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Coherence in multilevel systems. II. Description of a multilevel system as two levels in contact with a population reservoir

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The effects of incoherent feeding and decay on a coherently driven two-level system are investigated using density-matrix techniques. A closed-form solution that includes the effects of relaxation and spontaneous emission between the two levels is obtained for cases in which the sample size is both small and large compared to the wavelength of the coherent field. The formalism developed herein is somewhat more general than the original approach of Bloch, Bloembergen, Purcell, Pound, and Redfield, insofar as it is applicable to cases where the trace of density matrix is not constant in time. As such, several new phenomena are predicted. Specifically, the steady-state solutions reveal that under appropriate conditions a significant off-resonance coherent component can be maintained indefinitely by the combination of driving field and incoherent feeding. We term this effect "kinetic coherence" and develop its formalism for both optical and radio-frequency fields.

I. INTRODUCTION

The time evolution of an ensemble of two-state systems under the influence of a coherent radiation field is a problem of considerable importance and has been treated extensively for the case in which the ensemble is always composed of the same members. By this we mean that the identity of the individual members and the number of members in the ensemble is constant in time. Proton nuclear magnetic resonance is an example of this, where for a given sample size the number of protons is fixed, and the individual proton maintains its identity throughout an experiment.[1] In many cases, such as optical pumping in gases[2] and solids,[3,4] in some double resonance experiments,[5] in chemically induced nuclear and electron-spin polarization,[6,7] Stark-shift optical coherence,[8] and any process which involves excited states, the total population of the two-level ensemble is not constant in time. Although most of the processes mentioned above have been treated within their own context, little emphasis has been placed on how the creation and destruction of the states affect the properties of the ensemble in the presence of a coherent driving field.

It is the aim of this paper to treat an idealized two-level system which is coherently coupled and at the same time is being *incoherently* fed and is spontaneously decaying. In such a case, the number and identity of the individual members of the ensemble does not remain constant in time since the ensemble of excited states will not only evolve under the influence of the coherent radiation field, but will also decay to the ground state by radiative and nonradiative processes. In these cases the trace of the density matrix describing the ensemble is not constant in time, but rather decays from an initial $t=0$ value to zero. In addition, if the excitation source responsible for producing the excited states is left on after $t=0$, new excited states are continually created and also evolve under the influence of the coherent radiation field. The collection of excited-state two-level systems is therefore not a time-independent collection, but rather an ensemble which constantly has members feeding into and decaying from it.

Traditional treatments[9-11] that deal with coherent coupling experiments are inadequate for this purpose since they do not include the effects of feeding and decay. The theory and model system developed in this paper was specifically designed to incorporate these considerations for a proper description of zero-field excited-triplet-state coherence experiments,[12] and represents an extension of the basic theory presented in the first part of this series.[13] It must be emphasized, however, that the treatment is applicable to a broad range of phenomena owing to the general nature of the Hamiltonian that describes the time evolution of a two-level system. In some respects this problem has been considered by studies on lasers and laser amplifiers,[14-22] the paper by Icsevgi and Lamb[21] perhaps coming closest to this work. However, we wish to focus attention on the qualitative and quantitative effects of feeding and decay on a coherently driven system and we are not interested, at least at first, in the coherent field produced by the medium which is the primary concern of laser physicists. By considering "thin samples," the coupled Maxwell and Schrödinger equations may be avoided, and this allows exact

solutions to be obtained for both steady-state and transient response in the presence of feeding and decay for all strengths of the applied coherent driving field, and also in the presence of phenomenological relaxation terms.

In addition to providing a basis for understanding the modifications which occur when coherent coupling experiments are performed on systems undergoing feeding and decay, the mathematical formulation developed in the body of the paper suggests that an interesting and potentially important effect will be observed. This effect is related to the production of steady-state coherence in the ensemble of excited-state systems that results directly from a combination of incoherent feeding and the applied driving field. Drawing from magnetic-resonance analogies one concludes that, under certain conditions of relaxation parameters, the steady-state coherent component that is produced by the medium may be several orders of magnitude larger if the coherent driving field is applied off-resonance, and application of this phenomenon to laser amplifiers might prove fruitful. We will term this effect "kinetic coherence" insofar as its magnitude is directly related to the kinetic parameters associated with the creation of the ensemble of excited states.

If the transition involved in producing the kinetic coherent state is an electric dipole transition in the optical region, the coherent component is a precessing macroscopic electric dipole and is "superradiant"[22-27] in the same sense as the photon echo,[26] resulting in the emission of coherent radiation from what amounts to a linear combination of Dicke states.

The mathematical development is illustrated with three examples: the transient nutation,[28] spin locking,[11,29] and a discussion of the steady-state solutions in order to demonstrate the physical roles that feeding and decay play in the coherent coupling problem. Initially the problem is considered for the case in which the wavelength of the radiation field is large relative to the size of the sample. The equation of motion of the density matrix includes the Hamiltonian, a decay term which is analogous to adding decay to the Schrödinger equation, and a feeding term which is arranged to affect only the diagonal elements of the density matrix. T_1 and T_2 processes are not included at this point. The equation of motion for the density matrix, neglecting T_1 and T_2 processes, is solved exactly using matrix algebra techniques. The problem is then rewritten to include T_1, T_2, and T_{2e} processes.[11] It is shown that the qualitative results obtained in the absence of T_1 and T_2 are correct, provided that high-power applied fields are used. Finally, the case in which the wavelength of the radiation field is small relative to the size of the sample (the optical case) is considered. It is shown that except for the usual directional properties associated with coherent optical problems, the results obtained for the long-wavelength case also apply to the optical wavelength region, when the local coupling of dipoles is neglected.[30]

II. DISCUSSION

In order to discuss the role that feeding and decay play in the excited-state two-level coherent coupling problem, we must have a well-defined model for these processes. For this purpose, the entire experimental system is divided into two parts. The first part consists of the ensemble of excited two-level systems which are coupled by the field. The second part is taken to be an infinite reservoir that represents both a source and a sink for population to enter and leave the ensemble of two-level systems.

