Dissipation in Quantum Mechanics. Two-Level System. III

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Previous theory describing the behavior of a general two-level system (TLS) coupled to a loss mechanism and driven by an oscillating field near resonance is generalized to include an arbitrary driving field. Integro-differential equations of motion for the expectation values of the Pauli spin matrices, which describe completely the behavior of a TLS, are derived. The relaxation constants involved in these equations prove to be frequency-dependent, and reactive effects produced by the loss mechanism appear. It is shown that for an approximately monochromatic driving field near resonance, the general equations of motion can be approximated by differential equations. In the case of a magnetic TLS, these differential equations reduce to the Bloch equations if reactive effects are neglected. An exact solution of the general equations of motion for the special, simple case of a magnetic TLS being driven by a transverse rotating field of arbitrary frequency is obtained, and deviation from a Lorentzian resonance shape, caused by the frequency dependence of the relaxation and reactive constants, is found to exist.

I. INTRODUCTION

The usefulness of the study of a two-level system (TLS) coupled to a loss—or relaxation—mechanism (LM) was pointed out in the first article of the present series, and a theory describing this combination was developed in both the first and second articles[1]; the latter contains an analysis of the most general type of TLS coupled to an LM that consists of both a thermal reservoir and a large number of two-level systems identical to the one under consideration and randomly coupled to it. The existence of a prescribed (classical) driving field was taken into account in the above theory. Although no explicit restrictions were placed on the driving field, there was an implicit assumption—not unimportant in the derivation of the results—that the driving field was approximately an oscillating function with frequency near ω, the resonant frequency of the TLS. It is the purpose of the present article to remove this restriction on the driving field.

It should, perhaps, be made clear at the beginning that this generalization is not trivial or uninteresting. The present analysis does not consist of a rederivation of the previous results under less restrictive conditions, but rather, the derivation of new results which reduce, approximately, to those of II when the driving frequency is near ω. Its interest is illustrated by the fact that, whereas the previous results reduce to the Bloch equations if the TLS is a magnetic dipole, the present results do not reduce to these equations for a magnetic TLS, in general, and thus provide a more correct set of equations to be used with off-resonance driving fields. (As was shown in II, the equations for an electric dipole TLS are different from the Bloch equations even when the driving-field frequency is near resonance.) It may further be pointed out that experiments with driving fields far from resonance have acquired considerable interest recently.[2]

[1] I. R. Senitzky, Phys. Rev. **131**, 2827 (1963); **134**, A816 (1964); hereafter referred to as I and II, respectively.

[2] See, for instance, the discussion related to Raman lasers by N. Bloembergen and Y. R. Shen, Phys. Rev. **133**, A37 (1964), who give additional references.

In the development of the theory in II, the assumption concerning the driving field frequency was made not at the beginning but near the midpoint of the analysis, in going from Eqs. (II.27) to Eqs. (II.28). We will, however, rederive, in the present article, the theory preceding this assumption, because the derivation will be modified, to some extent. One purpose of the modification is the presentation of an alternate point of view of the approximations employed, and another, more important, purpose is the consideration of certain reactive effects of the loss mechanism which were previously neglected.

II. DERIVATION OF EQUATIONS OF MOTION

The notation of the present article is identical to that of II and will be briefly redefined. The dipole moment operator of the TLS (either electric or magnetic) is specified most generally as

$$\mathbf{d}=\mu\sum_{\alpha=1}^{4}\mathbf{a}_\alpha\sigma_\alpha\,, \tag{1}$$

where σ_4 is the unit matrix, σ_1, σ_2, and σ_3 are the three Pauli spin matrices, μ is a quantity having the dimensions of dipole moment, and the $\mathbf{a}$'s are four real 3-dimensional vectors characteristic of the TLS. For a spin-$\frac{1}{2}$ magnetic dipole, $\mathbf{a}_4=0$, and the other $\mathbf{a}$'s are orthogonal and equal in magnitude; it is convenient to take them to be orthonormal. For an electric dipole TLS, any set of values of the $\mathbf{a}$'s are possible, in principle. $\mathbf{a}_1$ and $\mathbf{a}_2$ determine the part of the dipole moment that oscillates (approximately) with frequency ω; $\mathbf{a}_3$ and $\mathbf{a}_4$ determine the "permanent" dipole moment. In the Heisenberg picture, which is used in the present treatment, σ_1, σ_2, and σ_3 are the dynamical variables which describe the TLS completely. The energy of the TLS, referred to the midpoint between the two levels, is given by $\frac{1}{2}\hbar\omega\sigma_3$.

As mentioned previously, the LM consists of two parts: a thermal reservoir (LM$^{(1)}$), and a large number of systems identical to the TLS (LM$^{(2)}$), all in similar environments, with random (but loose) coupling to each other and to the system under consideration. As discussed in detail in II, the essential qualitative difference between LM$^{(1)}$ and LM$^{(2)}$ is the fact that LM$^{(2)}$ does not have a fixed temperature. In all but the last stage of the analysis, LM$^{(1)}$ and LM$^{(2)}$ are treated similarly. At the end, however, the quantity that corresponds to the energy of a TLS in thermal equilibrium with LM$^{(2)}$ is set equal to the expectation value of $\frac{1}{2}\hbar\omega\bar{\sigma}_3$, the energy of the TLS under consideration averaged over several cycles. (This averaging was not performed in II. It is carried out so that any oscillations in σ_3 of a frequency of the order of ω or higher are removed from the expression for the thermal equilibrium energy of LM$^{(2)}$, the assumption being that thermal relaxation is much slower than a period of the TLS.) For the sake of simplicity, our initial discussion will refer to LM$^{(1)}$ only, that is, to a single thermal reservoir. At an appropriate later stage, it will be easy to bring into consideration LM$^{(2)}$ also.

A. Thermal Reservior Only

The field (operator) of the LM which acts on the TLS is designated, in units of $-\hbar/2\mu$, by $\mathbf{F}$, and the external (c number) field is designated in the same units by $\mathbf{f}$. Both $\mathbf{F}$ and $\mathbf{f}$ are regarded as small quantities compared to ω, which means that the TLS is coupled weakly to both LM and external field. The spatial components of $\mathbf{F}$ are assumed to behave independently, so that it is convenient to think of three independent (thermal reservoir) loss mechanisms, each responsible for the LM field along a different Cartesian axis (but all at the same temperature, of course). For the free (uncoupled) LM, the field is given by $F^{(0)}$. The only properties of $F^{(0)}$ that will be needed in the present discussion, and derived in I, are

$$\langle F_m^{(0)}(t)\rangle = 0, \tag{2}$$

$$\langle F_m^{(0)}(t_1)F_n^{(0)}(t_2)\rangle = \delta_{nm}(2/\pi)\int_0^\infty d\omega'[\eta_m(\omega')\cos\omega'(t_1-t_2) - i\xi_m(\omega')\sin\omega'(t_1-t_2)], \tag{3}$$

where

$$\xi_m(\omega') = \tfrac{1}{2}\pi\hbar Z_m^{-1}B_m(\omega')[1-\exp(-\hbar\omega'/kT)], \tag{3a}$$

$$\eta_m(\omega') = \tfrac{1}{2}\pi\hbar Z_m^{-1}B_m(\omega')[1+\exp(-\hbar\omega'/kT)], \tag{3b}$$

$$Z_m = \int_0^\infty dE\rho_m(E)\exp(-E/kT), \tag{3c}$$

$$B_m(\omega') = \int_0^\infty dE\rho_m(E+\hbar\omega')\rho_m(E) \times \tilde{F}_m^2(E+\hbar\omega',E)\exp(-E/kT), \tag{3d}$$

$\rho_m(E)$ being the density of energy states of the m axis LM (assumed closely spaced), $\tilde{F}^2(E_i,E_k)$ being the average over small ranges of E_i and E_k of $|F_{ik}^{(0)}(0)|^2$, and T being the LM temperature. Both $\eta_m(\omega')$ and $\xi_m(\omega')$ are assumed to be slowly varying functions of ω', except perhaps, in the neighborhood of $\omega'=0$. The quantity $\xi_m(0)$ obviously vanishes because of the vanishing of the factor in the square bracket of Eq. (3a) at $\omega'=0$. It is not unreasonable to expect also $\eta_m(0)$ to vanish, since the factor $B_m(0)$ depends on $\tilde{F}_m^2(E,E)$, and the diagonal matrix elements of $F_m^{(0)}$ vanish, according to Eq. (2). In the following theory, however, only the vanishing of $\xi_m(0)$ is necessary.

