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Dissipation in Quantum Mechanics. The Two-Level System

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The study of a two-level system (TLS) coupled to a loss mechanism (LM), the fluctuations of which are taken into account, is motivated by relating the problem to spin-lattice relaxation, radiation damping, and Brownian motion. A TLS of the electric dipole type (coupled to the LM through one variable only) is discussed first. The problem is formulated in terms of the Pauli spin matrices, and the Langevin equations for Brownian motion of a TLS are derived. Their solution—made possible by taking expectation values in LM space—contains the Weisskopf-Wigner exponential decay formula of radiation theory, as well as a second-order shift in frequency of the TLS produced by the (nondispersive) LM. A driving force is added to the problem and expressions are obtained for a driven lossy TLS of the electric dipole type. The same analysis is applied to a TLS of the magnetic dipole type and differential equations for the spin matrices—for which the expectation values in LM space has been taken—are obtained. Taking expectation values in TLS space converts these equations into the Bloch equations. Unlike the electric dipole case, no frequency shift is present.

INTRODUCTION

In two previous articles[1] the concept of dissipation in a quantum-mechanical system was discussed and an analysis was made of a lossy harmonic oscillator. The harmonic oscillator is, of course, one of the simplest as well as one of the most important systems in physics, and the analysis of its behavior has wide application. Another fundamental system is the two-level system (TLS), which is also an oscillator, and, like the harmonic oscillator, is described by a single frequency. It is, however, dissimilar to the harmonic oscillator in many other respects. Thus, the harmonic oscillator is one of the most important systems in classical as well as in quantum mechanics, while the TLS does not exist in classical mechanics; the harmonic oscillator is a linear system, while the TLS is a nonlinear system; the harmonic oscillator is related to Einstein-Bose statistics, while the TLS is related to Fermi-Dirac statistics.

The importance of the TLS lies not only in the fact that there are two-level systems in nature, but that any system with only one pair of levels spaced in correspondence with a given frequency (and a non-vanishing transition probability between these two levels) responds, approximately, to a perturbation having this frequency as though it were TLS. It is the purpose of the present article to discuss dissipation in the TLS.

It was easy to motivate the consideration of dissipation in a harmonic oscillator, even though one may think of dissipation as a macroscopic phenomenon, by pointing to a macroscopic system such as the electromagnetic field of a resonant cavity mode which needs, for certain purposes, quantum-mechanical treatment. There exists no macroscopic TLS and one does not, normally, think of lossy two-level oscillators. It was noted in I, however, that dissipation involves the weak interaction between a simple system and a large complex system that is described very incompletely, as in thermodynamics for instance, and is affected only slightly by the interaction. From this point of view a two-level oscillator with dissipation represents a two-level system in interaction with a thermal-reservoir type of environment. A good example of such an environment is a crystal lattice; another example is the radiation field of a large multimode cavity, or, in its limiting form, the radiation field of free space.[2] We may also include among the examples a system which subjects the TLS to sufficiently frequent weak collisions having random characteristics. Such a system will produce Brownian motion of the TLS. In II it was shown that the theory of the harmonic oscillator with dissipation is identical with the theory of Brownian motion of the harmonic oscillator, the fluctuations of the dissipation mechanism producing fluctuations of the harmonic oscillator coordinates. In the same manner, we can develop a theory of Brownian motion of a two-level system. Of course, there exists no position coordinate for a two-level system, so that the theory must be regarded as one of Brownian "motion" in a generalized sense.[3] The applicability of the theory of a TLS with dissipation is illustrated by the fact that in the following analysis there will be found a derivation of the Langevin equations for a TLS, of interest in the theory of Brownian motion; the Weisskopf-Wigner formula, of interest in radiation theory; and the Bloch

[1] I. R. Senitzky, Phys. Rev. **119**, 670 (1960); **124**, 642 (1961); referred to as I and II, respectively.

[2] The fact that the large complex system with which the TLS interacts is described very incompletely does not mean that more information about the system is not available. It merely means that this additional information will not be utilized, and certain detailed aspects of the interaction with a specific loss mechanism, which might serve to distinguish the effect of one loss mechanism from another, will not be considered.

[3] In Brownian motion of the TLS, the state of the TLS is considered to be changed at each collision by a random, but small, amount, so that the state after collision differs only slightly from that before collision. These are the weak collisions. There can also be, of course, strong collisions, after which the state of the TLS is unrelated to that before collision. If the strong collisions have a sufficiently low rate of occurrence, then the *average* behavior of a TLS in a large group of systems is also described by the present theory. R. Karplus and J. Schwinger [Phys. Rev. **73**, 1020 (1958)] have considered the strong collision case.

equations, of interest in the theory of spin-lattice coupling.

Our discussion will proceed, initially, in a manner somewhat similar to that of I, especially as far as the loss mechanism (LM) is concerned. Those considerations which are found in I will be summarized briefly for the sake of completeness; the earlier article should be consulted for a more detailed treatment of them, if such is desired. Section I is the basic part of the present article; it is an analysis of a TLS of the electric dipole type coupled to a loss mechanism, and includes the main ideas of the article. Section II contains the addition of a driving force to the problem under consideration, and Sec. III contains the consideration of a TLS of the magnetic dipole type.

I.

We consider a TLS coupled to a LM. The temporal development of the combined system will be described by the time-dependent operators of the Heisenberg picture. The representation will be that in which the energy of the uncoupled systems is diagonal.

The dynamical variables of the TLS are represented by 2×2 matrices, and it is well known that any 2×2 matrix can be specified as a linear superposition of the Pauli spin matrices together with the unit matrix. There are, therefore, only three linearly-independent dynamical variables available with which to describe the system, and the characteristics of the system determine their choice (as well as the relationship between them). Thus, if the TLS is an electric-dipole oscillator, it is usually described by its energy and dipole moment, the coupling to other systems taking place through the dipole moment. One need not refer to a third variable, but the time derivative of the dipole moment, or the current, which is proportional to the commutator of the above two variables, enters significantly into the calculation. On the other hand, if the TLS has a magnetic moment, it is described by the three components of angular momentum.

We will consider first a TLS of the electric dipole type, in which the coupling to the loss mechanism takes place through a single dynamical variable.[4] This variable and the energy of the TLS may be expressed as multiples of the Pauli spin operators σ_x and σ_z, respectively. For our purposes, it is important to realize that the well-known properties of Pauli spin matrices,[5]

$$\sigma_x^2=\sigma_y^2=\sigma_z^2=1\,, \tag{1a}$$

$$\{\sigma_x,\sigma_y\}=\{\sigma_x,\sigma_z\}=\{\sigma_y,\sigma_z\}=0\,, \tag{1b}$$

$$[\sigma_k,\sigma_l]=2i\sigma_m\,, \tag{1c}$$

apply not only when these matrices have their familiar time-independent form in the Schrödinger picture, but also when these matrices are time-dependent operators in the Heisenberg picture, the initial values being

$$\sigma_x(0)=\begin{pmatrix}0 & 1\\ 1 & 0\end{pmatrix},\quad \sigma_y(0)=\begin{pmatrix}0 & -i\\ i & 0\end{pmatrix},\quad \sigma_z(0)=\begin{pmatrix}1 & 0\\ 0 & -1\end{pmatrix}. \tag{2}$$

The proof that Eq. (1) applies in the time-dependent case follows from the observation that the Heisenberg operators are all derived from their initial values by the same unitary transformation, and Eqs. (1) clearly hold for the initial values.

The Hamiltonian for the coupled TLS and LM is

$$H=H_{\rm LM}+\tfrac{1}{2}\hbar\omega\sigma_z+\tfrac{1}{2}\hbar F\sigma_x\,, \tag{3}$$

where F is the coordinate (in appropriate units, and includes the coupling constant) through which the LM couples to the TLS, and where the two levels of the TLS are situated symmetrically about zero.

An important restriction of the situations considered is the requirement that the coupling between TLS and LM be weak. The condition of weak coupling may be expressed in several forms. One form is the statement that the forces exerted by the LM on the TLS be small compared to the internal forces of the TLS. This is a qualitative statement, since the internal details of the TLS are usually unknown. (Frequently, the consideration of a TLS is an idealization used to avoid involvment with internal details.) Another form, of phenomenological nature, is the requirement that the relative changes produced in the TLS by the LM in a single cycle (of the TLS) be small, or that significant changes in the TLS take place only during a time which is long compared to a period of oscillation of the TLS. In quantitative terms, this requirement means that the numbers associated with F be small compared to ω.

The LM is the same as that considered in I. It has many energy levels, closely spaced, and is affected only slightly by the TLS. It is described, for our purposes, by $H_{\rm LM}$ and F. In the uncoupled state, the diagonal matrix elements of F vanish. (This is a consequence of the requirement that the LM not exert a constant, or time-independent, force on the TLS.) The initial state of the LM is described by a diagonal density matrix

$$\rho_{mn}=\delta_{mn}Z^{-1}e^{-E_n/kT}\,, \tag{4}$$

[4] Although the electric dipole moment is a vector operator that consists of three components, these components are multiples of one another, and may be combined into a single operator in the description of the behavior of an electric dipole type of TLS. In the magnetic-dipole type of TLS, however, the three components of magnetic moment are linearly-independent (and non-commuting). It should be borne in mind that the components of $\boldsymbol{\sigma}$ in the electric dipole case are not related to spatial directions.