At a given instant of time, the ensemble of two-level systems is evolving under the influence of the applied radiation field; it is also decaying into the reservoir at a rate which is characteristic of the lifetimes of the two excited states. Population is also constantly being transferred from the reservoir into the ensemble. We assume that only the states which are affected by the radiation field are included in the ensemble, and that the reservoir is taken to be unaffected by the field. As a consequence, population which is transferred incoherently from the reservoir to the ensemble enters the ensemble in *one* of its two eigenstates, and not in a coherent superposition state; however, once the population has entered the ensemble it may evolve into a coherent superposition state, since it is now influenced by the radiation field. Later we will consider feeding into a coherent superposition of the two-level system.[31]

In terms of a density-matrix description of the ensemble, this implies that feeding only occurs to the diagonal elements of the density matrix. Off-diagonal elements occur only because of the effect of the radiation field on the population which is already in the ensemble. However, decay affects both the diagonal and off-diagonal elements. Since the reservoir is infinite in extent and unaffected by the two-level ensemble, the populating rate into the two eigenstates of the ensemble is taken to be constant. Thus, the model for feeding and decay processes contains the following features: (a) Feeding only occurs to the eigenstates of the ensemble of excited two-level systems and not to coherent superposition states; (b) the rates for feeding into the two eigenstates are constants,

and are independent of the state of the ensemble; (c) decay occurs from both the eigenstates and the superposition states of the ensemble; and (d) the rate of decay from the ensemble will depend on the state of the ensemble, and therefore the total population of the ensemble need not be constant in time.

A qualitative picture describing the examples to be discussed in Sec. III may be made in terms of a geometrical representation for the two-level system. The initial population difference between the two levels is represented by a vector that is aligned along the r_3 direction of the r space of the well-known Feynman, Vernon, and Hellwarth (FVH)[32] model. If a coherent radiation field with frequency equal to the frequency separation of the two levels is turned on, the vector, viewed in a reference frame rotating at the frequency of the applied field, will begin to precess about the field, resulting in a transient nutation.[28,33,34] In an idealized case in which there are no T_1 or T_2 relaxation processes and also where the composition of the ensemble remains constant in time, the vector will continue to precess about the applied field indefinitely. However, if we are dealing with an ensemble of excited states, the population vector, which began to precess when the radiation field was turned on, will decay with the lifetimes associated with the excited states. Further, population which enters the ensemble of excited states at times after the radiation field has been turned on will also precess about the field. This feeding and decay process can be viewed in the geometrical model as vectors which suddenly appear along r_3, immediately start precessing about the effective field, and shrink in length as they precess. These vectors have a different phase than the initial population vector, and have a random phase relationship among themselves. In the NMR problem one need only follow the precession of a single vector, whereas in the excited-state problem one must follow the precession of the initial vector, which is decaying in magnitude at a rate dependent upon its location in r space, in addition to following the precession of the entering vectors which also are decaying.

Another important experimental situation which demonstrates the necessity of including feeding and decay processes in the analysis of the experiments is spin locking.[11,29] In an NMR experiment where there is no feeding or decay, the initial population difference vector is made to precess about the applied radiation field, as in the transient nutation experiment discussed above. After it has precessed 90°, the applied radiation field is turned off. If nothing else were done at this point, the vector, which is now in the plane normal to the direction it was initially pointing, would rapidly vanish because of fanning in the rotating frame caused by the inhomogeneous nature of the line undergoing the transition. However, the field is immediately phase shifted and is reapplied along the same direction that the population vector is pointing in the rotating frame. The vector finds itself aligned along the rotating-frame static field, and the fanning does not occur. In such a case, the population is said to be spin locked in a superposition state. The vector will remain spin locked for a time corresponding to the $T_{1\rho}$ time in the rotating frame.[11,29]

If the analogous experiment is performed on the magnetic spin sublevels of an excited molecular triplet state, the initial spin-locked vector would vanish owing to both $T_{1\rho}$ processes and radiative and nonradiative relaxation to the ground state; however, new population would continually enter the ensemble of triplet states that are coupled by the radiation field. This additional population enters into the eigenstates of the triplet spins and not into the spin-locked superposition state.[35] We therefore encounter the situation in which the population that existed at time $t=0$, which we will refer to as the $t=0$ subensemble, is spin locked, whereas the entering population is not. The entering vectors are nonetheless driven by the applied radiation field and execute transient nutations in the plane normal to the spin-locked vector.[35] In NMR one has to deal only with the single initial population vector, whereas in the excited-state problem, both the $t=0$ subensemble and the entering vectors must be considered, in addition to the loss of the spin-locked vector because of radiative or nonradiative decay of the excited spin-locked states to the ground state.

As we will show, the kinetic coherent state is produced by essentially spin locking a set of vectors along an off-resonance effective field. The initial population difference vector executes an off-resonance transient nutation about the effective-field direction, and describes a cone around the effective field as illustrated in Fig. 1. Owing either to field inhomogeneity or sample inhomogeneity, this initial vector will fan out around the conical path producing a thin cone of vectors precessing around the effective field. (Inhomogeneity is not required, since even with a homogeneous field and sample the feeding process itself will cause a cone of vectors.) The cone of vectors has a net projection along the effective-field direction which can be resolved into an r_3 component and an r_1 coherent component. As the initial population decays, additional population is fed into the system, continually forming a new cone with a collinear r_1 component; thus, as the

r_1 vector arising from the first cone decays, it is replaced by the r_1 vector from succeeding cones through continual feeding. The cone, and therefore the coherent r_1 vector, is constantly replenished, and in this fashion coherence is maintained for times far exceeding the lifetime of the excited-state ensemble. Notice that for an on-resonance driving field the cone collapses into a disk whose vector sum is zero.[35] In order to maintain a coherent component under an on-resonance driving field, the power of the driving field must be reduced until competing relaxation mechanisms produce a "lopsided" disk by virtue of the fact that the individual population vectors vanish before a single nutation revolution can occur in the rotating frame. From this point of view, a maser or laser is seen to self-regulate at "saturation," for if the medium produces a higher field, the disk will tend to become more uniform and reduce the size of the coherent component. For the off-resonance condition this situation does not exist, and it may be possible to utilize this feature as a means of overcoming power broadening.[36]

III. MATHEMATICAL DEVELOPMENT

We consider the situation depicted in Fig. 2 in which a two-level system characterized by the states $|y\rangle$ and $|x\rangle$ is populated from the reservoir at a constant rate, decays back into the reservoir, and is also coherently driven by a sinusoidally oscillating field. In order to isolate and examine the effects of feeding and decay, we shall first consider the simplest case in which the wavelength of the radiation is much greater than the sample size, i.e., $\lambda^3 \gg$ vol. We shall also, at first, neglect all relaxation processes such as T_1, homogeneous and inhomogeneous T_2, and driving-field inhomogeneities. These considerations complicate the development considerably but at the same time do not significantly alter many of the qualitative features of the problem, and are thus reserved for later sections of the discussion.

