# Modified optical Bloch equations for solids

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Recently, DeVoe and Brewer [Phys. Rev. Lett. **50**, 1269 (1983)] observed a striking deviation from a prediction of the optical Bloch equations by monitoring optical free-induction decay in the impurity-ion crystal $Pr^{3+}$:$LaF_3$. At low optical fields, the $Pr^{3+}$ optical dephasing time $T_2$ arises from magnetic fluctuations of the local environment, but at elevated optical fields, $T_2$ is no longer a constant, as assumed in the Bloch equations, because the magnetic line-broadening process is quenched. Several theories have been developed to explain the phenomenon. In this paper we present a simple theory of relaxation in solids which allows for comparison with earlier work. A "strong-redistribution" model is proposed where the optically excited impurity ions experience frequency shifts $\epsilon$ induced by a thermal bath. Frequency jumps occur at an average rate $\Gamma$ and with an rms value of $\epsilon_0/\sqrt{2}$, where $\epsilon_0$ is the thermal width associated with the frequency shifts. Modified Bloch equations (MBE) follow that are solved explicitly for two limiting cases, $\epsilon_0 \ll \Gamma$ and $\epsilon_0 \gg \Gamma$, and qualitatively for the more general case where the ratio $\Gamma/\epsilon_0$ is arbitrary. Since many of the earlier theories are equivalent to the strong-redistribution model in the limit $\epsilon_0 \ll \Gamma$, we can assess the validity of the approximations made by these authors. Finally, we compare our MBE to the analogous transport equations that describe the effects of collisions in atomic vapors. We conclude that the problem addressed here is a general one, common to solids and gases alike, and is not restricted to impurity-ion crystals but will occur whenever frequency fluctuations are important.

## I. INTRODUCTION

In a recent experiment of DeVoe and Brewer,[1] it was concluded that the optical Bloch equations are incapable of describing the saturation phenomena observed. Optical free-induction decay (FID) measurements of the impurity-ion crystal $Pr^{3+}$:$LaF_3$ were conducted where the $Pr^{3+}$ ions are coherently prepared by a laser field under steady-state conditions and then freely precess when the driving field is removed. At low optical fields, the observed $Pr^{3+}$ optical linewidth is dominated by magnetic fluctuations arising from pairs of fluorine nuclear flip-flops. At high optical fields, this nuclear broadening mechanism is quenched and the Bloch equations are seriously violated. On physical grounds, this failure is due to a time averaging of the magnetic interaction as the optical nutation frequency increases.[2] The phenomenological dipole dephasing time $T_2$ of the Bloch equations is therefore not a true constant but lengthens with increasing field strength.

In order to explain these results, Schenzle *et al.*[2] formulated a microscopic theory which extends Redfield's arguments[3] for related saturation effects in nuclear magnetic resonance. Other theories[4–8] have also attempted to explain the DeVoe-Brewer experiments. In all of these theories, it is assumed that the net effect of the fluorine nuclear flip-flops is to produce frequency fluctuations $\epsilon(t)$ in the transition frequency of the ions. At each $Pr^{3+}$ site, frequency fluctuations occur at some effective rate $\Gamma$; the width of the distribution of frequency fluctuations produced by the fluorine nuclear flip-flops is designated by $\epsilon_0$. The different theories correspond to different mathematical models for the frequency fluctuations; Gaussian-Markovian models with $\epsilon_0 \ll \Gamma$,[2,4] Gaussian-Markovian models with the ratio of $\epsilon_0$ to $\Gamma$ arbitrary,[5,6] a non-Markovian model,[8] and a "random-telegraph" model[7] with arbitrary ratio of $\epsilon_0$ to $\Gamma$ have been considered. Some of these theories result in a set of modified Bloch equations (MBE) which characterize atoms in solids interacting with an optical field and a thermal bath that induces fluctuations in the optical transition frequency. In each case, the Bloch equations are modified to include relaxation rates which are intensity dependent.

It is the purpose of this paper to present a simple model of relaxation processes in solids that can be compared with the above theories. The starting point for the theory is a linear transport equation[9,10] in which the frequency shift $\epsilon$ is considered as a stochastic parameter. Within the restrictions of the "impact limit" of pressure-broadening theory,[9] the transport equation can provide a general description of Markovian frequency fluctuations in solids. Each of the Markovian theories mentioned above[2–7] can be obtained as some appropriate limit of the transport equation. In this work, we specialize the calculation to yet another limiting case of the transport equation, the so-called "strong-redistribution" model. In the strong-redistribution model, the frequency shift at each $Pr^{3+}$ site following a fluctuation is statistically determined by a Gaussian distribution having a width equal to the equilibrium width $\epsilon_0$ associated with the frequency fluctuations.

 

In other words, the frequency jumps occur as shown in Fig. 1 with an average time $\Gamma^{-1}$ between jumps, an average displacement of zero, and an rms displacement of $\epsilon_0/\sqrt{2}$. Using this model and treating the shift $\epsilon$ as a random variable, we derive *modified* Bloch equations which characterize the atom-field interaction in the presence of these frequency fluctuations. Use of the strong-redistribution model greatly simplifies the calculation, allows for a comparison with other theories, and can be solved without any restriction on the ratio of $\epsilon_0$ to $\Gamma$. The removal of the restriction $\epsilon_0 \ll \Gamma$ used in some earlier theories[2,4] is significant since the experimental results[1] and a Monte Carlo calculation of the low-field linewidth[11] seem to indicate that $\Gamma$ and $\epsilon_0$ are comparable.

The model for frequency fluctuations which we use is referred to as a "strong" redistribution model since each fluctuation, on average, leads to complete redistribution of the shift $\epsilon$ over the entire width $\epsilon_0$. The frequency fluctuations described by such a model are Markovian, but not Gaussian. In a Gaussian process, each frequency jump $\delta\epsilon$ is much less than the total width $\epsilon_0$. Although the strong redistribution and Markovian-Gaussian models differ in a fundamental manner, they are *mathematically* equivalent if $\epsilon_0 \ll \Gamma$, provided that one interprets the $\Gamma$ appearing in the Gaussian model as an effective collision rate. From a physical point of view, the condition $\epsilon_0 \ll \Gamma$ ensures that the fluctuation rate is sufficiently great to totally redistribute the shifts $\epsilon$ and it is unimportant whether this is achieved by a few "strong" or many "weak" frequency jumps. From a mathematical point of view, the equivalence of the two models follows from the fact that the two-time correlation functions $\langle \epsilon(t)\epsilon(t+\tau)\rangle$ are identical for both models and it is only this two-time correlation function which enters the theory when $\epsilon_0 \ll \Gamma$. Thus, for $\epsilon_0 \ll \Gamma$, we can directly compare our results with those of other authors[2–6] who employ Gaussian-Markovian models of frequency fluctuations. For $\epsilon_0 \gtrsim \Gamma$, our results will, in general, differ qualitatively from those of Gaussian-Markovian models[5,6]—the applicability of a given model must be determined by comparison with experiment. It might also be noted that our results should also agree with those of a "random-telegraph" model[7] if $\epsilon_0 \ll \Gamma$, but may differ from that model for $\epsilon_0 \gtrsim \Gamma$.

In the two limiting cases $\epsilon_0 \ll \Gamma$ and $\epsilon_0 \gg \Gamma$, it *is* possible to average over the variable $\epsilon$ to obtain *reduced* Bloch equations (RBE). In this manner, connection with previous work[2,4] is established. One conclusion is that two apparently equivalent methods, a second-order perturbation treatment[2] and a cumulant expansion approximation,[4] used previously for the $\epsilon_0 \ll \Gamma$ limit, can introduce some errors into the RBE, although these errors are not large for the specific problems discussed by these authors.[2,4] Our method is used to derive an expression for the free-induction decay signal emitted by impurity ions in a solid following steady-state preparation by an optical field. Explicit expressions are evaluated for both cases $\epsilon_0 \ll \Gamma$ and $\epsilon_0 \gg \Gamma$ while a qualitative discussion of the solution is given for the general case where the ratio $\Gamma/\epsilon_0$ is arbitrary. We will see that when $\Gamma \sim \epsilon_0$, it may be possible to observe an FID signal having two decay constants.[5,8,12]

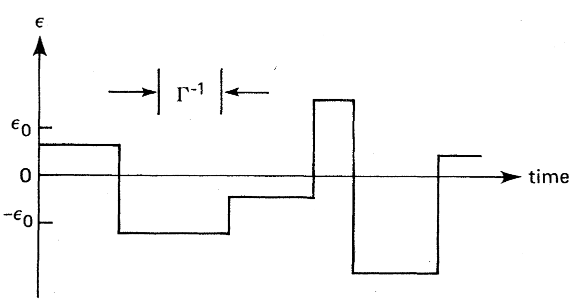


FIG. 1. A random frequency-jump model is shown where the frequency jumps $\epsilon$ occur at an average rate $\Gamma$. The value of $\epsilon$ following a jump is independent of its value before a jump and is determined statistically by a Gaussian distribution having an average value $\langle \epsilon \rangle = 0$ and an rms value $\langle \epsilon^2 \rangle^{1/2} = \epsilon_0/\sqrt{2}$.

The theory, as developed here, bears a marked resemblance to the quantum-mechanical transport equation[9] used to characterize a vapor in which active atoms interact with a radiation field while undergoing elastic velocity-changing collisions with a perturber bath. This result is not surprising, since the frequency fluctuations produced by the fluorine nuclear flip-flops in the solid are analogous to the changes in the Doppler-shifted atomic transition frequency (as viewed in the laboratory frame) produced by velocity-changing collisions in the vapor. It will come as no surprise, therefore, to find that the conventional Bloch equations are inadequate for describing frequency fluctuations in solids since it is well-known[9] that they cannot correctly model velocity-changing collisions in vapors.[13] The recent photon-echo experiment of Yodh and co-workers[14] appears to be an example of a case where it is possible to see effects in vapors which are analogous, but not identical, to those observed by DeVoe and Brewer[1] in solids.

This paper therefore supports the notion that the problem addressed here is a general one affecting solids and gases alike and is not restricted to impurity-ion crystals. In both cases, it is possible for a strong driving optical field to quench a line-broadening mechanism associated with frequency fluctuations produced by perturbations in the vapor or solid.

The approach followed in this work is by no means new. It is always possible to derive an integro-differential transport equation for density matrix elements which depend implicitly on a stochastic variable $\epsilon$ if the following three conditions are met.

(1) The perturber bath producing fluctuations in $\epsilon$ can be considered as an infinite, unchanging reservoir.

