MACROSCOPIC EQUATIONS FOR THE MAGNETIC MOMENT IN SOME MAGNETIC RESONANCE PROBLEMS

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A method is proposed for passing from Bloch's kinetic equations for the density matrix to the equations for the macroscopic magnetic moment of a nuclear system consisting of interacting spins. The method is applied to the problem of three equivalent particles of spin ½ coupled to each other by dipole-dipole interaction. The solution of the equations obtained is given.

THE Boltzmann equation for the density matrix of a spin system[1-3] is used for the description of nuclear magnetic absorption in the general case. Wangsness and Bloch have shown[4] that in certain cases Bloch's phenomenological equation can be applied to the case of noninteracting spins. In this paper we shall give a method for passing from the equations for the density matrix to the system of equations for the expectation values of the spin functions in the general case. By eliminating from this system all the spin functions other than the components of the macroscopic magnetic moment we can obtain an equation analogous to Bloch's phenomenological equation containing higher derivatives with respect to time. Although, apparently, it is not possible to make the transition to the macroscopic equations in the general case, nevertheless in one special case it turns out to be possible to establish certain properties of the relaxation matrix for a system consisting of an arbitrary number of interacting spins. Moreover, investigation of specific problems by means of these equations will perhaps facilitate the understanding of the physical meaning of the relaxation coefficients in the equation for the density matrix.

For the description of the transition to the macroscopic equations we shall need the differential equation for the expectation value of the spin function similar to the one obtained by Wangsness and Bloch in the discussion of a noninteracting system of spins. We shall start with the general equation for the density matrix for a spin system[2] (here and subsequently we shall use the notation introduced by Bloch):

$$\frac{d}{dt}(gv|\sigma|g'v') + i(gv|[E,\sigma]|g'v') = (gv|\Gamma(\sigma)|g'v'),$$

$$(gv|\Gamma(\sigma)|g'v') = \sum_p \sum_{v''v'''} \{2e^{\hbar p/kT}\Gamma^p_{gg'}(vv'v''v''')(g + p, v''|\sigma|g'+p, v''') - \Gamma^p_{gg}(vv'''v''v'')(gv'''|\sigma|gv') - \Gamma^p_{g'g'}(v'''v'v''v'')(gv|\sigma|g'v''')\},$$

$$\Gamma^p_{gg'}(vv'v''v''') = \pi\sum_{u,u'}\int \eta_u(f)\,\eta_{u'}(f-p)\,P(f)\,(gv, fu|G|g + p, v''; f-p, u')(g'+p, v'''; f-p, u'|G|g'v', fu)\,df. \qquad (1)$$

Here $(gv|\sigma|g'v')$ is the density matrix of the spin system; g, v are the quantum numbers of the states of the spin system; f, u are the quantum numbers of the states of the heat reservoir; $\eta_u(f)$ is the density of states of the heat reservoir with quantum numbers u; $P(f) = \exp(-\hbar f/kT)$; G is the operator describing the interaction of the spin system with the heat reservoir; E is the interaction energy of the spin system with the external magnetic field **H**.

In order to simplify the subsequent presentation we shall consider a spin system consisting of equivalent particles of spin I coupled by dipole-dipole interaction. Then G has the form

$$G = \sum_{\tau, s<t} I^{\tau}_{st} F^{-\tau}_{st}, \qquad \tau = 0, \pm 1, \pm 2, \qquad (2)$$

where s and t are numbers specifying the particles,

$$I^0_{st} = \sqrt{8/3}\,[I_{sz}I_{tz} - \tfrac{1}{4}(I^+_s I^-_t + I^-_s I^+_t)], \quad I^{\pm 1}_{st} = I^{\pm}_s I_{tz} + I_{sz}I^{\pm}_t,$$

$$I^{\pm 2}_{st} = I^{\pm}_s I^{\pm}_t; \qquad F^0_{st} = -\sqrt{3/8}\,C_{st}(1 - 3\cos^2\theta_{st}),$$

$$F^{\pm 1}_{st} = -\tfrac{3}{2}C_{st}\sin\theta_{st}\cos\theta_{st}e^{\pm i\varphi_{st}},$$

$$F^{\pm 2}_{st} = -\tfrac{3}{4}C_{st}\sin^2\theta_{st}e^{\pm 2i\varphi_{st}},$$

where $C_{st} = \hbar\gamma^2/r_{st}^3$, and θ_{st} and φ_{st} are the angles specifying the orientation of the vector $\mathbf{r}_{st}$, between the positions of the particles s and t.