We shall use a semiclassical approach for the driving-field Hamiltonian. Without loss of generality we assume that the driving field has real matrix elements and express the Hamiltonian as

$$\mathcal{H}' = \mathcal{H}_0 + V(t) , \tag{1a}$$

$$\mathcal{H}_0 = \tfrac{1}{2}\hbar\omega_0\sigma_3 , \tag{1b}$$

$$V(t) = \hbar\omega_1\sigma_1 \cos\omega t . \tag{2}$$

$\mathcal{H}_0$ is the time-independent Hamiltonian with eigenstates $|y\rangle$ and $|x\rangle$, separated in energy by $\hbar\omega_0$, and σ_n are the Pauli spin matrices. Invoking the rotating-field approximation and by performing a suitable transformation that is equivalent to transforming into a rotating frame,[37] we obtain an equation of motion in which the Hamiltonian is time independent. Let

$$U = \exp(\tfrac{1}{2}i\sigma_3\omega t) \tag{3}$$

and

$$|t\rangle = U|t\rangle' . \tag{4}$$

From the Schrödinger equation,

$$i\hbar \frac{d}{dt}|t\rangle' = \mathcal{H}'|t\rangle' . \tag{5}$$

We substitute for $|t\rangle'$ in terms of $|t\rangle$ and obtain

$$i\hbar \frac{d}{dt}|t\rangle = \mathcal{H}|t\rangle , \tag{6}$$

where

$$\mathcal{H} = \tfrac{1}{2}\hbar(\omega_0 - \omega)\sigma_3 + \tfrac{1}{2}\hbar\omega_1\sigma_1 . \tag{7}$$

The Hamiltonian in Eq. (7) is time independent for any value of the driving-field frequency ω, and re-

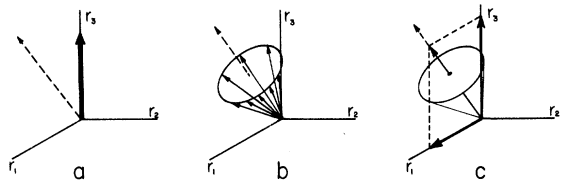

FIG. 1. (a) Initial population difference vector (heavy arrow) will precess about the effective field direction (dashed arrow in r_1-r_3 plane). (b) Feeding and dephasing processes produce a cone of vectors about the effective-field direction. (c) Vector sum of the cone yields a vector aligned along the effective-field direction and having a coherent component.

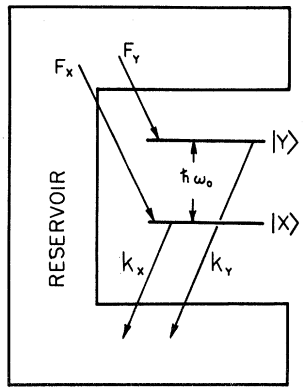

FIG. 2. Pictorial description of the model system presented in the discussion. F_y and F_x are constant feeding rates, whereas k_y and k_x are decay rate constants.

duces to the interaction representation[38] for $\omega = \omega_0$.

By considering a model system consisting of a reservoir, details of the feeding and decay processes are not considered explicitly and thus we allow the many-body problem to become tractable. The simplest way to include decay of a state is to assume that the amplitude for being in the state decays exponentially. For the two-level system, we have the state vector represented as a linear combination

$$|t\rangle = y|y\rangle + x|x\rangle \ , \tag{8}$$

where y and x are the usual time-dependent coefficients. We now let these amplitudes decay exponentially,

$$\dot{y} = -\tfrac{1}{2}k_y y \ , \tag{9a}$$

$$\dot{x} = -\tfrac{1}{2}k_x x \ ; \tag{9b}$$

k_y and k_x are physically observable rate constants associated with the decay of the states $|y\rangle$ and $|x\rangle$, respectively.

As discussed earlier, the populating process occurs only to the eigenstates $|y\rangle$ and $|x\rangle$ and cannot appear in a superposition state; thus, the equations describing the feeding process must deal only with the probabilities yy^* and xx^*, and they cannot affect the terms which define the relative phase factor as given by xy^* or yx^*. The result of this is that the feeding cannot be added to the amplitudes y and x but only to the probabilities.

At this point there are two possible ways to treat the problem. First, one could solve the coupled differential equations formed by combining Eqs. (6) and (9), take products of the solutions, and form integral equations that include the feeding process. Despite the fact that this method is exceedingly lengthy, it provides a certain amount of physical insight to the problem. The second approach is to use the density-matrix formulation which effectively deals with the coefficient products from the beginning. The solutions are much simpler from the computational point of view, and the development is mathematically less clumsy. We shall use this method in the following development.

A. Density-matrix solution

The equation of motion can be expressed as

$$i\hbar\dot{\rho} = [\mathcal{H}, \rho] - [K, \rho]_+ + F \ , \tag{10}$$

where $\mathcal{H}$ is the rotating-frame Hamiltonian, K is an imaginary decay matrix given explicitly in the y-x basis by

$$K = \frac{i\hbar}{2}\begin{bmatrix} k_y & 0 \\ 0 & k_x \end{bmatrix} \ , \tag{11}$$

and the symbol $[\ ]_+$ denotes anticommutation. The feeding matrix F is given by

$$F = i\hbar\begin{bmatrix} F_y & 0 \\ 0 & F_x \end{bmatrix} \ , \tag{12}$$

and affects only the diagonal elements of the density matrix.

Owing to the fact that the constant trace condition must be relaxed, one will need four, rather than the usual three, independent variables to completely describe the density matrix. This can be done easily by defining the components of the density matrix as follows:

$$\rho = \begin{bmatrix} r_y & \tfrac{1}{2}(r_1 - ir_2) \\ \tfrac{1}{2}(r_1 + ir_2) & r_x \end{bmatrix} \ . \tag{13}$$

These components have a geometrical significance which is only slightly different from the FVH model. The component r_y is represented by a vector which points "up" in a three-dimensional r space, whereas r_x points "down." They both share the same "in-plane" components r_1 and r_2. The FVH vector component r_3 is given by $r_y - r_x$. In terms of observables, r_y and r_x are proportional to the upper and lower level populations, respectively. The r_1 and r_2 components contain coherence information and are proportional to the expectation values of an induced or permanent electric or magnetic dipole, depending on the explicit form of $V(t)$.

Before solving Eq. (10) it is worthwhile to write it explicitly in terms of the r components:

$$\dot{r}_1 = -\Delta\omega r_2 - k_A r_1 \ , \tag{14a}$$

$$\dot{r}_2 = \Delta\omega r_1 - \omega_1(r_y - r_x) - k_A r_2 \ , \tag{14b}$$

$$\dot{r}_y = \tfrac{1}{2}\omega_1 r_2 - k_y r_y + F_y \ , \tag{14c}$$

$$\dot{r}_x = -\tfrac{1}{2}\omega_1 r_2 - k_x r_x + F_x \ , \tag{14d}$$

$$r_3 = r_y - r_x \ . \tag{15}$$

By comparing Eqs. (14a)–(14d) to the rotating-frame Bloch equations,[9] one can see immediately that k_A, the average of the decay rate constants k_x and k_y, will have the same effect as a T_2 process, and the combination of feeding and decay will appear to be a T_1 process. This is quite reasonable from a physical point of view since the in-plane components involve a superposition state which can be viewed as being "undecided" from which eigenstate it will eventually decay, thus giving rise to k_A. Also, an incoherent T_1 process will have a similar effect as decay from $|y\rangle$ or $|x\rangle$ into the reservoir with subsequent incoherent feeding into $|x\rangle$ and $|y\rangle$. The important differ-

ence between T_1 and feeding and decay, however, is that the final population difference in the levels is determined by a Boltzmann distribution in the T_1 case, as opposed to the feeding and decay process in which practically any polarization is possible, depending on the ratios of the feeding and decay constants and on the conditions of the experiment.