The Hamiltonian for the system consisting of the TLS and LM is given by

$$H = H_{\mathrm{LM}} + \tfrac{1}{2}\hbar\omega\sigma_3 + \tfrac{1}{2}\hbar\textstyle\sum_\alpha \sigma_\alpha \mathbf{a}_\alpha\cdot\boldsymbol{\mathfrak{F}}, \tag{4}$$

where

$$\boldsymbol{\mathfrak{F}} = \mathbf{F}+\mathbf{f}, \tag{4a}$$

and

$$H_{\mathrm{LM}} = \sum_{m=1}^{3} H_m, \tag{4b}$$

H_m being the Hamiltonian of the component LM acting along the mth axis. We allow, for the present, the possibility that the LM is not isotropic. The formal operator equations of motion, obtained from Eqs. (4) are

$$\dot{\sigma}_1 = -\omega\sigma_2 + \tfrac{1}{2}\textstyle\sum_m(a_{2m}\{\mathfrak{F}_m,\sigma_3\} - a_{3m}\{\mathfrak{F}_m,\sigma_2\}), \tag{5a}$$

$$\dot{\sigma}_2 = \omega\sigma_1 + \tfrac{1}{2}\textstyle\sum_m(a_{3m}\{\mathfrak{F}_m,\sigma_1\} - a_{1m}\{\mathfrak{F}_m,\sigma_3\}), \tag{5b}$$

$$\dot{\sigma}_3 = \tfrac{1}{2}\textstyle\sum_m(a_{1m}\{\mathfrak{F}_m,\sigma_2\} - a_{2m}\{\mathfrak{F}_m,\sigma_1\}), \tag{5c}$$

$$\dot{F}_m = -(i/\hbar)[F_m,H_m], \tag{5d}$$

$$\dot{H}_m = -(i/\hbar)\textstyle\sum_\alpha a_{\alpha m}\sigma_\alpha[H_m,F_m], \tag{5e}$$

where the index m designates the three spatial components resolved along Cartesian axes, and where the symmetrized product $(\{A,B\}=AB+BA)$ has been used for convenience. The last two equations yield

$$\dot{F}_m(t) = -\frac{i}{\hbar}[F_m(t),H_m(0)] + \frac{1}{2\hbar}\sum_\alpha a_{\alpha m}\int_0^t dt_1[F_m(t),[F_m(t_1),H_m(t_1)]\sigma_\alpha(t_1)], \tag{6}$$

which may be written as

$$F_m(t) = F_m^{(0)}(t) + \frac{1}{2\hbar}\sum_\alpha a_{\alpha m}\int_0^t dt_1\int_0^{t_1} dt_2 \times U_m(t-t_1)[F_m(t_1),[F_m(t_2),H_m(t_2)]\sigma_\alpha(t_2)] \times U_m^{-1}(t-t_1), \tag{7}$$

where

$$U(\tau) \equiv \exp[(i/\hbar)H_m(0)\tau],$$

and where $F_m^{(0)}(t)$ refers to the free LM; it is assumed that the coupling between TLS and LM has been turned on at $t=0$.

We consider first the $\alpha=4$ term in the summation of Eq. (7). Since σ_4 is the unit matrix, this term contains no dynamical variables referring to the TLS. It represents the reaction of the LM to the constant part of the dipole moment of the TLS, a reaction that is insensitive to the behavior of the TLS. We assume that, for sufficiently large t, this reaction becomes independent of the time, and we incorporate this constant into **f**, the external force acting on the TLS. Thus, only the terms with $\alpha=1, 2, 3$ are left for consideration.

An important approximation will now be performed. We ignore the noncommutativity of $\sigma_\alpha(t_2)$ and the LM variables, replace the LM variables by those for the free LM, and then replace the commutator by its expectation value. This approximation, discussed in I, is based on the neglect of quantum-mechanical correlation between TLS and LM in second-order terms, on the fact that the LM is perturbed only slightly by the TLS, and on the fact that final results will be (LM) expectation values. The result is

$$F_m(t)=F_m^{(0)}(t)+\frac{1}{2\hbar}\sum_\alpha a_{\alpha m}\int_0^t dt_1\int_0^{t_1} dt_2 \times\langle[F_m^{(0)}(t_1),[F_m^{(0)}(t_2),H_m^{(0)}]]\rangle\sigma_\alpha(t_2). \quad (8)$$

On the assumption that the energy states of the LM are densely spaced, it can be shown without much difficulty (see I) that

$$F_m(t)=F_m^{(0)}(t)-\frac{2}{\pi}\sum_\alpha a_{\alpha m}\int_0^t dt_1\int_0^\infty d\omega'\xi_m(\omega') \times\sin\omega'(t-t_1)\sigma_\alpha(t_1). \quad (9)$$

A substitution is now made for F_m from Eq. (9) into the first three of Eqs. (5), the equations of motion. The resulting equations contain both first- and second-order terms (F_m is considered a quantity of first order), the second-order terms having the form

$$-\frac{1}{\pi}\sum_m\sum_\alpha a_{\gamma m}a_{\alpha m}\int_0^t dt_1\int_0^\infty d\omega'\xi_m(\omega') \times\sin\omega'(t-t_1)\{\sigma_\beta(t),\sigma_\alpha(t_1)\}. \quad (10)$$

We leave the first-order terms unchanged, and approximate the second-order terms by replacing $\{\sigma_\beta(t),\sigma_\alpha(t_1)\}$ by the corresponding expression for the free TLS, $\{\sigma_\beta^{(0)}(t),\sigma_\alpha^{(0)}(t_1)\}$. (Approximations in second-order terms constitute, essentially, the spirit of the present analysis. One may regard the present approximation as the neglect of terms of higher than second order in the differential equations of motion.) In justification of this approximation, one might say, firstly, that it is reasonable to expect $\{\sigma_\beta(t),\sigma_\alpha(t_1)\}$ to depend mainly on $t-t_1$, since $\{\sigma_\beta(t),\sigma_\alpha(t)\}$ is independent of t (and is given by the anticommutator relationships for the Pauli spin matrices). Secondly, the integration over t_1 is weighted in favor of low values of $t-t_1$ by the integration over ω'. For $t-t_1$ small (compared to a relaxation time), and ignoring for the moment the effect of the external field, the expression for $\{\sigma_\beta(t),\sigma_\alpha(t_1)\}$ in the interval $t-t_1$ may be expected not to differ much from the corresponding expression for the case in which the coupling to the LM is removed at the beginning of the interval. As far as the effect of the external field is concerned, there are fields for which exact solutions for the σ's can be obtained in the absence of the LM. The effect of these fields could then be incorporated into the expression for $\{\sigma_\beta(t),\sigma_\alpha(t_1)\}$ to be used in the second-order terms. For the sake of keeping **f** general, however, we will forego, in the present discussion, this increase in accuracy and assume that **f** is sufficiently weak so that the approximation under discussion may be used.