[5] The notation used is $\{A,B\}\equiv AB+BA$, and k, l, m stand for the cyclic permutation of x, y, z.

where Z is the partition function

$$Z=\sum_j e^{-E_j/kT}, \tag{4a}$$

and T is the LM temperature.

The equations of motion for the combined system are

$$\dot{\sigma}_x=-\omega\sigma_y, \tag{5a}$$

$$\dot{\sigma}_y=\omega\sigma_x-F\sigma_z, \tag{5b}$$

$$\dot{\sigma}_z=F\sigma_y, \tag{5c}$$

$$\dot{F}=-(i/\hbar)[F,H_{\rm LM}], \tag{5d}$$

$$\dot{H}_{\rm LM}=-\tfrac{1}{2}i[H_{\rm LM},F]\sigma_x. \tag{5e}$$

Although σ_y does not occur in the initial description of the TLS, it is a convenient variable to use, and may be regarded as defined by Eq. (5a). Equations (5d) and (5e) can be combined into

$$\dot{F}=\frac{1}{i\hbar}[F,H_{\rm LM}(0)]+\frac{1}{2\hbar}\int_0^t dt_1\,[F(t),[F(t_1),H_{\rm LM}(t_1)]\sigma_x(t_1)], \tag{6}$$

which may be rewritten as an integral equation,

$$F(t)=F^{(0)}(t)+\frac{1}{2\hbar}\int_0^t dt_1\int_0^{t_1}dt_2\,U(t-t_1)\times[F(t_1),[F(t_2),H_{\rm LM}(t_2)]\sigma_x(t_2)]U^{-1}(t-t_1), \tag{7}$$

where

$$U(\tau)=\exp[(i/\hbar)H_{\rm LM}(0)\tau], \tag{7a}$$

and where $F^{(0)}(t)$ is defined by

$$F^{(0)}(0)=F(0), \tag{7b}$$

$$\dot{F}^{(0)}(t)=-(i/\hbar)[F^{(0)}(t),H_{\rm LM}(0)]. \tag{7c}$$

It is evident that $F^{(0)}(t)$ is the dynamical variable for the uncoupled, or free, LM [since $H_{\rm LM}(0)$ is the constant Hamiltonian for the uncoupled LM]. From Eq. (4) and the vanishing of the diagonal matrix elements of $F^{(0)}$, it follows that

$$\langle F^{(0)}(t)\rangle=0. \tag{8}$$

As in I, a significant approximation will now be performed in the expression for F of Eq. (7).[6] In the integrand of the last term, we replace the LM variables by their values for the uncoupled LM, ignore the noncommutativity of σ_x with the LM variables, and replace the commutator of the LM variables by its expectation value (times the unit LM operator). The result is

$$F(t)=F^{(0)}(t)+\frac{1}{2\hbar}\int_0^t dt_1\int_0^{t_1}dt_2\times\langle[F^{(0)}(t_1),[F^{(0)}(t_2),H_{\rm LM}(0)]]\rangle\sigma_x(t_2). \tag{9}$$

Setting

$$F_{mn}{}^{(0)}(t)=F_{mn}{}^{(0)}(0)e^{i\omega_{mn}t}\equiv\tilde{F}_{mn}e^{i\omega_{mn}t}, \tag{10}$$

where

$$\omega_{mn}=(E_m-E_n)/\hbar, \tag{10a}$$

we obtain

$$\langle[F^{(0)}(t_1),[F^{(0)}(t_2),H_{\rm LM}(0)]]\rangle=2Z^{-1}\sum_{i,k}e^{-E_i/kT}\hbar\omega_{ik}|\tilde{F}_{ik}|^2\cos\omega_{ik}(t_1-t_2), \tag{11}$$

so that

$$F=F^{(0)}+Z^{-1}\sum_{i,k}e^{-E_i/kT}|\tilde{F}_{ik}|^2\times\int_0^t dt_1\sin\omega_{ik}(t-t_1)\sigma_x(t_1). \tag{12}$$

Labeling the number of states per unit energy range by $\rho(E)$ and denoting the average of $|\tilde{F}_{ik}|^2$ (averaged over small ranges about E_i and E_k) by $\tilde{F}^2(E_i,E_k)$, we can convert the summation of Eq. (12) into an integration, obtaining in Appendix A the expression

$$F(t)=F^{(0)}(t)-\frac{2}{\pi}\int_0^t dt_1\int_0^\infty d\omega'\,\xi(\omega')\sin\omega'(t-t_1)\sigma_x(t_1), \tag{13}$$

where

$$\xi(\omega')\equiv\tfrac{1}{2}\pi\hbar Z^{-1}B(\omega')[1-\exp(-\hbar\omega'/kT)] \tag{13a}$$

and

$$B(\omega')\equiv\int_0^\infty dE\,\rho(E+\hbar\omega')\rho(E)\times\tilde{F}^2(E+\hbar\omega',E)e^{-E/kT}. \tag{13b}$$

The last term in Eq. (13) can be simplified. We define first the non-Hermitian spin operators

$$\sigma_+=2^{-1/2}(\sigma_x+i\sigma_y),\quad\sigma_-=2^{-1/2}(\sigma_x-i\sigma_y), \tag{14}$$

which, in turn, give

$$\sigma_x=2^{-1/2}(\sigma_++\sigma_-),\quad\sigma_y=-2^{-1/2}i(\sigma_+-\sigma_-). \tag{15}$$

The equations of motion become, with $F'\equiv2^{-1/2}F$,

$$\dot{\sigma}_+=i\omega\sigma_+-iF'\sigma_z, \tag{16a}$$

$$\dot{\sigma}_-=-i\omega\sigma_-+iF'\sigma_z, \tag{16b}$$

$$\dot{\sigma}_z=-iF'(\sigma_+-\sigma_-). \tag{16c}$$

If we set

$$\sigma_+(t)=\varphi_+(t)e^{i\omega t},\quad\sigma_-(t)=\varphi_-(t)e^{-i\omega t}, \tag{17}$$

[6] This approximation is discussed in detail in I. It involves the assumption that the LM is affected only slightly by its interaction with the TLS, the neglect of quantum-mechanical correlation effects in terms of higher order than the second, and the realization that final results will be expectation values with respect to the LM.

it is evident that φ_+ and φ_- are constants in the absence of coupling to the LM (which means $F=0$), and the requirement of weak coupling implies that $\varphi_\pm$ varies much more slowly than $\exp(\pm i\omega t)$. Substituting from Eq. (17) into Eq. (13), we have

$$
\begin{aligned}
F(t)=F^{(0)}(t)+\frac{i}{2^{1/2}\pi}\int_0^t dt_1\int_0^\infty d\omega'\,\xi(\omega') \\
\times\{\varphi_+(t_1)[\exp(i\omega' t-i\{\omega'-\omega\}t_1) \\
-\exp(-i\omega' t+i\{\omega'+\omega\}t_1)] \\
+\varphi_-(t_1)[\exp(i\omega' t-i\{\omega'+\omega\}t_1) \\
-\exp(-i\omega' t+i\{\omega'-\omega\}t_1)]\}. \quad (18)
\end{aligned}
$$

If the t_1 integration is considered first, it is seen that the main contribution to the integral, for

$$t\gg\omega^{-1}, \quad (19)$$

comes from those values of ω' that are in the neighborhood of ω. Based on this fact, several approximations can be made. We drop the terms in the integrand containing the factor $\exp[\pm i(\omega'+\omega)t_1]$, since these terms oscillate rapidly, and we take $\xi(\omega')$ outside of the ω' integral as $\xi(\omega)\equiv\xi$, assuming that $\xi(\omega')$ changes little in the significant range of ω'. The result is

$$
\begin{aligned}
F(t)&=F^{(0)}(t)+\frac{i\xi}{2^{1/2}\pi}\int_0^t dt_1\int_0^\infty d\omega' \\
&\quad\times\{\varphi_+(t_1)\exp[i\omega' t-i(\omega'-\omega)t_1] \\
&\quad-\varphi_-(t_1)\exp[-i\omega' t+i(\omega'-\omega)t_1]\} \\
&=F^{(0)}(t)+\frac{i\xi}{2^{1/2}\pi}\int_0^t dt_1\int_{-\omega}^\infty d\nu \\
&\quad\times\{\varphi_+(t_1)\exp[i\omega t-i\nu(t_1-t)] \\
&\quad-\varphi_-(t_1)\exp[-i\omega t+i\nu(t_1-t)]\}, \quad (20)
\end{aligned}
$$