The solution of Eq. (10) is given by the matrix equation

$$\rho(t) = Q^{\dagger}(\rho(0) - \rho_s)Q + \rho_s \ , \tag{16}$$

where the evolution operator Q is defined by

$$Q = \exp\{i[(\mathcal{H} + K)/\hbar]t\} \ . \tag{17}$$

Notice that since $\mathcal{H}$ is real and K imaginary, the adjoint of Q is not the inverse, and Q is therefore not unitary. For this reason the operations indicated in Eq. (16) do not result in a similarity transformation as would be expected for a process in which the trace of the density matrix is not constant in time. An explicit form for Q is obtained from Eq. (17) using Putzer's method,[39]

$$Q = \exp(-\tfrac{1}{2}k_A t)\begin{bmatrix} \cos\tfrac{1}{2}\Omega t + [(k_D + i\Delta\omega)/\Omega]\sin\tfrac{1}{2}\Omega t & (i\omega_1/\Omega)\sin\tfrac{1}{2}\Omega t \\ (i\omega_1/\Omega)\sin\tfrac{1}{2}\Omega t & \cos\tfrac{1}{2}\Omega t - [(k_D + i\Delta\omega)/\Omega]\sin\tfrac{1}{2}\Omega t \end{bmatrix} . \tag{18}$$

Equation (18) incorporates the following definitions:

$$k_A = \tfrac{1}{2}(k_x + k_y) \ , \tag{19a}$$

$$k_D = \tfrac{1}{2}(k_x - k_y) \ , \tag{19b}$$

$$\Delta\omega = \omega_0 - \omega \ , \tag{19c}$$

$$\Omega = [\omega_1^2 + (\Delta\omega - ik_D)^2]^{1/2} \ . \tag{19d}$$

In view of the similarity between the rotating-frame Bloch equations[9] and Eqs. (14a)–(14d), we cast the *steady-state solution* of the density matrix ρ_s into forms resembling NMR expressions. First, noting that the steady-state populations in the absence of a driving field ($\omega_1 = \Delta\omega = 0$) are given by

$$r_y^0 = F_y/k_y \ , \tag{20a}$$

$$r_x^0 = F_x/k_x \ , \tag{20b}$$

and

$$r_3^0 = r_y^0 - r_x^0 \ , \tag{21}$$

and by defining *effective* relaxation terms

$$t_2 = 1/k_A \ , \tag{22a}$$

$$t_1 = k_A/k_x k_y \ , \tag{22b}$$

we obtain

$$r_1^s = \frac{r_3^0 \Delta\omega\omega_1 t_2^2}{1 + \Delta\omega^2 t_2^2 + \omega_1^2 t_2 t_1} \ , \tag{23a}$$

$$r_2^s = \frac{-r_3^0 \omega_1 t_2}{1 + \Delta\omega^2 t_2^2 + \omega_1^2 t_2 t_1} \ , \tag{23b}$$

$$r_y^s = \frac{r_y^0(1 + \Delta\omega^2 t_2^2) + \omega_1^2 t_1 t_2^2[\tfrac{1}{2}(F_x + F_y)]}{1 + \Delta\omega^2 t_2^2 + \omega_1^2 t_2 t_1} \ , \tag{23c}$$

$$r_x^s = \frac{r_x^0(1 + \Delta\omega^2 t_2^2) + \omega_1^2 t_2^2 t_1[\tfrac{1}{2}(F_x + F_y)]}{1 + \Delta\omega^2 t_2^2 + \omega_1^2 t_2 t_1} \ , \tag{23d}$$

and

$$r_3^s = \frac{r_3^0(1 + \Delta\omega^2 t_2^2)}{1 + \Delta\omega^2 t_2^2 + \omega_1^2 t_2 t_1} \ . \tag{24}$$

These are the familiar forms for continuous-wave spectra in magnetic resonance. When the "power factor" $\omega_1^2 t_2 t_1$ is small, the components reduce to Lorentzian line shapes. It is interesting that the effective "transverse" relaxation, t_2, is determined by the average of the decay *rate constants*, whereas the effective spin-lattice relaxation time t_1 is determined by the average of the decay *lifetimes*.

The expression for F in Eq. (12) could be easily generalized to situations in which the feeding occurs to a superposition state, such as when a triplet state is optically pumped in the presence of a high magnetic field, and could also be made time dependent. The solution for ρ_s follows a similar format.

If one wishes to monitor the effects of feeding and decay more explicitly, the density matrix may be broken up into two parts corresponding to the "zero-time" subensemble mentioned in the discussion and the "fed" subensemble:

$$\rho(t) = Q^{\dagger}\rho(0)Q + \rho_s - Q^{\dagger}\rho_s Q \ . \tag{25}$$

The first term corresponds to the zero-time subensemble. Since feeding into $|y\rangle$ and $|x\rangle$ are independent processes, one could separate the last two terms of Eq. (25) into y-fed and x-fed subensembles by setting $F_x = 0$ and $F_y = 0$, respectively. This might prove useful if one wishes to determine the effects on the system of feeding into the individual states.

B. Transient solutions: Special cases

The simple form for Eq. (16) might lead one to think that it would be worthwhile to multiply the matrices explicitly and thereby obtain analytical expressions for the r-vector components. Unfortunately, the solutions are complicated enough to mask the physics contained within them, so we shall restrict our attention to various special cases which give some insight into the effects of feeding and decay. First, we consider the trivial case of no driving field. Setting $\omega_1 = \Delta\omega = 0$, we have $\Omega = ik_D$, and Q has a very simple form:

$$Q = \exp(-\tfrac{1}{2}k_A t)\begin{bmatrix} \exp(\tfrac{1}{2}k_A t) & 0 \\ 0 & \exp(-\tfrac{1}{2}k_A t) \end{bmatrix}$$

$$= \exp(-Kt/i\bar{h}) = Q^{\dagger} \ . \qquad (26)$$

The solutions are [from Eq. (16)]:

$$r_1 = r_1(0)e^{-k_A t} \ , \qquad (27a)$$

$$r_2 = r_2(0)e^{-k_A t} \ , \qquad (27b)$$

$$r_y = [r_y(0) - (F_y/k_y)]e^{-k_y t} + F_y/k_y \ , \qquad (27c)$$

$$r_x = [r_x(0) - (F_x/k_x)]e^{-k_x t} + F_x/k_x \ , \qquad (27d)$$

Notice that r_1 and r_2 are not fed, but merely decay from whatever initial values they had at the time $t = 0$. Equations (27a)–(27d) agree with simple rate equations that can be written by inspection from the two-level system pictured in Fig. 2.