The anticommutators for the free TLS are given by

$$\{\sigma_1^{(0)}(t),\sigma_1^{(0)}(t_1)\}=\{\sigma_2^{(0)}(t),\sigma_2^{(0)}(t_1)\} =2\cos\omega(t-t_1), \quad (11a)$$

$$\{\sigma_3^{(0)}(t),\sigma_\alpha^{(0)}(t_1)\}=2\delta_{\alpha 3}, \quad (11b)$$

$$\{\sigma_1^{(0)}(t),\sigma_2^{(0)}(t_1)\}=-2\sin\omega(t-t_1), \quad (11c)$$

and the expressions obtained by interchanging t and t_1 in Eqs. (11). The combination of integrals of the type (10) occurring in the equations of motion, together with the above approximation, lead to the following two integrals:

$$\kappa_m\equiv\frac{2}{\pi}\int_0^t dt_1\int_0^\infty d\omega'\xi_m(\omega')\sin\omega'(t-t_1) \times[1-\cos\omega(t-t_1)], \quad (12a)$$

and

$$\lambda_m\equiv\frac{2}{\pi}\int_0^t dt_1\int_0^\infty d\omega'\xi_m(\omega')\sin\omega'(t-t_1)\sin\omega(t-t_1). \quad (12b)$$

Carrying out the integration with respect to t_1 first, we obtain

$$\kappa_m=\frac{2}{\pi}\int_0^\infty d\omega'\xi_m(\omega')\left\{\frac{1-\cos\omega' t}{\omega'} -\frac{1}{2}\left[\frac{1-\cos(\omega'+\omega)t}{\omega'+\omega}+\frac{1-\cos(\omega'-\omega)t}{\omega'-\omega}\right]\right\}, \quad (13a)$$

$$\lambda_m=\frac{2}{\pi}\int_0^\infty d\omega'\xi_m(\omega')\frac{1}{2}\left[\frac{\sin(\omega'-\omega)t}{\omega'-\omega}-\frac{\sin(\omega'+\omega)t}{\omega'+\omega}\right]. \quad (13b)$$

For sufficiently large t, κ_m and λ_m become independent of t and are given by

$$\kappa_m=\frac{2}{\pi}\int_0^\infty d\omega'\xi_m(\omega')\left[\frac{1}{\omega'}-\omega'\frac{\mathcal{P}}{\omega'^2-\omega^2}\right], \quad (14)$$

$$\lambda_m=\xi_m(\omega). \quad (15)$$

Looking back at Eqs. (12), we see that κ_m is invariant and λ_m changes sign under time inversion. This indicates that κ_m is associated with reactive processes, while λ_m is associated with dissipative processes. It can also be said that λ_m refers to a resonant effect, the main contribution to the ω' integration coming from ω' near ω, while κ_m refers to a nonresonant effect, the contribution to the ω' integration coming from a broad range of values.

Substituting from Eq. (9) into the first three of Eqs. (5) (as mentioned previously), and using the above approximation, we obtain, as the equations of motion,

$$\dot{\sigma}_1=-\omega\sigma_2+\tfrac{1}{2}\sum_m[a_{2m}\{\mathfrak{F}_m^{(0)},\sigma_3\}-a_{3m}\{\mathfrak{F}_m^{(0)},\sigma_2\}+2a_{1m}a_{3m}\xi_m-2a_{2m}a_{3m}\kappa_m], \quad (16a)$$

$$\dot{\sigma}_2=\omega\sigma_1+\tfrac{1}{2}\sum_m[a_{3m}\{\mathfrak{F}_m^{(0)},\sigma_1\}-a_{1m}\{\mathfrak{F}_m^{(0)},\sigma_3\}+2a_{2m}a_{3m}\xi_m+2a_{1m}a_{3m}\kappa_m], \quad (16b)$$

$$\dot{\sigma}_3=\tfrac{1}{2}\sum_m[a_{1m}\{\mathfrak{F}_m^{(0)},\sigma_2\}-a_{2m}\{\mathfrak{F}_m^{(0)},\sigma_1\}-2(a_{1m}^2+a_{2m}^2)\xi_m], \quad (16c)$$

where

$$\mathfrak{F}_m^{(0)}\equiv F_m^{(0)}+f_m, \quad \xi_m\equiv\xi_m(\omega).$$

Except for the presence of the reactive constant κ_m, these equations are essentially identical to Eqs. (II.21).[3] It should be noted that the only unknowns occurring in these equations are operators referring to the TLS. The operators referring to the LM may be regarded as prescribed by Eqs. (2) and (3). Of course, the σ's, in their time development, become operators in both TLS and LM spaces. Such an operator solution of Eqs. (16) has not yet been found.

Defining the functions Φ_1, Φ_2, and Φ_3 by writing Eqs. (16) as

$$\dot{\sigma}_1=-\omega\sigma_2+\Phi_1(t), \quad (17a)$$

$$\dot{\sigma}_2=\omega\sigma_1+\Phi_2(t), \quad (17b)$$

$$\dot{\sigma}_3=\Phi_3(t), \quad (17c)$$

we have, as equivalent integral equations,[4]

$$\sigma_1=\int_{-\infty}^{t}dt_1\Phi_1(t_1)\cos\omega(t-t_1)-\int_{-\infty}^{t}dt_1\Phi_2(t_1)\sin\omega(t-t_1), \quad (18a)$$

$$\sigma_2=\int_{-\infty}^{t}dt_1\Phi_2(t_1)\cos\omega(t-t_1)+\int_{-\infty}^{t}dt_1\Phi_1(t_1)\sin\omega(t-t_1), \quad (18b)$$

$$\sigma_3=\int_{-\infty}^{t}dt_1\Phi_3(t_1). \quad (18c)$$

We now substitute from Eqs. (18) into only those terms of Eqs. (16) which contain the products $\{F_m^{(0)},\sigma_\alpha\}$, and then take expectation values (in both TLS and LM spaces) of both sides in the resulting equations. On the right sides we will have expectation values of the form

$$\langle\{F_m^{(0)}(t),\{F_n^{(0)}(t_1),\sigma_\alpha(t_1)\}\}\rangle, \quad (19a)$$

$$\langle\{F_m^{(0)}(t),\{f_n(t_1),\sigma_\alpha(t_1)\}\}\rangle, \quad (19b)$$

$$\langle F_m^{(0)}(t)\rangle. \quad (19c)$$

The expectation value (19c) vanishes, as a result of Eq. (2). As far as expressions (19a) and (19b) are concerned, we note that they are both second-order expressions, since $\mathbf{F}^{(0)}$ and $\mathbf{f}$ are considered small quantities of first order, and approximate by ignoring the operator aspects of σ_α with respect to the LM. This means that we are neglecting the quantum-mechanical correlation between the TLS and LM in second-order terms. [We are, in effect, disentangling the two systems at this point and restoring to them their individual identities. A similar approximation was performed in going from Eq. (7) to Eq. (8).] With this approximation, one obtains

$$\langle\{F_m^{(0)}(t),\{F_n^{(0)}(t_1),\sigma_\alpha(t_1)\}\}\rangle \approx 2\langle\{F_m^{(0)}(t),F_n^{(0)}(t_1)\}\rangle\langle\sigma_\alpha(t_1)\rangle =2\delta_{mn}\langle\{F_m^{(0)}(t),F_m^{(0)}(t_1)\}\rangle\langle\sigma_\alpha(t_1)\rangle, \quad (20)$$

$$\langle\{F_m^{(0)}(t),\{f_n(t_1),\sigma_\alpha(t_1)\}\}\rangle \approx 4f_n(t_1)\langle F_m^{(0)}(t)\rangle\langle\sigma_\alpha(t_1)\rangle=0. \quad (21)$$

For notational simplicity, we drop the expectation value sign from the σ's and consider all σ's, henceforth, to stand for their respective expectation values. We also go over, formally, to the isotropic case by writing

$$\sum_m L_m a_{\alpha m}a_{\beta m}=L\mathbf{a}_\alpha\cdot\mathbf{a}_\beta, \quad (22)$$

where L_m is any variable referring to the mth component LM and L is treated as an ordinary number.[5] One can return, explicitly, to the nonisotropic case by reversing Eq. (22). The language, henceforth, will likewise refer to the case of the isotropic LM (for which all component loss mechanisms are identical). Furthermore, we introduce the following symbolic notation:

$$\mathcal{O}\phi(t)\equiv\frac{1}{2}\int_{-\infty}^{t}dt_1\langle\{F^{(0)}(t),F^{(0)}(t_1)\}\rangle\phi(t_1), \quad (23a)$$

$$\mathcal{O}_+\phi(t)\equiv\frac{1}{2}\int_{-\infty}^{t}dt_1\langle\{F^{(0)}(t),F^{(0)}(t_1)\}\rangle\times\cos\omega(t-t_1)\phi(t_1), \quad (23b)$$

$$\mathcal{O}_-\phi(t)\equiv\frac{1}{2}\int_{-\infty}^{t}dt_1\langle\{F^{(0)}(t),F^{(0)}(t_1)\}\rangle\times\sin\omega(t-t_1)\phi(t_1), \quad (23c)$$

[3] The time from the commencement of coupling between TLS and LM until λ_m becomes constant is ignored in Eqs. (4), but is taken into account in Eqs. (II.21) by writing there $\bar{\xi}$ instead of ξ, $\bar{\xi}$ being a function of the time which starts from zero at $t=0$ and approaches the constant ξ in the manner in which λ_m becomes constant. Another trivial difference between Eqs. (II.21) and Eqs. (16) is the fact that in II, ξ_1, ξ_2, and ξ_3 are taken to be equal.

