A SYSTEM OF MAGNETIC MOMENTS IN A WEAK VARIABLE MAGNETIC FIELD

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A study is made of a system of magnetic moments subject to electric exchange and weak magnetic dipole-dipole interactions and situated in an external magnetic field $\mathbf{H}_0 + \mathbf{h}(t)$. Equations of motion for the magnetization vector are obtained up to terms of second order perturbation theory[5] and for weak variable fields $h(t) \ll H_0$. The limits of applicability of the equations obtained are discussed.

1. The microscopic theory of the behavior of nuclear magnetization in an external magnetic field has been developed in the paper by Wangsness and Bloch.[1] In that paper it was assumed that nuclear magnetic moments interact weakly with their molecular surroundings which are regarded as a heat reservoir.

In the cases when the nuclear spins are $I \leq 1$ the equation of motion in weak variable fields has the form:

$$d\mathbf{M}/dt + (\mathbf{i}M_x + \mathbf{j}M_y)/T_\perp + \mathbf{k}(M_z - M_0)/T_\parallel = \gamma[\mathbf{M}\times\mathbf{H}]. \quad (1)$$

For values of spin $I > 1$, Eqs. (1) are valid only in sufficiently weak constant fields H_0. In subsequent papers[2,3] Bloch took into account the direct and the indirect interactions of the nuclei and has investigated strong variable circularly polarized fields. The theory developed in references 1 — 3 in principle allows one to compute the relaxation times $T_\perp$ and $T_\parallel$ occurring in (1).

Bloch's equations have been widely used to describe the behavior not only of a system of nuclear magnetic moments but also of electronic magnetic moments in an external field. The phenomenon of ferromagnetic resonance in strong radio-frequency fields is also often discussed on the basis of the system of equations (1).[4]

Experimental data on the observation of relaxation and resonance phenomena in paramagnetic and ferromagnetic substances are analyzed on the basis of Bloch's equations; in such an analysis the relaxation times $T_\perp$ and $T_\parallel$ are obtained. However, the computation of these relaxation times is carried out on the basis of assumptions which are frequently different from those utilized in the derivation of equations (1). Therefore a comparison with experiment of the theoretically calculated values of $T_\perp$ and $T_\parallel$ based on the application of Bloch's equations is not consistent.

A more consistent method consists of finding the variation of magnetization with time (equation of motion) starting with a given form of the Hamiltonian, and of finding the coefficients appearing in these equations.

Kubo and Tomita[5] have developed a quite general and flexible method for determining the line shape of magnetic resonance absorption in radio-frequency fields. This method is more convenient than the method of Wangsness and Bloch for the description both of nuclear and of electronic magnetic resonance and relaxation. It was successfully applied to the study of line shape in nuclear and electronic resonance in a weak constant field ($\hbar\omega_0 \ll kT$).[6-8]

In this paper we obtain with the aid of the method of Kubo and Tomita the equation of motion for the magnetization vector of a system of magnetic moments coupled by electric exchange and weak magnetic dipole-dipole interactions. The coefficients occurring in these equations may be calculated in specific cases.

2. The expression for the components of the magnetization vector

$$\mathbf{M}(t) = \mathrm{Sp}\,\hat{\rho}(t)\,\hat{\mathbf{M}} \quad (2)$$

is determined by the density operator $\hat{\rho}(t)$ which obeys the equation of motion:

$$i\hbar\dot{\hat{\rho}}(t) = \hat{\mathscr{H}}(t)\,\hat{\rho}(t) - \hat{\rho}(t)\,\hat{\mathscr{H}}(t), \quad (3)$$

where

$$\hat{\mathscr{H}}(t) = \hat{\mathscr{H}} - \hat{\mathbf{M}}\cdot\mathbf{h}(t), \quad \hat{\mathbf{M}} = g\mu_0 \sum_j \hat{\mathbf{I}}_j, \quad (4)$$

and $\mathbf{h}(t)$ is a variable radio-frequency field which is assumed to be weak, $h(t) \ll H_0$.

