [H(s_2), \rho_A(0)]]ds_1 ds_2$$

We also that upto quadratic orders in the electric field,

$$\rho'_A(t) = -i[H(t), \rho_A(0)] - \int_0^t [H(t), [H(s), \rho_A(0)]]ds$$

$$= i\mu[(\sigma, E(t,r)), \rho_A(0)] - \mu^2 \int_0^t [(\sigma, E(t,r)), [(\sigma, E(s,r)), \rho_A(0)]]ds$$

and hence upto quadratic orders in the electric field, the average rate of increase of the dipole moment of the atom is given by

$$\partial_t p(t,r) = \mu.Tr(\sigma.\rho'_A(t)) =$$

$$(Tr(\sigma_x \rho'_A(t)), Tr(\sigma_y \rho'_A(t)), Tr(\sigma_z \rho'_A(t)))$$

If $N(r)$ denotes the number density of atoms in space, then the polarization field is given by

$$P(t,r) = N(r)p(t,r)$$

and its rate of change, i.e., the polarization current density, is given by

$$\partial_t P(t,r) = N(r).\partial_t p(t,r)$$

To evaluate this, we require

$$Tr(\sigma.[(\sigma, E(t,r)), \rho_A(0)]), Tr(\sigma.[(\sigma, E(t,r)), [(\sigma, E(s,r)), \rho_A(0)]])$$

or equivalently, in terms of components,

$$Tr(\sigma_k.[(\sigma, E(t,r)), \rho_A(0)]), Tr(\sigma_k.[(\sigma, E(t,r)), [(\sigma, E(s,r)), \rho_A(0)]])$$

$k = 1, 2, 3$, where

$$\sigma_1 = \sigma_x, \sigma_2 = \sigma_y, \sigma_3 = \sigma_z$$

Now,

$$Tr(\sigma_k[(\sigma, E(t,r)), \rho_A(0)]) =$$

$$\sum_{m=1}^{3} E_m(t,r) Tr(\rho_A(0)[\sigma_k, \sigma_m])$$

$$= 2i. \sum_{m,s=1}^{3} \epsilon(kms) E_m(t,r) Tr(\rho_A(0)\sigma_s)$$

where

$$\epsilon(123) = \epsilon(231) = \epsilon(312) = 1,$$

$$\epsilon(213) = \epsilon(132) = \epsilon(321) = -1$$

and $\epsilon(kms) = 0$ if any two of the three indices k, m, s are equal

Remark: Note the following identities for the Pauli spin matrices:

$$\sigma_k \sigma_m = i. \sum_s \epsilon(kms)\sigma_s + \delta(k,m) I_2$$

and hence,

$$[\sigma_k,\sigma_m]=2i.\sum_s \epsilon(kms)\sigma_s$$

with summation over the repeated index $s=1,2,3$ being implied. Thus,

$$[[\sigma_k,\sigma_m],\sigma_j]=-4\epsilon(kmr)\epsilon(rjs)\sigma_s$$

Further, note also

$$\sigma_k\sigma_m\sigma_s=(i\epsilon(kmr)\sigma_r+\delta(k,m)I)\sigma_s$$

$$=i\epsilon(kmr)(i\epsilon(rsj)\sigma_j+\delta(r,s)I)+\delta(k,m)\sigma_s$$

$$=-\epsilon(kmr)\epsilon(rsj)+\delta(km)\sigma_s+i\epsilon(kmr)\delta(rs).I$$

We now turn to evaluate the quadratic term:

$$Tr(\sigma_k.[(\sigma,E(t,r)),[(\sigma,E(s,r)),\rho_A(0)]])$$

$$=\sum_{j,m=1}^{3}[E_m(t,r)E_j(s,r)Tr(\sigma_k(\sigma_m\sigma_j\rho_A(0)+\rho_A(0)\sigma_j\sigma_m-\sigma_m\rho_A(0)\sigma_j$$

$$-\sigma_j\rho_A(0)\sigma_m))]$$

$$=\sum_{j,m}E_m(t,r)E_j(s,r)Tr[[\sigma_k,\sigma_m],\sigma_j]\rho_A(0)]$$

$$=-4\sum_{jmrs}E_m(t,r)E_j(s,r)\epsilon(kmr)\epsilon(rjs)Tr(\rho_A(0)\sigma_s)$$

where we have used

$$[\sigma_k,\sigma_m]=2i\epsilon(kms)\sigma_s$$

with summation over the repeated index s being implied.