where the substitution $\nu=\omega'-\omega$ has been made. Since the main contribution to the integral comes from $\nu\sim 0$, we change the lower limit of the ν integration to $-\infty$, obtaining,

$$
\begin{aligned}
F(t)=F^{(0)}(t)+2^{1/2}i\xi\int_0^t dt_1\,\delta(t_1-t) \\
\times[\varphi_+(t_1)e^{i\omega t}-\varphi_-(t_1)e^{-i\omega t}], \quad (21)
\end{aligned}
$$

which, from Eq. (15), gives the simple relationship

$$F(t)=F^{(0)}(t)-\xi\sigma_y(t). \quad (22)$$

It is useful to examine further the approximations by which Eq. (22) is derived from Eq. (18). In the latter, we have an integration over both ω' and t_1. The ω' integration selects those values of t_1 that are near t, and the t_1 integration selects those values of ω' that are near ω. Neither selection, or localization, is infinitely sharp, in the sense of a δ function; this is fortunate, for the two localizations work against each other. If there were complete localization in ω' there would be no localization in t_1, and vice versa. Thus, a small region about ω and a small region below t contribute to the double integral in Eq. (18). The δ function in Eq. (21) is obviously an idealization of a localized function which has both finite (rather than infinite) height and finite (rather than infinitesmal) width. It is reasonable to consider this width, which we label τ, and which may be regarded as the memory time of the LM, to have an order of magnitude large compared to ω^{-1} but small compared to the times during which secular changes take place, that is, times during which significant changes occur in the slowly varying quantities. If such a function is used in Eq. (21) instead of the δ function, then the term containing the integral goes continuously from zero to $-\xi\sigma_y(t)$ in a time of the order of τ. We write therefore, instead of Eq. (22), the relationship

$$F(t)=F^{(0)}(t)-\bar{\xi}(t)\sigma_y(t), \quad (23)$$

where $\bar{\xi}(t)$ is a (c-number) function (multiplied by the unit operator in LM space) that increases continuously from zero to ξ in a time τ and remains constant after that. Although, for most purposes, the difference between Eqs. (22) and (23) is insignificant, the latter satisfies the initial conditions for the σ's and will make it possible to satisfy the other initial conditions of our problem. (The initial conditions that have been imposed on F and the σ's imply that the coupling between the TLS and LM is turned on at $t=0$.)

There will be need later for an evaluation of $\langle F^{(0)}(t_1)F^{(0)}(t_2)\rangle$. Using the same methods as those employed in going from Eq. (9) to Eq. (13), we obtain in Appendix B

$$
\begin{aligned}
\langle F^{(0)}(t_1)F^{(0)}(t_2)\rangle=\frac{2}{\pi}\int_0^\infty d\omega'\,[\eta(\omega')\cos\omega'(t_1-t_2) \\
-i\xi(\omega')\sin\omega'(t_1-t_2)], \quad (24)
\end{aligned}
$$

where

$$\eta(\omega')=\tfrac{1}{2}\pi\hbar Z^{-1}B(\omega')[1+\exp(-\hbar\omega'/kT)]. \quad (24a)$$

[It is to be noticed that $\eta(\omega')$ differs from $\xi(\omega')$ only in the sign of the exponential.] In most of the later applications of Eq. (24) $\langle F^{(0)}(t_1)F^{(0)}(t_2)\rangle$ is one factor of an integrand, the other factor being approximately an oscillatory function of t_1 (or t_2) with angular frequency ω, and the integration being over t_1 (or t_2). As in the case of Eq. (13), the time integration localizes the region of ω' which contributes to the integral on the right side of Eq. (24). Using reasoning similar to that employed in going from Eq. (18) to Eq. (20), we have, for purposes of the above applications,

$$\langle F^{(0)}(t_1)F^{(0)}(t_2)\rangle = 2\left[\eta\delta(t_1-t_2) - \frac{i}{\pi}\xi\frac{\mathcal{P}}{t_1-t_2}\right], \quad (25)$$

and

$$\langle\{F^{(0)}(t_1),F^{(0)}(t_2)\}\rangle = 4\eta\delta(t_1-t_2), \quad (26)$$

where η stands for $\eta(\omega)$.

If the expression for $F(t)$ contained in Eq. (23) is substituted into the equations of motion for the TLS [Eqs. (5a)–(5c)], a complete set of equations of motion for the TLS alone is obtained, since the LM enters into resulting equations only through $F^{(0)}(t)$, which may be considered as a prescribed function (or operator), given by Eqs. (8) and (24) or (25). (Additional properties of $F^{(0)}(t)$, not needed for the present discussion, are given in II.) However, while $F(t)$ commutes with the TLS operators evaluated at time t, $F^{(0)}(t)$ does not necessarily commute with these operators, and to make full use of the equations, the commutators for $F^{(0)}$ and the σ's are needed. These commutators may be obtained from Eq. (23) together with the relationship

$$[F(t),\sigma_k(t)]=0, \quad (27)$$

where k stands for x, y, or z. The result is

$$[\sigma_y(t),F^{(0)}(t)]=0, \quad (28a)$$

$$[\sigma_x(t),F^{(0)}(t)]=2i\xi\sigma_z, \quad (28b)$$

$$[\sigma_z(t),F^{(0)}(t)]=-2i\xi\sigma_x. \quad (28c)$$

For the equations of motion of the TLS alone, we obtain

$$\dot{\sigma}_x=-\omega\sigma_y, \quad (29a)$$

$$\dot{\sigma}_y=\omega\sigma_x-\tfrac{1}{2}\{F^{(0)},\sigma_z\}, \quad (29b)$$

$$\dot{\sigma}_z=F^{(0)}\sigma_y-\xi. \quad (29c)$$

In the language of the theory of Brownian motion, Eqs. (29) are the *Langevin equations* for a TLS, with $F^{(0)}$ being the randomly fluctuating force (in appropriate units) acting on the TLS (see II). Equations (29), together with the initial conditions given by Eqs. (2), are sufficient, in principle, to determine the σ's. It is seen that the σ's, while being initially operators in TLS space only, become operators in both TLS and LM spaces as t increases, since the derivatives of the σ's contain $F^{(0)}$.

Before considering a solution of Eqs. (29), it is of interest to look at certain properties of the solutions which may be obtained easily from these equations. Using Eqs. (29) as the expressions for the time derivatives of the σ's, we obtain by means of Eqs. (28) and (1)

$$\sigma_k\dot{\sigma}_k+\dot{\sigma}_k\sigma_k=0, \quad (30a)$$

$$\sigma_k\dot{\sigma}_l+\dot{\sigma}_k\sigma_l=i\sigma_m, \quad (30b)$$

$$\sigma_k\dot{\sigma}_m+\dot{\sigma}_k\sigma_m=-i\dot{\sigma}_l, \quad (30c)$$

where k, l, and m stand for the cyclic permutations of x, y, and z. These equations may be combined to give

$$(d/dt)\sigma_k^2=0, \quad (d/dt)\{\sigma_p,\sigma_q\}=0, \quad (d/dt)[\sigma_k,\sigma_l]=2i\dot{\sigma}_m. \quad (31)$$

Now, Eqs. (31) are just the equations obtained by differentiating both sides of Eqs. (1). This means that if the solution of Eqs. (29) satisfy Eqs. (1) at time t, they will also satisfy them at time $t+\Delta t$. Since the initial values of the σ's, as given in Eqs. (2), satisfy Eqs. (1), the σ's must satisfy Eqs. (1) for all values of t. The solutions of Eqs. (29) having initial values given by Eqs. (2) are, therefore, true spin operators. It should be remembered, however, that they must now operate in both TLS and LM spaces. If the expectation value is taken in either space, these operator properties will be destroyed.