We note that each term in the sum (2) induces transitions only between levels which are separated from each other by $\tau\omega$ ($\omega = \gamma H_0$).

On substituting G in the form (2) into the expression for $\Gamma^p_{gg'}$ we obtain

$$\Gamma^p_{gg'} = \sum_{\tau, st, s't'} \Phi^\tau_{st, s't'} (gv \mid I_{st}^{-\tau} \mid g + \tau\omega, v'') (g' + \tau\omega, v''' \mid I^\tau_{s't'} \mid g'v'), \qquad (3)$$

$$\Phi^\tau_{st, s't'} = \pi \sum_{u, u'} \int \eta_u (f)\, \eta_{u'} (f - \tau\omega)\, P(f)\, (fu \mid F^\tau_{st} \mid f - \tau\omega, u') \times (f - \tau\omega, u' \mid F^{-\tau}_{s't'} \mid fu)\, df. \qquad (3')$$

On taking into account the remark made with respect to the operators I^τ_{st}, we obtain

$$\sum_{v''} (gv \mid I_{st}^{-\tau} \mid g + \tau\omega, v'') (g + \tau\omega, v'' \mid Q \mid g'v') = (gv \mid I_{st}^{-\tau} Q \mid g'v') \qquad (4)$$

(here Q is an arbitrary spin operator). This relation enables us to write Eq. (1) in operator form

$$\frac{d\sigma}{dt} + i\,[E, \sigma] = \sum_{\tau, st, s't'} \Phi^\tau_{st, s't'} \{2e^{\beta\tau} I_{st}^{-\tau} \sigma I^\tau_{s't'} - I_{st}^{-\tau} I^\tau_{s't'} \sigma - \sigma I_{st}^{-\tau} I^\tau_{s't'}\}, \qquad (5)$$

where $\beta = \hbar\omega/kT$. Then on multiplying both sides of (5) by the spin function and on obtaining the expectation values of the spin functions we can obtain the following equation

$$d\langle Q\rangle / dt = - i \langle [Q, E] \rangle + \sum_{\tau, st, s't'} \Phi^\tau_{st, s't'} \langle I_{st}^{-\tau} [Q, I^\tau_{s't'}] + [I_{st}^{-\tau}, Q]\, I^\tau_{s't'} \rangle, \qquad (6)$$

where

$$\langle Q \rangle = \mathrm{Tr}\,(Q\sigma).$$

If into this equation we substitute in place of Q the operator for the magnetic moment **M**, then on the right hand side we obtain the expectation value of a certain polynomial in the spin operators of the system under discussion. On substituting now into (6) these polynomials in place of Q we shall obtain a system of coupled equations which will contain expectation values of certain spin polynomials. It can be easily seen that for a finite number of spins under consideration the degree of the polynomial will be finite, and consequently the system of equations will be finite. Since for any spin system it is possible to obtain an equation analogous to (6),* then by the same method it is possible to obtain a system of macroscopic equations in the most general case. The number of linearly independent equations in the macroscopic system of equations will in the general case be of the same order as in the equation for the density matrix. However, in certain cases the number of spin functions related by the equations to the components of the magnetic moment may be greatly restricted by the conditions of the problem, while the system of equations for the density matrix contains the complete amount of information with regard to the spin system and, consequently, will be more complicated.

*Bloch[3] has obtained such an equation in the most general form in the case of an arbitrary external magnetic field.

