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Dissipation in Quantum Mechanics. Two-Level System. II

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The theory of a previous article, dealing with two types of two-level systems coupled to a loss mechanism (LM), is extended. The first extension consists of the consideration of the most general type of two-level system (TLS), in which the dipole moment is expanded in terms of the three Pauli spin matrices and unit matrix, the expansion coefficients being vectors (dipole vectors). The second extension consists of the addition to the thermal-reservoir type of LM of a large number of systems identical to the TLS under consideration. The TLS is described in terms of the time development of the Pauli matrices and differential equations are obtained for their expectation value in the presence of arbitrary driving fields. The Bloch equations for a magnetic dipole of spin $\frac{1}{2}$ are exhibited as a special case of these equations, corresponding to a particular combination of the dipole vectors. All other combinations describe electric dipole systems. Equations for two simple special cases of such systems are presented, one treated in the previous article and the other having permanent dipole moment. The frequency of oscillation of a freely decaying TLS is derived and shown to be shifted by an amount that depends on the relationship between the dipole vectors. It is pointed out that the commonly held belief that any TLS can be represented as a magnetic dipole of spin $\frac{1}{2}$ is only approximately correct in the presence of dissipation. The conditions under which the differential equations for the expectation values of the dynamical variables of the TLS can be converted into differential equations for macroscopic variables are discussed.

INTRODUCTION

IN a previous article,[1] it was pointed out that there are many important physical problems which may be considered, with some simplification, as that of a two-level system (TLS) coupled to a loss mechanism (LM), and a study was made of this problem. The LM was a thermal reservoir type of system, and two types of TLS were considered, the magnetic dipole type and the electric dipole type without permanent dipole moment that couples to its environment through one dynamical variable only. In the present article the analysis of I will be extended in two aspects. One extension is the consideration of the most general type of two-level dipole system, of which the two types considered in I are special cases.[2] The results will produce, on the one hand, a unified method of treating all two-level systems, and on the other hand, an explicit and systematic exhibition of differences among two-level systems. The other extension is the consideration of a more complicated LM, one which consists not only of a thermal reservoir but also—in addition to the reservoir—of a large number of systems identical to the TLS under consideration, loosely coupled to it, and surrounded by the same environment. (In the language of magnetic resonance, coupling of the "spin-spin" type as well as the "spin-lattice" type will be considered.) Part I and part II contain the first and second extensions, respectively. Some consequences of the results and several special cases are discussed in part III.

[1] I. R. Senitzky, Phys. Rev. **131**, 2827 (1963); hereafter referred to as I.

[2] The electric dipole TLS is far richer in possibilities than the magnetic dipole TLS, since the latter is restricted by the special properties of angular momentum. All cases other than the two considered in I are electric dipole types.

I.

The defining property of a dipole system may be given by the form of the interaction energy of the system with an electric or magnetic field,

$$H_{\text{int}} = -\mathbf{d}\cdot\mathbf{E},\ -\mathbf{d}\cdot\mathbf{H}, \tag{1}$$

respectively, where $\mathbf{d}$, the dipole moment, is an operator referring only to the system under consideration. Since our system is a TLS, the components of $\mathbf{d}$, that is d_x, d_y, and d_z are 2×2 Hermitian matrices, and for the most general TLS, these are arbitrary. Now, any 2×2 Hermitian matrix may be written as a linear superposition of the Pauli spin matrices together with the unit matrix. We may therefore write

$$\mathbf{d} = \mu \sum_{\alpha=1}^{4} \mathbf{a}_\alpha \sigma_\alpha, \tag{2}$$

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 dimension of dipole moment, and the $\mathbf{a}$'s are four real 3-dimensional vectors (determined by 12 real numbers, as many as determine three Hermitian 2×2 matrices) which determine completely the dipole moment operator of the TLS. The three Pauli spin matrices obey the well-known properties

$$\sigma_i^2 = 1, \quad \{\sigma_i, \sigma_j\} = 0, \quad [\sigma_k, \sigma_l] = 2i\sigma_m, \tag{3}$$

where $i \neq j$, $\{\ \}$ is the anticommutator bracket, and k, l, m stand for the cyclic permutation of 1, 2, 3. The $\mathbf{a}_i$'s may be normalized by the requirement that

$$d^2 (\equiv \mathbf{d}\cdot\mathbf{d}) = n\mu^2, \tag{4}$$

[the right side of Eq. (4) is to be considered multiplied by the unit matrix] which gives

$$\sum_{\alpha=1}^{4} a_\alpha^2 = n, \tag{5}$$

where n is a number of the order of unity chosen for convenience in the particular case under consideration. The representation of the σ's is such that the energy of the TLS is given by $\frac{1}{2}\hbar\omega\sigma_3$. For an electric dipole system, nothing more can be said about the **a**'s without additional information about the system. For a magnetic dipole TLS, however, the proportionality between magnetic moment and angular momentum requires that $\mathbf{a}_4=0$, and that $\mathbf{a}_1$, $\mathbf{a}_2$, and $\mathbf{a}_3$ be perpendicular to each other and equal in magnitude.

The TLS is subject to fields produced by the LM and to externally imposed fields, $\mathbf{E}_{\text{LM}}$ and $\mathbf{E}_{\text{ext}}$, respectively, for the electric system, and $\mathbf{H}_{\text{LM}}$ and $\mathbf{H}_{\text{ext}}$ for magnetic system. We introduce the notation

$$\mathbf{F} = -(2/\hbar)\boldsymbol{\mu}\mathbf{E}_{\text{LM}}, \quad \mathbf{f} = -(2/\hbar)\boldsymbol{\mu}\mathbf{E}_{\text{ext}} \tag{6a}$$

for the electric dipole case, and

$$\mathbf{F} = -(2/\hbar)\boldsymbol{\mu}\mathbf{H}_{\text{LM}}, \quad \mathbf{f} = -(2/\hbar)\boldsymbol{\mu}\mathbf{H}_{\text{ext}} \tag{6b}$$

for the magnetic dipole case. The Hamiltonian may now be written, for either case, as

$$H = H_{\text{LM}} + \tfrac{1}{2}\hbar\omega\sigma_3 + \tfrac{1}{2}\hbar \textstyle\sum_\alpha \sigma_\alpha \mathbf{a}_\alpha \cdot \mathfrak{F}, \tag{7}$$

where

$$\mathfrak{F} = \mathbf{F} + \mathbf{f}. \tag{7a}$$

Comparison of Eq. (7) with Eqs. (I.60) and (I.70) shows that the electric dipole case in I corresponds to $a_\alpha = \delta_{\alpha 1}$ and the magnetic case corresponds to $\mathbf{a}_1 = \hat{x}$, $\mathbf{a}_2 = \hat{y}$, $\mathbf{a}_3 = \hat{z}$, $\mathbf{a}_4 = 0$, $\hat{x}$, $\hat{y}$, $\hat{z}$ being unit Cartesian coordinate vectors.