The solution of Eqs. (29) will now be considered. Equations (29a) and (29b) yield

$$\ddot{\sigma}_x=-\omega^2\sigma_x+\tfrac{1}{2}\omega\{F^{(0)},\sigma_z\}, \quad (32)$$

$$\ddot{\sigma}_y=-\omega^2\sigma_y-\tfrac{1}{2}(d/dt)\{F^{(0)},\sigma_z\}. \quad (33)$$

These equations may be rewritten as integral equations. We have

$$\sigma_x(t)=\sigma_x^{(0)}(t)+\tfrac{1}{2}\int_0^t dt_1\,\sin\omega(t-t_1)\{F^{(0)}(t_1),\sigma_z(t_1)\}, \quad (34)$$

$$\sigma_y(t)=\sigma_y^{(0)}(t)-\tfrac{1}{2}\int_0^t dt_1\,\cos\omega(t-t_1)\{F^{(0)}(t_1),\sigma_z(t_1)\}, \quad (35)$$

where $\sigma_x^{(0)}(t)$ and $\sigma_y^{(0)}(t)$ describe the free TLS (and satisfy the initial conditions):

$$\sigma_x^{(0)}(t)=\begin{pmatrix}0 & e^{i\omega t}\\ e^{-i\omega t} & 0\end{pmatrix}, \quad (34a)$$

$$\sigma_y^{(0)}(t)=\begin{pmatrix}0 & -ie^{i\omega t}\\ ie^{-i\omega t} & 0\end{pmatrix}. \quad (35a)$$

Substituting from Eq. (35) into Eq. (29c) (where $F^{(0)}\sigma_y$ is rewritten as a symmetrized product for convenience), we obtain

$$\dot{\sigma}_z=-\xi-\tfrac{1}{4}\int_0^t dt_1\,\cos\omega(t-t_1)\{F^{(0)}(t),\{F^{(0)}(t_1),\sigma_z(t_1)\}\} + \int_0^t dt_1\,F^{(0)}(t_1)\sigma_y^{(0)}(t_1). \quad (36)$$

This is a differentio-integral equation for σ_z. In order to be able to solve it, we take the expectation value of both sides in LM space. The last term drops out, since

$$\langle F^{(0)}(t_1)\sigma_y^{(0)}(t_1)\rangle_{\mathrm{LM}}=\langle F^{(0)}(t_1)\rangle\sigma_y^{(0)}(t_1)=0, \quad (37)$$

according to Eq. (8), and the result is

$$\dot{\sigma}_z = -\bar{\xi} - \tfrac{1}{4}\int_0^t dt_1 \cos\omega(t-t_1) \times \langle\{F^{(0)}(t),\{F^{(0)}(t_1),\sigma_z(t_1)\}\}\rangle_{\mathrm{LM}}. \quad (38)$$

As illustrated by the form of Eq. (37), no special notation will be used, in general, to denote the expectation value of the σ's in LM space, in order not to give the impression that expectation values are taken in TLS space. When not evident from context and necessary for clarity, the notation $\langle\ \rangle_{\mathrm{LM}}$ will be used to indicate expectation value in LM space only. Without subscript, the brackets will indicate expectation value in both spaces when enclosing the σ's, and expectation value in LM space when enclosing the $F^{(0)}$'s.

An approximation will now be performed that may be regarded as the second essential approximation of the present theory. It is expressed by

$$\langle\{F^{(0)}(t),\{F^{(0)}(t_1),\sigma_z(t_1)\}\}\rangle_{\mathrm{LM}} \approx 2\langle\{F^{(0)}(t),F^{(0)}(t_1)\}\rangle_{\mathrm{LM}}\langle\sigma_z(t_1)\rangle_{\mathrm{LM}}, \quad (39)$$

which, according to the above notation convention, may be written as

$$2\langle\{F^{(0)}(t),F^{(0)}(t_1)\}\rangle\sigma_z(t_1). \quad (40)$$

(The factor 2 comes from the dropping of one symmetrizing bracket.) This approximation consists of the neglect of the noncommutativity of σ_z and the $F^{(0)}$'s, and the replacement of the expectation value of the product of σ_z and the LM variables by the product of the expectation values. It should be noted that the noncommutativity of the LM variables themselves has not been neglected and that the expectation value of the product of the LM variables is *not* split into a product of expectation values. The approximation of Eq. (39) is similar to—but less drastic than—the first essential approximation that was made in going from Eq. (7) to Eq. (9), the latter containing, in addition to the present approximation, the replacement of F by $F^{(0)}$ and the replacement of an LM operator by its expectation value. Replacing the expectation value in (38) by the expression (40) and making use of Eq. (26), we obtain

$$\dot{\sigma}_z = -\bar{\xi} - \eta\sigma_z. \quad (41)$$

This is an operator equation. σ_z is no longer an operator in LM space, but it is still an operator in TLS space (or spin space). It is, therefore, a 2×2 matrix. The first term on the right side of Eq. (41) is a multiple of the unit matrix. The equation for the matrix elements is thus

$$(d/dt)(\sigma_z)_{ij} = -\bar{\xi}\delta_{ij} - \eta(\sigma_z)_{ij}. \quad (42)$$

In solving this equation, we approximate by ignoring the short initial time variation of $\bar{\xi}$, and replace $\bar{\xi}$ by ξ. The solution is then, in view of the initial conditions of Eq. (2),

$$\sigma_z = \begin{pmatrix} (1-\sigma_0)e^{-\eta t}+\sigma_0 & 0 \\ 0 & (-1-\sigma_0)e^{-\eta t}+\sigma_0 \end{pmatrix}, \quad (43)$$

where

$$\sigma_0 = -\frac{\xi}{\eta} = -\frac{1-\exp(-\hbar\omega/kT)}{1+\exp(-\hbar\omega/kT)}. \quad (43a)$$

It is not possible to substitute this solution into Eqs. (34) and (35) in order to solve for σ_x and σ_y. The reason is the fact that in Eqs. (34) and (35) σ_z must still be an operator in LM space, since it enters there into first-order terms. From Eq. (29c) we have

$$\sigma_z = \sigma_z(0) - \xi t + \int_0^t dt_1\, F^{(0)}(t_1)\sigma_y(t_1). \quad (44)$$

Substituting from this expression into Eq. (35), we have an equation for σ_y that is the analog of Eq. (36) for σ_z. Performing the same operations as those which led from Eq. (36) to Eq. (41), we obtain

$$\sigma_y(t) = \sigma_y^{(0)}(t) - \eta\int_0^t dt_1\, \sigma_y(t_1)\cos\omega(t-t_1). \quad (45)$$

This integral equation may be rewritten in more familiar form as a differential equation,

$$\ddot{\sigma}_y + \eta\dot{\sigma}_y + \omega^2\sigma_y = 0, \quad (46)$$

with the initial conditions being determined by Eq. (45). Equation (46) is, of course, the differential equation for a damped harmonic oscillator, with solution

$$\sigma_y = \sigma_y(0)\exp[\pm i\tilde{\omega}t - \tfrac{1}{2}\eta t], \quad (47)$$

where

$$\tilde{\omega} = \omega[1-\tfrac{1}{4}(\eta^2/\omega^2)]^{1/2}, \quad (47a)$$

so that, in matrix form,

$$\sigma_y = e^{-(1/2)\eta t}\begin{pmatrix} 0 & -i\exp(i\tilde{\omega}t) \\ i\exp(-i\tilde{\omega}t) & 0 \end{pmatrix}. \quad (48)$$

The solution for σ_x can now be obtained most directly from Eq. (29a), which holds both for the operator form and for the (LM) expectation values of σ_x and σ_y. The result is

$$\sigma_x = \sigma_x(0) + \omega\sigma_y(0)\frac{\pm i\tilde{\omega}-\frac{1}{2}\eta}{\tilde{\omega}^2+\frac{1}{4}\eta^2}[1-\exp(\pm i\tilde{\omega}-\tfrac{1}{2}\eta)t] \approx \sigma_x(0) \mp i\sigma_y(0)[1-\exp(\pm i\tilde{\omega}-\tfrac{1}{2}\eta)t], \quad (49)$$

which becomes, in matrix form,

$$\sigma_x = e^{-(1/2)\eta t}\begin{pmatrix} 0 & \exp(i\tilde{\omega}t) \\ \exp(-i\tilde{\omega}t) & 0 \end{pmatrix}. \quad (50)$$

Equations (43), (48), and (50) are the solution of our problem. It can be verified directly that the σ's given by these equations are not true spin operators satisfying Eqs. (1), in accordance with previous discussion. They are, however, sufficient to yield answers to certain significant questions. In particular, "complete" expectation values (in both TLS and LM spaces) can be obtained immediately. We proceed to consider these.

Let the initial state of the TLS be given by

$$\psi=a_1\varphi_1+a_2\varphi_2, \tag{51}$$

where

$$|a_1|^2+|a_2|^2=1, \tag{51a}$$

and where φ_1 and φ_2 are the upper- and lower-energy states, respectively. Then the expectation value of the energy is given by

$$\tfrac{1}{2}\hbar\omega\langle\sigma_z\rangle=\tfrac{1}{2}\hbar\omega[(|a_1|^2-|a_2|^2)e^{-\eta t}+\sigma_0(1-e^{-\eta t})]. \tag{52}$$

It is seen that the energy expectation value decays from its initial value with a decay constant η to an equilibrium value

$$\tfrac{1}{2}\hbar\omega\sigma_0=-\tfrac{1}{2}\hbar\omega\frac{1-\exp(-\hbar\omega/kT)}{1+\exp(-\hbar\omega/kT)}, \tag{53}$$

which is in accordance with the Boltzman distribution law for a system with energy levels at $\frac{1}{2}\hbar\omega$ and $-\frac{1}{2}\hbar\omega$.