By eliminating from the system of the macroscopic differential equations of the first order all the spin functions with the exception of the magnetic moment we shall obtain an equation of high order with respect to the time derivatives, and nonlinear in the field. In the absence of an alternating field the equation for M_z, for example, must have the form

$$\frac{d^{(N)} M_z}{dt^{(N)}} + \frac{1}{T_1} \frac{d^{(N-1)} M_z}{dt^{(N-1)}} + \ldots + \frac{M_z - M^0}{T_N} = 0, \qquad (7)$$

where $T_1, T_2 \ldots T_N$ are expressed in terms of the relaxation coefficients Φ, while N is determined by the number of equations in the macroscopic system of equations. The form of Eq. (7) clearly demonstrates the well known fact that the relaxation process for the components of the magnetic moment of a complex spin system is, generally speaking, described by a large number of exponentials.

The macroscopic equations assume their simplest form in the case of a system of equivalent nuclei, when the relaxation coefficients do not depend on the index τ. This occurs, for example, in a liquid with a sufficiently short correlation time τ_c. Moreover, we shall assume that the temperature is sufficiently high to satisfy the condition $\beta \ll 1$ (this is almost always satisfied). In this case Eq. (5) for the density matrix assumes the form (cf. for example references 2 and 5)

$$\frac{d\sigma}{dt} = - i\,[E, \sigma] + \sum_{\tau, st, s't'} \Phi^\tau_{st, s't'} [I^\tau_{st}, [I^{-\tau}_{s't'}, \sigma - \sigma_0]]. \qquad (8)$$

Here σ_0 is the equilibrium density matrix.

The coefficients $\Phi^\tau_{st,s't'}$ [cf. formula (3)] no longer depend on ω, since we assume that $\omega\tau \ll 1$. For the expectation value of the spin function we obtain starting with Eq. (8)

$$d\langle Q \rangle / dt = - i \langle [Q, E] \rangle + S(Q),$$

$$S(Q) = \sum_{\tau, st, s't'} \Phi^\tau_{st, s't'} \{\langle [I^\tau_{st}, [Q, I^{-\tau}_{s't'}]] \rangle - \langle [I^\tau_{st}, [Q, I^{-\tau}_{s't'}]] \rangle_0\}; \qquad (9)$$

$\langle\ \rangle_0$ denotes averaging with respect to the equilibrium density matrix.

As is well known (cf., for example, Ayant's paper[6]), the coefficients $\Phi^{\tau}_{st,s't'}$ are the quantum analogue of the spectral density of the correlation function of the quantities F^{τ}_{st}. Assuming the motion of the medium to be classical we can evaluate these functions when the molecule (the spin system under discussion) undergoes Brownian rotation. Such calculations have been carried out by Aleksandrov[7] and by Hubbard.[5] By utilizing their calculations we can obtain in our case

$$\Phi^{\tau}_{st,\,s't'} = (3\hbar^2\gamma^4\tau_c / 10r^3_{st}r^3_{s't'})\left(1 - \tfrac{3}{2}\sin^2\vartheta\right) \equiv \Phi_{st,\,s't'}, \quad (10)$$

where ϑ is the angle between the vectors $\mathbf{r}_{st}$ and $\mathbf{r}_{s't'}$. It may be seen from this expression that the coefficients $\Phi^{\tau}_{st,s't'}$ do not depend on τ and S(Q) may now be written in the form

$$S(Q) = \sum_{st,\,s't'} \Phi_{st\,s't'}\,[\langle X_{st,\,s't'}(Q)\rangle - \langle X_{st,\,s't'}(Q)\rangle_0],$$

$$X_{st,\,s't'}(Q) = \sum_{\tau=-2}^{2} [I^{\tau}_{st},\,[Q,\,I^{-\tau}_{s't'}]]. \quad (11)$$

We now show that $X_{st,s't'}(Q)$ transform under rotations of the coordinate system in the same way as Q. In order to do this we consider the rotation of the coordinate system about any two axes (a rotation about a third axis may be expressed in terms of the rotations about the other two axes). We introduce $S_{\varphi} = \exp(-i\varphi I_z)$ the operator for the rotation about the z axis through an angle φ and $S_{\theta} = \exp(-i\theta I_y)$ the operator for the rotation about the y axis (here $\mathbf{I} = \Sigma_s \mathbf{I}_s$). Then, on taking into account the rules for the transformation of the spin operators I_x, I_y, I_z, it may be easily shown that