In short, we see that the polarization current density up to quadratic orders in the electric field is given by a formula of the form

$$J_{Pa}(t,r)=\partial_t P_a(t,r)=N(r)\sum_m \chi(1km)E_m(t,r)+\sum\chi(2kmj)E_m(t,r)\int_0^t E_j(\tau,r)d\tau$$

where

$$\chi(1km)=-2\mu^2.\sum_s \epsilon(kmr).Tr(\rho_A(0)\sigma_s)$$

$$\chi(2kmj)=4\mu^3\sum_{rs}\epsilon(mjr)\epsilon(rjs)Tr(\rho_A(0)\sigma_s)$$

More generally, we can expand $\rho'_A(t)$ as an infinite series:

$$\rho'_A(t)=-iad(H(t))(\rho_A(0))+\sum_{n\geq 1}(-i)^{n+1}\int_{0<t_n<...<t_1<t}ad(H(t)).ad(H(t_1)...ad(H(t_n))(\rho_A(0))dt_1...dt_n$$

$$= i\mu.ad(\sigma, E(t,r))(\rho_A(0))$$

$$+\sum_{n\geq 1}(i\mu)^{n+1}\int_{0<t_n<...<t_1<t} ad(((\sigma, E(t,r)))ad((\sigma, E(t_1,r)))...ad((\sigma, E(t_n,r)))(\rho_A(0))dt_1...dt_n$$

$$= i\mu.E_{m_0}(t,r)ad(\sigma_{m_0})(\rho_A(0))$$

$$+\sum_{n\geq 1}(i\mu)^{n+1}ad(\sigma_{m_0}).ad(\sigma_{m_1})...ad(\sigma_{m_n})(\rho_A(0))\int_{0<t_n<...<t_1<t} E_{m_0}(t,r)E_{m_1}(t_1,r)...E_{m_n}(t_n,r)dt_1...dt_n$$

summation over the repeated indices $m_0, m_1, ..., m_n$ being implied. Note that all these indices range over $1, 2, 3$.

2.2 spin j atoms interacting with the electric field taking bath noise into account

More generally, we can regard each atom as being described by a 3-vector valued spin-j operator, i.e., the atomic observables are $J = (J_x, J_y, J_z)$ where J_x, J_y, J_z are three $2j + 1 \times 2j + 1$ matrices satisfying the standard angular momentum commutation relations

$$[J_x, J_y] = iJ_z, [J_y, J_z] = iJ_x, [J_z, J_x] = iJ_y$$

The atomic dipole moment is then given by $\mu.J$ where μ is a constant, and if $E(t,r)$ is the electric field at position r at time t, where r is the location of the atom, then the interaction Hamiltonian between the electric dipole and this electric field is given by

$$H_0(t) = -\mu(E, J) = -\mu(J_xE_x + J_yE_y + J_zE_z)$$

The state of the atom at time t is then $\rho_A(t) = \rho_A(t|r)$ and this satisfies the Schrödinger-Von-Neumann-Liouville equation

$$d\rho_A(t) = -i[H_0(t), \rho_A(t)]dt$$

This is the mixed state version of the pure state Schrödinger equation

$$d|\psi(t)>= -iH_0(t)|\psi(t)> dt$$

However, now we also assume the presence of noise in the dynamics described by a white Gaussian noise potential of the form

$$V(t) = V_0(t)dB(t)/dt$$

where $B(.)$ is standard Brownian motion, i.e., a zero-mean Gaussian process with continuous sample paths and independent increments having variance

$$\mathbb{E}(B(t) - B(s))^2 = t - s, t \geq s$$

In order to guarantee unitary evolution of the atomic wave function, we must introduce an Itö correction term into the Schrödinger dynamics and describe it by an Itö stochastic differential equation:

$$d|\psi(t)>= (-(iH_0(t)+V_0(t)^2/2)dt - iV_0(t)dB(t))|\psi(t)>$$

Remark: It is a trivial exercise in the application of Itö's formula in the form $(dB(t))^2 = dt$ that

$$d<\psi(t)|\psi(t)>=<d\psi(t)|\psi(t)>+<\psi(t)|d\psi(t)>+<d\psi(t)|d\psi(t)>=0$$

guarantees unitary evolution. For mixed states, the corresponding evolution is now described by

$$d\rho_A(t) = [-i[H_0(t),\rho_A(t)] - P(t)\rho_A(t) - \rho_A(t)P(t) + V_0(t)\rho_A(t)V_0(t)]dt$$

$$-i(V_0(t)\rho_A(t) - \rho_A(t)V_0(t))dB(t)$$

where

$$P(t) = V_0(t)^2/2$$

The aim is to solve this stochastic differential equation approximately up to, say, quadratic orders in the electric field and quadratic orders in the Brownian noise process, and hence calculate its expected value and use this expected value to calculate the expected value of the atomic dipole moment of the atom, and hence the polarization field, and hence the polarization current density, which will be used in the Maxwell equations to obtain no.

Using the standard notation of Lie algebra theory,

$$L_XY = XY, R_XY = YX, ad(X)(Y) = [X,Y] = XY - YX = (L_X - R_X)(Y)$$

for two operators X, Y in a vector space, we can express the above SDE as

$$d\rho_A(t) = \theta_0(t)(\rho_A(t))dt + \theta_1(t)(\rho_A(t))dB(t)$$

where

$$\theta_0(t) = -i.ad(H_0(t)) - L_{P(t)} - R_{P(t)} + L_{V_0(t)}R_{V_0(t)}$$

and

$$\theta_1(t) = -i.ad(V_0(t))$$

We could solve this equation up to quadratic orders in the electric field and quadratic orders in $V_0(t)^2$, i.e., in the noise coefficients. In other words, we are assuming that the electric field and noise variance terms are of the same orders of smallness. This is because linear terms in the noise are proportional to $dB(t)/dt$ which has infinite variance, or equivalently, $dB(t) = O(\sqrt{dt})$ which is much larger than the corresponding$O(dt)$ term coming from the electric field. This gives us the approximate equation

$$\rho_A(t) = \rho_A(0) + \int_0^t \theta_0(s)(\rho_A(0))ds + \int_0^t \theta_1(s)(\rho_A(0))dB(s)$$

$$+\int_{0<s_2<s_1<t}\theta_0(s_1)(\theta_0(s_2)(\rho_A(0)))ds_1ds_2$$

$$+\int_{0<s_2<s_1<t}\theta_1(s_1)(\theta_1(s_2)(\rho_A(0)))dB(s_1)dB(s_2)$$

$$+\int_{0<s_2<s_1<t}\theta_0(s_1)(\theta_1(s_2))ds_1dB(s_2)$$

$$+\int_{0,s_2<s_1<t}\theta_1(s_1)\theta_0(s_2))dB(s_1)ds_2$$

The classical expected value of the atomic state $\rho_A(t)$ up to quadratic orders in the electric field and quadratic orders in the noise variance is then

$$\mathbb{E}(\rho_A(t)) = \rho_A(0) + \int_0^t \theta_0(s)(\rho_A(0))ds$$

$$+\int_{0<s_2<s_1<t}\theta_0(s_1)(\theta_0(s_2)(\rho_A(0)))ds_1ds_2$$

While the classical autocovariance of $\rho_A(t)$ is given up to quadratic terms, the noise variance is given by

$$C_A(t_1,t_2) = \mathbb{E}(\rho_A(t_1)\otimes\rho_A(t_2)) - \mathbb{E}(\rho_A(t_1))\otimes\mathbb{E}(\rho_A(t_2))$$

$$= \int_0^{min(t_1,t_2)} \theta_1(s)(\rho_A(0))\otimes\theta_1(s)(\rho_A(0))ds$$