A TLS can have only three linearly independent dynamical variables, since each variable corresponds to a 2×2 Hermitian matrix and, as mentioned previously, there are only three such linearly independent matrices possible, aside from the physically uninteresting unit matrix. In the present analysis, (as in I), the TLS will be described by the three Pauli spin matrices. Once we have these, we can obtain the dipole moment, the energy, and any other variable. The LM will be described (likewise as in I) by H_{LM} and **F**.

The expression for the Hamiltonian, Eq. (7), may be rewritten as

$$H = H_{\text{LM}} + \tfrac{1}{2}\hbar\omega\sigma_3 + \tfrac{1}{2}\hbar \sum_{\alpha=1}^{4}\sum_{m=1}^{3} \sigma_\alpha a_{\alpha m}\mathfrak{F}_m, \tag{8}$$

where the vectors have been resolved along Cartesian axes labeled 1, 2, and 3. As in I, we consider F_1, F_2, and F_3 to act independently of one another; we assume that they refer to three independent but identical LM's. Correspondingly, we write

$$H_{\text{LM}} = H_1 + H_2 + H_3. \tag{9}$$

(This separation of H_{LM} was not indicated explicitly in I, but is implicit in the argument used there.) The equations of motion for the TLS and LM, obtained from Eqs. (8) and (9), are

$$\dot{\sigma}_1 = -\omega\sigma_2 + \textstyle\sum_m (a_{2m}\mathfrak{F}_m\sigma_3 - a_{3m}\mathfrak{F}_m\sigma_2), \tag{10a}$$

$$\dot{\sigma}_2 = \omega\sigma_1 + \textstyle\sum_m (a_{3m}\mathfrak{F}_m\sigma_1 - a_{1m}\mathfrak{F}_m\sigma_3), \tag{10b}$$

$$\dot{\sigma}_3 = \textstyle\sum_m (a_{1m}\mathfrak{F}_m\sigma_2 - a_{2m}\mathfrak{F}_m\sigma_1), \tag{10c}$$

$$\dot{F}_m = -(i/\hbar)[F_m, H_m], \tag{10d}$$

$$\dot{H}_m = -(i/2)\textstyle\sum_\alpha a_{\alpha m}\sigma_\alpha [H_m, F_m]. \tag{10e}$$

The last two equations may be combined to give

$$\dot{F}_m(t) = -(i/\hbar)[F_m, H_m(0)] + (1/2\hbar)\sum_\alpha a_{\alpha m}\int_0^t dt_1 \times [F_m(t),[F_m(t_1),H_m(t_1)]\sigma_\alpha(t_1)], \tag{11}$$

which may be rewritten as an integral equation

$$F_m(t) = F_m^{(0)}(t) + (1/2\hbar)\sum_\alpha a_{\alpha m}\int_0^t dt_1 \int_0^{t_1} dt_2\, U_m(t-t_1) \times [F_m(t_1),[F_m(t_2),H_m(t_2)]\sigma_\alpha(t_2)]U_m^{-1}(t-t_1), \tag{12}$$

where

$$U(\tau) = \exp[(i/\hbar)H_m(0)\tau], \tag{12a}$$

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

$$F_m^{(0)}(t) = F_m(0), \tag{12b}$$

$$\dot{F}_m^{(0)}(t) = -(i/\hbar)[F_m^{(0)}(t), H_m(0)]. \tag{12c}$$

The important properties of $F^{(0)}(t)$ needed for present purposes, and derived in I, are the following:

$$\langle F_m^{(0)}(t)\rangle = 0, \tag{13}$$

$$\langle F_m^{(0)}(t_1)F_n^{(0)}(t_2)\rangle = \delta_{nm}(2/\pi)\int_0^\infty d\omega' [\eta(\omega')\cos\omega'(t_1-t_2) - i\xi(\omega')\sin\omega'(t_1-t_2)], \tag{14}$$

where

$$\xi(\omega') = \tfrac{1}{2}\pi\hbar Z^{-1}B(\omega')[1-\exp(-\hbar\omega'/kT)], \tag{14a}$$

$$\eta(\omega') = \tfrac{1}{2}\pi\hbar Z^{-1}B(\omega')[1+\exp(-\hbar\omega'/kT)], \tag{14b}$$

$$Z = \int_0^\infty dE\rho(E)\exp(-E/kT), \tag{14c}$$

$$B(\omega') = \int_0^\infty dE\rho(E+\hbar\omega')\rho(E) \times \tilde{F}^2(E+\hbar\omega', E)\exp(-E/kT), \tag{14d}$$

$\rho(E)$ being the density of energy states of the LM, $\tilde{F}^2(E_i,E_k)$ being the average over small ranges of E_i and E_k of $|F_{ik}^{(0)}|^2$, and T being the LM temperature. Both $\eta(\omega')$ and $\xi(\omega')$ are assumed to become vanishingly small as ω' approaches zero, and approximately constant in the neighborhood of $\omega'=\omega$, the values in this neighborhood being denoted by η and ξ (without argument) respectively. The expression $\langle F^{(0)}(t_1)F^{(0)}(t_2)\rangle$ will occur, in the present analysis, as a factor in an integrand, the integration being over t_1, or t_2, or both. Consider the expression

$$\int_0^t dt_1\varphi(t_1)\langle F^{(0)}(t_1)F^{(0)}(t_2)\rangle \tag{15}$$

for $t\gg\omega^{-1}$. Substituting from Eq. (14) into (15), and interchanging the order of integration, one sees that if $\varphi(t_1)$ is approximately an oscillating function with angular frequency ω'', the main contribution to the integration comes from the neighborhood $\omega'\sim\omega''$. The same argument applies if the integration in (15) is over t_2 [with $\varphi(t_2)$ instead of $\varphi(t_1)$]. We may therefore write for use in subsequent integration

$$\langle F^{(0)}(t_1)F^{(0)}(t_2)\rangle \approx 2\left[\eta(\omega'')\delta(t_1-t_2)-\frac{i}{\pi}\xi(\omega'')\frac{\mathcal{P}}{t_1-t_2}\right], \tag{16a}$$

and

$$\langle\{F^{(0)}(t_1),F^{(0)}(t_2)\}\rangle\approx 4\eta(\omega'')\delta(t_1-t_2). \tag{16b}$$

The values of ω'' to be encountered in the present article will be only ω and 0. For the latter value, it is clear that $\langle F^{(0)}(t_1)F^{(0)}(t_2)\rangle=0$.