$$S_{\varphi}I^{\tau}_{st}S^{-1}_{\varphi} = e^{-i\tau\varphi}I^{\tau}_{st}, \quad S_{\theta}I^{\tau}_{st}S^{-1}_{\theta} = \sum_{\mu} q^{\tau}_{\mu}(\theta)\, I^{\mu}_{st}, \quad (12)$$

where the following relations are satisfied by the $q^{\tau}_{\mu}(\theta)$:

$$q^{-\tau}_{-\mu}(\theta) = q^{\tau}_{\mu}(\theta) = q^{\mu}_{\tau}(-\theta), \quad \sum_{\mu} q^{\tau}_{\mu}(-\theta)\, q^{\mu}_{\lambda}(\theta) = \delta_{\tau\lambda}. \quad (13)$$

By utilizing (12) and (13), we obtain

$$S_{\theta}X_{st,\,s't'}(Q)\,S^{-1}_{\theta} = X_{st,\,s't'}(S_{\theta}QS^{-1}_{\theta}),$$
$$S_{\varphi}X_{st,\,s't'}(Q)\,S^{-1}_{\varphi} = X_{st,\,s't'}(S_{\varphi}QS^{-1}_{\varphi}).$$

Thus, under the rotations S_{φ} and S_{θ} the spin functions $X_{st,s't'}(I_{x,y,z})$ transform in the same way as $I_{x,y,z}$. Since $X_{st,s't'}(\mathbf{I})$ is the operator for a vector quantity, the commutation rules of $\mathbf{I}$ with this quantity are known.[8] By using them we obtain

$$-i\,\langle[\mathbf{X}_{st,\,s't'},\,E]\rangle = \gamma\,[\langle \mathbf{X}_{st,\,s't'}\rangle\,\mathbf{H}] \quad (14)$$

(γ is the gyromagnetic ratio for the nuclei under consideration). On substituting into (9) the vector spin functions in place of Q we obtain on the right hand side the expectation value also of vector spin functions. As a result, the following system of macroscopic equations is obtained:

$$d\mathbf{x}_i/dt = \gamma\,[\mathbf{x}_i\mathbf{H}] - \sum_{j=1}^{N}(\mathbf{x}_j - \mathbf{x}^0_j)/T_{ij},$$
$$i = 1, 2, \ldots, N, \quad (15)^*$$

where $\mathbf{x}_1 = \mathbf{M}$; $\mathbf{x}_j$ for $j \neq 1$ is the expectation value of a vector spin function ($\mathbf{x}^0_j$ is the equilibrium value of $\mathbf{x}_j$), such a function can only be a linear combination of quantities of the type

$$(\mathbf{I}_{s_1}\mathbf{I}_{t_1})(\mathbf{I}_{s_2}\mathbf{I}_{t_2})\ldots(\mathbf{I}_{s_n}\mathbf{I}_{t_n})\,\mathbf{I}_s;$$

N is the number of linearly independent equations in the system (15), which is determined not only by the number of particles, but also by the nature of their position with respect to each other; the quantities $1/T_{ij}$ consist of linear combinations of the relaxation coefficients $\Phi_{st,s't'}$.

In the case when the system (15) contains only one equation for $\mathbf{M}$ we obtain the usual Bloch equation with $T_1 = T_2$.

In solving the system (15) it is very convenient to utilize the matrix form of notation. Such a form of notation has been utilized, for example, in the papers by Jaynes[9] and Bloom[10] in investigating Bloch's phenomenological equation. If we introduce the matrices

$$X = \begin{pmatrix} \mathbf{x}_1 \\ \mathbf{x}_2 \\ \cdot \\ \cdot \\ \cdot \\ \mathbf{x}_N \end{pmatrix}, \quad \mathrm{B} = \begin{pmatrix} \beta & 0 & \ldots & 0 \\ 0 & \beta & \ldots & 0 \\ \cdot & \cdot & \cdot & \cdot \\ 0 & 0 & \ldots & \beta \end{pmatrix}, \quad \frac{1}{T} = \begin{pmatrix} \frac{1}{T_{11}} & \ldots & \frac{1}{T_{1N}} \\ \cdot & \cdot & \cdot \\ \frac{1}{T_{N1}} & \ldots & \frac{1}{T_{NN}} \end{pmatrix},$$