We next consider an on-resonance transient nutation. In this case, $\Delta\omega = 0$, and $\Omega = (\omega_1^2 - k_D)^{1/2}$. We assume initial random phases, i.e., $r_1(0) = 0$ and $r_2(0) = 0$, and let the initial values of the diagonal elements be steady-state values without the coherent driving field: $r_y(0) = r_y^0$, $r_x(0) = r_x^0$. We obtain from Eqs. (16) and (23),

$$r_1 = 0 \ , \qquad (28a)$$

$$r_2 = (e^{-k_A t}/\Omega^2)[r_s^2(k_D^2 - \omega_1^2\cos\Omega t) - (r_3^0 - r_3^s)\Omega\omega_1\sin\Omega t - (r_y^0 - r_y^s + r_x^0 - r_x^s)\omega_1 k_D(1 - \cos\Omega t)] + r_2^s \ , \qquad (28b)$$

$$r_y = (e^{-k_A t}/\Omega^2)\{(r_y^0 - r_y^s)[\Omega\cos(\tfrac{1}{2}\Omega t) + k_D\sin(\tfrac{1}{2}\Omega t)]^2$$
$$+ (r_x^0 - r_x^s)\omega_1^2\sin^2(\tfrac{1}{2}\Omega t) - r_2^s\omega_1\sin(\tfrac{1}{2}\Omega t)[\Omega\cos(\tfrac{1}{2}\Omega t) + k_D\sin(\tfrac{1}{2}\Omega t)]\} + r_y^s \ , \qquad (28c)$$

$$r_x = (e^{-k_A t}/\Omega^2)\{(r_x^0 - r_x^s)[\Omega\cos(\tfrac{1}{2}\Omega t) - k_D\sin(\tfrac{1}{2}\Omega t)]^2 + (r_y^0 - r_y^s)\omega_1^2\sin^2(\tfrac{1}{2}\Omega t)$$
$$+ r_2^s\omega_1\sin(\tfrac{1}{2}\Omega t)[\Omega\cos(\tfrac{1}{2}\Omega t) - k_D\sin(\tfrac{1}{2}\Omega t)]\} + r_x^s \ . \qquad (28d)$$

Despite the formidable appearance of these equations, the qualitative features are simple, since they are analogous to a damped harmonic oscillator. If $\Omega \leq k_A$, the curves will be dominated by the exponential term and will be highly damped. When $k_D > \omega_1$, the system behaves much like an overdamped oscillator. Any shifts in frequency or phase when $\omega_1 \gtrsim k_D$ will be masked by the exponential terms. When $\Omega \geq k_A$, it is also necessarily true that $\omega_1 \geq k_D$ and the observable oscillations will have nutation frequencies close to ω_1. If we allow the driving field to become very large, i.e., $\omega_1^2/k_x k_y \gg 1$, Eqs. (28a)–(28d) reduce to much simpler forms which are easier to relate to the geometrical model:

$$r_1 = 0 \ , \qquad (29a)$$

$$r_2 \cong r_3^0 e^{-k_A t}\left(\frac{k_x k_y}{\omega_1 k_A}\cos\omega_1 t - \sin\omega_1 t\right) - \frac{r_3^0 k_x k_y}{\omega_1 k_A} \ , \qquad (29b)$$

$$r_y \cong \tfrac{1}{2}r_3^0 e^{-k_A t}\left(\frac{k_D}{k_A} + \cos\omega_1 t\right) + \frac{F_y + F_x}{k_x + k_y} \ , \qquad (29c)$$

$$r_x \cong \tfrac{1}{2}r_3^0 e^{-k_A t}\left(\frac{k_D}{k_A} - \cos\omega_1 t\right) + \frac{F_y + F_x}{k_x + k_y} \ . \qquad (29d)$$

Notice that all the expressions contain the initial population polarization or alignment, $r_3^0 = r_y^0 - r_x^0$. As expected from the geometrical model, the vectors precess only in the r_2-r_3 plane. After the transient terms have died away, the populations in the two levels are approximately equal and r_2 is very small. This is to be expected from the vector model, since the "disk" that is ultimately formed has a vector sum of zero. In the absence of feeding or decay, Eqs. (28a)–(28d) and (29a)–(29d) reduce to the standard nutation described by the torque equation in the rotating frame. As is the case in NMR, we see that the ability to do well-defined pulse rotations of the r vector depends upon the relationship between the applied field strength and the effective relaxation $k_A = 1/t_2$. For sufficiently high power, a $\frac{1}{2}\pi$ pulse ($\omega_1 t = \frac{1}{2}\pi$) gives from Eqs. (29a)–(29d),

$$r_2(\tfrac{1}{2}\pi) \cong -r_3^0 \ , \qquad (30a)$$

$$r_y(\tfrac{1}{2}\pi) \cong r_x(\tfrac{1}{2}\pi) \cong r_3^0\frac{k_D}{k_A} + \frac{F_y + F_x}{k_x + k_y} \ . \qquad (30b)$$

The effects of feeding and decay on a spin-locked superposition state can be investigated by using Eqs. (30a) and (30b) as initial conditions for a phase-shifted transient nutation. Shifting the phase by 90° is tantamount to setting $r_1(0) = -r_2(\frac{1}{2}\pi) = r_3^0$ and $r_2(0) = 0$. r_y and r_x are unaffected by the phase shift. From these initial conditions, the expression for the spin-locked component is [from Eq. (16)]

$$r_1 \cong r_1(0)e^{-k_A t} = r_3^0 e^{-k_A t} . \tag{31}$$

From the vector model, one would predict that feeding subsequent to establishing the spin-locked component would contribute only a disk in the r_2-r_3 plane and thus could not affect the spin-locked component. Equation (31) shows that the spin-locked signal is indeed independent of feeding, and decays with the average of the decay rate constants for the two levels.

C. Long-term or "kinetic" coherence

Equation (31) demonstrates that a coherent component can be made to last on the order of the lifetime of the levels. In this section we shall propose that *this time is by no means an upper limit, and in fact it should be possible to maintain a significant coherent component for long periods of time, limited only by the coherence time of the driving field.* In many ways this is similar to dynamic equilibrium in which the component parts of the long-term coherence state are continually feeding and decaying, but a steady-state value is reached. The coherence is maintained by the driving field and is not destroyed by incoherent feeding or decay.