Equations (12) may be written as

$$F_m(t)=F_m^{(0)}(t)+\sum_\alpha a_{\alpha m}F_{\alpha m}^{(1)}(t), \tag{17}$$

where

$$F_{\alpha m}^{(1)}(t)=\frac{1}{2\hbar}\int_0^t dt_1\int_0^{t_1} dt_2 U_m(t-t_1) \times[F_m(t_1),[F_m(t_2),H_m(t_2)]\sigma_\alpha(t_2)] \times U_m^{-1}(t-t_1). \tag{17a}$$

By use of approximations based on the fact that the LM is affected only slightly by the TLS, it is shown in I that

$$F_{1m}^{(1)}(t)=-\bar{\xi}\sigma_2(t), \tag{18a}$$

$$F_{2m}^{(1)}(t)=\bar{\xi}\sigma_1(t), \tag{18b}$$

where $\bar{\xi}$ is a (c-number) function of t which is zero at $t=0$ and approaches the constant ξ in a time large compared to ω^{-1} but short compared to the time during which secular changes (in the TLS) take place. It is also shown in I [Eq. (I.76)] that

$$F_{3m}^{(1)}(t)=0, \tag{19a}$$

and by an identical argument[3] it can be shown that

$$F_{4m}^{(1)}(t)=0. \tag{19b}$$

[3] Equation (19a) follows from the fact that $\sigma_3(t)$ is a slowly varying function [while $\sigma_1(t)$ and $\sigma_2(t)$ are approximately oscillatory functions with (angular) frequency ω]. Equation (19b) follows from the fact that σ_4 is the unit matrix and does not vary at all.

Substituting from Eqs. (18) and (19) into (17), we obtain

$$F_m(t)=F_m^{(0)}(t)-a_{1m}\bar{\xi}\sigma_2(t)+a_{2m}\bar{\xi}\sigma_1(t). \tag{20}$$

This expression for $F_m(t)$ may now be substituted into Eqs. (10a)–(10c), the equations of motion for the TLS. Before we do that, however, it is convenient to rewrite the products of $\mathfrak{F}_m$ and σ_α in these equations as symmetrized products. (Note that $[F_m(t),\sigma_\alpha(t)]=0$.) Then, by substituting from Eq. (20) and utilizing the properties of the σ's given in Eq. (3), we get

$$\dot{\sigma}_1=-\omega\sigma_2+\sum_m(\tfrac{1}{2}a_{2m}\{\mathfrak{F}_m^{(0)},\sigma_3\} -\tfrac{1}{2}a_{3m}\{\mathfrak{F}_m^{(0)},\sigma_2\}+a_{1m}a_{3m}\bar{\xi}), \tag{21a}$$

$$\dot{\sigma}_2=\omega\sigma_1+\sum_m(\tfrac{1}{2}a_{3m}\{\mathfrak{F}_m^{(0)},\sigma_1\} -\tfrac{1}{2}a_{1m}\{\mathfrak{F}_m^{(0)},\sigma_3\}+a_{2m}a_{3m}\bar{\xi}), \tag{21b}$$

$$\dot{\sigma}_3=\sum_m[\tfrac{1}{2}a_{1m}\{\mathfrak{F}_m^{(0)},\sigma_2\}-\tfrac{1}{2}a_{2m}\{\mathfrak{F}_m^{(0)},\sigma_1\} -(a_{1m}^2+a_{2m}^2)\bar{\xi}], \tag{21c}$$

where

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

Equations (21) contain as unknowns, dynamical variables referring to the TLS only. $\mathbf{F}^{(0)}$ is a "prescribed" field, and so is, of course, $\mathbf{f}$. $\mathbf{F}^{(0)}$ is the quantum-mechanical version of a stochastic force. With $\bar{\xi}$ replaced by ξ, and with $f=0$, Eqs. (21) are of the type referred to in I as the Langevin equations for a TLS.

It should be noted that, while $F_m(t)$ and $\sigma_\alpha(t)$ commute, $F_m^{(0)}(t)$ and $\sigma_\alpha(t)$ do not. The commutation relations between the latter two operators may be obtained from Eq. (20), utilizing the fact that $[F_m(t),\sigma_\alpha(t)]=0$; the result is

$$[\sigma_1,F_m^{(0)}]=2ia_{1m}\bar{\xi}\sigma_3, \tag{22a}$$

$$[\sigma_2,F_m^{(0)}]=2ia_{2m}\bar{\xi}\sigma_3, \tag{22b}$$

$$[\sigma_3,F_m^{(0)}]=-2i\bar{\xi}(a_{1m}\sigma_1+a_{2m}\sigma_2). \tag{22c}$$

The problem may now be considered as formally defined by Eqs. (21)—Eqs. (22) being part of the prescription for $F^{(0)}$—with initial values for the σ's given by (their usual description when the Schrödinger picture is used)

$$\sigma_1(0)=\begin{pmatrix}0&1\\1&0\end{pmatrix},\quad \sigma_2(0)=\begin{pmatrix}0&-i\\i&0\end{pmatrix},\quad \sigma_3(0)=\begin{pmatrix}1&0\\0&-1\end{pmatrix}. \tag{23}$$

From Eqs. (21) and (22) it can be shown, in a manner similar to that employed in I, that the solutions of Eqs. (22) subject to the initial conditions (23) are true spin operators, satisfying Eqs. (3). These solutions, of course, are operators both with respect to the TLS and LM. It is easy to see that this satisfactory state of affairs is due to the fact that $\bar{\xi}$ vanishes at $t=0$, since $F(0)$ and $\sigma_\alpha(0)$ are taken to be the uncoupled operators, and the coupling between TLS and LM is assumed to begin at $t=0$. Having shown the consistency of the formalism, we now replace $\bar{\xi}$ by ξ, as approximation for compulational purposes.

A solution of Eqs. (21) that is an operator in both TLS and LM spaces has not been found (even when all $\mathbf{a}$'s but $\mathbf{a}_1$ vanish, as in the electric dipole case in I). In order to make progress with Eqs. (21), we take expectation values in LM space. This does not simplify the situation automatically since expectation values of products, $\langle\{F_m^{(0)},\sigma_\alpha\}\rangle$, occur, and they are certainly not equal to the product of the expectation values. The evaluation of expectation values of these products will involve a significant approximation.