The average dipole moment of the atom is then given by

$$< p(t) >= \mu\mathbb{E}[Tr(\rho_A(t)J)] = \mu.Tr(\mathbb{E}(\rho_A(t))J) = \mu.Tr(M_A(t)J)$$

where

$$M_A(t) = \mathbb{E}(\rho_A(t))$$

and the autocovariance function of the dipole moment is given by

$$C_p(t_1,t_2) =< p(t_1)\otimes p(t_2) > - < p(t_1) > \otimes < p(t_2) >$$

$$= \mathbb{E}(Tr(\rho_A(t_1)\mu J).Tr(\rho_A(t_2)\mu J))$$

$$-\mathbb{E}(Tr(\rho_A(t_1)\mu J).\mathbb{E}Tr(\rho_A(t_2)\mu J)$$

$$= \mu^2[\mathbb{E}(Tr((\rho_A(t_1)\otimes\rho_A(t_2))(J\otimes J))] - \mu^2 Tr((\mathbb{E}(\rho_A(t_1))\otimes\mathbb{E}(\rho_A(t_2))(J\otimes J))$$

$$= \mu^2 Tr(C_A(t_1,t_2)(J\otimes J))$$

Note that $\rho_A(t), M_A(t), C_A(t_1,t_2), < p(t) >, C_p(t_1,t_2)$ are all functions of r, the spatial location of the atom, because the electric field with which the atom interacts is a function of the spatial location, i.e., ideally we should write $\rho_A(t,r)$ for $\rho_A(t)$ etc. These formulae could readily be generalized to calculate the classical statistical correlations between the atomic state at two different space-time points and the statistical correlations between the atomic dipole moments at two different space-time points.

When we multiply the quantum-averaged dipole moment $\mu.Tr(\rho_A(t)J)$ by the number density $N(r)$ of atoms in space, we obtain the random polarization field $P(t,r)$ and, using the above formulas, we can calculate the mean and covariance of this polarization field at a given spatial location at any two times t_1, t_2 as

$$\mathbb{E}P(t,r) = N(r)\mu.Tr(M_A(t)J)) = N(r) < p(t) >,$$

$$Cov(P(t_1,r),P(t_2,r)) = \mathbb{E}(P(t_1,r)\otimes P(t_2,r)) - \mathbb{E}(P(t_1,r))\otimes\mathbb{E}(P(t_2,r))$$

$$= \mu^2 N(r)^2.Tr(C_A(t_1,t_2)(J\otimes J)) = N(r)^2 C_p(t_1,t_2)$$

We also require to calculate the covariance between the polarization field at two different points in space and, more generally, at two different space-time points, i.e., we require

$$C_P(t_1,r_1,t_2,r_2) = Cov(P(t_1,r_1),P(t_2,r_2)) =$$

$$\mathbb{E}(P(t_1,r_1)\otimes P(t_2,r_2)) - \mathbb{E}P(t_1,r_1)\otimes\mathbb{E}P(t_2,r_2)$$

For this, we assume that the number density of atoms $N(r)$ is a random variable with mean $N_0(r)$ and covariance $C_N(r_1,r_2)$. We also assume the incident electric field $E(t,r)$ to have a mean value of $E_0(t,r)$ and a space-time covariance of $C_E(t_1,r_1,t_2,r_2)$. Then, we can use this data to evaluate C_P. In this expression,

$$\mathbb{E}(P(t_1,r_1)\otimes P(t_2,r_2)) = N(r_1)N(r_2)\mathbb{E}(Tr(\rho_A(t_1,r_1)J).\otimes(Tr(\rho_A(t_2,r_2)J))$$

$$= N(r_1)N(r_2)\mathbb{E}(Tr(\rho_A(t_1,r_1)\otimes\rho_A(t_2,r_2))(J\otimes J))$$

$$= \mathbb{E}P(t_1,r_1)\otimes\mathbb{E}P(t_2,r_2)$$

$$+N(r_1)N(r_2)Tr(C_A(t_1,r_1,t_2,r_2)(J\otimes J))$$

where

$$C_A(t_1,r_1,t_2,r_2) = Cov(\rho_A(t_1,r_1),\rho_A(t_2,r_2))$$

$$= \mathbb{E}(\rho_A(t_1,r_1)\otimes\rho_A(t_2,r_2)) - \mathbb{E}(\rho_A(t_1,r_1))\otimes\mathbb{E}(\rho_A(t_2,r_2))$$