$$\mathbf{x}_j = \begin{pmatrix} x^+_j \\ x^-_j \\ x_{jz} \end{pmatrix}, \quad \beta = i\gamma \begin{pmatrix} H_z & 0 & -H^+ \\ 0 & -H_z & H^- \\ -\tfrac{1}{2}H^- & \tfrac{1}{2}H^+ & 0 \end{pmatrix},$$

$$\frac{1}{T_{ij}} = \frac{1}{T_{ij}}\begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}, \quad (16)$$

where, as usual, $x^{\pm}_j = x_{jx} \pm ix_{jy}$, $H^{\pm} = H_x \pm iH_y$, then the system (15) can be written in the form

$$\left(\frac{d}{dt} + \frac{1}{T} + \mathrm{B}\right)X = \frac{1}{T}X^0. \quad (17)$$

It may be easily seen that the matrices 1/T and B commute. This fact greatly simplifies the solution of (17). Indeed, in this case one can immediately obtain the general solution of (17) (similar to Jaynes' paper[9]) in the form

$$X(t) = u(t,0)\,X(0) + \int_0^t u(t,t')\,dt'\,\frac{1}{T}X^0,$$
$$u(t,t') = R(t,t')\exp\left\{-\frac{t-t'}{T}\right\},$$
$$R(t,t') = \exp\left\{-\int_{t'}^{t}\mathrm{B}(t'')\,dt''\right\}. \quad (18)$$

From this last expression it may be seen that the

*$[\mathbf{x}_i\mathbf{H}] = \mathbf{x}_i \times \mathbf{H}$

determination of the operator R(t,t′), associated with the external magnetic field, reduces to the similar problem for the Bloch equation. The evaluation of the operator exp $[-(t-t')/T]$ reduces to the purely algebraic problem of the diagonalization of the matrix 1/T.

As an illustration of the preceding discussion we consider the derivation of the macroscopic system of equations and its solution in the case of a molecule containing three protons which have the same gyromagnetic ratio and which are separated from each other by the same distance r (CH_3 – group). The correlation time in the medium is assumed to be small,* $\tau_c \omega \ll 1$. We shall start with Eq. (9) and expression (11) for S(Q). Since in our system the distance between any pair of particles is equal to r, while the angles between any pair of vectors joining any one particle to the other two are equal to $\vartheta = 60°$, then it follows from formula (10) that

$$\Phi_{st,\,st'} = \Phi\begin{cases} 1, & t = t' \\ -1/8, & t \neq t' \end{cases}, \quad \Phi = \frac{3\hbar^2\gamma^4\tau_c}{10r^6}. \qquad (19)$$

If in (19) we also take into account the fact that

$$\sum_{\tau}\sum_{st,\,s't'}[I^{\tau}_{st}, [Q, I^{-\tau}_{s't'}]] = \frac{1}{4}\sum_{\tau}[I^{\tau}[Q, I^{-\tau}]] = \frac{1}{4}X(Q),$$

where

$$I^0 = \sqrt{8/3}\left[I_z^2 - \frac{1}{4}(I^+I^- + I^-I^+)\right], \quad I^{\pm 1} = I^{\pm}I_z + I_zI^{\pm},$$

$$I^{\pm 2} = I^{\pm}I^{\pm}, \quad \mathbf{I} = \sum_s \mathbf{I}_s,$$

then we can obtain for S(Q) the following expression

$$S(Q) = \Phi\left\{\frac{9}{8}\sum_{st}\left[\langle X_{st,\,st}(Q)\rangle - \langle X_{st,\,st}(Q)\rangle_0\right] - \frac{1}{32}\left[\langle X(Q)\rangle - \langle X(Q)\rangle_0\right]\right\}.$$