We consider $\mathfrak{F}^{(0)}$ (that is, both $F^{(0)}$ and f) to be a small quantity (compared to ω) of first order. Equations (21) can be rewritten as integral equations for the σ's in which the kernels are of first order. Defining the symbols $\Phi_1(t)$, $\Phi_2(t)$, $\Phi_3(t)$ by rewriting Eqs. (21) as

$$\dot{\sigma}_1(t)=-\omega\sigma_2(t)+\Phi_1(t)\,, \tag{24a}$$

$$\dot{\sigma}_2(t)=\omega\sigma_1(t)+\Phi_2(t)\,, \tag{24b}$$

$$\dot{\sigma}_3(t)=\Phi_3(t)\,, \tag{24c}$$

we obtain, as equivalent integral equations,

$$\sigma_1=\sigma_1^{(0)}+\int_0^t dt_1\cos\omega(t-t_1)\Phi_1(t_1)-\int_0^t dt_1\sin\omega(t-t_1)\Phi_2(t_1)\,, \tag{25a}$$

$$\sigma_2=\sigma_2^{(0)}+\int_0^t dt_1\cos\omega(t-t_1)\Phi_2(t_1)+\int_0^t dt_1\sin\omega(t-t_1)\Phi_1(t_1)\,, \tag{25b}$$

$$\sigma_3=\sigma_3^{(0)}+\int_0^t dt_1\Phi_3(t_1)\,, \tag{25c}$$

where the $\sigma^{(0)}$'s are the solution for the free TLS satisfying the initial conditions. A substitution is now made from Eqs. (25) into all products $\{F_m^{(0)},\sigma_\alpha\}$ occurring in Eqs. (21). Thus

$$\begin{aligned}\langle\{F_m^{(0)}(t_1),\sigma_1(t_1)\}\rangle_{\text{LM}} =\tfrac{1}{2}\sum_n\int_0^t dt_1\langle[a_{1n}\sin\omega(t-t_1)&+a_{2n}\cos\omega(t-t_1)]\{F_m^{(0)}(t),\{\mathfrak{F}_n^{(0)}(t_1),\sigma_3(t_1)\}\}\\ &-a_{3n}\{F_m^{(0)}(t),\{\mathfrak{F}_n^{(0)}(t_1),\sigma_2(t_1)\}\}\cos\omega(t-t_1)\\ &-a_{3n}\{F_m^{(0)}(t),\{\mathfrak{F}_n^{(0)}(t_1),\sigma_1(t_1)\}\}\times\sin\omega(t-t_1)\rangle_{\text{LM}}\,,\end{aligned} \tag{26}$$

where use has been made of the fact that $\langle F_m^{(0)}\sigma_\alpha^{(0)}\rangle_{\text{LM}}=\langle F_m^{(0)}\rangle\sigma_\alpha^{(0)}$, and of Eq. (13).[4] The significant approximation consists of ignoring the noncommutativity of σ_α and $F_m^{(0)}$ and replacing the product of the LM variables by its expectation value, both operations to be performed only in terms of higher order than the first. (See I for a discussion of this approximation.) Utilizing Eq. (13) and the Krönecker delta of Eq. (14), we obtain, with the above approximation,

$$\begin{aligned}\langle\{F_m^{(0)},\sigma_1\}\rangle_{\text{LM}} &=\int_0^t dt_1\langle\{F_m^{(0)}(t),F_m^{(0)}(t_1)\}\rangle\\ &\times\{\sigma_3(t_1)[a_{1m}\sin\omega(t-t_1)+a_{2m}\cos\omega(t-t_1)]\\ &-a_{3m}[\sigma_1(t_1)\sin\omega(t-t_1)+\sigma_2(t_1)\cos\omega(t-t_1)]\}\,.\end{aligned} \tag{27}$$

The considerations related to Eqs. (15) and (16) may now be applied. $\sigma_3(t_1)$ is a slowly varying function of t_1, so that Eq. (16b) with $\omega''=\omega$ must be used for the integration associated with first square bracket in Eq. (27). The second square bracket is shown in I [Eq. (I.81); note that σ_1 and σ_2 approximately oscillate with frequency ω] to be itself a slowly varying function of t_1, so that Eq. (16b) with $\omega''=0$ must be used for the integration associated with the second square bracket, and the contribution of this integration vanishes. The result is

$$\langle\{F_m^{(0)},\sigma_1\}\rangle_{\text{LM}}=2a_{2m}\eta\langle\sigma_3\rangle_{\text{LM}}\,. \tag{28a}$$

In an entirely similar manner, one obtains

$$\langle\{F_m^{(0)},\sigma_2\}\rangle_{\text{LM}}=-2a_{1m}\eta\langle\sigma_3\rangle_{\text{LM}}\,, \tag{28b}$$

and

$$\langle\{F_m^{(0)},\sigma_3\}\rangle_{\text{LM}}=2\eta(a_{1m}\langle\sigma_2\rangle_{\text{LM}}-a_{2m}\langle\sigma_1\rangle_{\text{LM}})\,. \tag{28c}$$

Substituting from Eqs. (28) into Eqs. (21), we have

$$\dot{\sigma}_1=(-\omega+\mathbf{a}_1\cdot\mathbf{a}_2\eta)\sigma_2-a_2^2\eta\sigma_1+\mathbf{a}_1\cdot\mathbf{a}_3\eta(\sigma_3-\sigma_0)+\mathbf{a}_2\cdot\mathbf{f}\sigma_3-\mathbf{a}_3\cdot\mathbf{f}\sigma_2\,, \tag{29a}$$

$$\dot{\sigma}_2=(\omega+\mathbf{a}_1\cdot\mathbf{a}_2\eta)\sigma_1-a_1^2\eta\sigma_2+\mathbf{a}_2\cdot\mathbf{a}_3\eta(\sigma_3-\sigma_0)+\mathbf{a}_3\cdot\mathbf{f}\sigma_1-\mathbf{a}_1\cdot\mathbf{f}\sigma_3\,, \tag{29b}$$

$$\dot{\sigma}_3=-(a_1^2+a_2^2)\eta(\sigma_3-\sigma_0)+\mathbf{a}_1\cdot\mathbf{f}\sigma_2-\mathbf{a}_2\cdot\mathbf{f}\sigma_1\,, \tag{29c}$$

where the notation

$$\sigma_0\equiv-\xi/\eta \tag{29d}$$

is used, and where LM expectation value brackets are omitted. (Since these are operator equations in TLS space, σ_0 is to be understood as being multiplied by the unit matrix.) It is easily seen that the results of I for the magnetic and electric dipole cases treated there are obtained by setting $a_{im}=\delta_{im}$ in the former case and $a_{im}=\delta_{i1}\delta_{m1}$ in the latter. Equations (29) describe the

[4] Where brackets are used to indicate expectation value with respect to the LM, the subscript "LM" will be added if the enclosed expression refers to both the TLS and LM, while no subscript will be used if the enclosed expression refers only to the LM.

behavior of the most general TLS coupled to a thermal-reservoir type LM.