With the expectations being carried out w.r.t. the probability distribution of the bath Brownian motion noise. We could also assume the electric field to be a random field and further average the above space-time moments of the density operator w.r.t. the probability distribution of the electric field. However, these models implicitly assume that the joint state of an atom at (t_1,r_1) and another at (t_2,r_2) is given by $\rho_A(t_1,r_1)\otimes\rho_A(t_2,r_2)$. A still more general model will involve assuming that the atoms at two different spatial points interact according to an interaction potential $c.J_1.J_2$, where J_1, J_2 are the spins of the atoms at these two spatial locations, and then use something like the quantum Boltzmann equation to obtain the single atom state by a quantum averaging over the other atoms.

However, that would make things too complicated, and we refer the interested reader to the next section and the literature.

Evaluating these terms explicitly gives

$$M_A(t) = \rho_A(0) + \int_0^t \mathbb{E}\theta_0(s)(\rho_A(0))ds$$

$$+ \int_{0<s_2<s_1<t} \mathbb{E}(\theta_0(s_1)(\theta_0(s_2)(\rho_A(0))))ds_1ds_2$$

where

$$\mathbb{E}\theta_0(t) = i\mu.ad((E_0(t,r),J)) - L_{V_0^2} - R_{V_0^2} + L_{V_0}R_{V_0}$$

and

$$\mathbb{E}[\theta_0(t_1)o\theta_0(t_2)] = \mathbb{E}[\theta_0(t_1)] \otimes \mathbb{E}[\theta_0(t_2)]$$

$$-\mu^2 ad((C_E(t_1,r,t_2,r), J \otimes J))$$

Note the notation used:

$$E_0(t,r) = \mathbb{E}(E(t,r)), C_E(t_1,r_1,t_2,r_2) = Cov(E(t_1,r_1),E(t_2,r_2)) = \mathbb{E}(E(t_1,r_1) \otimes E(t_2,r_2)) - E_0(t_1,r_1) \otimes E_0(t_2,r$$

These equations can be used in the Maxwell equations to get the change in the electric field caused by interaction with matter:

$$curl(\delta E(t,r)) = -\mu_0 \partial_t \delta H(t,r), curl\delta H = \partial_t P(t,r) + \epsilon_0.\partial_t \delta E(t,r)$$

$$\epsilon_0.div\delta E(t,r) + divP(t,r) = 0, div\delta H(t,r) = 0$$

By manipulating these equations after taking *curl* of the first, we easily derive the wave propagation equation for the change in the electric field caused by its interaction with matter (Note that we are neglecting the magnetic dipole moment of matter, i.e., the magnetization):

$$(\nabla^2 - \mu_0\epsilon_0\partial_t^2)\delta E(t,r) = \mu_0\partial_t^2 P(t,r) - \epsilon_0^{-1}\nabla(divP(t,r))$$

where $P(t,r)$ is the random classical polarization field discussed above and given by quantum averaging of the dipole moment per unit volume:

$$P(t,r) = \mu.Tr(\rho_A(t)J)$$

where $\rho_A(t) = \rho_A(t,r)$ is the state of an atom located at r.

3 A remark on how the atomic dynamics in an electric field can get coupled to the noisy bath

Atom in an electric field coupled to a noisy Brownian bath. How does the noise coupling arise? The atomic dipole moment operator is $\mu.J$ where J is a vector of three angular momentum matrices of size $2j+1 \times 2j+1$ satisfying the canonical angular momentum commutation relations. The electric field operator generated by this electric dipole is $E_d(R) = -\nabla_R(K\mu J.(R-r)/|R-r|^3)$ where r is the location of the atom. The bath consists of charged particles whose velocities execute Brownian motion (because the random force on them is described by white noise process and hence their accelerations are also white noise processes), and hence the dipole moment of any one bath particle is proportional to $\int_0^t B(s)ds$ where $B(t)$ is Brownian motion. Thus the total polarization field produced by the bath particles (i.e., dipole moment per unit volume) has the form $P_b(t,R) = \sum_k f_k(R)\int_0^t B_k(s)ds$ and hence the corresponding bath polarization current density is $\partial_t P_b(t,R) = \sum_k f_k(R)B_k(t)$ with the $B_k's$ being standard independent Brownian motion processes. The magnetic vector potential field generated by this bath polarization current is given by (in the non-relativistic approximation)