[4] A slight modification is made here, for convenience, in comparison with II; the lower limit of the integrals is taken to be $-\infty$ instead of zero, Eqs. (16) are assumed to hold for all time, and initial conditions are ignored. (Because of coupling to the LM, the initial conditions for the TLS become eventually unimportant.)

[5] The left side of Eq. (21) may be written as $\mathbf{a}_\alpha L\mathbf{a}_\beta$, where L is a diagonal tensor with elements L_1, L_2, and L_3. Since it is much simpler, intuitively, to think of the isotropic case, however, we use the isotropic case notation.

where $\phi(t)$ is an arbitrary function. (The reason for the "+" and "−" will become apparent later.) With the above substitutions, approximations, and notational simplifications, Eqs. (16) yield

$$\begin{aligned}\dot{\sigma}_1=&(-\omega+\mathbf{a}_1\cdot\mathbf{a}_2\,\mathcal{O}+a_3^2\,\mathcal{O}_-)\sigma_2+\mathbf{a}_2\cdot\mathbf{f}\sigma_3-\mathbf{a}_3\cdot\mathbf{f}\sigma_2\\&-(a_2^2\,\mathcal{O}+a_3^2\,\mathcal{O}_+)\sigma_1+\mathbf{a}_1\cdot\mathbf{a}_3(\mathcal{O}_+\sigma_3+\xi)\\&-\mathbf{a}_2\cdot\mathbf{a}_3(\mathcal{O}_-\sigma_3+\kappa),\end{aligned}\tag{24a}$$

$$\begin{aligned}\dot{\sigma}_2=&(\omega+\mathbf{a}_1\cdot\mathbf{a}_2\,\mathcal{O}-a_3^2\,\mathcal{O}_-)\sigma_1+\mathbf{a}_3\cdot\mathbf{f}\sigma_1-\mathbf{a}_1\cdot\mathbf{f}\sigma_3\\&-(a_1^2\,\mathcal{O}+a_3^2\,\mathcal{O}_+)\sigma_2+\mathbf{a}_2\cdot\mathbf{a}_3(\mathcal{O}_+\sigma_3+\xi)\\&+\mathbf{a}_1\cdot\mathbf{a}_3(\mathcal{O}_-\sigma_3+\kappa),\end{aligned}\tag{24b}$$

$$\begin{aligned}\dot{\sigma}_3=&\mathbf{a}_1\cdot\mathbf{f}\sigma_2-\mathbf{a}_2\cdot\mathbf{f}\sigma_1-(a_1^2+a_2^2)(\mathcal{O}_+\sigma_3+\xi)\\&+\mathbf{a}_3\cdot(\mathbf{a}_1\,\mathcal{O}_++\mathbf{a}_2\,\mathcal{O}_-)\sigma_1+\mathbf{a}_3\cdot(\mathbf{a}_2\,\mathcal{O}_+-\mathbf{a}_1\,\mathcal{O}_-)\sigma_2.\end{aligned}\tag{24c}$$

The significance of the operators $\mathcal{O}$, $\mathcal{O}_+$, and $\mathcal{O}_-$ will now be examined. We consider first the effect of operating with these operators on a sinusoidal function of the time, $\sin(\nu t+\theta)$. We have, noting Eq. (3),

$$\begin{aligned}\mathcal{O}\sin(\nu t+\theta)&\equiv\frac{1}{2}\int_{-\infty}^{t}dt_1\langle\{F^{(0)}(t),F^{(0)}(t_1)\}\rangle\sin(\nu t_1+\theta),\\&=\frac{2}{\pi}\int_{-\infty}^{t}dt_1\int_0^\infty d\omega'\eta(\omega')\cos\omega'(t-t_1)\sin(\nu t_1+\theta),\quad(25)\\&\equiv A_1+A_2,\end{aligned}$$

where

$$A_1=\frac{2}{\pi}\int_{-\infty}^{t}dt_1\int_0^\infty d\omega'\eta(|\nu|)\cos\omega'(t-t_1)\sin(\nu t_1+\theta),\tag{25a}$$

and

$$A_2=\frac{2}{\pi}\int_{-\infty}^{t}dt_1\int_0^\infty d\omega'[\eta(\omega')-\eta(|\nu|)]\times\cos\omega'(t-t_1)\sin(\nu t_1+\theta).\tag{25b}$$

For simplicity of notation, we drop the absolute value bars about the argument of η [η is defined by Eq. (3b) only for positive argument] and use henceforth the definition

$$\eta(\nu)\equiv\eta(|\nu|).\tag{26}$$

The value of A_1 is given immediately by

$$\begin{aligned}A_1&=2\eta(\nu)\int_{-\infty}^{t}dt_1\delta(t-t_1)\sin(\nu t_1+\theta),\\&=\eta(\nu)\sin(\nu t+\theta).\end{aligned}\tag{27}$$

For A_2, we have, carrying out the t_1 integration first,

$$\begin{aligned}A_2=&\frac{2\nu}{\pi}\cos(\nu t+\theta)\int_0^\infty d\omega'\frac{\eta(\omega')-\eta(\nu)}{\omega'^2-\nu^2}\\&+\lim_{\tau\to\infty}\frac{1}{\pi}\int_0^\infty d\omega'[\eta(\omega')-\eta(\nu)]\left\{\frac{\cos[(\nu-\omega')\tau+\omega' t+\theta]}{\nu-\omega'}\right.\\&\left.+\frac{\cos[(\nu+\omega')\tau-\omega' t+\theta]}{\nu+\omega'}\right\}.\end{aligned}\tag{28}$$

The second integral in Eq. (28) vanishes in the limit, and we have, from Eqs. (25), (27), and (28),

$$\mathcal{O}\sin(\nu t+\theta)=\eta(\nu)\sin(\nu t+\theta)+\chi(\nu)\cos(\nu t+\theta),\tag{29}$$

where

$$\chi(\nu)=\frac{2}{\pi}\nu\int_0^\infty d\omega'\frac{\eta(\omega')-\eta(\nu)}{\omega'^2-\nu^2}.\tag{29a}$$

The effect of operating with $\mathcal{O}$ on a general function $\varphi(t)$ can now be obtained easily. Since $\mathcal{O}$ is a linear operator, all we need do is express $\varphi(t)$ as a sum, discrete or continuous, of sinusoidal functions of the time, and apply Eq. (29) to each member of the sum. In view of the interest that usually exists in the steady state, we will consider explicitly only discrete sums. Introducing complex notation, Eq. (29) yields

$$\mathcal{O}e^{i\nu t}=[\eta(\nu)+i\chi(\nu)]e^{i\nu t},\tag{30}$$

and setting

$$\varphi(t)=\sum_j a_je^{i\nu_jt},\tag{31}$$

we obtain

$$\mathcal{O}\varphi(t)=\sum_j a_j[\eta(\nu_j)+i\chi(\nu_j)]e^{i\nu_jt}.\tag{32}$$

We can therefore say that the operator $\mathcal{O}$ decomposes a function into its (complex) harmonic components, multiplies each component by a complex number which depends on the frequency, and then superposes the components again. In the language of electrical circuitry, the effect of $\mathcal{O}$ is that of a frequency-sensitive filter. It is clear that η is associated with resistive effects, and χ is associated with reactive effects.