(2) The jump time from one value $\epsilon$ to another value $\epsilon'$ (represented as *instantaneous* in Fig. 1) is small compared to all other relevant time scales in the problem.

(3) The value of $\epsilon'$ after a jump depends at most only on the value of $\epsilon$ before the jump.

In the last twenty years, advances in the use of stochastic equations to model physical systems have appeared to follow two parallel but somewhat independent tracks. The first of these is the use of transport equations to describe the time evolution of atoms in a vapor interacting with radiation fields.[9] The second is a somewhat more general development of theories of stochastic equations as applied to both atomic vapors and solids.[10] There have also appeared systematic treatments[15] of the cumulant expansion which leads directly to reduced Bloch equations.

Whereas the method of stochastic equations is not new, we apply it for the first time to the case of optical free-induction decay in solids. Previous treatments[2,4–8] of this problem employed an averaging procedure using properties of the correlation function $\langle \epsilon(t)\epsilon(t-\tau)\rangle$. While this averaging procedure can be formulated in a manner that is equivalent to the transport-equation approach, it is not as convenient. Moreover, we have solved the transport equation without any assumption of the ratio of $\Gamma$ to $\epsilon_0$, within the limits of the strong-redistribution model. Our method enables us to examine the form of the free-induction decay signal for various relaxation schemes (i.e., different decay rates for ionic states, spontaneous emission from one state into another) in the limits $\epsilon_0 \ll \Gamma$ and $\epsilon_0 \gg \Gamma$. For $\epsilon_0 \gtrsim \Gamma$ our predictions can differ qualitatively from those of Gaussian-Markovian[5,6] and random-telegraph models.[7]

A discussion of the model and a derivation of the MBE is given in Sec. II. In Sec. III, the limiting cases under which the MBE can be averaged over bath variables to produce the reduced Bloch equations are discussed. A calculation of the FID signal emitted by atoms after steady-state preparation is presented in Sec. IV, and the relationship between the MBE in vapors and solids is explored in Sec. V. Section VI contains a final discussion of our results.

## II. MODIFIED BLOCH EQUATIONS

To arrive at the modified Bloch equations, we consider a two-state atom (upper state, 2; lower state, 1) interacting with a radiation field (Fig. 2). The frequency separation of the atomic levels is $\omega$, and each state is incoherently pumped at rate $\Lambda_i$ and decays with decay rate $\gamma_i$ $(i=1,2)$. The branching ratio for decay of level 2 back to level 1 is $\gamma_2'/\gamma_2$. The radiation field is characterized by a frequency $\Omega$ and a Rabi frequency $\chi=p_{12}E/\hbar$ where $E$ is the electric field amplitude and $p_{12}$ is an optical dipole matrix element.

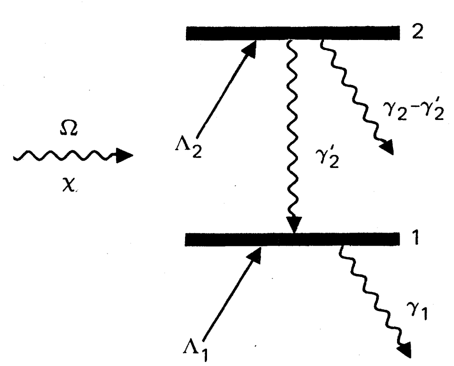


FIG. 2. The two-level quantum system considered shows that levels 1 and 2 are incoherently pumped by a thermal bath at rates $\Lambda_1$ and $\Lambda_2$ and have decay rates $\gamma_1$ and $\gamma_2$. The single-channel $2\rightarrow 1$ decay rate is $\gamma_2'$. The incident field has frequency $\Omega$ and Rabi frequency $\chi$.

The atom is conveniently described by the density matrix elements $\rho_{11}$, $\rho_{22}$, $\tilde{\rho}_{12}$, and $\tilde{\rho}_{21}$, where the tilde indicates an interaction representation. The linear combinations of these elements $u=\tilde{\rho}_{12}+\tilde{\rho}_{21}$, $v=i(\tilde{\rho}_{21}-\tilde{\rho}_{12})$, and $w=\rho_{22}-\rho_{11}$ are customarily referred to as elements of the Bloch vector.[16] In this paper, we choose to work with equations of the density matrix elements since they are somewhat more general than the conventional Bloch equations. However, we shall still refer to these density matrix equations as the "Bloch" equations. Those who prefer the Bloch variables $u$, $v$, and $w$ may refer to Appendix A where all relevant equations are written in this notation.

A field-interaction representation $\rho_{12}=\tilde{\rho}_{12}e^{i\Omega t}\rho_{21}^*$ is used to transform into a frame rotating at the optical frequency $\Omega$. Then, neglecting terms varying as $e^{\pm i(\Omega+\omega)t}$ in this frame, one finds that the density matrix elements obey the equations of motion[17]

$$\dot{\rho}=-\underline{A}\rho+\Lambda\;, \tag{1}$$

where $\rho$ and $\Lambda$ are the column vectors

$$\rho=\begin{bmatrix}\rho_{11}\\ \rho_{22}\\ \tilde{\rho}_{12}\\ \tilde{\rho}_{21}\end{bmatrix},\quad \Lambda=\begin{bmatrix}\Lambda_1\\ \Lambda_2\\ 0\\ 0\end{bmatrix} \tag{2}$$

and the matrix

$$\underline{A}=\begin{bmatrix}\gamma_1 & -\gamma_2' & i\chi/2 & -i\chi/2\\ 0 & \gamma_2 & -i\chi/2 & i\chi/2\\ i\chi/2 & -i\chi/2 & \mu_{12} & 0\\ -i\chi/2 & i\chi/2 & 0 & \mu_{12}^*\end{bmatrix}. \tag{3}$$

Here

$$\mu_{12}=\gamma_{12}-i\Delta\;, \tag{4}$$

$$\Delta=\omega-\Omega\;, \tag{5}$$

$$\gamma_{12}=(\gamma_1+\gamma_2)/2\;, \tag{6}$$

and

$$\chi=p_{12}E/\hbar\;. \tag{7}$$

Equation (1) can be further modified by various broadening or relaxation processes occurring in the solid. In this paper we consider two such processes. First, there is a large scale *inhomogeneous* broadening produced by crystalline strains. The atomic frequency varies from one atomic site to another and the overall distribution of frequencies is governed by a distribution function $g(\omega)$ centered at $\omega=\omega_0$, the unperturbed transition frequency, or equivalently by a distribution function $g(\Delta)$ centered at

$$\Delta_0=\omega_0-\Omega\;. \tag{8}$$

The distribution $g(\Delta)$ is assumed to be of the Gaussian form

$$g(\Delta)=(\pi^{1/2}\Delta^*)^{-1}\exp[-(\Delta-\Delta_0)^2/(\Delta^*)^2]\ , \tag{9}$$

where the linewidth $\Delta^*$ is assumed to be larger than all other relevant frequencies in the problem.

In addition to this large-scale inhomogeneous broadening mechanism, it is assumed that local magnetic field variations (due to the fluorine nuclei) at the impurity-ion sites ($Pr^{3+}$ ions) provide an *independent* and second source of *inhomogeneous* broadening leading to shifts in the tuning parameter

$$\Delta\to\Delta+\epsilon(t)\ , \tag{10}$$

where $\epsilon(t)$ is the frequency displacement produced by the local-field variations.

The model for $\epsilon(t)$ which we adopt is shown in Fig. 1. The frequency displacement $\epsilon(t)$ is assumed to undergo jumps at some average rate $\Gamma$. The jumps occur "instantaneously" and the value of $\epsilon$ following any jump is *independent* of its value before the jump, being determined statistically by the thermal-equilibrium distribution of frequency fluctuations,

$$F(\epsilon)=(\pi^{1/2}\epsilon_0)^{-1}\exp[-(\epsilon/\epsilon_0)^2]\ . \tag{11}$$

Thus the root-mean-square (rms) value of $\epsilon$ is $\epsilon_0/\sqrt{2}$. As discussed in the Introduction, this strong-redistribution model is mathematically equivalent to Gaussian-Markovian models[2–6] in the limit $\epsilon_0\ll\Gamma$, but can differ from those models if $\epsilon_0\gtrsim\Gamma$.

Instead of considering $\epsilon$ as a function of time at each atomic site, one can formulate an equivalent theory by assuming that $\epsilon$ is a stochastic variable that characterizes the *ensemble* density matrix $\rho(\epsilon,t)$. Owing to the local-field variations, there is a contribution $(d\rho/dt)_{\mathrm{lf}}$ to $\dot\rho$ of the form

$$(\partial\rho(\epsilon,t)/\partial t)_{\mathrm{lf}}=-\Gamma\rho(\epsilon,t)+\int W(\epsilon'\to\epsilon)\rho(\epsilon',t)d\epsilon'\ , \tag{12}$$

where $\Gamma$ is the average rate at which frequency jumps occur and $W(\epsilon'\to\epsilon)$ is the probability density per unit time that a jump changes the local-field frequency from $\epsilon'$ to $\epsilon$. The first term on the right-hand side (rhs) of Eq. (12) gives the loss of element $\rho(\epsilon,t)$ resulting from frequency shifts taking $\epsilon$ to any other value $\epsilon'$, while the second term adds to $\rho(\epsilon,t)$ resulting from jumps that take $\epsilon'$ to $\epsilon$. In the model that we have adopted, the kernel $W(\epsilon'\to\epsilon)$ is independent of the initial frequency displacement $\epsilon'$, and the distribution of $\epsilon$ following a jump is given by Eq. (11). In other words, the kernel appropriate to the adopted model for $\epsilon(t)$ is simply

$$W(\epsilon'\to\epsilon)=\Gamma F(\epsilon)\ . \tag{13}$$

Treating $\epsilon$ as a stochastic variable rather than a time-dependent function simplifies the analysis.