$$A_b(t,R) = \int_{bath} \partial_t P(t,r)d^3r/|R-r| = \sum_k (\int_{bath} f_k(r)d^3r/|R-r|)B_k(t) = \sum_k g_k(R)B_k(t)$$

The corresponding bath electric field is given by

$$E_b(t,R) = -\partial_t A_b(t,R) = -\sum_k g_k(R)B'_k(t)$$

where $B'_k(t) = dB_k(t)/dt$. Thus, the interaction energy between the bath and the atomic dipole is proportional to the cross term in $\int |E_d(R) + E_b(t,R)|^2 d^3R$ and this cross term is $2\int (E_d(R), E_b(t,R))d^3R$. It is clear that this interaction energy between the dipole and the bath the dipole can be expressed as a bilinear combination of J and the $B'_k(t)'s$, ie, this energy has the form

$$H_I(t) = \sum_k (J, h_k(r))B'_k(t) = \sum_k V_k(r)dB_k(t)/dt$$

where the $h'_k s$ are real 3-vector valued functions of the spatial location of the atom. If we simplify to the case of just one bath particle, then this expression for the interaction energy becomes

$$H_I(t) = V_0 dB(t)/dt$$

and this results after taking into account the Ito correction term in the stochastic Schrödinger equation

$$dU(t) = (-(iH_0(t) + V_0^2/2)dt - iV_0 dB(t))U(t)$$

for the unitary evolution operator $U(t)$.

4 A brief discussion of the quantum Boltzmann equation for taking into account spin interactions between atoms at two different spatial points

Let $\rho_A(t,r)$ denote the mixed state of an atom located at the position r and at time t.

We use as an approximation to the joint state $\rho_{12}(t,r_1,r_2)$ of a pair of atoms located at positions r_1 and r_2 respectively $\rho_A(t,r_1)\otimes\rho_A(t,r_2)$. The interaction potential energy between these two atoms is given by the spin coupling formula $a.J_1, J_2$ where a is a constant. Then by taking the partial trace of the Schrödinger equation over all the remaining $N=\int N(r)d^3r$ atoms in the medium, we obtain the following nonlinear Schrödinger equation for a single atom at r:

$$\partial_t\rho_A(t,r)=-i[H_0(t),\rho_A(t,r)]-ia\int Tr_2[J\otimes J,\rho_A(t,r)\otimes\rho_A(t,r')]N(r')d^3r'$$

If we wish to be more accurate, then we assume that we would write without any approximations (with $\rho_A(t,r)=\rho_k(t,r), k=1,2,3$ in the sense that these are all identical copies of the first density marginal but act in the different component Hilbert spaces)

$$\rho_{12}(t,r_1,r_2)=\rho_1(t,r_1)\otimes\rho_2(t,r_2)+g_{12}(t,r_1,r_2)$$

where $\rho_2(t,r)=\rho_1(t,r)$ by indistinguishability and g_{12} is of the first order of smallness with ρ_1,ρ_2 being of zeroth order of smallness. Since $\rho_1(t,r_1)$ and $\rho_2(t,r_2)$ are the marginals of $\rho_{12}(t,r_1,r_2)$, it follows that

$$Tr_1g_{12}(t,r_1,r_2)=Tr_2g_{12}(t,r_1,r_2)=0$$

Likewise, the joint state $\rho_{123}(t,r_1,r_2,r_3)$ would be expressed as

$$\rho_{123}(t,r_1,r_2,r_3)=\rho_1(t,r_1)\otimes\rho_2(t,r_2)\otimes\rho_3(t,r_3)$$