The steady-state expressions in Eqs. (23a)–(23d) can be somewhat deceptive if one does not keep in mind the fact that the effective relaxation term t_1 was constructed only to show the analogy to T_1, and is not related to the actual thermalization of the two levels. The ratios of feeding and decay constants determine the initial polarization of the system, and the population difference can thus be highly non-Boltzmann. With this in mind we reexamine Eq. (23a). The r_1 component represents the "dispersion spectrum" or the real part of the susceptibility in the language of NMR, and reaches a maximum "off resonance." Owing to the fact that r_3^0 can be significantly larger than a Boltzmann distribution of population, the steady-state coherent component can be orders of magnitude larger than the thermally populated case. From the vector model, one would expect the condition $\Delta\omega = \omega_1$ to give a maximum in-plane component. The special form of Eq. (23a) suggests that the problem is identical in form to the one treated long ago by Bloch, when he calculated the maximum nuclear induction signal in an NMR experiment.[9] The off-resonance value which corresponds to a maximum value of r_1^s is

$$\Delta\omega_{\max} = (1/t_2)(1+\omega_1^2 t_1 t_2)^{1/2} , \tag{32}$$

giving a value for r_1^s of

$$r_1^s(\max) = \omega_1 t_2 r_3^0/2(1+\omega_1^2 t_1 t_2)^{1/2} , \tag{33}$$

and for sufficiently high power, i.e., $\omega_1^2 t_1 t_2 \gg 1$,

$$r_1^s(\max) \cong \tfrac{1}{2} r_3^0 (t_2/t_1)^{1/2} . \tag{34}$$

The maximum value for r_2^s, on resonance, is equal in magnitude to the high-power value for r_1^s given in Eq. (34):

$$r_2^s(\max) = -\tfrac{1}{2} r_3^0 (t_2/t_1)^{1/2} . \tag{35}$$

If the lifetimes of the two levels were equal, the long-term coherent component would be half the initial polarization and $\Delta\omega_{\max} \cong \omega_1$. Since r_1 in Eq. (34) is linearly dependent on r_3^0, the coherent component could be doubled by doubling the feeding rates, unless, of course, this results in a significant depletion of the "infinite" reservoir in which case the assumptions that lead to Eq. (34) are no longer valid.

Of course, the expected value for r_1^s in Eq. (34) is not realistic owing to omission of the effects of relaxation. These will be dealt with analytically at the end of Sec. III D. However, the similarity of Eq. (34) to the Bloch-equation solution allows one to speculate that if the field is strong enough to "overcome" relaxation effects, i.e., if one can observe a transient nutation, the long-term coherent component will be present and can approach the value given by Eq. (34).

D. Relaxation

At this point we shall investigate the effects of relaxation on the steady-state components of the r vector. These terms may be obtained in a reasonably simple analytical form if we restrict ourselves to Bloch-type[9] relaxation terms T_1 and T_2, and Redfield-type[11] T_{2e} processes. The transient solutions will be dealt with at the end of this section.

In terms of the r-vector components, the complete equations of motion that include driving field, feeding and decay, and relaxation are given by

$$\dot{r}_1 = -\Delta\omega r_2 - (k_A + 1/T_{2e}) r_1 , \tag{36a}$$

$$\dot{r}_2 = \Delta\omega r_1 - \omega_1 (r_y - r_x) - (k_A + 1/T_2) r_2 , \tag{36b}$$

$$\dot{r}_y = \tfrac{1}{2}\omega_1 r_2 - (k_y + 1/T_y) r_y + r_x/T_x + F_y , \tag{36c}$$

$$\dot{r}_x = -\tfrac{1}{2}\omega_1 r_2 - (k_x + 1/T_x) r_x + r_y/T_y + F_x . \tag{36d}$$

T_x and T_y are related to the probability per unit time for a transition from $|x\rangle$ to $|y\rangle$ and from $|y\rangle$ to $|x\rangle$, respectively. Notice that this form allows for spontaneous emission from $|y\rangle$ to $|x\rangle$ in addition to "spin-lattice relaxation." Specifically, we could break T_y into two terms,

$$1/T_y = 1/T_{ys} + 1/T_{1y} \ , \tag{37}$$

in which T_{ys} is related to spontaneous emission from $|y\rangle$ to $|x\rangle$ and T_{1y} is related to the normal thermal probability for a transition from $|y\rangle$ to $|x\rangle$. If spontaneous emission is negligible, as is the case in a rf region of the applied field, we have the normal spin-lattice relaxation (SLR) time encountered in NMR,

$$1/T_1 = 1/T_x + 1/T_{1y} \ . \tag{38}$$

T_x and T_{1y} are related by the Boltzmann factor,

$$T_{1y}/T_x = \exp(-\hbar\omega_0/kT) \ . \tag{39}$$

For high-power driving fields in solids, Redfield suggested[11] an alternative form for the Bloch equations, separating T_2 into T_{2e}, a transverse spin-lattice relaxation time applied to the driving-field direction, and T_2, the normal transverse relaxation time. This distinction becomes necessary, for example, in the spin-lock experiment in which the decay of the spin-locked vector is not an energy-conserving process, and thus cannot be due to "spin-spin" relaxation. The steady-state solutions are readily solved by setting the time derivatives equal to zero and solving for the components. As was done earlier, we choose to define relaxation terms T and τ such that the functional forms for the components can be recognized as being similar to the NMR expressions

$$r_1^s = \frac{r_3^0\omega_1\Delta\omega TT_e}{1+\Delta\omega^2T^2+\omega_1^2T\tau} \ , \tag{40a}$$

$$r_2^s = \frac{-r_3^0\omega_1 T}{1+\Delta\omega^2T^2+\omega_1^2T\tau} \ , \tag{40b}$$

$$r_y^s = \frac{r_y^0(1+\Delta\omega^2TT_e)+(\omega_1^2T\tau/k_A)[\frac{1}{2}(F_x+F_y)]}{1+\Delta\omega^2T^2+\omega_1^2T\tau} \ , \tag{40c}$$

$$r_x^s = \frac{r_x^0(1+\Delta\omega^2TT_e)+(\omega_1^2T\tau/k_A)[\frac{1}{2}(F_x+F_y)]}{1+\Delta\omega^2T^2+\omega_1^2T\tau} \ . \tag{40d}$$