Following Kubo and Tomita[5] we write the time-independent part of the Hamiltonian $\hat{\mathscr{H}}$ in the form

$$\mathcal{H} = \mathcal{H}_1 + \mathcal{H}_2 + \mathcal{H}', \tag{5}$$

in such a way that

$$[\mathcal{H}_2, \hat{\mathbf{M}}] = [\mathcal{H}_1, \mathcal{H}_2] = 0. \tag{6}$$

In order to do this we assume that $\mathcal{H}_2$ contains, in addition to terms which do not depend on the spins $\hat{\mathbf{I}}_j$, other terms that describe electric exchange interactions, while the operator

$$\mathcal{H}_1 = -\hbar\omega_0 \sum_j \hat{\mathbf{I}}_{jz}, \quad \hbar\omega_0 = g\mu_0 H_0 \tag{7}$$

determines the interaction of the magnetic moments with the external constant field $H_0 = H_2$.

The operator

$$\mathcal{H}' = g^2\mu_0^2 \sum_{j>k} r_{jk}^{-3} (\hat{\mathbf{I}}_j \hat{\mathbf{I}}_k - 3(\hat{\mathbf{I}}_j \mathbf{r}_{jk})(\hat{\mathbf{I}}_k \mathbf{r}_{jk}) / r_{jk}^2) \tag{8}$$

is regarded as a perturbation.

To solve the equation of motion (3) we set

$$\hat{\rho}(t) = \exp(-i\mathcal{H}t/\hbar)\, \hat{\rho}'(t) \exp(i\mathcal{H}t/\hbar). \tag{9}$$

Then in the new representation we shall have for $\hat{\rho}'(t)$

$$i\hbar\dot{\hat{\rho}}' = [\hat{\rho}'(t), \ \hat{\mathbf{M}}(t)]\, \mathbf{h}(t), \tag{10}$$

where

$$\hat{\mathbf{M}}(t) = \exp(i\mathcal{H}t/\hbar)\, \hat{\mathbf{M}} \exp(-i\mathcal{H}t/\hbar). \tag{11}$$

On integrating (10) by the method of successive approximations we obtain, up to terms linear in $\mathbf{h}(t)$,

$$\hat{\rho}'(t) = \hat{\rho}'(-\infty) + \frac{1}{i\hbar} \int_{-\infty}^{t} [\hat{\rho}'(-\infty), \hat{\mathbf{M}}(t')]\, \mathbf{h}(t')\, dt'. \tag{12}$$

Further, in the same approximation we obtain from (9)

$$\hat{\rho}(t) = \hat{\rho}_0 + \frac{1}{i\hbar} \int_{-\infty}^{t} [\hat{\rho}_0, \hat{\mathbf{M}}(t'-t)]\, \mathbf{h}(t')\, dt', \tag{13}$$

where

$$\hat{\rho}_0 = \exp\{(F - \mathcal{H})/kT\} \tag{14}$$

is the equilibrium density operator in the absence of the radio-frequency field $\mathbf{h}(t)$ which is switched on at $t = -\infty$.

We introduce the following notation:

$$\hat{M}_{\pm 1} = \mp(\hat{M}_x \pm i\hat{M}_y)/\sqrt{2}, \quad \hat{M}_0 = \hat{M}_z,$$
$$h_{\pm 1} = \mp(h_x \pm ih_y)/\sqrt{2}, \qquad h_0 = h_z, \tag{15}$$

then

$$\hat{\mathbf{M}}\cdot\mathbf{h}(t) = \sum_\alpha (-1)^\alpha M_\alpha h_{-\alpha} = \sum_\alpha M_\alpha h_\alpha^*,$$

where $\alpha = 0, \pm 1$.

Now by using (13) expression (2) can be written in terms of its components

$$M_\alpha(t) = M_{0\alpha} + \sum_\beta \int_{-\infty}^{t} \frac{i}{\hbar} \operatorname{Sp} \hat{\rho}_0 [M_\alpha(t-t'), M_\beta]\, h_\beta^*(t')\, dt', \tag{16}$$

where

$$M_{0\alpha} = \operatorname{Sp} \hat{\rho}_0 \hat{M}_\alpha \delta_{\alpha 0},$$

and where we have taken into account the fact that

$$\operatorname{Sp}[\hat{\rho}_0, \hat{M}_\beta(t'-t)]\, \hat{M}_\alpha = -\operatorname{Sp} \hat{\rho}_0 [\hat{M}_\alpha(t-t'), \ \hat{M}_\beta]. \tag{17}$$