We can take expectation values with respect to the TLS in Eqs. (29). This leaves the equations formally unaltered, and the σ's now stand for their respective expectation values in both LM and TLS spaces. Henceforth, Eqs. (29) will be understood to refer to these expectation values. Discussion of the physical significance of these equations will be postponed until the second generalization has been carried out.

II.

The second generalization refers directly to the LM rather than to the TLS. Instead of assuming the LM to be only a thermal reservoir of given temperature, we consider the LM to consist of two parts: The first part (to be referred to as LM_1) is the same thermal reservoir considered heretofore; the second part (to be referred to as LM_2) consists of a large number of two-level systems identical to the TLS under consideration, situated in a similar environment, and loosely and randomly coupled to our system and to each other. The individual two-level systems of LM_2 are thus coupled (in addition to the coupling to each other and to the TLS under consideration) to a thermal reservoir and to external forces. The thermal reservoir may be LM_1 itself, but since the possibility of the transmission of effects through LM_1 will be excluded, it is simpler to consider the systems of LM_2 coupled to a thermal reservoir that is identical to LM_1 but independent. In the notation to be used, LM_2 consists of the mutually coupled two-level systems only, the thermal reservoir and external forces being described separately.

The Hamiltonian for our problem is obtained by adding several terms to the expression in Eq. (7), the result being

$$H=\tfrac{1}{2}\hbar\omega\sigma_3+\tfrac{1}{2}\hbar\sum_\alpha \sigma_\alpha \mathbf{a}_\alpha\cdot(\mathbf{F}^{(1)}+\mathbf{F}^{(2)}+\mathbf{f}) +H_{\mathrm{LM1}}+H_{\mathrm{LM2}}+H^{(2)}, \quad (30)$$

where H_{LM1} and H_{LM2} are the energy operators of LM_1 and LM_2, respectively, $F^{(1)}$ and $F^{(2)}$ are the coordinates through which LM_1 and LM_2, respectively, couple to the TLS, and $H^{(2)}$ contains the coupling term of LM_2 to any other systems, the energy operators for those systems, and the coupling term of LM_2 with prescribed external forces; that is, $H^{(2)}$ refers to everything that couples to LM_2 except the TLS under consideration.

We pass now from the Heisenberg picture, which has been used in the preceding discussion, to the interaction picture. All the operators are transformed into primed quantities in the manner[5]

$$A'=UAU^*, \quad (31)$$

where U is a unitary operator satisfying the differential equation and initial condition

$$i\hbar\dot{U}(t)=U(t)H^{(2)}(t), \quad (31a)$$

$$U(0)=1. \quad (31b)$$

All the primed operators now satisfy the equation of motion

$$i\hbar\dot{A}'=[A', H'-H^{(2)\prime}]+i\hbar(\partial A'/\partial t); \quad (32)$$

in other words, the effective Hamiltonian that enters into the equations of motion for the primed operators no longer contains any reference to the influence of external systems or forces on LM_2. Formally, LM_1 and LM_2 appear equivalently in these equations. In this transformation, the effect of the external influences on LM_2 has been removed from the operators and transferred to the state vector, which becomes

$$\Psi'(t)=U(t)\Psi. \quad (33)$$

It should be remembered that Ψ is independent of the time and would be the initial state if the Schrödinger picture were used; Eq. (33) may be written as

$$\Psi'(t)=U(t)\Psi'(0). \quad (34)$$

Now, if there were no coupling between the TLS and LM_2, Eq. (34) would yield the entire time development of LM_2, and leave all the other systems to which reference is made in the Hamiltonian of Eq. (30) unaffected, that is in their Heisenberg (initial) state. The presence of the $\frac{1}{2}\hbar\sigma_\alpha\mathbf{a}_\alpha\cdot\mathbf{F}^{(2)}$ coupling term affects U only indirectly through the time development of $H^{(2)}(t)$ [note that $H^{(2)}(0)$ does not contain this coupling term] and does not produce lowest order effects. The difference between $\Psi'(t)$ and the corresponding state in which there is no coupling between the TLS and LM_2 is due only to the effect on LM_2 of the TLS, and is slight. We approximate by ignoring this effect in $\Psi'(t)$ and consider $\Psi'(t)$ to denote the state in which the TLS and LM_1 are unaffected while LM_2 has developed under the influence of those systems (and external forces) explicitly entering into $H^{(2)}$. (This approximation is similar in spirit to approximations previously made for LM_1, where LM variables in interaction terms were replaced by their uncoupled values. In the present instance, LM_2 may be uncoupled only from the TLS but not from the effect of $H^{(2)}$. This is the essential difference between LM_1 and LM_2.)

Analytically, the approximation just performed may be described as follows: Set

$$\Psi=\psi_{\mathrm{TLS}}\psi_{\mathrm{LM1}}\psi_{\mathrm{LM2}}, \quad (35a)$$

(this implies that the coupling is turned on at $t=0$), which is the same as

$$\Psi'(0)=\psi_{\mathrm{TLS}}'(0)\psi_{\mathrm{LM1}}'(0)\psi_{\mathrm{LM2}}'(0). \quad (35b)$$

Consider $H^{(2)}(t)$ to be obtained from $H^{(2)}(0)$ (by means of the Heisenberg equations of motion) by neglecting

[5] It should be noted that the interaction picture is obtained here by starting with the Heisenberg picture. This accounts for the order of the factors in the right side of Eq. (31a), the opposite of that used when starting with the Schrödinger picture.

the $F^{(2)}$ term in Eq. (30). Then $H^{(2)}(t)$ and, therefore—as is evident from Eq. (31a)—$U(t)$ contain only LM_2 variables. Thus,

$$\Psi'(t)=\psi_{TLS}'(0)\psi_{LM1}'(0)U(t)\psi_{LM2}'(0) =\psi_{TLS}'(0)\psi_{LM1}'(0)\psi_{LM2}'(t). \quad (36)$$

Since LM_1 and LM_2 are large and incompletely described systems, we will describe their initial states, as has been done previously for LM_1, by ensemble averages. For this purpose, it is more convenient to use density matrices instead of state vectors. We therefore write, in place of Eq. (36),

$$P'(t)=\rho_{TLS}'(0)\rho_{LM1}'(0)\rho_{LM2}'(t), \quad (37)$$

where P is the density matrix for the combination of systems, and the ρ's are individual density matrices. Both $\psi_{LM2}'(t)$ and $\rho_{LM2}'(t)$ describe the development of LM_2 in the absence of coupling to the TLS under consideration.