The expectation value of the "electric-dipole moment" (in dimensionless units) is, from Eq. (50),

$$\langle\sigma_x\rangle=2|a_1a_2|e^{-(1/2)\eta t}\cos(\tilde{\omega}t+\alpha), \tag{54}$$

where we have set

$$a_1=|a_1|e^{i\alpha_1},\quad a_2=|a_2|e^{i\alpha_2},\quad \alpha\equiv\alpha_2-\alpha_1. \tag{54a}$$

Similarly, the expectation value of the "current" is obtained from Eq. (48) as

$$-\omega\langle\sigma_y\rangle=-2\omega|a_1a_2|e^{-(1/2)\eta t}\sin(\tilde{\omega}t+\alpha). \tag{55}$$

We see that the dipole moment and current expectation values decay one half as fast as the energy. It is also interesting to note that the LM shifts the TLS frequency slightly, the shift being given by Eq. (47a).[7]

A comparison of the above results with those for the harmonic oscillator, found in I, shows some similarities. The ratio of the decay constant of the TLS energy to that of the dipole moment or current may be compared to the same ratio of the decay constant of the harmonic oscillator energy to that of the position or momentum. It is to be noted, however, that the energy decay constants themselves have a different dependence on the temperature. If the coupling between the harmonic oscillator and LM is such that the decay constants are the same at low temperature ($kT\ll\hbar\omega$), the ratio of η to the harmonic oscillator (energy) decay constant β is

$$\frac{\eta}{\beta}=\frac{1+\exp(-\hbar\omega/kT)}{1-\exp(-\hbar\omega/kT)}. \tag{56}$$

It is interesting to note that the ratio of the loss constants is the inverse of the ratio of the corresponding thermal-equilibrium energies when the ground-state energy of both systems is taken to be zero. Another similarity between the TLS (of the electric dipole type) and harmonic oscillator is the frequency shift introduced by the loss [Eq. (47a)]. In terms of the energy decay constant, this shift is identical for both systems.

As mentioned in the introduction, the radiation field of a large cavity—and in the limit, free space—may be represented by our LM. Consider now zero temperature and the TLS initially in the upper state. Then

$$a_2=0,\quad |a_1|=1,\quad \xi=\eta,\quad \text{and}\quad \langle\sigma_z\rangle=2e^{-\eta t}-1. \tag{57}$$

In the Schrödinger picture, using superposition constants $a_1^s(t)$ and $a_2^s(t)$, we have

$$\langle\sigma_z\rangle=|a_1^s(t)|^2-|a_2^s(t)|^2. \tag{58}$$

Equating the right side of Eqs. (57) and (58), and making use of the normalization condition of Eq. (51a), we obtain

$$|a_1^s(t)|^2=e^{-\eta t}, \tag{59}$$

which is essentially the result of Weisskopf and Wigner[8] for the time dependence of the wave function of a "two-level" atom radiating into space.

II.

We come now to the problem of a driven TLS. The Hamiltonian of Eq. (3) is supplemented by an additional term and becomes

$$H=H_{\mathrm{LM}}+\tfrac{1}{2}\hbar\omega\sigma_z+\tfrac{1}{2}\hbar\sigma_xF+\tfrac{1}{2}\hbar\sigma_xf, \tag{60}$$

where the c-number $f(t)$ is the driving force (in appropriate units), and obviously couples to the TLS through the same coordinate as the LM. We impose on f a condition of weak coupling similar to that imposed on F. This condition is expressed by the requirement

$$f\ll\omega, \tag{61}$$

and implies that significant secular changes produced by the driving force can occur only in a time large compared to a period. Quantities which previously were slowly varying compared to $\exp(i\omega t)$ continue to be slowly varying.

The relationships referring to the LM derived

[7] It should be borne in mind that the LM properties are *non-dispersive.* This is implicit in the assumption that the variation of $\xi(\omega')$ and $\eta(\omega')$ in the significant neighborhood of ω is sufficiently small to be neglected [in going from Eq. (18) to Eq. (20)]. Dispersive characteristics would produce a lower-order shift than that given by Eq. (47a). In the language of electrical circuits, the LM of the present article acts as a pure resistance (without reactance).

[8] V. Weisskopf and E. Wigner, Z. Physik **63**, 54 (1930); see also W. Heitler, *The Quantum Theory of Radiation* (Oxford University Press, New York, 1954), 3rd ed., p. 182.

previously remain unaffected. In particular, Eqs. (8), (24), (25), and (22) are unchanged. Instead of Eqs. (29) (the Langevin equations) we now obtain in exactly the same manner,

$$\dot{\sigma}_x = -\omega\sigma_y, \tag{62a}$$

$$\dot{\sigma}_y = \omega\sigma_x - \tfrac{1}{2}\{F^{(0)},\sigma_z\} - f\sigma_z, \tag{62b}$$

$$\dot{\sigma}_z = F^{(0)}\sigma_y + f\sigma_y - \bar{\xi}. \tag{62c}$$

The commutation relationships of Eq. (28) are unchanged and Eqs. (31) still apply, so that the solutions of Eqs. (62) are true spin operators in accordance with Eqs. (1).

As in the case of the solution of Eqs. (29), we proceed to solve Eqs. (62) by taking expectation values in LM space and making the same type of approximation in the second order terms as those used in the previous solution. From Eqs. (62a) and (62b) we have

$$\sigma_y = \sigma_y^{(0)} - \tfrac{1}{2}\int_0^t dt_1 \cos\omega(t-t_1) \times \{F^{(0)}(t_1) + f(t_1), \sigma_z(t_1)\}, \tag{63}$$

which is substituted into the $F^{(0)}\sigma_y$ term only (and not into the $f\sigma_y$ term) of Eq. (62c). Writing $F^{(0)}\sigma_y$ as a symmetrized product, this term is

$$\tfrac{1}{2}\{F^{(0)}(t),\sigma_y(t)\} = F^{(0)}(t)\sigma_y^{(0)}(t) - \tfrac{1}{4}\int_0^t dt_1 \cos\omega(t-t_1)\{F^{(0)}(t),\{F^{(0)}(t_1),\sigma_z(t_1)\}\} - \tfrac{1}{2}\int_0^t dt_1 \cos\omega(t-t_1)\{F^{(0)}(t), f(t_1)\sigma_z(t_1)\}. \tag{64}$$

The last term on the right side of Eq. (64) is treated as a second-order term, since both $F^{(0)}$ and f are small quantities of first order. In accordance with our approximation scheme, this term vanishes when expectation values are taken in LM space. The first term also vanishes in this process (without approximation), and the second term is approximated according to Eq. (39), yielding

$$\tfrac{1}{2}\langle\{F^{(0)}(t),\sigma_y(t)\}\rangle_{\mathrm{LM}} = -\eta\sigma_z(t), \tag{65}$$

where use has been made of Eq. (26). We similarly substitute from the expression for σ_z obtained by integrating Eq. (62c) into the $\frac{1}{2}\{F^{(0)},\sigma_z\}$ term of Eq. (62b) to obtain in exactly the same manner

$$\tfrac{1}{2}\langle\{F^{(0)}(t),\sigma_z(t)\}\rangle_{\mathrm{LM}} = \eta\sigma_y(t). \tag{66}$$

Equations (62) therefore become, in terms of expectation values in LM space,

$$\dot{\sigma}_x = -\omega\sigma_y, \tag{67a}$$

$$\dot{\sigma}_y = \omega\sigma_x - \eta\sigma_y - f\sigma_z, \tag{67b}$$

$$\dot{\sigma}_z = -\xi - \eta\sigma_z + f\sigma_y, \tag{67c}$$

where, as an approximation for computational purposes, $\bar{\xi}$ has been replaced by ξ. [For $f=0$, we have here, of course, the case previously treated and the transformation of Eqs. (29) into those for expectation values in LM space.] Equations (67), subject to the initial conditions given by Eqs. (2), are now the equations of motion in TLS (spin) space only. They are equations for three 2×2 matrices, or twelve equations for the twelve matrix elements. The first two equations [(67a) and (67b)] have the same form for all the matrix elements, but the third equation has a different form for the diagonal elements than for the off-diagonal elements, since the ξ term is a multiple of the unit matrix. We can convert Eqs. (67) into three equations for three unknowns by taking expectation values in TLS space. Since the equations are linear in the σ's they remain formally unchanged, and now all the σ's stand for the corresponding expectation values.