We have used the definitions

$$1/T = k_A + 1/T_2 \ , \tag{41a}$$

$$1/T_e = k_A + 1/T_{2e} \ , \tag{41b}$$

$$\tau = \frac{k_A}{k_xk_y + k_y/T_x + k_x/T_y} \ , \tag{41c}$$

$$r_y^0 = \frac{F_y(k_x+1/T_x)+F_x(1/T_x)}{k_xk_y+k_y/T_x+k_x/T_y} \ , \tag{41d}$$

$$r_x^0 = \frac{F_x(k_y+1/T_y)+F_y(1/T_y)}{k_xk_y+k_y/T_x+k_x/T_y} \ , \tag{41e}$$

$$r_3^0 = r_y^0 - r_x^0 \ . \tag{41f}$$

Thus far only the homogeneous relaxation time T_2 has been considered. The inhomogeneous relaxation time T_2^*, can be included by assuming some line-shape distribution, usually Lorentzian or Gaussian, centered about some average Larmor frequency $\bar{\omega}_0$. Here we treat the case for a Lorentz distribution given by the normalized shape function

$$g(\omega_0) = \frac{T_2^*}{\pi}\frac{1}{1+(\omega_0-\bar{\omega}_0)^2T_2^{*2}} \ , \tag{42a}$$

$$\int_{-\infty}^{\infty} g(\omega_0)\,d\omega_0 = 1 \ . \tag{42b}$$

Integration of the coherent components over all Larmor frequencies ω_0, yields

$$\bar{r}_1^s = \frac{r_3^0\omega_1\Delta\bar{\omega}}{\Delta\bar{\omega}^2+\{1/T_2^*+[(1+\omega_1^2T\tau)/TT_e]^{1/2}\}^2} \ , \tag{43a}$$

$$\bar{r}_2^s = \frac{-r_3^0\omega_1(1/T_e+1/T_2^*\{[(T/T_e)/(1+\omega_1^2T\tau)]^{1/2}\})}{\Delta\bar{\omega}^2+\{1/T_2^*+[(1+\omega_1^2T\tau)/TT_e]^{1/2}\}^2} \ , \tag{43b}$$

where $\Delta\bar{\omega} \equiv \bar{\omega}_0 - \omega$. We are now in a position to look again at the long-term coherent components. The off-resonance value that corresponds to a maximum value of $\bar{r}_1^s$ is

$$\Delta\omega_{max} = 1/T_2^* + [(1+\omega_1^2T\tau)/TT_e]^{1/2} \tag{44}$$

yielding

$$\bar{r}_1^s(\max) = \frac{r_3^0\omega_1}{2\{1/T_2^*+[(1+\omega_1^2T\tau)/TT_e]^{1/2}\}} \ . \tag{45}$$

If one has sufficient driving-field strength to "exceed the linewidth," i.e., $\omega_1T_2^* \gg 1$, $\omega_1T \gg 1$, and $\omega_1^2T\tau \gg 1$, Eq. (45) reduces to an expression similar to Eq. (34):

$$\bar{r}_1^s(\max) \cong \tfrac{1}{2}r_3^0(T_e/\tau)^{1/2}; \tag{46}$$

under the influence of inhomogeneous broadening the maximum value of $\bar{r}_2^s$ on resonance is *not* equal to the high-power limit for $\bar{r}_1^s$ given in Eq. (46), in constrast to the cases treated in Eqs. (34) and (35) in which relaxation was neglected. To see this we rewrite Eq. (43b) for

on resonance, $\Delta\bar{\omega}=0$.

$$\bar{r}_2^s=\frac{-r_3^0\omega_1 T}{1+\omega_1^2 T\tau+(1/T_2^*)[(TT_e)(1+\omega_1^2 T\tau)]^{1/2}}. \quad (47)$$

Notice that any nonzero value for $1/T_2^*$ will reduce the size of $\bar{r}_2^s$. Figure 3 gives the ratio of the maximum values of $\bar{r}_2^s$ and $\bar{r}_1^s$ as a function of the parameter T/T_2^*. As is seen from Fig. 3, significant differences between the maximum values of $\bar{r}_1^s$ and $\bar{r}_2^s$ become observable when the inhomogeneous relaxation time is greater than or equal to the homogeneous relaxation time. This is what one would expect physically, since the high-power conditions required to obtain Eq. (46) imply that all isochromats in the line behave identically, whereas the low-power conditions required to obtain a maximum for $\bar{r}_2^s$ imply that each isochromat in the inhomogeneous line will have a different effective field direction and the vector sum over the isochromats will necessarily be smaller.

Some additional points can be made about Eq. (46). Bloch noted[9] that an excessively long T_1 could be troublesome if one attempted to observe the coherent component. With feeding and decay this is not a problem; in fact, one would like to have T_1 as long as possible, for then $\tau\cong t_1$, and the "recovery" of the system is due to feeding, decay, and T_2 processes.

In the absence of inhomogeneous relaxation the maximum values of the coherent components can still differ appreciably. It can easily be shown that the maximum value of r_2^s in the absence of inhomogeneous relaxation is given by

$$r_2^s(\max)=\tfrac{1}{2}r_2^0(T/\tau)^{1/2}. \quad (48)$$

The difference between this and Eq. (6) lies in the field-dependent transverse relaxation time T_{2e}, which may be orders of magnitude longer than T_2, again favoring the off-resonance method for establishing coherence.

In summary, the maximum steady-state coherent component that may be maintained is achieved by applying a high-power off-resonance driving field. The conditions favoring this method are insensitivity to inhomogeneous broadening and the field-dependent transverse relaxation time. Even under the most unfavorable conditions, the off-resonance method would yield a component that is equal to but never less than that which could be obtained by using an on-resonance driving field.

A solution for the transient behavior of the density matrix including relaxation is not simple from an operational point of view. One concise representation is in the form of the Liouville operator.[40-42] In this case we treat the elements of the 2×2 density matrix as being the components of a four-vector. The time-dependent density matrix is written as

$$i\hbar\dot{\rho}=\hbar L\rho, \quad (49)$$

where the Liouville operator is defined by a 4×4 matrix with elements,

$$\hbar L_{mn,m'n'}=\mathcal{H}_{mm'}\delta_{nn'}-\delta_{mm'}\mathcal{H}_{n'n}. \quad (50)$$

Relaxation and decay are easily incorporated into the L matrix, owing to the fact that terms which multiply only the off-diagonal or diagonal elements of ρ can be inserted by inspection whereas it is clumsy to perform this operation in the matrix representation of the density matrix. Explicitly, the L matrix corresponding to Eqs. (49) and (50) is given by

$$L=\begin{bmatrix} -i\left(k_y+\frac{1}{T_y}\right) & \frac{-\omega_1}{2} & \frac{\omega_1}{2} & \frac{i}{T_x} \\ \frac{-\omega_1}{2} & \Delta\omega-\frac{i}{2}\left[\left(\frac{1}{T_{2e}}+\frac{1}{T_2}\right)+2k_A\right] & -\frac{i}{2}\left(\frac{1}{T_{2e}}-\frac{1}{T_2}\right) & \frac{\omega_1}{2} \\ \frac{\omega_1}{2} & -\frac{i}{2}\left(\frac{1}{T_{2e}}-\frac{1}{T_2}\right) & -\Delta\omega-\frac{i}{2}\left[\left(\frac{1}{T_{2e}}+\frac{1}{T_2}\right)+2k_A\right] & \frac{-\omega_1}{2} \\ \frac{i}{T_y} & \frac{\omega_1}{2} & \frac{-\omega_1}{2} & -i\left(k_x+\frac{1}{T_x}\right) \end{bmatrix}, \quad (51)$$

and the equation of motion corresponding to Eq. (10) becomes

$$i\hbar\dot{\rho}=\hbar L\rho+i\hbar F. \quad (52)$$

F is a feeding vector which in this case has two nonzero elements, F_y and F_x. The solution to Eq. (52) is straightforward:

$$\rho(t)=e^{-iLt}[\rho(0)-\rho_s]+\rho_s. \quad (53)$$

The exponential operator can be calculated in matrix form using Putzer's method,[39] and a closed-form solution may be obtained. It might be noted