The effects of operating with $\mathcal{O}_+$ and $\mathcal{O}_-$ can be obtained either by a method similar to that used for $\mathcal{O}$, or by utilizing the result already derived for $\mathcal{O}$. We obtain

$$\begin{aligned}\mathcal{O}_+\sin(\nu t+\theta)=&\tfrac{1}{2}[\eta(\nu+\omega)+\eta(\nu-\omega)]\sin(\nu t+\theta)\\&+\tfrac{1}{2}[\chi(\nu+\omega)+\chi(\nu-\omega)]\cos(\nu t+\theta),\end{aligned}\tag{33}$$

$$\begin{aligned}\mathcal{O}_-\sin(\nu t+\theta)=&-\tfrac{1}{2}[\chi(\nu+\omega)-\chi(\nu-\omega)]\sin(\nu t+\theta)\\&+\tfrac{1}{2}[\eta(\nu+\omega)-\eta(\nu-\omega)]\cos(\nu t+\theta).\end{aligned}\tag{34}$$

In complex notation, this becomes

$$\begin{aligned}\mathcal{O}_+e^{i\nu t}=\tfrac{1}{2}\{&[\eta(\nu+\omega)+\eta(\nu-\omega)]\\&+i[\chi(\nu+\omega)+\chi(\nu-\omega)]\}e^{i\nu t},\end{aligned}\tag{35}$$

$$\begin{aligned}\mathcal{O}_-e^{i\nu t}=\tfrac{1}{2}\{&-[\chi(\nu+\omega)-\chi(\nu-\omega)]\\&+i[\eta(\nu+\omega)-\eta(\nu-\omega)]\}e^{i\nu t}.\end{aligned}\tag{36}$$

The result of operating with $\mathcal{O}_+$ or $\mathcal{O}_-$ on $\varphi(t)$ can be read off immediately from Eqs. (35) and (36), and need not be written down.

The results of operating with the above three operators on a constant are also useful. These may be obtained by setting $\nu=0$ in Eqs. (30), (35), and (36). Since $\chi(0)=0$, from Eq. (29a), we have

$$\mathcal{O}\cdot 1=\eta(0). \tag{37}$$

Recalling that η is an even function of its argument and χ is an odd function, we obtain from Eqs. (35) and (36),

$$\mathcal{O}_+\cdot 1=\eta(\omega), \tag{38}$$

$$\mathcal{O}_-\cdot 1=-\chi(\omega). \tag{39}$$

Before proceeding further, it is useful to have a steady-state solution of Eqs. (24) for the case $\mathbf{f}=0$, that is, the case of thermal equilibrium between TLS and LM. By setting the time derivatives of the σ's equal to zero, we obtain such a solution. Equations (24) yield three algebraic (inhomogeneous) equations with constant coefficients for the three σ's and can be solved in a routine fashion. Rather than derive the exact solution, we will be satisfied with an approximate one, the approximation being based on the smallness of ξ and η (and, therefore, χ and κ) compared to ω. Such an approximate solution is given by

$$\sigma_1=\sigma_2=0,$$

$$\sigma_3=-\frac{\xi(\omega)}{\eta(\omega)}=-\frac{1-\exp(-\hbar\omega/kT)}{1+\exp(-\hbar\omega/kT)}\equiv\sigma_0. \tag{40}$$

It is easy to see that the exact σ's differ from those of Eq. (40) by an amount that is of the order of magnitude of η/ω. If $\mathbf{a}_1$ and $\mathbf{a}_2$ are perpendicular to $\mathbf{a}_3$, or if $\mathbf{a}_3=0$, Eqs. (40) are an exact solution of Eqs. (24) with $\mathbf{f}=0$. σ_0 is, obviously, the energy (in units of $\frac{1}{2}\hbar\omega$) of a TLS in thermal equilibrium with the LM.

B. Thermal Reservoir and Large Number of Identical Systems

We can now generalize the analysis so that it applies to the case in which the LM consists of the two parts mentioned at the beginning of the article, the thermal reservoir and the large number of systems identical to the TLS. If we consider, at first, two thermal reservoirs [denoted by superscripts (1) and (2)], it follows from the derivation of the present results that the only changes necessary are the substitutions

$$\eta(\nu)\rightarrow\eta^{(1)}(\nu)+\eta^{(2)}(\nu), \tag{41a}$$

$$\xi(\nu)\rightarrow\xi^{(1)}(\nu)+\xi^{(2)}(\nu), \tag{41b}$$

$$\chi(\nu)\rightarrow\chi^{(1)}(\nu)+\chi^{(2)}(\nu), \tag{41c}$$

$$\kappa\rightarrow\kappa^{(1)}+\kappa^{(2)}. \tag{41d}$$

If, now, instead of the second thermal reservoir, we have a loss mechanism ($\mathrm{LM}^{(2)}$) consisting of a large number of systems identical to our TLS (randomly and weaking coupled to each other) then the temperature of $\mathrm{LM}^{(2)}$ is no longer prescribed and necessarily constant, but σ_0 for this system may be replaced by $\bar{\sigma}_3$, the average of σ_3 over several cycles. (See II for a more detailed discussion of matters related to the second LM.) Writing $\xi(\omega)$ as $-\eta(\omega)\sigma_0$, from Eq. (40), we have the substitution,

$$\begin{aligned}\xi_1(\omega)+\xi_2(\omega)\rightarrow&-[\eta^{(1)}(\omega)\sigma_0^{(1)}+\eta^{(2)}(\omega)\sigma_0^{(2)}]\\ &=-[\eta^{(1)}(\omega)\sigma_0^{(1)}+\eta^{(2)}(\omega)\bar{\sigma}_3].\end{aligned} \tag{42}$$

$\xi(\omega)$ occurs explicitly in Eq. (24) [it should be recalled that ξ without the argument stands for $\xi(\omega)$], and there the substitution of Eq. (41b) and Eq. (42) should be made. It is also true that $\xi(\omega')$ occurs under the integral sign in the definition of $\kappa^{(2)}$, with ω' being the variable of integration [see Eq. (14)]. There, however, the contribution to the integral comes from a wide range of frequencies, and we approximate by leaving the form of the definition of $\kappa^{(2)}$ the same as that for the thermal reservoir, the temperature in the definition of $\xi^{(2)}(\omega')$ being replaced by a reasonable average value. (The same consideration applies to $\eta^{(2)}$.) Using the notation

$$\tilde{\eta}=\eta^{(1)}+\eta^{(2)},\quad \tilde{\chi}=\chi^{(1)}+\chi^{(2)} \tag{43}$$

we redefine the operators $\mathcal{O}$, $\mathcal{O}_+$, and $\mathcal{O}_-$ by

$$\mathcal{O}e^{i\nu t}=[\tilde{\eta}(\nu)+i\tilde{\chi}(\nu)]e^{i\nu t}, \tag{44a}$$

$$\begin{aligned}\mathcal{O}_+e^{i\nu t}=\tfrac{1}{2}\{&[\tilde{\eta}(\nu+\omega)+\tilde{\eta}(\nu-\omega)]\\ &+i[\tilde{\chi}(\nu+\omega)+\tilde{\chi}(\nu-\omega)]\}e^{i\nu t},\end{aligned} \tag{44b}$$

$$\begin{aligned}\mathcal{O}_-e^{i\nu t}=\tfrac{1}{2}\{&-[\tilde{\chi}(\nu+\omega)-\tilde{\chi}(\nu-\omega)]\\ &+i[\tilde{\eta}(\nu+\omega)-\tilde{\eta}(\nu-\omega)]\}e^{i\nu t}.\end{aligned} \tag{44c}$$