The solution of Eqs. (67) depends, of course, on the driving field $f(t)$. If we consider the situation in which

$$f(t) = f_0\cos(\nu t+\theta), \quad |\nu-\omega| \ll \omega,$$

then the (approximate) solution of these equations poses no problem in principle, but is tedious for arbitrary ν and t. The result consists of transients (exponentially damped) and a steady-state term. It is shown in Appendix C that for the case of resonance ($\nu=\tilde{\omega}$) we have, approximately,

$$\langle\sigma_z\rangle = \langle\sigma_z(0)\rangle e^{-(3/4)\eta t}[\cos\Omega t - (\eta/4\Omega)\sin\Omega t] + \frac{\xi}{\eta^2+\frac{1}{2}f_0^2}\left\{-\eta + e^{-(3/4)\eta t}\left[\eta\cos\Omega t - \tfrac{1}{2}(f_0^2+\tfrac{1}{2}\eta^2)\frac{\sin\Omega t}{\Omega}\right]\right\} + 2|a_1a_2|\sin(\alpha-\theta)\left[-(\eta/f_0)e^{-(1/2)\eta t} + e^{-(3/4)\eta t}\left(\frac{\eta}{f_0}\cos\Omega t + \frac{f_0^2+\frac{1}{2}\eta^2}{2f_0}\frac{\sin\Omega t}{\Omega}\right)\right], \tag{68}$$

where

$$\Omega \equiv \tfrac{1}{2}[f_0^2 - \tfrac{1}{4}\eta^2]^{(1/2)}; \tag{68a}$$

in the more general case $\nu \neq \tilde{\omega}$ but $|\nu-\tilde{\omega}| \ll \tilde{\omega}$, we have for the steady-state solution

$$\langle\sigma_z\rangle = -\frac{\xi}{\eta}\frac{\eta^2+4(\Delta\omega)^2}{\frac{1}{2}f_0^2+\eta^2+4(\Delta\omega)^2}, \tag{69}$$

where

$$\Delta\omega \equiv \nu - \tilde{\omega}. \tag{69a}$$

Equations (68) and (69) give the expectation value of the TLS energy in units of $\frac{1}{2}\hbar\omega$. The approximations used in obtaining these equations remove the distinction between $\tilde{\omega}$ and ω.

It is seen from Eq. (69) that the steady state value of the energy is a maximum (for constant driving field amplitude) when $\nu=\tilde{\omega}$, and approaches the arithmetic mean of the two levels as the driving field increases. This is the usual saturation phenomenon. If the TLS is initially in an energy state, then the change from the initial value to the steady-state value at resonance can be one of two types: A monotonic approach if $f_0^2 \leqslant \frac{1}{4}\eta^2$ and a damped oscillatory approach if $f_0^2 > \frac{1}{4}\eta^2$. The last term in Eq. (68) gives the effect of the phase relationships between the initial TLS oscillation and the driving field. [α determines the phase of the TLS oscillation, according to Eqs. (51) and (54a), and θ determines that of the driving field.]

III.

We consider now a TLS of the magnetic dipole type. This means that the coupling to the LM takes place, in general, through all the σ's (rather than through one, as in the electric dipole case). The Hamiltonian is given by

$$H = H_{\rm LM} + \tfrac{1}{2}\hbar\omega\sigma_z + \tfrac{1}{2}\hbar\boldsymbol{\sigma}\cdot\boldsymbol{\mathfrak{F}} \tag{70}$$

where

$$\boldsymbol{\mathfrak{F}} = \mathbf{F} + \mathbf{f}, \tag{70a}$$

F_x, F_y, F_z being dynamical variables of the loss mechanism, and f_x, f_y, f_z being the components of the driving field acting on the TLS. The same condition of weak coupling is imposed on the problem as previously: The numbers associated with $\mathfrak{F}$ are small compared to ω. In addition, we assume that there is no correlation among the different components of $\mathbf{F}$; that is F_x, F_y, F_z behave as though they referred to independent loss mechanisms. For the sake of simplicity, these mechanisms are taken to be identical, so that ξ and η are the same for all. The independence of the F's means that for $i \neq j$,

$$[F_i, F_j] = [F_i, \dot{F}_j] = 0, \tag{71a}$$

$$\begin{aligned}\langle F_i(t_1)\cdots F_i(t_n)F_j(t_1)\cdots F_j(t_m)\rangle \\ = \langle F_i(t_1)\cdots F_i(t_n)\rangle\langle F_j(t_1)\cdots F_j(t_m)\rangle.\end{aligned} \tag{71b}$$

The equations of motion for the combined system are now

$$\dot{\sigma}_x = -\omega\sigma_y - \mathfrak{F}_z\sigma_y + \mathfrak{F}_y\sigma_z, \tag{72a}$$

$$\dot{\sigma}_y = \omega\sigma_x + \mathfrak{F}_z\sigma_x - \mathfrak{F}_x\sigma_z, \tag{72b}$$

$$\dot{\sigma}_z = \mathfrak{F}_x\sigma_y - \mathfrak{F}_y\sigma_x, \tag{72c}$$

$$\dot{\mathbf{F}} = -(i/\hbar)[\mathbf{F}, H_{\rm LM}], \tag{72d}$$

$$\dot{H}_{\rm LM} = -\tfrac{1}{2}i[H_{\rm LM}, \mathbf{F}]\cdot\boldsymbol{\sigma}. \tag{72e}$$

Because of the independence of the components of $\mathbf{F}$, we obtain a relationship identical to Eq. (13) for each component of $\mathbf{F}$ in exactly the same manner as that in which Eq. (13) was obtained:

$$\mathbf{F} = \mathbf{F}^{(0)} - \frac{2}{\pi}\int_0^t dt_1 \int_0^\infty d\omega'\ \xi(\omega')\ \sin\omega'(t-t_1)\boldsymbol{\sigma}(t_1). \tag{73}$$

This equation leads, in the same way as Eq. (13) led to Eq. (23), to

$$F_x = F_x^{(0)} - \xi\sigma_y, \tag{74}$$

$$F_y = F_y^{(0)} + \xi\sigma_x. \tag{75}$$

As far as F_z is concerned, the analysis is different. σ_z has only a slowly varying time dependence, and the main contribution to the integral of Eq. (73) comes from the low values of ω' (rather than from $\omega' \sim \omega$, as in the case of σ_x and σ_y). This brings us to a matter that is encountered more than once in the analysis of a TLS of the magnetic dipole type, namely, the effect of the LM at very low frequencies. Since we now have coupling through σ_z, secular effects due to this coupling will come from the low-frequency properties of the LM. For present purposes, we assume that we are dealing with a LM the effect of which vanishes as the frequency approaches zero. [The vanishing of the diagonal matrix elements of $F^{(0)}$, which led to Eq. (8), is required by this assumption.] This assumption implies that $\xi(\omega')$ and $\eta(\omega')$ become negligible for very low frequencies, and we approximate by dropping the integral in the z component of Eq. (73). We therefore have, in addition to Eqs. (74) and (75),

$$F_z = F_z^{(0)}. \tag{76}$$

From Eqs. (74)–(76) we obtain the commutation relations

$$[\sigma_x, \mathbf{F}^{(0)}] = 2i\xi\sigma_z\mathbf{i}, \tag{77a}$$

$$[\sigma_y, \mathbf{F}^{(0)}] = 2i\xi\sigma_z\mathbf{j}, \tag{77b}$$

$$[\sigma_z, \mathbf{F}^{(0)}] = -2i\xi(\sigma_x\mathbf{i} + \sigma_y\mathbf{j}), \tag{77c}$$

where $\mathbf{i}$, $\mathbf{j}$, $\mathbf{k}$ are unit vectors in the x, y, z direction, respectively. The substitution of Eqs. (74)–(76) into the equations of motion [Eqs. (72a)–(72c)] results in

$$\dot{\sigma}_x = -\omega\sigma_y - \tfrac{1}{2}\{\mathfrak{F}_z^{(0)}, \sigma_y\} + \tfrac{1}{2}\{\mathfrak{F}_y^{(0)}, \sigma_z\}, \tag{78a}$$

$$\dot{\sigma}_y = \omega\sigma_x + \tfrac{1}{2}\{\mathfrak{F}_z^{(0)}, \sigma_x\} - \tfrac{1}{2}\{\mathfrak{F}_x^{(0)}, \sigma_z\}, \tag{78b}$$

$$\dot{\sigma}_z = \tfrac{1}{2}\{\mathfrak{F}_x^{(0)}, \sigma_y\} - \tfrac{1}{2}\{\mathfrak{F}_y^{(0)}, \sigma_x\} - 2\xi, \tag{78c}$$

where

$$\boldsymbol{\mathfrak{F}}^{(0)} = \mathbf{F}^{(0)} + \mathbf{f},$$

and where symmetrized products are used for the sake of uniformity even though the factors may commute. For $\mathbf{f} = 0$, Eqs. (78) are the Langevin equations for a TLS of the magnetic dipole type. By means of Eqs. (77) it can be shown easily that Eqs. (31) are satisfied, so that the σ's determined by Eqs. (2) and (78) are true spin operators.