Then by utilizing the commutation relations

$$[I_z, I^{\tau}_{st}] = \tau I^{\tau}_{st}, \quad [I_z, I^{\tau}] = \tau I^{\tau},$$

$$[I^{\tau}_{st}, I^{-\tau}_{st}] = \frac{\tau}{2}(I_{sz} + I_{tz}), \qquad \sum_{\tau=1}^{2}\tau[I^{\tau}, I^{-\tau}] = 8I^2I_z - 6I_z,$$

we evaluate $X_{st,st}(\gamma\mathbf{I})$ and $X(\gamma\mathbf{I})$. As a result of this we obtain for $S(\gamma\mathbf{I})$

$$S(\gamma\mathbf{I}) = -(\mathbf{M} - \mathbf{M}^0)/T_{11} + \mathbf{x}/T_{12}.$$

Here

$$\mathbf{M} = \gamma\langle\mathbf{I}\rangle, \quad \mathbf{x} = \gamma\langle\mathbf{s}\rangle, \quad \mathbf{s} = \left(\frac{1}{2}I^2 - \frac{13}{8}\right)\mathbf{I},$$

$$1/T_{11} = 10\Phi, \quad 1/T_{12} = \Phi.$$

We have chosen the spin functions **s** in such a way that its expectation value with respect to the equilibrium density matrix is equal to zero. Then, on substituting into equation (9) $\mathbf{Q} = \gamma\mathbf{s}$ and on evaluating the commutators, we finally obtain the following system of equations

$$\frac{d\mathbf{M}}{dt} = \gamma[\mathbf{M}\ \mathbf{H}] - \frac{\mathbf{M} - \mathbf{M}^0}{T_{11}} + \frac{\mathbf{x}}{T_{12}},$$

$$\frac{d\mathbf{x}}{dt} = \gamma[\mathbf{x}\ \mathbf{H}] + \frac{\mathbf{M} - \mathbf{M}^0}{T_{21}} - \frac{\mathbf{x}}{T_{22}}, \qquad (20)$$

where $1/T_{21} = 5\Phi/16$, $1/T_{22} = 17\Phi/4$.

If from these equations we eliminate **x** then the equation for the magnetic moment will have the form

$$\frac{d^2\mathbf{M}}{dt^2} = \gamma\frac{d}{dt}[\mathbf{M}\ \mathbf{H}] + \gamma\left[\frac{d\mathbf{M}}{dt}\mathbf{H}\right] - \gamma^2[[\mathbf{M}\ \mathbf{H}]\mathbf{H}] - \left(\frac{1}{T'} + \frac{1}{T''}\right)\frac{d\mathbf{M}}{dt} - \frac{\mathbf{M} - \mathbf{M}^0}{T'T''} + \gamma\left(\frac{1}{T'} + \frac{1}{T''}\right)[\mathbf{M}\ \mathbf{H}] - \frac{\gamma}{T_{11}}[\mathbf{M}^0\ \mathbf{H}],$$

where 1/T′ and 1/T″ are the eigenvalues of the matrix

$$\begin{pmatrix} 1/T_{11} & -1/T_{12} \\ -1/T_{21} & 1/T_{22} \end{pmatrix}.$$

By utilizing the matrix form of notation we can easily obtain the solution of the system (20) in the case of "slow passage" (the stationary solution when the external field is of the form $H_x = H_1\cos\omega t$, $H_y = -H_1\sin\omega t$, $H_z = H_0$) and of the spin-echo signal (external transverse field consists of a series of consecutive pulses at the resonance frequency). In both cases the solution may be written in the form

$$\mathbf{M} = \frac{T'T''}{(T' - T'')T_{11}}\left[\frac{T_{11} - T''}{T''}\mathbf{M}_{T'} - \frac{T_{11} - T'}{T'}\mathbf{M}_{T''}\right], \qquad (21)$$

where $\mathbf{M}_T$ is the solution of the phenomenological Bloch equation for the corresponding problem (cf., for example, references 9 and 10).

It may be seen from formula (21) that by determining experimentally the relative contribution to the relaxation of the times T′ and T″ we should obtain the same ratio given by $l = (T_{11} - T') \times T''/(T_{11} - T'')T'$, both in the slow passage experiments, and also in methods associated with the spin echo technique. In the example considered by us $l \sim 1/200$, and, therefore, it will hardly be possible to observe two relaxation times in experiments based on utilizing the solution (21).

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*The problem of the relaxation of the magnetic moment of such a system was considered in references 5 and 7.

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