We introduce the tensor relaxation function

$$G_{\alpha\beta}(\tau) = \operatorname{Sp} \hat{\rho}_0 \int_0^{1/kT} \frac{1}{2}\{M_\alpha(\tau - i\hbar\sigma) M_\beta$$
$$+ M_\beta(-\tau - i\hbar\sigma) M_\alpha\}\, d\sigma - \frac{1}{kT} M_{0\alpha} M_{0\beta}, \tag{18}$$

which, as can be easily seen, satisfies the following relations

$$G_{\alpha\beta}(\tau) = G_{\beta\alpha}(-\tau), \tag{19}$$

$$-dG_{\alpha\beta}(\tau)/d\tau = (i/\hbar) \operatorname{Sp} \hat{\rho}_0 [\hat{M}_\alpha(\tau), \hat{M}_\beta]. \tag{20}$$

By noting that at $\tau = t - t'$ the right-hand side of the expression coincides with the integrand in (16), we obtain

$$M_\alpha(t) - M_{0\alpha} = -\sum_\beta \int_0^\infty \frac{d}{d\tau} G_{\alpha\beta}(\tau) \cdot h_\beta^*(t-\tau)\, d\tau. \tag{21}$$

Thus, to calculate the components of the magnetization $M_\alpha(t)$ it is enough to find the components of the tensor $G_{\alpha\beta}(\tau)$.

3. To calculate the components of the tensor $G_{\alpha\beta}(\tau)$ we shall write the expression for the operator $\hat{M}_\alpha(t)$ occurring in it in the form of the following expansion:

$$\hat{M}_\alpha(t) = \hat{M}_\alpha^0(t) + (1/i\hbar) \int_0^t [\hat{M}_\alpha^0(t), \ \mathcal{H}'(t_1)]\, dt_1$$
$$+ (i\hbar)^{-2} \int_0^t dt_1 \int_0^{t_1} dt_2 [[\hat{M}_\alpha^0(t), \mathcal{H}'(t_1)], \mathcal{H}'(t_2)], + \ldots, \tag{22}$$

where

$$\hat{M}_\alpha^0(t) = \exp(i\mathcal{H}_0 t/\hbar)\, \hat{M}_\alpha \exp(-i\mathcal{H}_0 t/\hbar), \tag{23}$$

$$\mathcal{H}'(t) = \exp(i\mathcal{H}_0 t/\hbar)\, \mathcal{H}' \exp(-i\mathcal{H}_0 t/\hbar). \tag{24}$$

Then

$$G_{\alpha\beta}(\tau) = G_{\alpha\beta}^{(0)}(\tau) + G_{\alpha\beta}^{(1)}(\tau) + G_{\alpha\beta}^{(2)}(\tau) + \ldots. \tag{25}$$

In the following we shall restrict ourselves to the approximation of second order in $\mathcal{H}'$.

We obtain for $G_{\alpha\beta}^{(0)}$:

$$G_{\alpha\beta}^{(0)} = \int_0^{1/kT} \operatorname{Sp} \hat{\rho}_0 \frac{1}{2} \{\hat{M}_\alpha^0(\tau - i\hbar\sigma)\, \hat{M}_\beta$$
$$+ \hat{M}_\beta^0(-\tau - i\hbar\sigma)\, \hat{M}_\alpha\}\, d\sigma - \frac{1}{kT} M_{0\alpha} M_{0\beta}. \tag{26}$$

By utilizing (6), (7), (23), we obtain:

$$\hat{M}^0_\alpha(\tau - i\hbar\sigma) = \exp(-i\alpha\omega_0\tau - \alpha\hbar\omega_0\sigma)\hat{M}_\alpha. \quad (27)$$