Incorporating Eqs. (9)–(13) into Eq. (1) and labeling $\boldsymbol{\rho}$ by the parameters $\epsilon$ and $\Delta$ in addition to $t$, one obtains the modified Bloch equations

$$\begin{aligned}\partial\boldsymbol{\rho}(\Delta,\epsilon,t)/\partial t &= -\underline{A}\boldsymbol{\rho}(\Delta,\epsilon,t)+i\epsilon\underline{B}\boldsymbol{\rho}(\Delta,\epsilon,t)\\ &\quad -\underline{\Gamma}\boldsymbol{\rho}(\Delta,\epsilon,t)+\Gamma F(\epsilon)\int\boldsymbol{\rho}(\Delta,\epsilon',t)d\epsilon'\\ &\quad +\boldsymbol{\Lambda}(\Delta,\epsilon)\ ,\end{aligned} \tag{14a}$$

where

$$\underline{B}=\begin{bmatrix}0&0&0&0\\0&0&0&0\\0&0&1&0\\0&0&0&-1\end{bmatrix} \tag{14b}$$

and

$$\boldsymbol{\Lambda}(\Delta,\epsilon)=F(\epsilon)g(\Delta)\begin{bmatrix}\lambda_1\\\lambda_2\\0\\0\end{bmatrix}\ . \tag{14c}$$

Equations (14) are the modified Bloch Equations (MBE) for the specific local-field model which has been adopted. Note that the source term (14c) depends on both distribution functions $F(\epsilon)$ and $g(\Delta)$ corresponding to the two inhomogeneous line shapes. In experiments, one does not measure $\boldsymbol{\rho}(\Delta,\epsilon,t)$, but rather its value averaged over the variable $\epsilon$, i.e.,

$$\mathbf{Q}(\Delta,t)=\int\boldsymbol{\rho}(\Delta,\epsilon,t)d\epsilon\ . \tag{15}$$

In certain limiting cases to be discussed in the next section, it is possible to average (14a) over $\epsilon$ to arrive at a corresponding equation for $\mathbf{Q}(\Delta,t)$. In that case, we shall refer to the equations for $\mathbf{Q}(\Delta,t)$ as the reduced Bloch equations (RBE).

The MBE or RBE can be solved once the initial conditions are specified. In typical coherent transient experiments, the applied-field amplitude and frequency are kept constant during a specified time interval and then changed to a new value for a second time interval. Thus, for any *one* of these intervals the MBE or RBE are a set of coupled first-order linear differential equations with constant coefficients. A general solution of the entire coherent transient problem can be obtained by solving the equations in each time interval and matching the boundary conditions. In Sec. III, we consider the solution for an arbitrary time interval in which the applied-field amplitude and frequency remain constant. In Sec. IV, an expression for an FID signal is derived.

## III. REDUCED BLOCH EQUATIONS

The MBE can be reduced if either $\epsilon_0\ll\Gamma$ or $\epsilon_0\gg\Gamma,\chi,\gamma_{12}$, cases which we now consider.

### A. Case $\epsilon_0\ll\Gamma$

We expand $\boldsymbol{\rho}(\Delta,\epsilon,t)$ as a power series in $\epsilon$ and write

$$\boldsymbol{\rho}(\Delta,\epsilon,t)=F(\epsilon)\boldsymbol{\psi}(\Delta,\epsilon,t) \tag{16}$$

with

$$\boldsymbol{\psi}(\Delta,\epsilon,t)\simeq\boldsymbol{\psi}^{(0)}(\Delta,t)+\epsilon\boldsymbol{\psi}^{(1)}(\Delta,t)+\epsilon^2\boldsymbol{\psi}^{(2)}(\Delta,t)\ , \tag{17}$$

$F(\epsilon)$ being the Gaussian distribution (11). Consequently, the variable $\mathbf{Q}(\Delta,t)$ defined by (15) is given by

$$\mathbf{Q}(\Delta,t)\simeq\boldsymbol{\psi}^{(0)}(\Delta,t)+\langle\epsilon^2\rangle\boldsymbol{\psi}^{(2)}(\Delta,t)\ , \tag{18}$$

where

$$\langle\epsilon^2\rangle=\int \epsilon^2 F(\epsilon)d\epsilon=\epsilon_0^2/2 \tag{19}$$

and we have used the fact that $\langle\epsilon\rangle=0$.

Using (14a) and (16)—(19) one obtains the following sequence of equations:

$$\frac{\partial\boldsymbol{\psi}^{(0)}(\Delta,t)}{\partial t}=-(\underline{A}+\underline{\Gamma})\boldsymbol{\psi}^{(0)}(\Delta,t)+\Gamma\mathbf{Q}(\Delta,t)+\boldsymbol{\Lambda}(\Delta)\ , \tag{20a}$$

$$\frac{\partial\boldsymbol{\psi}^{(1)}(\Delta,t)}{\partial t}=-(\underline{A}+\underline{\Gamma})\boldsymbol{\psi}^{(1)}(\Delta,t)+i\underline{B}\boldsymbol{\psi}^{(0)}(\Delta,t)\ , \tag{20b}$$

$$\frac{\partial\boldsymbol{\psi}^{(2)}(\Delta,t)}{\partial t}=-(\underline{A}+\underline{\Gamma})\boldsymbol{\psi}^{(2)}(\Delta,t)+i\underline{B}\boldsymbol{\psi}^{(1)}(\Delta,t)\ , \tag{20c}$$

where the elements of the incoherent source term (14c) are

$$\Lambda_i(\Delta)=\begin{cases}\lambda_i g(\Delta), & i=1,2\\ 0, & i=3,4\ .\end{cases} \tag{21}$$

Taking the time derivative of (18), we find to second order in $\epsilon$ that

$$\frac{\partial\mathbf{Q}(\Delta,t)}{\partial t}=-\underline{A}\mathbf{Q}(\Delta,t)+i\langle\epsilon^2\rangle\underline{B}\boldsymbol{\psi}^{(1)}(\Delta,t)+\boldsymbol{\Lambda}(\Delta)\ . \tag{22}$$

Because of the $\langle\epsilon^2\rangle$ factor of $\boldsymbol{\psi}^{(1)}(\Delta,t)$ in (22), $\boldsymbol{\psi}^{(1)}(\Delta,t)$ of (20b) need only be solved to zeroth order in $\epsilon$. To zeroth order in $\epsilon$, it follows from (18) that $\boldsymbol{\psi}^{(0)}(\Delta,t)=\mathbf{Q}(\Delta,t)$. Using this approximation to solve (20a) and (20b) for $\boldsymbol{\psi}^{(1)}(\Delta,t)$, we obtain to zeroth order in $\epsilon$,

$$\boldsymbol{\psi}^{(1)}(\Delta,t)=e^{-(\underline{A}+\underline{\Gamma})(t-t_0)}\boldsymbol{\psi}^{(1)}(\Delta,t_0)+i\int_{t_0}^{t}\{\exp[-(\underline{A}+\underline{\Gamma})(t-t')]\}\underline{B}\mathbf{Q}(\Delta,t')dt'\ , \tag{23}$$

where $t_0$ is some arbitrary initial time at which $\boldsymbol{\psi}^{(1)}(\Delta,t)$ and $\mathbf{Q}(\Delta,t)$ are specified. Furthermore, to zeroth order in $\epsilon$ it follows from (20a) and (18) that

$$\mathbf{Q}(\Delta,t')\simeq\boldsymbol{\psi}^{(0)}(\Delta,t')\simeq e^{-\underline{A}(t'-t)}\mathbf{Q}(\Delta,t)+\int_t^{t'}e^{-\underline{A}(t'-t'')}\boldsymbol{\Lambda}(\Delta)dt''\ . \tag{24}$$

Combining (22)—(24), one arrives at the RBE

$$\frac{\partial\mathbf{Q}(\Delta,t)}{\partial t}=-[\underline{A}+\underline{R}(t-t_0)]\mathbf{Q}(\Delta,t)+[\underline{1}-\underline{S}(t-t_0)]\boldsymbol{\Lambda}(\Delta)+i\langle\epsilon^2\rangle\underline{B}e^{-(\underline{A}+\underline{\Gamma})(t-t_0)}\boldsymbol{\psi}^{(1)}(\Delta,t_0)\ , \tag{25}$$

where

$$\underline{R}(\tau)=\langle\epsilon^2\rangle\underline{B}\int_0^{\tau}e^{-\Gamma\tau'}e^{-\underline{A}\tau'}\underline{B}e^{\underline{A}\tau'}d\tau'\ , \tag{26}$$

$$\underline{S}(\tau)=\langle\epsilon^2\rangle\underline{B}\int_0^{\tau}e^{-\Gamma\tau'}e^{-\underline{A}\tau'}\underline{B}(1-e^{\underline{A}\tau'})\underline{A}^{-1}d\tau'\ , \tag{27}$$

and $\boldsymbol{\psi}^{(1)}(\Delta,t_0)$ must be obtained from (20b). Equation (25) is the RBE in the limit $\epsilon_0\ll\Gamma$.

If in (25), one arbitrarily sets $\underline{S}(\tau)=\underline{0}$, neglects the $\boldsymbol{\psi}^{(1)}(\Delta,t)$ term, and evaluates the matrix $\underline{R}(t-t_0)$ at $t-t_0=\infty$, there results

$$\frac{\partial\mathbf{Q}(\Delta,t)}{\partial t}=-(\underline{A}+\underline{R})\mathbf{Q}(\Delta,t)+\boldsymbol{\Lambda}(\Delta)\ , \tag{28a}$$

$$\underline{R}\equiv\underline{R}(\infty)\ , \tag{28b}$$

which reproduces the equations of Schenzle *et al.*[2] and Yamanoi and Eberly.[4] The $\underline{R}$ matrix

$$\underline{R}=\begin{bmatrix}0&0&0&0\\0&0&0&0\\R_2&-R_2&R_1&0\\R_2^*&-R_2^*&0&R_1\end{bmatrix} \tag{29}$$

calculated in the limit $\Gamma\gg\gamma_1,\gamma_2$ then has elements

$$R_1=\frac{\langle\epsilon^2\rangle}{\Gamma\beta^2}\left[\Delta^2+\frac{\chi^2\Gamma^2}{\Gamma^2+\beta^2}\right]\ , \tag{30}$$

$$R_2=\frac{\langle\epsilon^2\rangle}{2}\frac{\chi}{\Gamma}\left[\frac{-\Delta+i\Gamma}{\Gamma^2+\beta^2}\right]\ , \tag{31}$$

where

$$\beta^2=\Delta^2+\chi^2\ . \tag{32}$$

Equations (28)—(32) agree with the results of these authors, which are given in Appendix B in their notation for easy comparison.