We come now to the essential aspect, and significant approximation, of the analysis of LM_2. LM_2 is obviously not a thermal reservoir, since the individual two-level systems of which it is composed can interchange an amount of energy comparable to their own energy with the thermal reservoir and external forces to which they are coupled. LM_2 cannot, therefore, be described by a fixed temperature. We assume, however, that the randomness associated with LM_2 (due, mainly, to the random coupling among the individual two-level systems) is sufficiently great so that at any given time LM_2 may be described by a temperature, this temperature being, in general, a function of the time.[6] According to this assumption, the density matrix for LM_2 is given by

$$\rho_{LM2}'(t)=Z_2^{-1}\exp[-H_{LM2}/kT(t)], \quad (38)$$

where

$$Z_2(t)=\text{Trace}\exp[-H_{LM2}/kT(t)]. \quad (38a)$$

$T(t)$ is left unspecified for the time being, but we assume that it changes slowly compared to $\exp(i\omega t)$.

Now, LM_2, like LM_1, is a large system with many energy levels closely spaced. It may therefore be treated, in the analysis of its interaction with the TLS, in the same manner as LM_1, but with cognizance of the time dependence of the temperature. (It is to be noted that the temperature enters into the analysis only through the density matrix.) Our problem thus reduces itself to the analysis of the interaction of a TLS with external fields and with two LM's, one of which has a time-dependent temperature.

Before this problem is considered, it is useful to look at a simpler situation, one in which there are two LM's with different, but *fixed*, temperatures. Very little analysis is required to notice that the change required in the results obtained for a single LM is the replacement of η by $\eta^{(1)}+\eta^{(2)}$, and the replacement of ξ by $\xi^{(1)}+\xi^{(2)}$, where the superscripts (1) and (2) refer to LM_1 and LM_2, respectively, and $\xi^{(i)}$ and $\eta^{(i)}$ are defined for each LM in the same manner as ξ and η, respectively, by Eqs. (14a) and (14b). From Eq. (29d) we have

$$\sigma_0^{(i)}=-\xi^{(i)}/\eta^{(i)}; \quad (39)$$

thus Eqs. (29a)–(29c) remain valid provided we set

$$\eta=\eta^{(1)}+\eta^{(2)}, \quad (40a)$$

and

$$\sigma_0=(\sigma_0^{(1)}\eta^{(1)}+\sigma_0^{(2)}\eta^{(2)})(\eta^{(1)}+\eta^{(2)})^{-1}. \quad (40b)$$

We return now to situation in which $T^{(2)}$ is time-dependent. This time dependence affects several calculations that lead to Eqs. (29). First, there is the computation of expectation values of LM_2 variables. The expectation value of an operator $A'(t)$ referring to a single time (and uncoupled from the TLS) poses no problem since it is given by Trace $\rho_{LM}'(t)A'(t)$. The expectation value of an operator of the type $\{A_1'(t_1),A_2'(t_2)\}$, referring to two different times, requires, in general, another method of evaluation. In the present analysis, however, this type of expectation value is needed only in cases where the result is negligible unless t_1 is very close to t_2. [See, for instance, Eqs. (14) and (16).] Bearing in mind that $T^{(2)}(t)$ is a slowly varying function, we can use previous results for this type of expectation value, with $\xi^{(2)}$ and $\eta^{(2)}$—which are functions of $T^{(2)}$—evaluated at either t_1, t_2, or an intermediate value. Then, there is the question of time integration where $\xi^{(2)}$ or $\eta^{(2)}$ is part of the integrand. [See, for instance, Eq. (27).] Here too, no formal modification of the final result is necessary, for in this integration the main contribution comes only from a very small neighborhood of a definite time, so that $\xi^{(2)}$ or $\eta^{(2)}$ may be evaluated at that time and taken outside the integral. It is seen, therefore, that Eqs. (29a)–(29c) and (40) remain formally unchanged, but with $\eta^{(2)}$ and $\sigma_0^{(2)}$ being considered now functions of t (the same argument as that of the σ's and $\mathbf{f}$).

Our final task is the evaluation of $T^{(2)}$, or, more directly, of $\sigma_0^{(2)}$ and $\eta^{(2)}$, through which the behavior of the TLS depends on $T^{(2)}$. From Eqs. (39), (14a), and (14b), we have

$$\sigma_0^{(2)}=\frac{\exp(-\hbar\omega/kT^{(2)})-1}{\exp(-\hbar\omega/kT^{(2)})+1}. \quad (41)$$

This is the expectation value of the energy, in units of $\frac{1}{2}\hbar\omega$, of a TLS of the type under consideration when it is in thermal equilibrium with a reservoir at temperature $T^{(2)}$. We may therefore regard $\sigma_0^{(2)}$ as the average energy (at time t) of the two-level systems that constitute LM_2. Since these two-level systems are identical to the TLS under consideration and are situated in a

[6] Since LM_2 is composed of two-level systems, its temperature may assume negative as well as positive values, depending on the average energy of the two-level systems.

similar environment, this energy is also equal to $\sigma_3(t)$.[7] Thus, we have the important result

$$\sigma_0^{(2)}=\sigma_3(t). \tag{42}$$

As far as $\eta^{(2)}$ is concerned, no such simple consideration applies. The best that can be done within the scope of the present analysis is to observe that $\eta^{(2)}$ is not as strongly dependent on $T^{(2)}$ as $\sigma_0^{(2)}$ (or $\xi^{(2)}$)—as can be seen from Eqs. (14)—and to approximate it by an average value.