$$+g_{12}(t,r_1,r_2)\otimes\rho_3(t,r_3)+\rho_1(r_1)\otimes g_{23}(t,r_2,r_3)$$

$$+\rho_2(t,r_2)\otimes g_{13}(t,r_1,r_3)+g_{123}(t,r_1,r_2,r_3)$$

where $\rho_1(t,r)=\rho_2(t,r)=\rho_3(t,r)$, by indistinguishability, and likewise, $g_{12}(t,r,r')=g_{23}(t,r,r')=g_{13}(t,r,r')$, again by indistinguishability. Since ρ_{123} has first order marginals $\rho_1=\rho_2=\rho_3$ and second order marginals $\rho_{12}=\rho_{23}=\rho_{13}$, and the partial traces of $g_{12}=g_{23}=g_{13}$ are zero, it follows that

$$Tr_3g_{123}(t,r_1,r_2,r_3)=Tr_2g_{123}(t,r_1,r_2,r_3)=Tr_1g_{123}(t,r_1,r_2,r_3)=0$$

It should be mentioned that g_{123} is of the second order of smallness. Then, we have the following exact equations obtained by taking partial traces,

$$\partial_t\rho_1(t,r)=-i[H_0(t,r),\rho_1(t,r)]-i\int Tr_2[V_{12}(r,r'),\rho_{12}(t,r,r')]d^3r'$$

$$= -i[H_0(t), \rho_1(t,r)] - i\int Tr_2[V_{12}(r,r'), \rho_1(t,r)\otimes\rho_1(t,r') + g_{12}(t,r,r')]N(r')d^3r'$$

where

$$\partial\rho_{12}(t,r,r') = -i[H_0(t,r) + H_0(t,r') + V_{12}(r,r'), \rho_{12}(t,r,r')]$$

$$-i\int Tr_3[V_{13}(r,r'') + V_{23}(r',r''), \rho_{123}(t,r,r',r'')]N(r'')d^3r''$$

where

$$V_{12}(r,r') = a.(J_1.J_2) = a.J\otimes J$$

(since $J_1 = J\otimes I, J_2 = I\otimes J$).

where $H_0(t,r) = -\mu(E(r,r),J)$ is the interaction Hamiltonian with the external electric field, with explicit dependence of this interaction on the location r of the atom displayed.

By neglecting g_{123}, these equations result in two first-order, nonlinear matrix differential equations for $\rho_1(t,r)$ and $g_{12}(t,r,r')$, and then we reconstruct the second-order marginals:

$$\rho_{12}(t,r,r') = \rho_1(t,r)\otimes\rho_1(t,r') + g_{12}(t,r,r')$$

Assume that these equations have been solved. Then, we obtain the following formula for the spatial correlations of the polarization field $P(t,r)$ at the same time:

$$< P(t,r) >= N(r)Tr(\rho_1(t,r)J),$$

$$< P(t,r_1)\otimes P(t,r_2) >= N(r_1)N(r_2).Tr(\rho_{12}(t,r_1,r_2)(J\otimes J))$$

$$= N(r_1)N(r_2).[Tr(\rho_1(t,r_1)J)\otimes Tr(\rho_1(t,r_2)J) + Tr(g_{12}(t,r_1,r_2)(J\otimes J))$$

where by $J\otimes J$, we mean the 9-vector operator

$$(J_x\otimes J_x, J_x\otimes J_y, J_x\otimes J_z, J_y\otimes J_x, J_y\otimes J_y, J_y\otimes J_z$$

$$, J_z\otimes J_x, J_z\otimes J_y, J_z\otimes J_z)$$

and by $P(t,r_1)\otimes P(t,r_2)$, we mean the standard tensor product between 3-vectors. Note that if $N(r)$ is a random field, then we have to further average these expressions w.r.t. the probability distribution of $N(.)$.