The RBE (25) differs from the RBE (28) of Schenzle *et al.*[2] and Yamanoi and Eberly.[4] These results differ basically for two reasons. First, if one considers only those times $(t-t_0)\gg\Gamma^{-1}$ (which is an implicit assumption in the work of Schenzle *et al.*[2] and Yamonoi and Eberly[4]), the last term in Eq. (25) can be neglected owing to the $\exp[-\Gamma(t-t_0)]$ factor, and the upper integration limits in Eqs. (26) and (27) can be set equal to infinity. In this limit, Eqs. (25) and (28) differ only by the term $-\mathbf{S}(\infty)\boldsymbol{\Lambda}(\Delta)$ appearing in Eq. (25). This term is absent in the work of Schenzle *et al.*[2] and Yamanoi and Eberly,[4] since they effectively neglected the inhomogeneous term in calculating the effects of the frequency fluctuations. Although this term is absent in their equations and it is not negligible compared to the $\underline{R}\mathbf{Q}$ term, an analysis of the overall effect of the $\underline{S}\boldsymbol{\Lambda}$ term on the FID solutions indicates its contribution is only of order $(\gamma_i/\Gamma)^2$, which is implicitly chosen to be a small parameter in these earlier calculations.[2,4,18] Thus, the RBE of Refs. 2 and 4 are a correct starting point within the limits of these approximations. Quite generally, one can show that (25) and (28) are approximately equal if $\Gamma$ is much greater than $\gamma_i$, $|\Delta|$, and $\chi$; this result follows if one neglects the $\boldsymbol{\psi}^{(1)}$ term in Eq. (25) owing to the $e^{-\Gamma(t-t_0)}$ factor and evaluates $\underline{S}(t-t_0)$ and $\underline{R}(t-t_0)$ asymptotically for large $\Gamma$. Additional analysis of Eqs. (25) and (28) is required to determine if any more general conclusions concerning the differences between these equations can be established. At this stage, however, we must consider Eq. (25) as the correct RBE in the limit $\epsilon_0\ll\Gamma$ for the model problem under consideration.

For the steady-state problem $[d\mathbf{Q}(\Delta,t)/\partial t=0;\ t_0\sim-\infty]$, it is possible to simplify Eq. (25). The

$\{\exp[-(\underline{A}+\underline{\Gamma})(t-t_0)]\}\boldsymbol{\psi}^{(1)}(\Delta,t_0)$ term can be dropped as $t_0 \sim -\infty$ and the upper integration limit in Eqs. (26) and (27) can be set equal to infinity. In addition, one can use (18) and (20a) to show that the $\underline{S}\Lambda(\Delta)$ term in (25) can be replaced by $\underline{S}\underline{A}\mathbf{Q}(\Delta)$ with an error of order $\epsilon_0^4$. With these modifications, Eqs. (25)—(27) can be combined and the integrations carried out to obtain

$$\frac{\partial \mathbf{Q}(\Delta)}{\partial t}=\mathbf{0}=-(\underline{A}+\underline{H})\mathbf{Q}(\Delta)+\boldsymbol{\Lambda}(\Delta)\ , \tag{33}$$

where

$$\underline{H}=\langle \epsilon^2\rangle \underline{B}(\underline{A}+\underline{\Gamma})^{-1}\underline{B}\ . \tag{34}$$

We invert the $(\underline{A}+\underline{\Gamma})$ matrix to obtain the explicit result

$$\underline{H}=\begin{bmatrix} 0 & 0 & 0 & 0\\ 0 & 0 & 0 & 0\\ 0 & 0 & H_1 & H_2\\ 0 & 0 & H_2 & H_1^* \end{bmatrix} \tag{35}$$

with

$$H_1=\frac{1}{D^t}\left[(\mu_{12}^t)^*+\frac{\chi^2}{4\gamma_L^t}\right]\langle \epsilon^2\rangle\ , \tag{36}$$

$$H_2=-\frac{\chi^2\langle \epsilon^2\rangle}{4D^t\gamma_L^t}\ . \tag{37}$$

The superscript $t$ denotes the addition of $\Gamma$ as follows:

$$\gamma_1^t=\gamma_1+\Gamma,\ \ \gamma_2^t=\gamma_2+\Gamma\ , \tag{38}$$

$$\gamma_{12}^t=\gamma_{12}+\Gamma,\ \ \mu_{12}^t=\mu_{12}+\Gamma\ , \tag{39}$$

$$D^t=|\mu_{12}^t|^2+\tfrac{1}{2}\chi^2\gamma_{12}^t/\gamma_L^t\ , \tag{40}$$

and

$$\frac{1}{\gamma_L^t}=\frac{1}{\gamma_2^t}+\frac{1}{\gamma_1^t}\left[1-\frac{\gamma_2'}{\gamma_2^t}\right],\ \ \frac{1}{\gamma_L}=\frac{1}{\gamma_2}+\frac{1}{\gamma_1}\left[1-\frac{\gamma_2'}{\gamma_2}\right]. \tag{41}$$

Equation (33) is the correct RBE to use for the steady-state problem in the limit $\epsilon_0 \ll \Gamma$.

### B. Case $\epsilon_0 \gg \Gamma,\chi,\gamma_{12}$

If $\epsilon_0$ is much greater than all relevant relaxation rates in the problem but is still much less than the large inhomogeneous width $\Delta^*$, it is again possible to obtain RBE of a certain type. To write RBE in this limit, one must first separate out the zero-field solution for $\rho_{11}$ and $\rho_{22}$ by writing

$$\rho_{ii}(\Delta,\epsilon,t)=\rho_{ii}^{(0)}(\Delta,\epsilon,t)+\delta\rho_{ii}(\Delta,\epsilon,t),\ \ i=1,2 \tag{42a}$$

where $\rho_{ii}^{(0)}(\Delta,\epsilon,t)$ is the solution of Eq. (14a) with the field $\chi$ set equal to zero. Then $\delta\boldsymbol{\rho}=(\delta\rho_{11},\delta\rho_{22},\tilde{\rho}_{12},\tilde{\rho}_{21})$ satisfies

$$\begin{aligned}\partial[\delta\boldsymbol{\rho}(\Delta,\epsilon,t)]/\partial t &= -\underline{A}\delta\boldsymbol{\rho}(\Delta,\epsilon,t)+i\epsilon\underline{B}\delta\boldsymbol{\rho}(\Delta,\epsilon,t)-\underline{\Gamma}\delta\boldsymbol{\rho}(\Delta,\epsilon,t)\\ &\quad+\Gamma F(\epsilon)\int \delta\boldsymbol{\rho}(\Delta,\epsilon',t)d\epsilon'+\delta\boldsymbol{\Lambda}(\Delta,\epsilon)\ ,\end{aligned} \tag{42b}$$

where

$$\delta\boldsymbol{\Lambda}(\Delta,\epsilon)=\frac{i\chi}{2}[\rho_{22}^{(0)}(\Delta,\epsilon,t)-\rho_{11}^{(0)}(\Delta,\epsilon,t)]\begin{bmatrix}0\\0\\1\\-1\end{bmatrix}. \tag{42c}$$

The $\delta\rho_{ii}(\Delta,\epsilon,t)$ as well as $\tilde{\rho}_{12}(\Delta,\epsilon,t)$ and $\tilde{\rho}_{21}(\Delta,\epsilon,t)$ result solely from atom-field interactions. Consequently, they are nonvanishing in a frequency range of order $\gamma_{12}$ (or $\Gamma$ or $\chi$) about $(\Delta+\epsilon)=0$ ("hole burning"). If $\epsilon_0 \gg \gamma_{12},\Gamma,\chi$, the "out" (loss terms at rate $\Gamma$) are no longer approximately compensated by the "in" terms, whose contributions are relatively small [of order $\max(\gamma_{12},\Gamma,\chi)/\epsilon_0$]. If the "in" terms in Eq. (42b) are dropped, one finds that $\delta\boldsymbol{\rho}$ is essentially equal to $F(\epsilon)$ times a function of $(\Delta+\epsilon)$. If the resulting equation is averaged over *both* $\epsilon$ and $\Delta$ using the fact that the inhomogeneous width $\Delta^*$ is larger than all relevant frequency scales in the problem, then one can obtain the RBE

$$\partial[\delta\mathbf{Q}(\Delta,t)]/\partial t=-(\underline{A}+\underline{\Gamma})[\delta\mathbf{Q}(\Delta,t)]+\delta\boldsymbol{\Lambda}(\Delta) \tag{42d}$$

with the understanding that $\delta\mathbf{Q}(\Delta,t)$ must ultimately be averaged over $\Delta$ for Eq. (42d) to be valid. The local-field fluctuations result solely in a decay of ionic density matrix elements that have been created by the external laser field.

## IV. FREE-INDUCTION DECAY

We consider the case where the radiation field interacts with atoms for times $-\infty < t \le 0$ and creates a "steady-state" density matrix distribution $\boldsymbol{\rho}(\Delta,\epsilon)$. At time $t=0$, the field is suddenly switched off and the sample radiates a free-induction decay signal whose amplitude

$$Q_3(t)=\int Q_3(\Delta,t)d\Delta\ , \tag{43}$$

represents an average over the inhomogeneous line shape function $g(\Delta)$, Eq. (9). The quantity $Q_3(\Delta,t)$, is the third element of the vector $\mathbf{Q}$ defined by (15) and corresponds to the third element ($\tilde{\rho}_{12}$) of $\boldsymbol{\rho}$ in (2). In this section, we calculate $Q_3(t)$ for the two limits, $\epsilon_0 \ll \Gamma$ and $\epsilon_0 \gg \Gamma$, in which the RBE is valid and also for the more general MBE.

### A. RBE: $\epsilon_0 \ll \Gamma$

For times $t>0$, there is no applied radiation field ($\chi=0$) and the matrix $\underline{A}$ is diagonal. Setting $t_0=0$ in (25), evaluating $\boldsymbol{\psi}^{(1)}(\Delta)$ needed in (25) from (20a) and (20b), and making use of (34), one finds that

$$\begin{aligned}\partial Q_3(\Delta,t)/\partial t &= -\left[\mu_{12}+\frac{\langle\epsilon^2\rangle}{\Gamma}(1-e^{-\Gamma t})\right]Q_3(\Delta,t)\\ &\quad -e^{-(\mu_{12}+\Gamma)t}[\underline{H}\mathbf{Q}(\Delta)]_3\ .\end{aligned} \tag{44}$$

Here, $\mathbf{Q}(\Delta)$ is the steady-state value of $\mathbf{Q}$ obtained as a solution of (33). Since $\underline{H}$ is of order $\langle\epsilon^2\rangle/\Gamma$ [see Eq. (34)], the second term in Eq. (44) is down by order $\langle\epsilon^2\rangle/\Gamma^2$ from the first and can be neglected, leading to the solution

$$Q_3(\Delta,t)=Q_3(\Delta)\exp\left[-\left[\mu_{12}+\frac{\langle\epsilon^2\rangle}{\Gamma}\right]t+\frac{\langle\epsilon^2\rangle}{\Gamma^2}(1-e^{-\Gamma t})\right]. \quad (45)$$