We proceed now, as previously, to obtain equations for the expectation values in LM space. Equations

(78a) and (78b) may be combined to give

$$\sigma_x(t)=\sigma_x^{(0)}(t)-\tfrac{1}{2}\int_0^t dt_1\cos\omega(t-t_1)[\{\mathfrak{F}_z^{(0)}(t_1),\sigma_y(t_1)\}-\{\mathfrak{F}_y^{(0)}(t_1),\sigma_z(t_1)\}]-\tfrac{1}{2}\int_0^t dt_1\sin\omega(t-t_1)\times[\{\mathfrak{F}_z^{(0)}(t_1),\sigma_x(t_1)\}-\{\mathfrak{F}_x^{(0)}(t_1),\sigma_z(t_1)\}],\quad(79a)$$

$$\sigma_y(t)=\sigma_y^{(0)}(t)+\tfrac{1}{2}\int_0^t dt_1\cos\omega(t-t_1)[\{\mathfrak{F}_z^{(0)}(t_1),\sigma_x(t_1)\}-\{\mathfrak{F}_x^{(0)}(t_1),\sigma_z(t_1)\}]-\tfrac{1}{2}\int_0^t dt_1\sin\omega(t-t_1)\times[\{\mathfrak{F}_z^{(0)}(t_1)\ \sigma_y(t_1)\}-\{\mathfrak{F}_y^{(0)}(t_1),\sigma_z(t_1)\}],\quad(79b)$$

and Eq. (78c) gives

$$\sigma_z(t)=\sigma_z(0)-2\bar{\xi}t+\tfrac{1}{2}\int_0^t dt_1[\{\mathfrak{F}_x^{(0)}(t_1),\sigma_y(t_1)\}-\{\mathfrak{F}_y^{(0)}(t_1),\sigma_x(t_1)\}].\quad(79c)$$

These expressions are now to be substituted into the $F_i^{(0)}\sigma_j$ terms of Eqs. (78) and LM expectation values taken. Making use of the independence of the $F^{(0)}$'s as given by Eqs. (71), and of previously discussed approximations [those employed in going from Eqs. (62) to Eqs. (67)] we obtain, in analogy with Eqs. (65) and (66),

$$\langle\tfrac{1}{2}\{F_y^{(0)},\sigma_z\}\rangle_{\rm LM}=-\eta\sigma_x,\quad(80a)$$

$$\langle\tfrac{1}{2}\{F_x^{(0)},\sigma_z\}\rangle_{\rm LM}=\eta\sigma_y,\quad(80b)$$

$$\langle\tfrac{1}{2}\{F_x^{(0)},\sigma_y\}\rangle_{\rm LM}=-\eta\sigma_z,\quad(80c)$$

$$\langle\tfrac{1}{2}\{F_y^{(0)},\sigma_x\}\rangle_{\rm LM}=\eta\sigma_z.\quad(80d)$$

The evaluation of $\langle F_z^{(0)},\sigma_x\rangle_{\rm LM}$ and $\langle\{F_z^{(0)},\sigma_y\}\rangle_{\rm LM}$ requires some additional consideration. From Eqs. (79a), (15), and (17) we have

$$\langle\{F_z^{(0)},\sigma_x\}\rangle_{\rm LM}=-\int_0^t dt_1\ \{F_z^{(0)}(t),F_z^{(0)}(t_1)\}\rangle\times[\cos\omega(t-t_1)\sigma_y(t_1)+\sin\omega(t-t_1)\sigma_x(t_1)]=\frac{i}{2^{1/2}}\int_0^t dt_1\langle\{F_z^{(0)}(t),F_z^{(0)}(t_1)\}\rangle\times[\varphi_+(t_1)e^{i\omega t}-\varphi_-(t_1)e^{-i\omega t}].\quad(81)$$

The expression to be inserted for $\langle\{F_z^{(0)}(t),F_z^{(0)}(t_1)\}\rangle$ in Eq. (81) is that of Eq. (24) rather than Eq. (25), since the square-bracketed expression in the integrand of Eq. (81) varies slowly with respect to t_1 (and does *not* oscillate with approximate frequency ω). Thus,

$$\langle\{F_z^{(0)},\sigma_x\}\rangle_{\rm LM}=\frac{2^{3/2}i}{\pi}\int_0^t dt_1\int_0^\infty d\omega'\,\eta(\omega')\cos\omega'(t-t_1)\times[\varphi_+(t_1)e^{i\omega t}-\varphi_-(t_1)e^{-i\omega t}].\quad(82)$$

The main contribution to this integral, as far as the ω' integration is concerned, comes from small values of ω'. In accordance with our previous assumption about the magnitude of $\eta(\omega')$ as ω' approaches zero, the right side of Eq. (82) is negligible, and we obtain

$$\langle\{F_z^{(0)},\sigma_x\}\rangle_{\rm LM}=0.\quad(83a)$$

In an identical manner we have

$$\langle\{F_z^{(0)},\sigma_y\}\rangle_{\rm LM}=0.\quad(83b)$$

The substitution of Eqs. (80) and (83) into Eqs. (78) gives, for the expectation values in LM space,

$$\dot\sigma_x=-\omega\sigma_y-\eta\sigma_x-f_z\sigma_y+f_y\sigma_z,\quad(84a)$$

$$\dot\sigma_y=\omega\sigma_x-\eta\sigma_y+f_z\sigma_x-f_x\sigma_z,\quad(84b)$$

$$\dot\sigma_z=-2\xi-2\eta\sigma_z+f_x\sigma_y-f_y\sigma_x,\quad(84c)$$

where $\bar{\xi}$ has again been approximated by ξ. These equations are the analog of Eqs. (67), and like those, are operator equations in TLS space. The discussion concerning the matrix elements and expectation values of Eqs. (67) applies also here.

If the TLS is a magnetic dipole and the separation between energy levels is due to a dc magnetic field $\boldsymbol{\omega}$ (in frequency units) along the z axis, then Eqs. (84) may be written as

$$\dot{\boldsymbol{\sigma}}=(\mathbf{f}+\boldsymbol{\omega})\times\boldsymbol{\sigma}-\eta(\sigma_x\mathbf{i}+\sigma_y\mathbf{j})-2\eta(\sigma_z-\sigma_0)\mathbf{k},\quad(85)$$

where σ_0 is defined by Eq. (43a). If we now take expectation values in TLS space, Eq. (85) remains formally unchanged, and the σ's represent their expectation values. In this last form, Eq. (85) is equivalent to the Bloch equations,[9] η^{-1} being the transverse relaxation time T_2, $(2\eta)^{-1}$ being the longitudinal relaxation time T_1, and σ_0 being the equilibrium value of σ_z in absence of a driving field. (The ratio of the two relaxation times is that to be expected when the relaxation mechanism has the properties of our LM. Spin-lattice coupling refers to such a mechanism, but spin-spin coupling is, in general, a more complicated problem.)

We will not discuss the solutions of Eqs. (84) or (85) in detail, since the solution of the Bloch equations is available.[10] It is, however, of interest to compare some aspects of the behavior of a TLS of the magnetic dipole type with those of a TLS of the electric dipole

[9] F. Bloch, Phys. Rev. **70**, 460 (1946); R. K. Wangsness and F. Bloch, *ibid.* **89**, 728 (1953).

[10] H. C. Torrey, Phys. Rev. **76**, 1059 (1949); see also A. Abragam, *The Principles of Nuclear Magnetism* (Oxford University Press, New York, 1961), Chap. III.

type. In the absence of a driving field, that is, for $f=0$, we have for the LM expectation values,

$$\sigma_x=e^{-\eta t}\begin{pmatrix} 0 & e^{i\omega t} \\ e^{-i\omega t} & 0 \end{pmatrix}, \tag{86a}$$

$$\sigma_y=e^{-\eta t}\begin{pmatrix} 0 & -ie^{i\omega t} \\ ie^{-i\omega t} & 0 \end{pmatrix}, \tag{86b}$$

$$\sigma_z=\begin{pmatrix} (1-\sigma_0)e^{-2\eta t}+\sigma_0 & 0 \\ 0 & (-1-\sigma_0)e^{-2\eta t}+\sigma_0 \end{pmatrix}. \tag{86c}$$

It is seen that here, as in the case of the electric dipole-type TLS, σ_z decays twice as fast as either σ_x or σ_y, but, unlike that case, the frequency of oscillation of σ_x and σ_y is unchanged by the LM.

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APPENDIX A

We indicate the derivation of Eq. (13) from Eq. (12). Designating by $\rho(E)$ the density of energy states of the LM, we have

$$\sum_{i,k}\rightarrow\int_0^\infty \rho(E_i)dE_i\int_0^\infty \rho(E_k)dE_k; \tag{A1}$$

setting

$$E=\tfrac{1}{2}(E_i+E_k),\quad \omega'=\omega_{ik}, \tag{A2}$$

we obtain

$$\int_0^\infty dE_i\int_0^\infty dE_k\rightarrow\int_0^\infty \hbar d\omega'\int_{(1/2)\hbar\omega'}^\infty dE + \int_{-\infty}^0 \hbar d\omega'\int_{-(1/2)\hbar\omega'}^\infty dE \equiv \int \hbar d\omega'\int dE. \tag{A3}$$

The last term of Eq. (12) can be written as

$$Z^{-1}\int \hbar d\omega'\int dE\,\rho(E+\tfrac{1}{2}\hbar\omega')\rho(E-\tfrac{1}{2}\hbar\omega') \times\exp[-(E+\tfrac{1}{2}\hbar\omega')/kT]\tilde{F}^2(E+\tfrac{1}{2}\hbar\omega',E-\tfrac{1}{2}\hbar\omega') \times\int_0^t \sin\omega'(t-t_1)\sigma_x(t_1)dt_1. \tag{A4}$$

Noting that

$$\int_{\pm(1/2)\hbar\omega'}^\infty dE\,\rho(E+\tfrac{1}{2}\hbar\omega')\rho(E-\tfrac{1}{2}\hbar\omega')e^{-E/kT} \times\tilde{F}^2(E+\tfrac{1}{2}\hbar\omega',E-\tfrac{1}{2}\hbar\omega') =\exp(\mp\tfrac{1}{2}\hbar\omega'/kT)B(\pm\omega'), \tag{A5}$$

where

$$B(\omega')=\int_0^\infty dE\,\rho(E+\hbar\omega')\rho(E)\tilde{F}^2(E+\hbar\omega',E)e^{-E/kT}, \tag{A6}$$

we obtain, after change in sign of variable of integration in one of the integrals, an expression for (A4) which is the last term of Eq. (13). Thus, Eq. (13) is derived.