Now by noting that $\mathrm{Sp}\,\hat{\rho}_0\hat{M}_\alpha\hat{M}_\beta = 0$ for $\alpha \neq -\beta$, we obtain

$$G^{(0)}_{\alpha\beta} = (-1)^\alpha \chi_\alpha \exp(-i\alpha\omega_0\tau)\,\delta_{\alpha,-\beta} - \frac{1}{kT} M^2_{0z}\delta_{\alpha 0}\delta_{\beta 0}, \quad (28)$$

where

$$\chi_\alpha = \mathrm{Sp}\,\hat{\rho}_0 |M_\alpha|^2 \sinh(\alpha\hbar\omega_0/kT)/\alpha\hbar\omega_0. \quad (29)$$

In the case of weak fields $\hbar\omega_0 \ll kT$ as a result of the isotropic nature of the operator (14) in spin space we have

$$\chi_{+1} = \chi_{-1} \equiv \chi_\perp = \chi_0 \equiv \chi_\parallel. \quad (30)$$

In strong fields, generally speaking, $\chi_\perp \neq \chi_\parallel$.

In the case of isotropic surroundings $G^{(1)}_{\alpha\beta} = 0$. This follows from the fact that $G^{(1)}_{\alpha\beta}$ contains only the first power of $\mathcal{H}'$.

For the computation of $G^{(2)}_{\alpha\beta}$ it is convenient to write the operator $\hat{\mathcal{H}}'$ in the form

$$\hat{\mathcal{H}}' = \sum_\lambda \sum_{j>k} \Phi^{-\lambda}_{jk}\{jk\}_\lambda, \quad (31)$$

where

$$\begin{aligned}
\{jk\}_{\pm 2} &= 2\hat{I}_{j\pm 1}\hat{I}_{k\pm 1},\\
\{jk\}_{\pm 1} &= \mp\sqrt{2}(\hat{I}_{j\pm 1}\hat{I}_{k0} + \hat{I}_{j0}\hat{I}_{k\pm 1}), \qquad (32)\\
\{jk\}_0 &= \hat{I}_{j0}\hat{I}_{k0} + {}^1/_2(\hat{I}_{j+1}\hat{I}_{k-1} + \hat{I}_{j-1}\hat{I}_{k+1});
\end{aligned}$$

$$\begin{aligned}
\Phi^{\pm 2}_{jk} &= -\sqrt{6\pi/5}\,g^2\mu_0^2 r_{jk}^{-3} Y_{2,\pm 2}(\vartheta_{jk},\varphi_{jk}),\\
\Phi^{\pm 1}_{jk} &= -\sqrt{6\pi/5}\,g^2\mu_0^2 r_{jk}^{-3} Y_{2,\pm 1}(\vartheta_{jk},\varphi_{jk}), \qquad (33)\\
\Phi^{0}_{jk} &= -\sqrt{16\pi/5}\,g^2\mu_0^2 r_{jk}^{-3} Y_{2,0}(\vartheta_{jk},\varphi_{jk}),
\end{aligned}$$

Y_{lm} is the normalized spherical harmonic, and

$$\hat{I}_{\pm 1} = \mp(\hat{I}_x \pm i\hat{I}_y)/\sqrt{2},\quad \hat{I}_0 = \hat{I}_z. \quad (34)$$

By utilizing (6), (7) and (24) we obtain:

$$\hat{\mathcal{H}}'(t - i\hbar\sigma) = \sum_\lambda \exp(-i\lambda\omega_0 - \lambda\hbar\omega_0\sigma)\hat{\mathcal{H}}'_\lambda(t - i\hbar\sigma), \quad (35)$$

where

$$\hat{\mathcal{H}}'_\lambda(t - i\hbar\sigma) = \exp(i\hat{\mathcal{H}}_2 t/\hbar + \hat{\mathcal{H}}_2\sigma)\hat{\mathcal{H}}'_\lambda \exp(-i\hat{\mathcal{H}}_2 t/\hbar - \hat{\mathcal{H}}_2\sigma). \quad (36)$$