For $\langle\epsilon^2\rangle/\Gamma^2 \ll 1$, the final term in the exponent can be discarded and one has

$$Q_3(\Delta,t)=Q_3(\Delta)\exp\left[-\left[\gamma_{12}+\frac{\langle\epsilon^2\rangle}{\Gamma}-i\Delta\right]t\right]. \quad (46)$$

The steady-state solution $\mathbf{Q}(\Delta)=[1/(\underline{A}+\underline{H})]\mathbf{\Lambda}(\Delta)$ of (33) for $Q_3(\Delta)$ is straightforward, but tedious. After some algebra, one finds

$$Q_3(\Delta)=\frac{1}{2}\frac{i\chi N_{21}(\Delta)[\mu_{12}^*+(\delta H)^*]}{|\mu_{12}+\delta H|^2+\frac{1}{2}\frac{\chi^2\gamma_{12}}{\gamma_L}\left[1+\frac{\langle\epsilon^2\rangle}{D^t}\left[\frac{\gamma_{12}^t}{\gamma_{12}}+\frac{\gamma_L}{\gamma_L^t}\right]\right]}, \quad (47)$$

where the zero-field population difference

$$N_{21}(\Delta)=\left[\frac{\Lambda_2}{\gamma_2}-\left[\frac{\Lambda_1}{\gamma_1}+\frac{\gamma_2'\Lambda_2}{\gamma_1\gamma_2}\right]\right]g(\Delta) \quad (48)$$

and

$$\delta H=(\mu_{12}^t)^*\langle\epsilon^2\rangle/D^t. \quad (49)$$

One is faced with the task of evaluating

$$Q_3(t)=\int d\Delta\, Q_3(\Delta)\exp\left[-\left[\gamma_{12}+\frac{\langle\epsilon^2\rangle}{\Gamma}-i\Delta\right]t\right], \quad (50)$$

the FID signal in the limit $\langle\epsilon^2\rangle/\Gamma^2 \ll 1$.

Substituting (47) into (50) and factoring the broad inhomogeneous distribution $N_{21}(\Delta)$ outside the integral, allows one to carry out contour integration in the upper-half plane. The integration can be simplified somewhat with the additional restriction

$$\frac{\langle\epsilon^2\rangle}{\Gamma\gamma_{12}}<1. \quad (51)$$

In that case, the poles in Eq. (47) can be found using an iterative procedure in which (1) the poles are first obtained by neglecting all terms containing $\langle\epsilon^2\rangle$, (2) the approximate values for the poles are inserted in the $\langle\epsilon^2\rangle$ terms, and finally, (3) the poles are recalculated including the modified $\langle\epsilon^2\rangle$ terms. In this manner, one finds that, to order $\epsilon_0^2$ and with the restriction (51), the FID signal is given by

$$Q_3(t)=-i\pi\frac{\chi}{2}N_{21}(0)\times\left[1-\frac{\gamma_{12}}{\bar\gamma}-\frac{\langle\epsilon^2\rangle}{D_0}\left[1+\frac{\gamma_{12}^t}{\bar\gamma}\right]\right]\exp(-\gamma_F t), \quad (52)$$

where

$$\gamma_F=\gamma_{12}+\frac{\langle\epsilon^2\rangle}{\Gamma}+\bar\gamma, \quad (53)$$

$$\bar\gamma^2=\gamma_{12}^2+\chi^2\gamma_{12}/(2\gamma_L)+[\langle\epsilon^2\rangle\chi^2/(2D_0\gamma_L)]\times[3\gamma_{12}+\Gamma+(\gamma_L/\gamma_L^t)\gamma_{12}]+(2\langle\epsilon^2\rangle\gamma_{12}/D_0)(2\gamma_{12}+\Gamma), \quad (54)$$

and

$$D_0=(\gamma_{12}^t)^2-\gamma_{12}^2+\frac{\chi^2}{2}\left[\frac{\gamma_{12}^t}{\gamma_L^t}-\frac{\gamma_{12}}{\gamma_L}\right]. \quad (55)$$

We now see that when $\chi\sim 0$, $Q_3(t)$ is of order $\chi^3$ and there is no linear FID, in contrast to the findings of Schenzle *et al.*[2] In weak fields, the FID decay rate is

$$\gamma_F\sim\left[\gamma_{12}+\frac{\langle\epsilon^2\rangle}{\Gamma}+\left[\gamma_{12}^2+\frac{2\langle\epsilon^2\rangle\gamma_{12}(2\gamma_{12}+\Gamma)}{(\gamma_{12}^t)^2-\gamma_{12}^2}\right]^{1/2}\right]. \quad (56)$$

In intense fields, the decay constant is

$$\gamma_F\sim\left[\gamma_{12}+\frac{\langle\epsilon^2\rangle}{\Gamma}+\left[\gamma_{12}^2+\tfrac{1}{2}\chi^2\frac{\gamma_{12}}{\gamma_L}+\frac{\tfrac{1}{2}\chi^2\langle\epsilon^2\rangle\left[\frac{\gamma_{12}^t}{\gamma_L}+\frac{2\gamma_{12}}{\gamma_L}+\frac{\gamma_{12}}{\gamma_L^t}\right]}{(\gamma_{12}^t)^2-\gamma_{12}^2+\tfrac{1}{2}\chi^2\left[\frac{\gamma_{12}^t}{\gamma_L^t}-\frac{\gamma_{12}}{\gamma_L}\right]}\right]^{1/2}\right]. \quad (57)$$

It is interesting to examine two limits of the high-field result, Eq. (57). First consider the case of an "open system" with equal decay rates ($\gamma_1=\gamma_2=\gamma_{12}$ and $\gamma_2'=0$) so that $\gamma_L=\gamma_{12}/2$, $(\gamma_{12}^t/\gamma_L^t-\gamma_{12}/\gamma_L)=0$, and

$$\gamma_F\sim\left\{\gamma_{12}+\frac{\langle\epsilon^2\rangle}{\Gamma}+\left[\gamma_{12}^2+\chi^2\left[1+\frac{\langle\epsilon^2\rangle\left[\frac{\gamma_{12}^t}{\gamma_{12}}+2+\frac{\gamma_{12}}{\gamma_{12}^t}\right]}{(\gamma_{12}^t)^2-\gamma_{12}^2}\right]\right]^{1/2}\right\}. \quad (58)$$

If, furthermore, $\Gamma \gg \gamma_{12}$, then

$$\gamma_F \sim \left\{ \gamma_{12} + \frac{\langle \epsilon^2 \rangle}{\Gamma} + \left[ \gamma_{12}^2 + \chi^2 \left[ 1 + \frac{\langle \epsilon^2 \rangle}{\Gamma \gamma_{12}} \right] \right]^{1/2} \right\}$$
$$\simeq \chi (1 + \langle \epsilon^2 \rangle / \Gamma \gamma_{12})^{1/2} \,. \tag{59}$$

It is interesting to note that (59) *is* the prediction of the conventional Bloch equations in the limit (51). On the other hand, for a "closed system" ($\gamma_2 = \gamma_2' + \gamma_1 \equiv 2\gamma$; $\gamma_1 = 0$), one has $\gamma_L = \gamma$, $\gamma_{12} = \gamma$, and

$$\gamma_F \sim \left\{ \gamma + \frac{\langle \epsilon^2 \rangle}{\Gamma} + \left[ \gamma^2 + \frac{\chi^2}{2} \left[ 1 + \frac{\langle \epsilon^2 \rangle \left[ \dfrac{\gamma_{12}^t}{\gamma} + 2 + \dfrac{\gamma}{\gamma_L^t} \right]}{(\gamma_{12}^t)^2 - \gamma^2 + \frac{1}{2}\chi^2 \left[ \dfrac{\gamma_{12}^t}{\gamma_L^t} - 1 \right]} \right] \right]^{1/2} \right\} . \tag{60}$$

In addition, when $\Gamma \gg \gamma$ and $\chi \gg \Gamma$,

$$\gamma_F \sim \left[ \gamma + \frac{\langle \epsilon^2 \rangle}{\Gamma} + \left[ \gamma^2 + \frac{\chi^2}{2} + \frac{\langle \epsilon^2 \rangle \Gamma}{\gamma} \right]^{1/2} \right]$$
$$\simeq \chi / \sqrt{2} \,. \tag{61}$$

Thus, the high-intensity value for $\gamma_F$ differs for the open system [Eq. (59)] and the closed system [Eq. (61)]. In general, the value of $\gamma_F$ for $\chi \gg \Gamma$ and $\Gamma \gg \gamma_{12}$ follows from (57) as

$$\gamma_F \sim \chi \left[ \frac{1}{2} \frac{\gamma_{12}}{\gamma_L} + \frac{1}{2} \frac{\langle \epsilon^2 \rangle \Gamma}{\gamma_L \left[ \Gamma^2 + \chi^2 \left[ 1 - \dfrac{1}{2} \dfrac{\gamma_{12}}{\gamma_L} \right] \right]} \right]^{1/2} \tag{62}$$

and is seen to depend on the ratio $\gamma_{12}/\gamma_L$.

The contour integral of (50) was carried out in the limit $\langle \epsilon^2 \rangle / \Gamma \gamma_{12} \ll 1$ [Eq. (51)]. This restriction can be removed if need be. For example, if $\langle \epsilon^2 \rangle / \Gamma \gamma_{12} \gg 1$ and $\langle \epsilon^2 \rangle / \Gamma \gamma_L \gg [(\chi^2 + \Gamma^2)/\chi\Gamma]^2$, then the poles in $\Delta$ in (47) in the upper-half plane occur at

$$\Delta = \frac{1}{\sqrt{2}} (\pm 1 + i) \left[ \frac{\langle \epsilon^2 \rangle \Gamma}{2\gamma_L} \right]^{1/4} \chi^{1/2} \,. \tag{63}$$

These poles would lead to an FID signal that decays with rate

$$\gamma_F \sim \frac{1}{\sqrt{2}} \left[ \frac{\langle \epsilon^2 \rangle \Gamma}{2\gamma_L} \right]^{1/4} \chi^{1/2} \tag{64}$$

and is modulated at a frequency equal to $\gamma_F$.[19] If $\langle \epsilon^2 \rangle / \Gamma \gamma_{12} \gg 1$, $\chi \gg \Gamma$ and $\langle \epsilon^2 \rangle / \Gamma \gamma_L \ll \chi^2 / \Gamma^2$, then $\gamma_F \sim \chi (\gamma_{12}/2\gamma_L)^{1/2}$. For intermediate values of $\langle \epsilon^2 \rangle / \Gamma \gamma_{12}$, there are still two poles in the lower-half plane and these will lead to an FID signal that is characterized by two decay constants.[5,8]