The expression $\mathcal{O}_+\sigma_3+\xi$ in Eq. (24) becomes, under the substitutions of Eqs. (41) and (42) (with the new definition of $\mathcal{O}_+$),

$$\mathcal{O}_+\sigma_3-\eta^{(1)}(\omega)\sigma_0^{(1)}-\eta^{(2)}(\omega)\bar{\sigma}_3. \tag{45}$$

If we assume that $\bar{\sigma}_3$ varies sufficiently slowly so that [see Eq. (38)]

$$\mathcal{O}_+\bar{\sigma}_3\approx\tilde{\eta}(\omega)\bar{\sigma}_3, \tag{46}$$

expression (45) may be written as

$$\mathcal{O}_+(\sigma_3-\bar{\sigma}_3)+\eta^{(1)}(\omega)(\bar{\sigma}_3-\sigma_0^{(1)}). \tag{47}$$

Dropping the superscript (1) as a notational simplification, and setting $\tilde{\kappa}=\kappa^{(1)}+\kappa^{(2)}$, the equations of motion, with the LM consisting of both $\mathrm{LM}^{(1)}$ and $\mathrm{LM}^{(2)}$, now become,

$$\begin{aligned}\dot{\sigma}_1=&(-\omega+\mathbf{a}_1\cdot\mathbf{a}_2\,\mathcal{O}+a_3^2\,\mathcal{O}_-)\sigma_2\\ &+\mathbf{a}_2\cdot\mathbf{f}\sigma_3-\mathbf{a}_3\cdot\mathbf{f}\sigma_2-(a_2^2\,\mathcal{O}+a_3^2\,\mathcal{O}_+)\sigma_1\\ &+\mathbf{a}_1\cdot\mathbf{a}_3[\mathcal{O}_+(\sigma_3-\bar{\sigma}_3)+\eta(\omega)(\bar{\sigma}_3-\sigma_0)]\\ &-\mathbf{a}_2\cdot\mathbf{a}_3(\mathcal{O}_-\sigma_3+\tilde{\kappa}),\end{aligned} \tag{48a}$$

$$\begin{aligned}\dot{\sigma}_2=&(\omega+\mathbf{a}_1\cdot\mathbf{a}_2\,\mathcal{O}-a_3^2\,\mathcal{O}_-)\sigma_1\\&+\mathbf{a}_3\cdot\mathbf{f}\sigma_1-\mathbf{a}_1\cdot\mathbf{f}\sigma_3-(a_1^2\,\mathcal{O}+a_3^2\,\mathcal{O}_+)\sigma_2\\&+\mathbf{a}_2\cdot\mathbf{a}_3[\mathcal{O}_+(\sigma_3-\bar{\sigma}_3)+\eta(\omega)(\bar{\sigma}_3-\sigma_0)]\\&+\mathbf{a}_1\cdot\mathbf{a}_3(\mathcal{O}_-\sigma_3+\tilde{\kappa}),\end{aligned}\tag{48b}$$

$$\begin{aligned}\dot{\sigma}_3=&\mathbf{a}_1\cdot\mathbf{f}\sigma_2-\mathbf{a}_2\cdot\mathbf{f}\sigma_1\\&-(a_1^2+a_2^2)[\mathcal{O}_+(\sigma_3-\bar{\sigma}_3)+\eta(\omega)(\bar{\sigma}_3-\sigma_0)]\\&+\mathbf{a}_3\cdot(\mathbf{a}_1\,\mathcal{O}_++\mathbf{a}_2\,\mathcal{O}_-)\sigma_1+\mathbf{a}_3\cdot(\mathbf{a}_2\,\mathcal{O}_+-\mathbf{a}_1\,\mathcal{O}_-)\sigma_2,\end{aligned}\tag{48c}$$

where the operators are defined by Eqs. (44), and refer to both parts of the LM, while $\eta(\omega)$ and σ_0 refer only to the thermal reservoir. Equations (48) reduce, of course, to Eqs. (24), equations for the case in which the LM consists of the thermal reservoir only, if we ignore $\mathrm{LM}^{(2)}$. This is accomplished by setting $\tilde{\eta}=\eta$, $\tilde{\chi}=\chi$ in Eqs. (44) and (46), and $\tilde{\kappa}=\kappa$ in Eqs. (48).

A considerable simplification occurs in the appearance of the equations of motion for the case of a magnetic dipole, where $\mathbf{a}_1=\hat{x}$, $\mathbf{a}_2=\hat{y}$, $\mathbf{a}_3=\hat{z}$, and for the case of a spatially linear electric dipole without permanent dipole moment, where we can set $\mathbf{a}_1=\hat{x}$, $\mathbf{a}_2=\mathbf{a}_3=0$. These cases are of sufficient interest to be described explicitly. For the magnetic dipole we have

$$\dot{\sigma}_1=-(\omega-\mathcal{O}_-)\sigma_2-(\mathcal{O}+\mathcal{O}_+)\sigma_1+f_2\sigma_3-f_3\sigma_2,\tag{49a}$$

$$\dot{\sigma}_2=(\omega-\mathcal{O}_-)\sigma_1-(\mathcal{O}+\mathcal{O}_+)\sigma_2+f_3\sigma_1-f_1\sigma_3,\tag{49b}$$

$$\dot{\sigma}_3=-2[\mathcal{O}_+(\sigma_3-\bar{\sigma}_3)+\eta(\omega)(\bar{\sigma}_3-\sigma_0)]+f_1\sigma_2-f_2\sigma_1,\tag{49c}$$

and, for the spatially linear electric dipole, we have

$$\dot{\sigma}_1=-\omega\sigma_2,\tag{50a}$$

$$\dot{\sigma}_2=\omega\sigma_1-f_1\sigma_3-\mathcal{O}\sigma_2,\tag{50b}$$

$$\dot{\sigma}_3=f_1\sigma_2-[\mathcal{O}_+(\sigma_3-\bar{\sigma}_3)+\eta(\omega)(\bar{\sigma}_3-\sigma_0)].\tag{50c}$$

III. DISCUSSION

Equations (48)–(50) are integrodifferential equations of motion, since the $\mathcal{O}$'s are integral operators. These equations may be reduced to differential equations only by approximations that will be discussed later. It should be pointed out that there is good reason for not being able to describe completely the behavior of our system by means of differential equations. Such equations can refer only to interactions that are localized in time. Although, in principle, any system is describable by such equations, the incompleteness of the description of the LM is compensated, somewhat, by endowing it with a memory. (For example, a single classical particle is described completely by instantaneous position and velocity. If we ignore the velocity, then a memory of position is helpful.) Systems with memory cannot be described, generally, by differential equations, but require integral—or integrodifferential—equations. Another view of the necessity of integral operators in the equations of motion is afforded by the observation that the constants referring to the loss mechanism, that occur as the result of operating with the $\mathcal{O}$'s, depend on the frequencies contained in the σ's. Only integral operators can distinguish between different frequency components and produce the frequency-dependent relaxation constants that are, obviously, required by the present theory.[6]

Equation (49c) shows that the term responsible for relaxation of the energy consists of two parts, one being $\mathcal{O}_+(\sigma_3-\bar{\sigma}_3)$, which refers to both $\mathrm{LM}^{(1)}$ and $\mathrm{LM}^{(2)}$, and the other being $\eta(\omega)(\bar{\sigma}-\sigma_0)$, which refers to $\mathrm{LM}^{(1)}$ only. The first part tends to produce a damping only in the rapid oscillation of σ_3, and does not affect the average. The second part tends to relax the average energy to thermal equilibrium. This division is intuitively reasonable, since all the identical two-level systems comprising $\mathrm{LM}^{(2)}$ have approximately the same average energy, and the total energy cannot be affected by the coupling among the individual systems, even though oscillations may be damped. The coupling to the thermal reservoir, however, will tend to relax this total energy to that of thermal equilibrium with the reservoir. Note that in the relaxation of the average energy, only the natural frequency ω is involved, and not the driving field frequency. This is due to the fact that the average energy tends to relax through spontaneous emission and thermally induced emission, and both of these processes have narrow frequency ranges approximately centered at ω.