We wish to obtain more generally the space-time correlations of the polarization field, i.e.,

$$Cov(P(t_1,r_1), P(t_2,r_2)) = \mathbb{E}(P(t_1,r_1)\otimes P(t_2,r_2)) - \mathbb{E}(P(t_1,r_1))\otimes\mathbb{E}(P(t_2,r_2))$$

That can be obtained by the following procedure: We start with the joint state $\rho_{12}(t_1,r_1,r_2)$ at time t_1 of the atoms at r_1 and r_2. We measure the spin component $(J,n) = \sum_{a=1}^3 J_a n_a$ of the atom at r_1 at time t_1, where $n = ((n_a))$ is a unit vector and $J_1 = J_x, J_2 = J_y, J_3 = J_z$. Let $E_n(a), a = -j, -j+1, ..., j-1, j$ be the spectral projections of the operator (J,n) corresponding to the eigenvalues

$-j, -j+1, ..., j-1, j$ respectively. After making this measurement, let a denote the outcome. The collapsed state is then

$$\tilde{\rho}_{12}(t_1, r_1, r_2|a, n) = (E_n(a) \otimes I).\rho_{12}(t_1, r_1, r_2)(E_n(a) \otimes I)/Tr(\rho_{12}(t_1, r_1, r_2)(E_n(a) \otimes I))$$

$$= (E_n(a) \otimes I).\rho_{12}(t_1, r_1, r_2)(E_n(a) \otimes I)/Tr(\rho_1(t_1, r_1)E_n(a))$$

Let $T_{t,s}$ denote the nonlinear evolution operator of the quantum Boltzmann equation, as explained above. Then, the joint state at time $t_2 > t_1$ after making the above measurement is given by

$$T_{t_2,t_1}(\tilde{\rho}_{12}(t_1, r_1, r_2|a, n))$$

and hence the joint probability of obtaining the outcome a for the measurement of (J, n) at time t_1 on the atom at r_1 followed by the outcome b for the measurement of the spin component (J, m) $(m = ((m_a)))$ on the the atom at r_2 at time t_2, taking into account state collapse after the first measurement, is given by

$$Pr(a, t_1, n; b, t_2, m) = Tr(T_{t_2,t_1}(\tilde{\rho}_{12}(t_1, r_1, r_2|a))(I \otimes E_m(b)))$$

and this probability can be used to evaluate the correlation between the n^{th} and m^{th} components of the polarization field at (t_1, r_1) and (t_2, r_2) as

$$< (n, P(t_1, r_1)).(m, P(t_2, r_2) >=< n \otimes m, P(t_1, r_1) \otimes P(t_2, r_2) >=$$

$$\sum_{a,b=-j}^{j} ab.Pr(a, t_1, n; b, t_2, m)$$

We can further average this correlation w.r.t. the probability distribution of the random electric field to obtain the space-time correlations of the polarization field.

5 Conclusions

In this paper, we have derived approximate equations for the polarization field and its first two space-time statistical moments based on a quantum mechanical model for a finite state atom described by its spin interacting with an external electric field. We have explained how bath noise is to be taken into account in these equations and have also considered nonlinear effects when the atomic spins of two atoms at two different spatial locations interact with each other, leading thus to a nonlinear quantum Boltzmann equation for the evolution of the first two marginals of the quantum mixed state of a large collection of atoms. This work has applications in nonlinear quantum optics because the averaged atomic dipole moment, and hence the polarization field, as well as their space-time correlations obtained by solving the Schrödinger/master equation perturbatively for the atomic states and then calculating the averaged dipole moment of the atom in such a state, are nonlinear functionals of the electric field. When Maxwell's equations for the perturbation in the electric field caused by such a polarization field coming from light interacting with matter are set up, these equations are therefore highly nonlinear in the incident electric field and thus lead to phenomena such as the generation of higher harmonic frequencies in the scattered radiation pattern. In short, such higher harmonics can thus be attributed to quantum mechanical effects. Using the quantum Boltzmann equation for two-particle states, we are then able to calculate the correlation in the polarization field at two different space-time points based on sequential measurements of atomic spin components, taking into account state collapse, thus overcoming the difficulty in defining temporal correlations owing to the impossibility of simultaneous measurement of an observable at two different times, or equivalently, the difficulty in defining the joint state of two atoms at two different times, namely Heisenberg uncertainty. This paper also contains an account of how one can compute the Lindblad noise bath coupling operator to the atomic dynamics when the bath noise is produced by charged particles executing Brownian motion.

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