### B. RBE: $\epsilon_0 \gg \Gamma, \chi, \gamma_{12}$

The FID signal follows directly from Eqs. (42) and (43). In practice, the solution is obtained from (52)—(54) by setting $\langle \epsilon^2 \rangle = 0$ and replacing all $\gamma$'s by the corresponding $\gamma^t$'s, Eqs. (38) and (39), in these equations. In this way, one finds[20]

$$Q_3(t) = -\tfrac{1}{2} i \pi \chi N_{21}(0) \left[ 1 - \frac{\gamma_{12}^t}{\gamma_B^t} \right] \epsilon^{-\gamma_F' t} \,, \tag{65}$$

where

$$\gamma_F' = \gamma_{12} + \Gamma + \gamma_B^t \tag{66}$$

and

$$(\gamma_B^t)^2 = (\gamma_{12} + \Gamma)^2 + \tfrac{1}{2}\chi^2 \frac{\gamma_{12}^t}{\gamma_L^t} \,. \tag{67}$$

In weak fields,

$$\gamma_F' \sim 2(\gamma_{12} + \Gamma) = 2\gamma_{12}^t \,, \tag{68}$$

while in intense fields,

$$\gamma_F \sim \frac{\chi}{\sqrt{2}} \left[ \frac{\gamma_{12}^t}{\gamma_L^t} \right]^{1/2} . \tag{69}$$

The formal structure of the solution is very similar to that obtained in the $\epsilon_0 \ll \Gamma$ limit if the correspondence $(\langle \epsilon^2 \rangle / \Gamma) \leftrightarrow \Gamma$ is made. Significant differences between the models emerge when one observes the dependence of $\gamma_F$ and $\gamma_F'$ on $\Gamma$. Thus according to (66), for the case $\epsilon_0 \gg \Gamma$ and weak fields, $\gamma_F$ grows with $\Gamma$, whereas according to (46) for the case $\epsilon_0 \ll \Gamma$, $\gamma_F$ decreases with $\Gamma$, as in motional narrowing.[10]

Our result (65) for the strong-redistribution model in the limit $\epsilon_0 \gg \Gamma, \chi, \gamma_{12}$ differs from that found using a Gaussian-Markovian model[5,6] for frequency fluctuations. One might expect that, in the Gaussian-Markovian limit with $\epsilon_0 \gg \Gamma$, the effects of frequency fluctuations would be minimal since the frequency fluctuations are not strong enough to remove the ionic frequency from the "hole" created by the ion-field interaction (in contrast to the strong-redistribution model). Hanamuru[5] found that the FID decay rate was, indeed, independent of the rate $\Gamma$; however, he found an FID signal linear in $\chi$ (for small $\chi$) that decayed as $\exp(-\langle \epsilon^2 \rangle t^2)$. From some very general considerations (see Sec. IV C), one can rule out a linear

FID signal, so this result is somewhat surprising and may have resulted from an error in integrating over $\Delta$.[21] Javanainen[6] found no evidence for a linear FID signal. As noted earlier, the validity of a given model must be determined by comparison with experiment.

### C. MBE: General solution

We now present a formal solution to the MBE (14a) which is valid for any ratio of $\Gamma/\epsilon_0$. For $t>0$, $\tilde{\rho}_{12}(\Delta,\epsilon,t)$ satisfies the equation

$$\frac{\partial\tilde{\rho}_{12}(\Delta,\epsilon,t)}{\partial t}=(-\mu_{12}^t+i\epsilon)\tilde{\rho}_{12}(\Delta,\epsilon,t)+\Gamma F(\epsilon)\int\tilde{\rho}_{12}(\Delta,\epsilon',t)d\epsilon' \ . \tag{70}$$

From this equation, it follows that

$$Q_3(\Delta,t)=\int\tilde{\rho}_{12}(\Delta,\epsilon,t)d\epsilon \tag{71}$$

satisfies the integral equation

$$Q_3(\Delta,t)=\int d\epsilon\, e^{(-\mu_{12}^t+i\epsilon)t}\tilde{\rho}_{12}(\Delta,\epsilon)+\int_0^t G(t-t')Q_3(\Delta,t')dt' \ , \tag{72}$$

where

$$G(\tau)=\Gamma\int d\epsilon\, F(\epsilon)e^{(-\mu_{12}^t+i\epsilon)\tau} \tag{73}$$

and $\tilde{\rho}_{12}(\Delta,\epsilon)$ is the steady-state value of $\tilde{\rho}_{12}$. Equation (72) is easily solved by Fourier-transform techniques enabling one to arrive at

$$Q_3(\Delta,t)=\frac{1}{2\pi}\int d\omega\frac{e^{-i\omega t}}{1-G(\omega)}\int d\epsilon\frac{\tilde{\rho}_{12}(\Delta,\epsilon)}{\mu_{12}^t-i(\epsilon+\omega)} \ , \tag{74}$$

where

$$G(\omega)=\Gamma\int d\epsilon\, F(\epsilon)[\mu_{12}^t-i(\epsilon+\omega)]^{-1} \ . \tag{75}$$

The quantity $\tilde{\rho}_{12}(\Delta,\epsilon)$ is obtained as the third component $[\tilde{\rho}_{12}=(\rho)_3]$ of the solution of the steady-state equation [see Eq. (14a)]

$$\underline{L}(\Delta,\epsilon)\rho(\Delta,\epsilon)-\Gamma F(\epsilon)\int\rho(\Delta,\epsilon')d\epsilon'=\Lambda(\Delta,\epsilon) \ , \tag{76}$$

where

$$\underline{L}(\Delta,\epsilon)=\underline{A}+\underline{\Gamma}-i\epsilon\underline{B} \ . \tag{77}$$

This equation may be solved to give

$$\rho(\Delta,\epsilon)=\underline{L}^{-1}(\Delta,\epsilon)\left[1-\Gamma\int\underline{L}^{-1}(\Delta,\epsilon')F(\epsilon')d\epsilon'\right]^{-1}\Lambda(\Delta,\epsilon) \ . \tag{78}$$

Equations (74) and (78), together with

$$I(t)=\mathrm{Im}[Q_3(t)] \ , \tag{79}$$

$$Q_3(t)=\int Q_3(\Delta,t)d\Delta \tag{80}$$

determine the FID signal.

No numerical evaluation of Eqs. (74)—(80) is given in this paper. Instead, some general features of the solution are discussed. If the variable $\omega$ in Eq. (74) is changed to $\omega'=\omega+\epsilon+\Delta+i\gamma_{12}$, Eqs. (74), (75), and (80) are transformed into

$$Q_3(t)=\int d\Delta\int d\epsilon\int d\omega\frac{e^{-(\gamma_{12}-i\Delta-i\epsilon-i\omega)t}\tilde{\rho}_{12}(\Delta,\epsilon)}{\left[1-\Gamma\int d\epsilon' F(\epsilon')[\Gamma-i(\omega+\epsilon-\epsilon')]^{-1}\right](\Gamma-i\omega)} \ . \tag{81}$$

When the contour integration is carried out in the upper-half plane, it is only the poles of $\omega$, $\Delta$, and $\epsilon$ in the upper-half plane which will contribute. Since $F(\epsilon)$ is Gaussian, the integral in the denominator of Eq. (81) can be related to the complex error function. A simple analysis then reveals that there are exactly two poles of $\omega$ in the upper-half plane, leading to two FID decay rates. One of these poles is always at $\omega=i\Gamma$ and the other varies from $\omega=i\langle\epsilon^2\rangle/\Gamma$ if $\epsilon_0\ll\Gamma$ to $i\epsilon_0\{\ln[\epsilon_0/(2\Gamma\sqrt{\pi})]\}^{1/2}$ for $\epsilon_0\gg\Gamma$. In the limiting cases discussed above, one of the terms is dominant; if $\epsilon_0\ll\Gamma$, it is the pole at $\omega\simeq i\langle\epsilon^2\rangle/\Gamma$ which provides the major contribution [see Eq. (46)], while for $\epsilon_0\gg\Gamma$, it is the pole at $\omega=i\Gamma$ [see Eq. (66)] which is dominant. For $\epsilon_0\approx\Gamma$ the two terms may be comparable. The integration over $\Delta$ can increase the number of decay constants if there are several poles in the upper-half plane. However, it is possible to use Eq. (78) to show that, in the linear-field regime, there are no $\Delta$ poles in the upper-half plane—i.e., there is no linear FID. The contribution from the $\epsilon$ integration has not been analyzed, in general.

### D. Qualitative discussion of FID signal

The decay constants observed in the FID signal can be understood in terms of the coherence time

$$\tau(\eta)=\eta^{-1} \tag{82}$$

associated with each of the relevant frequencies $\eta$ (e.g., $\eta=\chi,\gamma,\gamma_{12},\Gamma,\Delta$) in the problem. The trouble is that there are *so* many relevant frequencies which must be considered for *both* the transient $(t>0)$ and preparatory $(t<0)$ domains, that it is difficult to keep track of the various coherence times $\tau(\eta)$ without a score card. Nevertheless, we note the following features. (i) The large-scale inhomogeneous broadening gives rise to a *linear* FID signal that decays very rapidly with a rate $\Delta^*$. This component of the FID signal has been neglected in this work. (ii) *Neglecting the contributions of the local field variations,* one finds that the decay rate of the nonlinear contribution to the FID signal is determined by the two decay times $\tau(\gamma_{12})$ and $\tau(\chi^*)$ where

$$\chi^* = \chi(\gamma_{12}/\gamma_1)^{1/2} . \tag{83}$$

The quantity $\chi^*$ is the effective rate at which the field drives the optical coherence $\tilde{\rho}_{12}$, as may be seen by solving (14a) for $\rho_{11}$ and $\rho_{22}$, substituting the solution in the $\tilde{\rho}_{12}$ and $\tilde{\rho}_{21}$ equations, and noting the effective rate constant $\chi^*$. In weak fields, $\tau(\gamma_{12}) < \tau(\chi^*)$ and the FID signal decays with rate $\gamma_{12}$. In intense fields, $\tau(\chi^*) < \tau(\gamma_{12})$ and the signal decays with rate $\chi^*$. Note that if homogeneous broadening had been present such that $\gamma_{12} \gg \gamma_L$, the decay rate $\chi^* = \chi(\gamma_{12}/\gamma_L)^{1/2}$ would, in fact, be much greater than the value $\chi^* \simeq \chi$ obtained when $\gamma_{12}$ and $\gamma_L$ are comparable. (iii) The local-field variations modify the decay rate in a manner that depends critically on the phase buildup