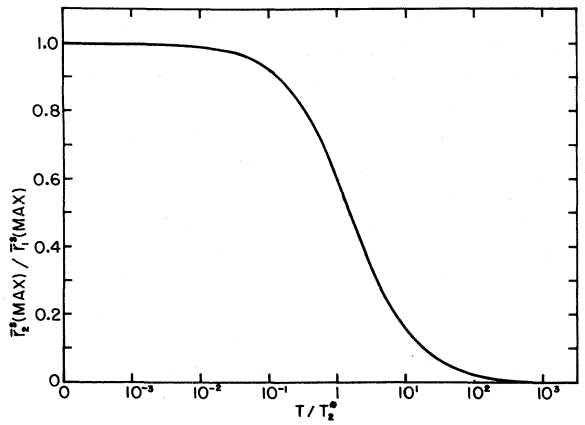

FIG. 3. Comparison of the two in-plane coherent components under the influence of inhomogeneous decay. The maximum value of the r_1 component is obtained under conditions of high-power applied field and is thus independent of inhomogeneous decay. The maximum value of r_2 is obtained for relatively low power and is strongly affected when the homogeneous and inhomogeneous relaxation times become comparable.

that the characteristic equation for L yields a quartic polynomial with real coefficients. A strict algorithm for calculating the eigenvalues and the resulting exponential matrix can be made, and one may avoid iterative methods that generally restrict calculations to time regions that lie relatively close to $t=0$.

E. Case in which $\lambda^3 \ll$ vol

When the wavelength of the driving field becomes much smaller than the sample size, the phase of the radiation field is no longer constant throughout the sample, and therefore the jth molecule within the sample which is at a position $\vec{r}_j$ will experience an interaction Hamiltonian $V_j(t)$ given by

$$V_j(t) = \hbar\omega_1 \cos(\omega t - \vec{k}\cdot\vec{r}_j), \tag{54}$$

where $\vec{k}$ is the wave vector of the radiation with frequency ω. For the same reasons that prompted a rotating-frame transformation, we may perform a suitable unitary transformation on the density matrix which will remove the explicit time and space dependence from the Hamiltonian. Defining

$$U_{k,j} = \exp[\tfrac{1}{2} i\sigma_3(\omega t - k\cdot r^j)], \tag{55}$$

we transform the laboratory-frame density matrix for the jth molecule to

$$\rho_j(t) = U_{k,j}\rho_j'(t)U_{k,j}^{-1}. \tag{56}$$

This leads to an equation of motion,

$$i\hbar\dot{\rho}_j = [\mathcal{H}, \rho_j], \tag{57}$$

where the Hamiltonian $\mathcal{H}$ is identical to the rotating-frame Hamiltonian in Eq. (7). If we assume that we have a sample of identical noninteracting systems, the form of $\mathcal{H}$, which is both time and space independent, renders the j index superfluous. The equation of motion is thus identical, and the development follows the same lines. The spatial transformation is not as trivial as the rotating-frame transformation and will depend strongly on the shape of the sample, how it is driven by the applied field, and how it is observed. This type of effect is well known theoretically and experimentally.

We have shown above that the position-dependent phase factor introduced into the Hamiltonian for the short-wavelength case in essence does not alter the development of the previous sections; in particular, in the absence of strong local dipole-dipole interactions, it does not hinder the production of a long-term coherent state. Through the use of a spatially dependent unitary transformation, one can relate the optical case to the simple and highly useful geometrical picture. It must be noted, however, that for optical-frequency energy separations, the long-term coherent component will manifest itself as a precessing macroscopic electric dipole, and therefore the sample itself will produce a coherent radiation field. If this field becomes comparable to the driving field, it must be included in the Hamiltonian. This problem and other considerations inherent in a practical optical case, such as specific spatial effects, noise arising from on-resonance spontaneous emission, nonlinear effects, and specific relaxation mechanisms have not been considered here. However, it is tempting to consider a situation in which a properly chosen system and experimental arrangement could produce coherent radiation fields that are orders of magnitude larger than the fields that could be produced by on-resonance excitation, by initiating stimulated emission off-resonance in an inhomogeneous distribution and taking advantage of the fact that the off-resonance effect allows one to couple the entire line into the radiation field in the absence of collisions or other processes which tend to yield a homogeneous line on the time scale of the stimulated emission output.

IV. SUMMARY

We have presented a discussion of coherence in an ensemble of excited-state two-level systems for the case in which population is being fed into the ensemble at a constant rate and decay is occurring from the ensemble at a rate dependent upon the state of the ensemble. The problem was ini-

tially treated in the absence of conventional T_1 and T_2 processes, and an exact solution was obtained using the density-matrix formalism. Several examples were treated to illustrate modifications which must be considered when a coherent coupling experiment is performed on an excited ensemble.

We have shown that it is possible to produce and maintain a coherent state in the excited ensemble for times which are only limited by the coherence time of the driving field, despite the fact that this time may greatly exceed the lifetimes of the excited states. Conventional T_1 and T_2 processes were added to the development and exact solutions were obtained for the steady-state case which is important when examing the possibility of producing long-term or kinetic coherence. It was demonstrated that if the modified Bloch equations are applicable, T_1 and T_2 processes do not modify the *"qualitative"* results obtained in their absence. Furthermore, even when rapid T_2 processes occur under low-power conditions, it was shown that for high power in some instances, sizable long-term coherent components may nonetheless be maintained, since the coherent component is effectively "spin-locked" along the rotating-frame static field.

Finally, it was shown that for thin samples the development applies to the short-wavelength optical case in addition to the long-wavelength case. It was pointed out that the steady-state coherent component in optical systems will produce coherent radiation for both on- and off-resonance driving fields, and that it may be possible to employ the off-resonance technique in a coherent light amplifier.

Experimental verification of long-term kinetic coherence has been established for electron spins in excited triplet states in zero field and will be presented in a forthcoming publication.[43]

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