A. Differential Equations

We consider now possible simplifications of the equations of motion through approximation. An obvious approximation consists of the neglect of the high-frequency variation of σ_3 in the dissipation terms. (This high-frequency variation of σ_3 is, at most, a second-order quantity. If η and the $\mathcal{O}$ operators are considered first-order quantities, then, at most, third-order quantities are being neglected.) This is equivalent to replacing σ_3 by $\bar{\sigma}_3$ in the dissipation terms, and the terms $\mathcal{O}_+(\sigma_3-\bar{\sigma}_3)$ drop out of Eqs. (48)–(50). Another, and more significant, approximation is the assumption that, as far as their values in the dissipation terms are concerned, σ_1 and σ_2 are approximately monochromatic. Such an assumption is justified only if $\mathbf{f}(t)$ is approximately monochromatic, and is equivalent to the neglect of harmonics in the dissipation terms. For a monochromatic function $\varphi_\nu(t)$ of frequency ν we have, from Eqs. (44),

$$\mathcal{O}\varphi_\nu(t)=\left[\tilde{\eta}(\nu)+\frac{1}{\nu}\tilde{\chi}(\nu)\frac{d}{dt}\right]\varphi_\nu(t),\tag{51a}$$

$$\begin{aligned}\mathcal{O}_+\varphi_\nu(t)=\frac{1}{2}\Big\{&[\tilde{\eta}(\nu+\omega)+\tilde{\eta}(\nu-\omega)]\\&+\frac{1}{\nu}[\tilde{\chi}(\nu+\omega)+\tilde{\chi}(\nu-\omega)]\frac{d}{dt}\Big\}\varphi_\nu(t),\end{aligned}\tag{51b}$$

[6] The memory effects in relaxation of a spin system are emphasized by P. N. Argyres and P. L. Kelley, Phys. Rev. 134, A98 (1964).

$$\mathcal{O}_-\varphi_\nu(t)=\frac{1}{2}\left\{-[\tilde{\chi}(\nu+\omega)-\tilde{\chi}(\nu+\omega)]+\frac{1}{\nu}[\tilde{\eta}(\nu+\omega)-\tilde{\eta}(\nu-\omega)]\frac{d}{dt}\right\}\varphi_\nu(t). \quad (51c)$$

These equations are equivalent to Eqs. (44), and may be taken as alternative definitions of the operators. Assuming that σ_1, σ_2, and σ_3 are approximately monochromatic—the first two with frequency ν and the third with frequency zero—for purposes of approximating the dissipation terms, we obtain *differential equations* in place of Eqs. (48), since the operators are now differential operators given by Eqs. (51), and $\bar{\sigma}_3$ has been replaced by σ_3. These differential equations will not, in general, apply to a transient situation that is obtained, for example, when the driving field is suddenly turned off, for then, the only frequency-determining element is the natural frequency ω of the TLS (and not the driving field frequency ν), and we can expect σ_1 and σ_2 to be approximately oscillatory with frequency ω. If, however, the driving field frequency is in the neighborhood of ω, then the differential equations resulting from the assumption of approximate monochromaticity will apply to both the steady and transient states. We write down the equations of motion for just this case, and we consider the driving field frequency to be sufficiently near resonance so that

$$\tilde{\eta}(\nu-\omega)\approx\tilde{\eta}(0), \quad (52a)$$

$$\tilde{\chi}(\nu-\omega)\approx\tilde{\chi}(0)=0. \quad (52b)$$

The permissible range of ν for this simplification of the equations of motion is thus determined by the spectral properties of the LM. A further approximation in the dissipation terms may be introduced by the fact that, near resonance, we have the approximate relationships[7]

$$\sigma_1\approx-\omega\sigma_2, \quad (53a)$$

$$\dot{\sigma}_2\approx\omega\sigma_1. \quad (53b)$$

Utilizing Eqs. (51)–(53), and making the assumption, discussed earlier, that $\tilde{\eta}(0)$ vanishes, it is easy to show that

$$\mathcal{O}_+\sigma_1-\mathcal{O}_-\sigma_2\approx0, \quad (54a)$$

$$\mathcal{O}_+\sigma_2+\mathcal{O}_-\sigma_1\approx0. \quad (54b)$$

Using all of the above approximations, the equations of motion for a near-resonance driving field—as well as for free decay—become

$$\dot{\sigma}_1=[-\omega+\mathbf{a}_1\cdot\mathbf{a}_2\eta(\omega)+a_2{}^2\tilde{\chi}(\omega)]\sigma_2+\mathbf{a}_2\cdot\mathbf{f}\sigma_3-\mathbf{a}_3\cdot\mathbf{f}\sigma_2-[a_2{}^2\tilde{\eta}(\omega)-\mathbf{a}_1\cdot\mathbf{a}_2\tilde{\chi}(\omega)]\sigma_1+\mathbf{a}_1\cdot\mathbf{a}_3\eta(\omega)(\sigma_3-\sigma_0)+\mathbf{a}_2\cdot\mathbf{a}_3[\tilde{\chi}(\omega)\sigma_3-\tilde{\kappa}], \quad (55a)$$

$$\dot{\sigma}_2=[\omega+\mathbf{a}_1\cdot\mathbf{a}_2\eta(\omega)-a_1{}^2\tilde{\chi}(\omega)]\sigma_1+\mathbf{a}_3\cdot\mathbf{f}\sigma_1-\mathbf{a}_1\cdot\mathbf{f}\sigma_3-[a_1{}^2\tilde{\eta}(\omega)+\mathbf{a}_1\cdot\mathbf{a}_2\tilde{\chi}(\omega)]\sigma_2+\mathbf{a}_2\cdot\mathbf{a}_3\eta(\omega)(\sigma_3-\sigma_0)-\mathbf{a}_1\cdot\mathbf{a}_3[\tilde{\chi}(\omega)\sigma_3-\tilde{\kappa}], \quad (55b)$$

$$\dot{\sigma}_3=\mathbf{a}_1\cdot\mathbf{f}\sigma_2-\mathbf{a}_2\cdot\mathbf{f}\sigma_1-(a_1{}^2+a_2{}^2)\eta(\omega)(\sigma_3-\sigma_0), \quad (55c)$$

for a general TLS. For a magnetic dipole, we have

$$\dot{\sigma}_1=-[\omega-\tilde{\chi}(\omega)]\sigma_2-\tilde{\eta}(\omega)\sigma_1+f_2\sigma_3-f_3\sigma_2, \quad (56a)$$

$$\dot{\sigma}_2=[\omega-\tilde{\chi}(\omega)]\sigma_1-\tilde{\eta}(\omega)\sigma_2+f_3\sigma_1-f_1\sigma_3, \quad (56b)$$

$$\dot{\sigma}_3=f_1\sigma_2-f_2\sigma_1-2\eta(\omega)(\sigma_3-\sigma_0), \quad (56c)$$

and for a spatially linear electric dipole, we have

$$\dot{\sigma}_1=-\omega\sigma_2, \quad (57a)$$

$$\dot{\sigma}_2=[\omega-\tilde{\chi}(\omega)]\sigma_1-f_1\sigma_3-\tilde{\eta}(\omega)\sigma_2, \quad (57b)$$

$$\dot{\sigma}_3=f_1\sigma_2-\eta(\omega)(\sigma_3-\sigma_0). \quad (57c)$$

If we neglect the reactive constants $\tilde{\chi}(\omega)$ and $\tilde{\kappa}$,[8] then Eqs. (55)–(57) are identical with those derived in II [Eqs. (II.43)–(II.45), respectively]. Except for the reactive constants $\tilde{\chi}(\omega)$, Eqs. (56) are identical with the Bloch equations.