$$\varphi_\epsilon = \int_0^\tau \epsilon(t)dt \tag{84}$$

at a particular atomic site [$\tau$ is some relevant coherence time and $\epsilon(t)$ is shown in Fig. 1]. If $\varphi_\epsilon \ll 1$, the local-field variations do not modify the FID decay, while for $\varphi_\epsilon \gtrsim 1$, they can be significant. In intense fields ($\chi^* \gg \epsilon_0$ and $\tau = \chi^{*-1}$), the phase $\varphi_\epsilon$ always is much less than unity and the effects of the local-field variations can be neglected. This is the reason why the high-field FID decay is relatively insensitive to the form of the local-field variations [see (59), (61), and (69)]. In weak fields, the value of $\varphi_\epsilon$ depends on the coherence time $\tau$. If $\epsilon_0 \ll \gamma_{12} < \Gamma$, then $\tau = \gamma_{12}^{-1}$ or $\Gamma^{-1}$ and $\varphi_\epsilon$ is always much less than unity implying the local-field variations do not contribute significantly to the FID signal decay. This result is seen in Eq. (56) where the FID signal decays with a rate approximately equal to $2\gamma_{12}$ if $\epsilon_0 \ll \gamma_{12} \ll \Gamma$. [Not included in (56) is a rapidly decaying component associated with $\tau(\Gamma)$ which decays at rate $\Gamma$.] If $\epsilon_0 > \gamma_{12}^t$, however, $\tau = (\gamma_{12}^t)^{-1}$, and the dominant contribution to the FID signal is associated with $\tau(\gamma_{12}^t)$ and decays with decay rate $2\gamma_{12}^t$ [see Eq. (68)]. [There is also a rapidly decaying component, associated with $\tau(\epsilon_0)$ that decays with a decay rate of order $\epsilon_0$ that is not included in (68).] Finally, if $\epsilon_0 \simeq \Gamma \simeq \gamma_{12}$, then two components in the FID should be seen, one which decays at rate $2\gamma_{12}^t$ and the other which decays with a rate of order $\epsilon_0$. Then results are in qualitative agreement with the discussion following (80).

## V. MODIFIED BLOCH EQUATIONS IN VAPORS

It is perhaps useful to relate the MBE derived in this paper (14a) with the corresponding MBE appropriate to an atomic or molecular vapor. Of course, the MBE used in this work actually represents a highly simplified *model* of relaxation in solids. In vapors, collisions between the "active" atoms of interest and a bath of perturber atoms is the relaxation mechanism. As long as the impact approximation is valid (inverse duration of a collision much greater than all relevant frequency scales in the problem), it is possible to obtain the macroscopic MBE starting with a *microscopic* theory of the collisional processes occurring in the vapor. In other words, the MBE in vapors result from a detailed quantum-mechanical theory rather than from an approximate model of the relaxation phenomena.

In neutral vapors, there is only *one* source of inhomogeneous broadening. The moving atoms see the radiation field at a frequency that is Doppler shifted by an amount $\mathbf{k}\cdot\mathbf{v}$ from the field frequency $\Omega$ ($\mathbf{k}$ is the radiation field propagation vector; $\mathbf{v}$ is the atomic velocity). The atoms are characterized by an equilibrium distribution

$$W_0(\mathbf{v}) = (\pi^{1/2}u)^{-3}e^{-v^2/u^2} , \tag{85}$$

where $u$ is the most probable speed of the active atoms. This velocity distribution is the source of the inhomogeneous broadening. Thus, the factor $\mathbf{k}\cdot\mathbf{v}$ is analogous to the frequency displacement $\epsilon$. "Velocity-changing collisions" are analogous to the jumps in the frequency parameter $\epsilon$ in solids. Models for the collision kernels $W_{ij}(\mathbf{v}'\to\mathbf{v})$ and rates $\Gamma_{ij}(v)$ $(i,j=1,2)$ appropriate to vapors are discussed in the literature.[9] In general, these kernels are not of the same form as that adopted for $W(\epsilon'\to\epsilon)$ in this work [the corresponding kernel $W(\mathbf{v}'\to\mathbf{v}) = W_0(\mathbf{v})$ is referred to as the "strong-collision" kernel], so that the quantitative details of the solutions for the vapor and the solid will differ. However, the overall qualitative structure of the solutions may be similar.

Since there is only one inhomogeneous width for vapors, it is possible to remove the "bath" variable $\mathbf{v}$ in vapors only if the collision rates are much greater than the Doppler width $ku$. (In the case of a solid, recall that one never removes the variable $\Delta$ from the equations.) Thus, formal reduction of the MBE to the RBE is possible only in this limit. However, the equations for a vapor can be greatly simplified in certain limits. Again there is a critical phase

$$\varphi_d = k\delta v(\tau)\tau$$

which enters. The quantity $\delta v(\tau)$ is the rms velocity change resulting from collisions acquired in the effective coherence time $\tau$ of the problem. If $\varphi_d \ll 1$, the velocity-changing aspects of collisions can be neglected and if $\varphi_d \geq 1$, the velocity-changing aspects of collisions can modify the line shapes.

As in the case of solids, the coherence time can be reduced by applying a strong radiation field. As such, the effects of velocity-changing collisions can be suppressed at strong-field strengths if $\varphi_d \ll 1$. This effect has been reported recently by Yodh and co-workers in a photon-echo experiment in Yb.[14] Thus, both solids and vapors can exhibit similar "deviations" from the conventional Bloch equations. Both solids and vapors can be described by MBE, but the theoretical basis for the MBE in vapors is much firmer than it is for solids.

## VI. CONCLUSIONS

In this paper we presented a rather simple model to describe the way in which local-field variations shift the transition frequency at atomic sites in a crystal. The frequency shift $\epsilon$ at any site undergoes jumps at some average rate $\Gamma$. The value of $\epsilon$ following any jump is independent of its value before the jump and is determined statistically by a Gaussian distribution having average value $\langle\epsilon\rangle = 0$ and an rms value $\langle\epsilon^2\rangle^{1/2} = \epsilon_0/\sqrt{2}$, where $\epsilon_0$ is the thermal width associated with the fluctuations. Using this strong-redistribution model, we derived a set of modified Bloch equations (MBE) that characterize atoms in a solid interacting with an external optical field and perturbed by local-field fluctuations. The atoms are also subject to a second inhomogeneous broadening due to crystalline strains. All effects of homogeneous broadening were neglected, but these could be included easily by a suitable modification of decay rates associated with the coherence.

Under certain limiting conditions ($\epsilon_0 \ll \Gamma$, $\epsilon_0 \gg \Gamma$) the MBE can be averaged over the bath variable $\epsilon$, enabling one to obtain reduced Bloch equations (RBE). When $\epsilon_0 \ll \Gamma$, our MBE are similar to those obtained by other authors.[2,4–8] However, our more general approach enables us to take the limit $\epsilon_0 \ll \Gamma$ in a systematic manner, allowing us to easily assess the accuracy of the RBE for this limiting case. For the $\epsilon_0 \ll \Gamma$ case, a contribution to the decay rate of $\langle \epsilon^2 \rangle / \Gamma$ is found for the free-induction decay signal and is a signature of theories in which $\epsilon_0 \ll \Gamma$, namely, the contribution to the decay rate decreases with increasing $\Gamma$. For the other limit, $\epsilon_0 \gg \Gamma, \chi, \gamma_{12}$, an additional decay rate $\Gamma$ appears in the FID signal. This type of dependence is typical of "strong" relaxation theories in which perturbations remove atoms from the frequency range in which they can effectively interact with the radiation field and differs from that predicted using Gaussian-Markovian[5,6] models of frequency fluctuations. Comparison with experiment could help decide on the validity of a particular model. We note that no attempt has been made to improve the theoretical fit of Refs. 2 and 4 to the DeVoe-Brewer experiments. Numerical methods would be required if in fact $\Gamma \simeq \epsilon_0$.

Although the modified Bloch equations were applied to a calculation of the FID signal, they could equally well be applied to a wide variety of steady-state and coherent transient problems. Moreover, one has the freedom to use "kernels" $W(\epsilon' \to \epsilon)$ [recall that $W(\epsilon' \to \epsilon)$ is the probability density per unit time that the frequency displacement changes from $\epsilon'$ to $\epsilon$] other than the one used in this work.[21]

The relaxation mechanism in solids and vapors was compared. It was shown that the frequency displacement $\epsilon$ is analogous to the Doppler shift $\mathbf{k}\cdot\mathbf{v}$ in vapors. While there are similarities between the equations characterizing the vapor and the solid, certain differences were noted. In particular, there is only one source of inhomogeneous broadening in the vapor as compared to two in the solid. In addition, the collision kernel $W(\mathbf{v}' \to \mathbf{v})$ appropriate to vapors invariably depends on the initial velocity $\mathbf{v}'$. A more detailed discussion of saturation behavior in vapors is planned for a future paper. Finally, we conclude from the arguments presented here that the saturation effects described constitute a general phenomenon, appropriate to impurity-ion crystals and gases alike. It is not restricted to impurity-ion crystals and will occur whenever frequency fluctuations appear.

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## APPENDIX A

Instead of using the vector $\boldsymbol{\rho}$, one may use a vector $\mathbf{M}$ having components

$$M_1 = u = \tilde{\rho}_{12} + \tilde{\rho}_{21} , \tag{A1a}$$

$$M_2 = v = i(\tilde{\rho}_{21} - \tilde{\rho}_{12}) , \tag{A1b}$$

$$M_3 = w = \rho_{22} - \rho_{11} , \tag{A1c}$$

$$M_4 = m = \rho_{11} + \rho_{22} . \tag{A1d}$$

The column vector $\mathbf{M}_B = (M_1, M_2, M_3)^T$ is conventionally referred to as the Bloch vector. In terms of $\mathbf{M}$, Eq. (1) of the text may be rewritten as

$$\frac{\partial \mathbf{M}}{\partial t} = -\underline{A}'\mathbf{M} + \mathbf{\Lambda}' , \tag{A2}$$

where

$$\underline{A}' = \begin{bmatrix} \gamma_{12} & \Delta & 0 & 0 \\ -\Delta & \gamma_{12} & -\chi & 0 \\ 0 & \chi & \frac{1}{2}(\gamma_1+\gamma_2+\gamma_2') & \frac{1}{2}(\gamma_2+\gamma_2'-\gamma_1) \\ 0 & 0 & \frac{1}{2}(\gamma_2-\gamma_2'-\gamma_1) & \frac{1}{2}(\gamma_1+\gamma_2-\gamma_2') \end{bmatrix} \tag{A3}$$

and

$$\mathbf{\Lambda}' = \begin{bmatrix} 0 \\ 0 \\ \Lambda_2 - \Lambda_1 \\ \Lambda_1 + \Lambda_2 \end{bmatrix} . \tag{A4}$$