From Eqs. (29a)–(29c), (40), and (42) we finally obtain

$$\dot{\sigma}_1=(-\omega+\tilde{\eta}\mathbf{a}_1\cdot\mathbf{a}_2)\sigma_2-\tilde{\eta}a_2^2\sigma_1+\eta\mathbf{a}_1\cdot\mathbf{a}_3(\sigma_3-\sigma_0)+\mathbf{a}_2\cdot\mathbf{f}\sigma_3-\mathbf{a}_3\cdot\mathbf{f}\sigma_2, \tag{43a}$$

$$\dot{\sigma}_2=(\omega+\tilde{\eta}\mathbf{a}_1\cdot\mathbf{a}_2)\sigma_1-\tilde{\eta}a_1^2\sigma_2+\eta\mathbf{a}_2\cdot\mathbf{a}_3(\sigma_3-\sigma_0)+\mathbf{a}_3\cdot\mathbf{f}\sigma_1-\mathbf{a}_1\cdot\mathbf{f}\sigma_3, \tag{43b}$$

$$\dot{\sigma}_3=-(a_1^2+a_2^2)\eta(\sigma_3-\sigma_0)+\mathbf{a}_1\cdot\mathbf{f}\sigma_2-\mathbf{a}_2\cdot\mathbf{f}\sigma_1, \tag{43c}$$

where

$$\tilde{\eta}\equiv\eta^{(1)}+\eta^{(2)},\quad \eta\equiv\eta^{(1)},\quad \sigma_0\equiv\sigma_0^{(1)}. \tag{43d}$$

In summary, these equations describe the behavior (in terms of expectation values) of a general TLS subject to external fields and coupled both to a thermal reservoir and a large number of systems identical to itself. The dipole moment is given in terms of the σ's by Eq. (2), and the energy is $\frac{1}{2}\hbar\omega\sigma_3$. Two relaxation constants, $\tilde{\eta}$ and η (with η being a better "constant" than $\tilde{\eta}$, which may be regarded as depending to some extent on the energy of the TLS) and an equilibrium energy σ_0 enter into these equations. The equilibrium energy is that associated with the temperature T_1 of the thermal reservoir, and is obtained by replacing superscript (2) with superscript (1) in Eq. (41).[8]

III.

A few special types of TLS are of immediate interest. As mentioned previously, the magnetic dipole type has $\mathbf{a}_1=\hat{x}$, $\mathbf{a}_2=\hat{y}$, $\mathbf{a}_3=\hat{z}$, $\mathbf{a}_4=0$. Equations (43) become

$$\dot{\sigma}_x=-\omega\sigma_y-\tilde{\eta}\sigma_x+f_x\sigma_z-f_z\sigma_x, \tag{44a}$$

$$\dot{\sigma}_y=\omega\sigma_x-\tilde{\eta}\sigma_y+f_z\sigma_x-f_x\sigma_z, \tag{44b}$$

$$\dot{\sigma}_z=-2\eta(\sigma_3-\sigma_0)+f_x\sigma_y-f_y\sigma_x. \tag{44c}$$

[7] Strictly speaking, there is an approximation involved in this statement, since LM expectation values are averages both with respect to a thermal (canonical) ensemble—implicit in the density matrix employed—and a quantum-mechanical ensemble, while TLS expectation values are averages only with respect to the latter.

[8] Equations (43), involving two dissipation parameters, $\tilde{\eta}$ and η, are more general than the LM considered in the above discussion. More general loss mechanisms may be obtained by replacing LM_1 with several thermal reservoirs at different temperatures, some of these temperatures being, possibly, prescribed slowly varying functions of the time. Equations of the form of Eq. (40) can then be used to obtain new values for the two dissipation parameters with Eqs. (43) remaining formally unchanged.

These are essentially the Bloch equations,[9] with $(2\eta)^{-1}$ being the longitudinal relaxation time T_1, and $\tilde{\eta}^{-1}$ being the transverse relaxation time T_2. If the spin-lattice coupling is regarded as coupling between the TLS and a thermal reservoir, and spin-spin coupling as coupling among identical two-level systems, then Eqs. (44) and (43d) show the contribution of each type of coupling to the relaxation process. It should be noticed that it is not, in general, correct to refer to T_2 as the spin-spin relaxation time (as is often done) unless $\eta^{(1)}$ is negligible compared to $\eta^{(2)}$. If $\eta^{(2)}$ is zero, Eqs. (44) reduce to the results of I [Eqs. (I.84)].

As far as the electric dipole TLS is concerned, there are many possible types, depending on the choice of the $\mathbf{a}$'s. $\mathbf{a}_4$ does not enter into the equations of motion and, therefore, does not affect the behavior of the TLS. $\mathbf{a}_4$ and $\mathbf{a}_3$ determine the "permanent" dipole moment, or the nonoscillating part of the (expectation value of the) dipole moment of the free TLS, for only σ_3 can have a nonoscillating value different from zero when $f=\tilde{\eta}=\eta=0$ in Eqs. (43). Since $\mathbf{a}_4$ plays no dynamic role, we will ignore it henceforth, and refer to the constant part of σ_3 (times $\mu\mathbf{a}_3$) as the permanent dipole moment. A case in which the dipole moment has a particularly simple appearance is that in which $\mathbf{a}_2=\mathbf{a}_3=0$, so that $\mathbf{d}=\mu\mathbf{a}_1\sigma_1$. This is the electric dipole case treated in I. Taking $a_1^2=1$, $\mathbf{a}_1\cdot\mathbf{f}\equiv f$, Eqs. (43) become

$$\sigma_1=-\omega\sigma_2, \tag{45a}$$

$$\dot{\sigma}_2=\omega\sigma_1-\tilde{\eta}\sigma_2-f\sigma_3, \tag{45b}$$

$$\dot{\sigma}_3=-\eta(\sigma_3-\sigma_0)+f\sigma_2, \tag{45c}$$

which, for $\eta^{(2)}=0$, reduce to Eqs. (I.67). A permanent dipole moment may be added to this case, while still maintaining a relatively simple form of the equations, by considering $\mathbf{a}_2=0$, $\mathbf{a}_3=\epsilon\mathbf{a}_1$. (Here, the permanent dipole moment has the same spatial direction as the oscillating dipole moment.) The result is

$$\dot{\sigma}_1=-\omega\sigma_2-\epsilon\dot{\sigma}_3, \tag{46a}$$

$$\dot{\sigma}_2=\omega\sigma_1-\tilde{\eta}\sigma_2+\epsilon f\sigma_1-f\sigma_3, \tag{46b}$$

$$\dot{\sigma}_3=-\eta(\sigma_3-\sigma_0)+f\sigma_2, \tag{46c}$$

where Eq. (46a) has been simplified in appearance by substitution from Eq. (46c).