It is important to bear in mind the conditions and approximations that led from the integrodifferential equations of motion Eqs. (48)–(50), to the differential equations of motion, Eqs. (55)–(57). These are: (1) the driving field is approximately monochromatic; (2) the fundamental frequency is near resonance; (3) harmonics in σ_1 and σ_2 and high-frequency oscillations (of fundamental frequency or higher) in σ_3 are neglected. Without the second condition, differential equations can be obtained for the steady state, but they are different from those for free decay.

It is natural to ask how many relaxation constants are contained in the equations of motion. The answer obviously depends on the number of frequencies that are contained in the σ's. Besides $\eta(\omega)$, we have $\tilde{\eta}(\omega)$, $\tilde{\eta}(\nu\pm\omega)$, $\tilde{\chi}(\omega)$, and $\tilde{\chi}(\nu\pm\omega)$ for each frequency ν. Of course, not all frequencies are of equal importance, and, furthermore, not all constants referring to a given frequency are of equal importance. In the case in which the driving field consists of the superposition of several frequencies we can expect the components of σ_1 and σ_2 having these frequencies, the fundamentals, to be important. If some linear combination of the fundamentals

[7] This can be seen from Eqs. (48) by noting that f/ω, η/ω, and $\tilde{\eta}/\omega$ are much smaller than unity. At resonance (in the steady state), σ_1 and σ_2 are of the order of magnitude of $(f/\tilde{\eta})\sigma_3$, so that $\omega\sigma_1$ and $\omega\sigma_2$ are much larger than $f\sigma_3$. Off resonance, on the other hand, σ_1 and σ_2 are of the order of magnitude of $(f/\omega)\sigma_3$, so that $\omega\sigma_1$ and $\omega\sigma_2$ are of the same order of magnitude as $f\sigma_3$. It is only near resonance, therefore, that the approximate equalities $\dot{\sigma}_1\approx-\omega\sigma_2$, $\dot{\sigma}_2\approx\omega\sigma_1$ hold.

[8] One effect of the reactive constant $\tilde{\chi}(\omega)$—for the two special cases considered in Eqs. (56) and (57), it is the only effect—is the introduction of a small frequency shift of relative magnitude $\tilde{\chi}(\omega)/\omega$. This shift is due to the reactive effect of the LM, and should be differentiated from that which is produced by the purely resistive effect of the LM and is of the order of magnitude of $\tilde{\eta}^2/\omega^2$. For a discussion of the latter, see Ref. 9.

equals the resonant frequency, we can also expect these resonant harmonics to be important. Thus there will be important relaxation constants corresponding to the fundamentals and the resonant harmonics.

B. Special Solution of the General Equations of Motion

It is not the purpose of the present article to study in detail the solution of the equations of motion derived here. For the case of an approximately monochromatic driving field near resonance, the solutions have already been discussed[9] and the more general case, especially the interesting situation of a driving field containing two frequencies that are linearly related to the resonant frequency, will be discussed in a forthcoming article. We will, however, treat here a particularly simple special case for which there exists an exact steady-state solution of the integrodifferential equations of motion.

We consider a magnetic dipole TLS subject to a rotating transverse driving field of arbitrary frequency ν. The equations that must be satisfied are Eqs. (49), with

$$f_1=if_0(e^{i\nu t}-e^{-i\nu t}), \tag{58a}$$

$$f_2=f_0(e^{i\nu t}+e^{-i\nu t}), \tag{58b}$$

$$f_3=0. \tag{58c}$$

We try a solution of the form

$$\sigma_1=Ae^{i\nu t}+A^*e^{-i\nu t}, \tag{59a}$$

$$\sigma_2=-i(Ae^{i\nu t}-A^*e^{-i\nu t}), \tag{59b}$$

$$\sigma_3=\bar{\sigma}_3=\text{constant}. \tag{59c}$$

Noting that for this form of the solution, the relationships

$$\dot{\sigma}_1=-\nu\sigma_2, \tag{60a}$$

$$\dot{\sigma}_2=\nu\sigma_1, \tag{60b}$$

apply, we obtain easily, by use of Eqs. (51),

$$\mathcal{O}\sigma_1=\tilde{\eta}(\nu)\sigma_1-\tilde{\chi}(\nu)\sigma_2, \tag{61a}$$

$$\mathcal{O}\sigma_2=\tilde{\eta}(\nu)\sigma_2+\tilde{\chi}(\nu)\sigma_1, \tag{61b}$$

$$\mathcal{O}_+\sigma_1=\tfrac{1}{2}\{[\tilde{\eta}(\nu+\omega)+\tilde{\eta}(\nu-\omega)]\sigma_1 -[\tilde{\chi}(\nu+\omega)+\tilde{\chi}(\nu-\omega)]\sigma_2\}, \tag{61c}$$

$$\mathcal{O}_+\sigma_2=\tfrac{1}{2}\{[\tilde{\eta}(\nu+\omega)+\tilde{\eta}(\nu-\omega)]\sigma_2 +[\tilde{\chi}(\nu+\omega)+\tilde{\chi}(\nu-\omega)]\sigma_1\}, \tag{61d}$$

$$\mathcal{O}_-\sigma_1=\tfrac{1}{2}\{-[\tilde{\chi}(\nu+\omega)-\tilde{\chi}(\nu-\omega)]\sigma_1 -[\tilde{\eta}(\nu+\omega)-\tilde{\eta}(\nu-\omega)]\sigma_2\}, \tag{61e}$$

$$\mathcal{O}_-\sigma_2=\tfrac{1}{2}\{-[\tilde{\chi}(\nu+\omega)-\tilde{\chi}(\nu-\omega)]\sigma_2 +[\tilde{\eta}(\nu+\omega)-\tilde{\eta}(\nu-\omega)]\sigma_1\}. \tag{61f}$$

Substitution into Eqs. (49) produces a cancellation of all terms containing the constants $\tilde{\eta}(\nu+\omega)$ and $\tilde{\chi}(\nu+\omega)$, and only terms containing the constants $\tilde{\eta}(\nu-\omega)$ and $\tilde{\chi}(\nu-\omega)$ are left, besides of course, the term containing the constant $\eta(\omega)$ which occurs explicitly in the equations of motion. The resulting algebraic equations for the σ's are

$$(\omega'-\nu)\sigma_2=-\eta'\sigma_1+f_2\sigma_3, \tag{62a}$$

$$-(\omega'-\nu)\sigma_1=-\eta'\sigma_2-f_1\sigma_3, \tag{62b}$$

$$\eta(\omega)(\sigma_3-\sigma_0)=-f_0(A+A^*), \tag{62c}$$

where

$$\omega'\equiv\omega-[\tilde{\chi}(\nu-\omega)+\tilde{\chi}(\nu)],$$

$$\eta'\equiv\tilde{\eta}(\nu-\omega)+\tilde{\eta}(\nu).$$

The solution of these algebraic equations yields

$$A=\frac{f_0\sigma_3[\eta'-i(\nu-\omega')]}{(\nu-\omega')^2+\eta'^2}, \tag{63a}$$

$$\sigma_3=\sigma_0\left[1+\frac{2f_0^2\eta'/\eta(\omega)}{(\nu-\omega')^2+\eta'^2}\right]^{-1}. \tag{63b}$$

We note that the resonance[10] is not exactly Lorentzian, owing to the fact that both η' and ω' vary as the driving field frequency ν passes through resonance. Although $\tilde{\eta}(\nu)$ and $\tilde{\chi}(\nu)$ may have little relative variation in the neighborhood of $\nu=\omega$, $\tilde{\eta}(\nu-\omega)$ may be expected to increase from zero on both sides of resonance, and $\tilde{\chi}(\nu-\omega)$ changes sign in going through resonance.

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[9] I. R. Senitzky, Phys. Rev. **135**, A1498 (1964). The solution of Eqs. (54) and (55), with the reactive constants neglected, is discussed in some detail in this reference.

[10] The rotating driving field described by Eqs. (58) rotates in the same sense as the free magnetic dipole for $\nu>0$, and in the opposite sense for $\nu<0$. Resonance results, of course, only when the sense of rotation of the field and dipole are the same.