Note that, in general, Eq. (A2) cannot be reduced to an independent equation for the conventional Bloch vector $\mathbf{M}_B = (M_1, M_2, M_3)^T$. This reduction *is* possible if either (i) $\gamma_2 - \gamma_1 = 0$ or (ii) $\Lambda_1 \sim 0$, $\gamma_1 \sim 0$, $\Lambda_2 = 0$, $\gamma_2' = \gamma_2$, which implies $m = \text{const} \equiv m_0$. For case (i), the Bloch equations are

$$\frac{\partial \mathbf{M}_B}{\partial t} = -\begin{bmatrix} \frac{1}{T_2} & \Delta & 0 \\ -\Delta & \frac{1}{T_2} & -\chi \\ 0 & \chi & \frac{1}{T_1} \end{bmatrix} \mathbf{M}_B + \begin{bmatrix} 0 \\ 0 \\ \Lambda_2 - \Lambda_1 \end{bmatrix} \tag{A5}$$

with

$$\frac{1}{T_2} = \gamma_{12}, \quad \frac{1}{T_1} = \gamma_1 = \gamma_2 . \tag{A6}$$

For case (ii) the Bloch equations are

$$\frac{\partial \mathbf{M}_B}{\partial t} = -\begin{bmatrix} \frac{1}{T_2} & \Delta & 0 \\ -\Delta & \frac{1}{T_2} & -\chi \\ 0 & \chi & \frac{1}{T_1} \end{bmatrix} \mathbf{M}_B + \begin{bmatrix} 0 \\ 0 \\ -\frac{m_0}{T_1} \end{bmatrix} \quad \text{(A7)}$$

with

$$\frac{1}{T_2} = \gamma_{12}, \quad \frac{1}{T_1} = \gamma_2 . \quad \text{(A8)}$$

In terms of $\mathbf{M}$, Eq. (14a) becomes

$$\frac{\partial \mathbf{M}(\Delta,\epsilon,t)}{\partial t} = -\underline{A}'\mathbf{M}(\Delta,\epsilon,t) + \epsilon\underline{B}'\mathbf{M}(\Delta,\epsilon,t) - \underline{\Gamma}\mathbf{M}(\Delta,\epsilon,t) + \Gamma F(\epsilon)\int \mathbf{M}(\Delta,\epsilon',t)d\epsilon' + \mathbf{\Lambda}'(\Delta,\epsilon) \quad \text{(A9)}$$

with

$$\mathbf{\Lambda}'(\Delta,\epsilon) = F(\epsilon)g(\Delta)\begin{bmatrix} 0 \\ 0 \\ \lambda_2-\lambda_1 \\ \lambda_1+\lambda_2 \end{bmatrix}, \quad \underline{B}' = \begin{bmatrix} 0 & -1 & 0 & 0 \\ 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{bmatrix}, \quad \text{(A10)}$$

and Eqs. (25) and (27) become

$$\frac{\partial \mathbf{Q}'(\Delta,t)}{\partial t} = -[\underline{A}' + \underline{R}'(t-t_0)]\mathbf{Q}'(\Delta,t) + [\underline{1} - \underline{S}'(t-t_0)]\mathbf{\Lambda}'(\Delta,\epsilon) + i\langle\epsilon^2\rangle\underline{B}'e^{-(\underline{A}'+\underline{\Gamma})(t-t_0)}\boldsymbol{\psi}'^{(1)}(\Delta,t_0) , \quad \text{(A11)}$$

$$\frac{\partial \mathbf{Q}'(\Delta,t)}{\partial t} = -(\underline{A}' + \underline{R}')\mathbf{Q}(\Delta,t) + \mathbf{\Lambda}'(\Delta) , \quad \text{(A12)}$$

where

$$\mathbf{Q}'(\Delta,t) = \int \mathbf{M}(\Delta,\epsilon,t)d\epsilon , \quad \text{(A13)}$$

$$\underline{R}' = \underline{T}^{-1}\underline{R}\underline{T}, \quad \underline{S}' = \underline{T}^{-1}\underline{S}\underline{T} , \quad \text{(A14)}$$

$$\boldsymbol{\psi}'(1) = \underline{T}^{-1}\boldsymbol{\psi}^{(1)} , \quad \text{(A15)}$$

and

$$\underline{T} = \frac{1}{2}\begin{bmatrix} 0 & 0 & -1 & 1 \\ 0 & 0 & 1 & 1 \\ 1 & i & 0 & 0 \\ 1 & -i & 0 & 0 \end{bmatrix}, \quad \underline{T}^{-1} = \begin{bmatrix} 0 & 0 & 1 & 1 \\ 0 & 0 & -i & i \\ -1 & 1 & 0 & 0 \\ 1 & 1 & 0 & 0 \end{bmatrix} . \quad \text{(A16)}$$

Similarly, Eq. (33) becomes

$$\frac{\partial \underline{Q}'(\Delta)}{\partial t} = 0 = -(\underline{A}' + \underline{H}')\mathbf{Q}'(\Delta) + \mathbf{\Lambda}'(\Delta) , \quad \text{(A17)}$$

where

$$\underline{H}' = \underline{T}^{-1}\underline{H}\underline{T} = \begin{bmatrix} H_1' & H_2' & 0 & 0 \\ -H_2' & H_3' & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{bmatrix}, \quad \text{(A18)}$$

$$H_1' = \mathrm{Re}(H_1) + H_2 , \quad \text{(A19a)}$$

$$H_2' = -\mathrm{Im}(H_1) , \quad \text{(A19b)}$$

$$H_3' = \mathrm{Re}(H_1) - H_2 , \quad \text{(A19c)}$$

and

$$\mathbf{\Lambda}'(\Delta) = \mathbf{\Lambda}'(\Delta,\epsilon)/F(\epsilon) , \quad \text{(A20)}$$

and Eq. (42d) becomes

$$\frac{[\delta\mathbf{Q}'(\Delta,t)]}{\partial t} = -(\underline{A}' + \underline{\Gamma})\delta\mathbf{Q}'(\Delta,t) + \delta\mathbf{\Lambda}'(\Delta) , \quad \text{(A21)}$$

where

$$\delta\mathbf{Q}'(\Delta,t) = \underline{T}^{-1}\delta\mathbf{Q}(\Delta,t); \quad \delta\mathbf{\Lambda}'(\Delta) = \underline{T}^{-1}\delta\mathbf{\Lambda}(\Delta) . \quad \text{(A22)}$$

## APPENDIX B

This is a comparison of Eq. (28) with Schenzle *et al.*[2] and Yamanoi and Eberly.[4] Yamanoi and Eberly[4] derive MBE of the form of Eq. (A12) for the case of a closed system represented in Eqs. (A7) and (A8) with

$$\frac{1}{T_2} = \frac{1}{2T_1} \equiv \gamma . \quad \text{(B1)}$$

Explicit evaluation of Eq. (A14) for $\underline{R}'$ using Eq. (29) then allows one to write the first three components $\mathbf{Q}_B = (Q_1', Q_2' Q_3')^T = (Q_u, Q_v, Q_w)^T$ of Eq. (A12) as

$$\frac{\partial \mathbf{Q}_B}{\partial t} = -\begin{bmatrix} \gamma+\Gamma_{11} & \Delta & \Gamma_{13} \\ -\Delta & \gamma+\Gamma_{22} & -\Omega+\Gamma_{23} \\ 0 & \Omega & 2\gamma \end{bmatrix}\mathbf{Q}_B + \begin{bmatrix} 0 \\ 0 \\ 2\gamma w_{\text{eq}} \end{bmatrix}, \quad \text{(B2)}$$

where the following change of notation has been made:

$$\Omega = \chi , \quad \text{(B3a)}$$

$$\Gamma_{11} = \Gamma_{22} = R_1 = \frac{(\delta\omega)^2\tau_c}{(\Omega')^2}\left[\Delta^2 + \frac{\Omega^2}{1+(\Omega')^2\tau_c^2}\right], \quad \text{(B3b)}$$

$$\Gamma_{13} = -2\,\mathrm{Re}(R_2) = \frac{(\delta\omega)^2\tau_c(\Delta\Omega)\tau_c^2}{1+(\Omega')^2\tau_c^2} , \quad \text{(B3c)}$$

$$\Gamma_{23} = -2\,\mathrm{Im}(R_2) = -\frac{(\delta\omega)^2\tau_c^2\Omega}{1+(\Omega')^2\tau_c^2} , \quad \text{(B3d)}$$

$$\tau_c = 1/\Gamma , \quad \text{(B3e)}$$

$$(\delta\omega)^2 = \langle\epsilon^2\rangle , \quad \text{(B3f)}$$

$$\Omega' = (\Delta^2+\chi^2)^{1/2} \rightarrow (\Delta^2+\Omega^2)^{1/2} . \quad \text{(B3g)}$$

Equation (B2) agrees identically with the result of Yamanoi and Eberly.[4]

Schenzle *et al.*[2] derive MBE of the form of Eq. (A12) for an open system represented by Eqs. (A5) and (A6) with

$$\frac{1}{T_2}=\gamma , \tag{B4a}$$

$$\Lambda_2-\Lambda_1=w_0/T_1 . \tag{B4b}$$

Making the change of variables

$$\Gamma\rightarrow\Sigma , \tag{B5a}$$

$$\langle\epsilon^2\rangle\rightarrow 4\pi N^2/\sigma^2 , \tag{B5b}$$

one finds that Eq. (A14) may be written

$$\dot{Q}_u=-\Delta Q_v-\frac{4\pi N^2}{\sigma\beta^2}\left[\frac{\Delta^2}{\sigma^2}+\frac{\chi^2}{\beta^2+\sigma^2}\right]Q_u - \frac{4\pi N^2}{\sigma}\frac{\Delta\chi}{\beta^2+\sigma^2}Q_w-\gamma Q_u , \tag{B6a}$$

$$\dot{Q}_v=\Delta Q_u+\chi w-\frac{4\pi N^2}{\sigma\beta^2}\left[\frac{\Delta^2}{\sigma^2}+\frac{\chi^2}{\beta^2+\sigma^2}\right]Q_v-\gamma Q_v + \frac{4\pi N^2}{\sigma}\frac{\chi\sigma}{\beta^2+\sigma^2}Q_w , \tag{B6b}$$

$$\dot{Q}_w=-\chi Q_v-(Q_w-w_0)/T_1 \tag{B6c}$$

which is the result of Schenzle *et al.*,[2] except that they do *not* have the last term in Eq. (B6b).


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