APPENDIX B

Our purpose is the derivation of Eq. (24). From Eqs. (4) and (10) we have

$$\langle F^{(0)}(t_1)F^{(0)}(t_2)\rangle=Z^{-1}\sum_{j,k}e^{-E_j/kT}|\tilde{F}_{j,k}|^2e^{i\omega_{jk}(t_1-t_2)}. \tag{A7}$$

Equations (A1)–(A3) and (A6) allow us to write

$$\begin{aligned}\langle F^{(0)}(t_1)F^{(0)}(t_2)\rangle &=Z^{-1}\int_0^\infty dE_j\,\rho(E_j)\int_0^\infty dE_k\,\rho(E_k) \times e^{-E_j/kT}\tilde{F}^2(E_j,E_k)e^{i\omega_{jk}(t_1-t_2)}\\ &=Z^{-1}\bigg[\int_0^\infty \hbar d\omega'\,B(\omega')\exp(-\hbar\omega'/kT)e^{i\omega'(t_1-t_2)} +\int_{-\infty}^0\hbar d\omega'\,B(-\omega')e^{i\omega'(t_1-t_2)}\bigg]\\ &=\hbar Z^{-1}\int_0^\infty d\omega' B(\omega')\{\cos\omega'(t_1-t_2)[\exp(-\hbar\omega'/kT)+1] +i\sin\omega'(t_1-t_2)[\exp(-\hbar\omega'/kT)-1]\}. \end{aligned}\tag{A8}$$

Since

$$\xi(\omega')=\tfrac{1}{2}\pi\hbar Z^{-1}B(\omega')[1-\exp(-\hbar\omega'/kT)] \tag{A9a}$$

and

$$\eta(\omega')=\tfrac{1}{2}\pi\hbar Z^{-1}B(\omega')[1+\exp(-\hbar\omega'/kT)], \tag{A9b}$$

Eq. (24) follows from Eq. (A8).

APPENDIX C

We obtain expressions for $\langle\sigma_z\rangle$ from Eqs. (67). The combination of Eqs. (67a) and (67b) yields

$$\ddot{\sigma}_y+\eta\dot{\sigma}_y+\omega^2\sigma_y=-(d/dt)(f\sigma_z), \tag{A10}$$

which is equivalent to

$$\sigma_y=\sigma_y^{[0]}-\frac{1}{\tilde{\omega}}\int_0^t dt_1\,e^{-(1/2)\eta(t-t_1)} \times\sin\tilde{\omega}(t-t_1)\frac{d}{dt_1}[f(t_1)\sigma_z(t_1)], \tag{A11}$$

and which may be approximated by

$$\sigma_y=\sigma_y^{[0]}-\int_0^t dt_1\, e^{-(1/2)\eta(t-t_1)}\cos\tilde{\omega}(t-t_1)f(t_1)\sigma_z(t_1)\,, \quad \text{(A12)}$$

where $\sigma_y^{[0]}$ refers to the undriven TLS ($f=0$) and is given by Eq. (48). The substitution from Eq. (A12) into Eq. (67c) followed by integration yields

$$\sigma_z=\sigma_z(0)-\xi t+\int_0^t dt_1\, f(t_1)\sigma_y^{[0]}(t_1)-\eta\int_0^t dt_1\,\sigma_z(t_1) -\int_0^t dt_1\int_0^{t_1} dt_2\,\sigma_z(t_2)f(t_1)f(t_2)e^{-(1/2)\eta(t_1-t_2)} \times\cos\tilde{\omega}(t_1-t_2)\,. \quad \text{(A13)}$$

We consider a sinusoidal driving field, $f(t)=f_0\cos(\nu t+\theta)$, with the driving frequency near the resonant frequency, so that

$$|\nu-\tilde{\omega}|\ll\tilde{\omega}. \quad \text{(A14)}$$

Under these conditions, the double integral term in Eq. (A13) may be approximated by dropping, first, the t_1+t_2 term in combining $f(t_1)$ and $f(t_2)$, and then the $\tilde{\omega}+\nu$ term in combining the result with $\cos\tilde{\omega}(t_1-t_2)$. The double integral term thus becomes

$$-\tfrac{1}{4}f_0^2\int_0^t dt_1\int_0^{t_1} dt_2\,\sigma_z(t_2)e^{-(1/2)\eta(t_1-t_2)}\cos\bar{\nu}(t_1-t_2)\,, \quad \text{(A15)}$$

where

$$\bar{\nu}\equiv\nu-\tilde{\omega}\,.$$

Carrying out the t_1 integration first, we obtain for Eq. (A13)

$$\sigma_z(t)=\psi(t)+\int_0^t dt_1\, K(t-t_1)\sigma_z(t_1)\,, \quad \text{(A16)}$$

where

$$\psi(t)\equiv\sigma_z(0)-\xi t+\int_0^t dt_1\, f(t_1)\sigma_y^{[0]}(t_1)\,, \quad \text{(A16a)}$$

and

$$K(t)=-\eta-\frac{\frac{1}{4}f_0^2}{\bar{\nu}^2+\frac{1}{4}\eta^2}\left(\tfrac{1}{2}\eta-\tfrac{1}{2}\eta e^{-(1/2)\eta t}\cos\bar{\nu}t +\bar{\nu}e^{-(1/2)\eta t}\sin\bar{\nu}t\right). \quad \text{(A16b)}$$

Equation (A16) is an integral equation of the Volterra type and may be solved by Laplace transformation. Using the notation $\mathfrak{L}\{\varphi(t)\}\equiv\bar{\varphi}(s)$, the solution is given by

$$\sigma_z(t)=\mathfrak{L}^{-1}\left\{\frac{\bar{\psi}(s)}{1-\bar{K}(s)}\right\}. \quad \text{(A17)}$$

Some calculation shows that

$$[1-\bar{K}(s)]^{-1}=\frac{s[(\frac{1}{2}\eta+s)^2+\bar{\nu}^2][4\bar{\nu}^2+\eta^2]}{[(\frac{1}{2}\eta+s)^2+\bar{\nu}^2][s(4\bar{\nu}^2+\eta^2)+\eta(4\bar{\nu}^2+\eta^2)+\frac{1}{2}\eta f_0^2]+f_0^2 s[\bar{\nu}^2-\frac{1}{2}\eta(\frac{1}{2}\eta+s)]}. \quad \text{(A18)}$$

From the properties of the inverse Laplace transform, it is clear that the steady state solution (averaged over the time) is given by $2\pi i$ times the residue of $\bar{\psi}(s)[1-\bar{K}(s)]^{-1}$ at the pole $s=0$. Only the $-\xi t$ term in $\psi(t)$, [or the $-\xi/s^2$ term in $\bar{\psi}(s)$] contributes at this pole and the result is given by Eq. (69).

In the case of resonance, that is $\bar{\nu}=0$, Eq. (A18) simplifies to

$$\frac{1}{1-\bar{K}(s)}=\frac{s(\frac{1}{2}\eta+s)}{s^2+\frac{3}{2}\eta s+(\frac{1}{2}\eta^2+\frac{1}{4}f_0^2)}. \quad \text{(A19)}$$

Also, carrying out the integration in the last term of Eq. (A16a), we utilize Eq. (55) and drop the oscillatory term of the integrand, obtaining

$$\bar{\psi}(s)_{\bar{\nu}=0}=\frac{\langle\sigma_z(0)\rangle}{s}-\frac{\xi}{s^2}+\frac{|a_1a_2|f_0\sin(\alpha-\theta)}{(s+\frac{1}{2}\eta)^2}. \quad \text{(A20)}$$

The substitution of Eqs. (A19) and (A20) into Eq. (A17) leads to Eq. (68). Once σ_z is known, σ_y may be obtained from Eq. (A12) and σ_x may be obtained from a corresponding equation, namely,

$$\sigma_x=\sigma_x^{[0]}+\int_0^t dt_1\, e^{-(1/2)\eta(t-t_1)}\sin\tilde{\omega}(t-t_1)f(t_1)\sigma_z(t_1)\,. \quad \text{(A21)}$$