It is not the purpose of the present article to present equations for large number of special cases, although there are many others than the above three particularly simple ones that are of interest. Neither is it the purpose of the present article to discuss the solution of the above equations for given driving fields, which will be done in a forthcoming article. We will, however, discuss the solution of the equations in the absence of a driving field, that is, the free decay of a TLS from given initial conditions. For the sake of simplicity, we consider only

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

the case in which $\mathbf{a}_3$ is either perpendicular to both $\mathbf{a}_1$ and $\mathbf{a}_2$, or is equal to zero. From Eqs. (43) we have, under the above conditions,

$$\sigma_1 = (-\omega + \tilde{\eta}\mathbf{a}_1 \cdot \mathbf{a}_2)\sigma_2 - \tilde{\eta} a_2^2 \sigma_1, \qquad (47a)$$

$$\dot{\sigma}_2 = (\omega + \tilde{\eta}\mathbf{a}_1 \cdot \mathbf{a}_2)\sigma_1 - \tilde{\eta} a_1^2 \sigma_2, \qquad (47b)$$

$$\dot{\sigma}_3 = -(a_1^2 + a_2^2)\eta(\sigma_3 - \sigma_0). \qquad (47c)$$

The last equation may be solved immediately, the result being

$$\sigma_3 = \sigma_0 + [\sigma_3(0) - \sigma_0] \exp[-(a_1^2 + a_2^2)\eta t]. \qquad (48)$$

Equations (47a) and (47b) yield the same equation for both σ_1 and σ_2:

$$\ddot{\sigma}_{1,2} + \tilde{\eta}(a_1^2 + a_2^2)\dot{\sigma}_{1,2} + \{\omega^2 + \tilde{\eta}^2[a_1^2 a_2^2 - (\mathbf{a}_1 \cdot \mathbf{a}_2)^2]\}\sigma_{1,2} = 0. \qquad (49)$$

The solution of Eq. (49) is

$$\sigma_1 = \exp[-\tilde{\eta}(a_1^2 + a_2^2)t][a_1 \cos\Omega t + B_1 \sin\Omega t], \qquad (50a)$$

$$\sigma_2 = \exp[-\tilde{\eta}(a_1^2 + a_2^2)t][a_2 \cos\Omega t + B_2 \sin\Omega t], \qquad (50b)$$

where A_i and B_i are constants determined by the initial conditions, and

$$\Omega^2 = \omega^2\{1 - (\tilde{\eta}^2/\omega^2)[(\mathbf{a}_1 \cdot \mathbf{a}_2)^2 + \tfrac{1}{4}(a_1^2 - a_2^2)^2]\}. \qquad (50c)$$

We see that σ_3 approaches exponentially its equilibrium value, and that σ_1 and σ_2 undergo exponentially damped oscillation. The frequency of oscillation Ω is of interest. For $\mathbf{a}_1$ and $\mathbf{a}_2$ equal in magnitude and perpendicular to each other, Ω is equal to ω, the frequency of the free TLS; otherwise Ω is less than ω. Equation (50c) puts into perspective the results of I, where it was found that the frequency of the undriven damped oscillator was equal to that of the free oscillator for the magnetic dipole case but not for the electric dipole case. It is seen, now, that the magnetic dipole TLS has just that combination of $\mathbf{a}$'s which yields $\Omega = \omega$, but the electric dipole TLS treated in I has $\mathbf{a}_2 = 0$, $a_1^2 = 1$, giving

$$\Omega^2 = \omega^2[1 - \tfrac{1}{4}(\tilde{\eta}^2/\omega^2)], \qquad (51)$$

in accordance with the results of I.[10]

One encounters occasionally the statement that any TLS is equivalent to a system of spin $\frac{1}{2}$ in an appropriate magnetic field.[11] While this statement is true in the absence of dissipation, it is only an approximation when the TLS is coupled to an LM, as the above frequency consideration reveals, and as can be seen from the dependence of Eqs. (43) on the type of TLS considered.

It was shown previously [Eqs. (44)] that the Bloch equations are a special case of Eqs. (43). The Bloch equations were developed originally for macroscopic matter. They also apply to the expectation values for a microscopic system, and, in this sense, correspond to a special case of Eqs. (43). One might ask how the present theory applies to macroscopic matter, or to a large number of systems identical to—but possibly, with different orientation than—the TLS under consideration. Can one, as in the case of the Bloch equations, regard a linear combination of the (four) σ's as the component of dipole moment along a given direction for macroscopic matter containing N systems similar to our TLS? The macroscopic dipole moment $\mathbf{D}$ is given by

$$\mathbf{D} = \sum_{j=1}^{N} \mathbf{d}^{(j)} = \sum_{j=1}^{N} \sum_{\alpha=1}^{4} \mathbf{a}_\alpha^{(j)} \sigma_\alpha^{(j)}, \qquad (52)$$

where the superscript j refers to the jth TLS. The above question is equivalent to asking if $\sigma_\alpha^{(j)}$ is independent (except for a multiplicative constant) of j. It is clear that if the systems are all oriented in the same manner, then $\mathbf{a}_\alpha^{(j)} = \mathbf{a}_\alpha$, $\sigma_\alpha^{(j)} = \sigma_\alpha$,

$$\mathbf{D} = N\mathbf{d}, \qquad (53)$$

and the answer to the above question is, obviously, in the affirmative. If the microscopic systems are not all oriented similarly, then the coefficients $\mathbf{a}_i^{(j)} \cdot \mathbf{f}$ in Eq. (43a) are different for differently oriented systems. If, because of symmetry conditions, the solutions of Eqs. (43) are essentially unaffected by a particular distribution of orientations, then the answer is still in the affirmative, but if no such symmetry exists, then Eqs. (43) cannot be regarded as equations for macroscopic polarization. The macroscopic result must be obtained through Eq. (52), that is, by solving first the microscopic problem, and then superposing the solutions.

The essential aspects of the orientation of a magnetic dipole TLS are determined by the field that is responsible for the level separation. If this field is the same for all the two-level systems, Eqs. (43)—which reduce in this instance to the Bloch equations—may be regarded as equations for macroscopic polarization. Such is the case for macroscopic matter in an external (laboratory) field. If, however, the field responsible for level separation is an internal field and has different orientation for different dipoles, the above equations may be applied only on a microscopic scale.

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[10] A comparison might be made between Eqs. (47a) and (47b) on the one hand, and equations for the coordinate and momentum of a harmonic oscillator with dissipation on the other. If both coordinate and momentum couple to the loss mechanism, then the equations of motion for the harmonic oscillator variables (in appropriate units) are

$$\dot{p} = -\omega q - \eta_1 p, \quad \dot{q} = \omega p - \eta_2 q.$$

It is seen that if $\mathbf{a}_1$ and $\mathbf{a}_2$ are orthogonal, σ_2 and σ_1 correspond to the coordinate and momentum, respectively, of the harmonic oscillator. It can also be seen readily that the oscillatory frequency of the freely decaying harmonic oscillator is equal to ω only if $\eta_1 = \eta_2$. Comparison with the dissipation terms in Eqs. (47a) and (47b) shows that this corresponds to our requirement (in addition to that of orthogonality) that $a_1^2 = a_2^2$.

[11] N. Bloembergen and Y. R. Shen, Phys. Rev. **133**, A37 (1964); A. Abragam, *The Principles of Nuclear Magnetism* (Oxford University Press, New York, 1961), p. 36.