Now

$$\begin{aligned}
G^{(2)}_{\alpha\beta}(\tau) = &-\frac{1}{2\hbar^2}\int_0^{1/kT} d\sigma \exp(-i\alpha\omega_0\tau)\sum_{\lambda\lambda'}\int_0^\tau dt_1\int_0^{t_1} dt_2\\
&\times \exp(-i\lambda\omega_0 t_1 - i\lambda'\omega_0 t_2)\,\mathrm{Sp}\,\hat{\rho}_0\{\exp(-(\alpha+\lambda+\lambda')\hbar\omega_0\sigma)\\
&\times([[\hat{M}_\alpha, \hat{\mathcal{H}}'_\lambda(t_1 - i\hbar\sigma)], \hat{\mathcal{H}}'_{\lambda'}(t_2 - i\hbar\sigma)]\hat{M}_\beta)\\
&+\exp((\alpha+\lambda+\lambda')\hbar\omega_0\sigma) \qquad (37)\\
&\times(\hat{M}_\beta[\hat{\mathcal{H}}'_{\lambda'}(t_2 + i\hbar\sigma), [\hat{\mathcal{H}}'_\lambda(t_1 + i\hbar\sigma), \hat{M}_\alpha]])\}.
\end{aligned}$$

The magnitude of the trace does not depend on the choice of the origin of time, since the trace is invariant with respect to a unitary transformation of an operator which follows the trace sign:

$$\exp(i\hat{\mathcal{H}}_0\Delta T/\hbar)\ldots\exp(-i\hat{\mathcal{H}}_0\Delta T/\hbar).$$

Therefore

$$\begin{aligned}
&\mathrm{Sp}\,\hat{\rho}_0([[\hat{M}_\alpha, \hat{\mathcal{H}}'_\lambda(t_1 - i\hbar\sigma)], \hat{\mathcal{H}}'_{\lambda'}(t_2 - i\hbar\sigma)]\hat{M}_\beta)\\
&= \exp(-i(\alpha+\lambda+\lambda'+\beta)\omega_0\Delta T)\\
&\times \mathrm{Sp}\,\hat{\rho}_0([[\hat{M}_\alpha, \hat{\mathcal{H}}'_\lambda(t_1 - i\hbar\sigma)], \hat{\mathcal{H}}'_{\lambda'}(t_2 - i\hbar\sigma)]\hat{M}_\beta),
\end{aligned}$$

from which we obtain

$$\alpha + \lambda + \lambda' + \beta = 0. \quad (38)$$

Since expression (31) for $\hat{\mathcal{H}}'$ contains spherical harmonics, only the terms with $\lambda = -\lambda'$ differ from zero in averaging at $t_1 = t_2$ in the case of isotropic surroundings. For $t_1 \neq t_2$ the terms with $\lambda = -\lambda'$ give the principal contribution to (37). Therefore, in accordance with (38) we may set $\alpha = -\beta$.

We then have:

$$\begin{aligned}
G^{(2)}_{\alpha\beta}(\tau) = &-\frac{1}{2\hbar^2}\int_0^{1/kT} d\sigma \exp(-i\alpha\omega_0\tau)\sum_\lambda\int_0^\tau d\vartheta\,(\tau - \vartheta)\exp(i\lambda\omega_0\vartheta)\\
&\times \mathrm{Sp}\,\hat{\rho}_0\{\exp(-\alpha\hbar\omega_0\sigma)[[\hat{M}_\alpha, \hat{\mathcal{H}}'_\lambda(-\vartheta - i\hbar\sigma)], \hat{\mathcal{H}}'_{-\lambda}(-i\hbar\sigma)]\hat{M}_{-\alpha}\\
&+\exp(\alpha\hbar\omega_0\sigma)\hat{M}_{-\alpha}[\hat{\mathcal{H}}'_{-\lambda}(\vartheta + i\hbar\sigma), [\hat{\mathcal{H}}'_\lambda(i\hbar\sigma), \hat{M}_\alpha]]\}\,\delta_{\alpha,-\beta},
\end{aligned} \quad (39)$$

where $\vartheta = t_2 - t_1$.

We introduce the notation

$$\begin{aligned}
&\frac{1}{2\hbar^2}\int_0^{1/kT} d\sigma\,\mathrm{Sp}\,\hat{\rho}_0\{\exp(-\alpha\hbar\omega_0\sigma)\\
&\times[[\hat{M}_\alpha, \hat{\mathcal{H}}'_\lambda(-\vartheta - i\hbar\sigma)], \hat{\mathcal{H}}'^{*}_\lambda(i\hbar\sigma)]\hat{M}^*_\alpha + \exp(\alpha\hbar\omega_0\sigma)\hat{M}^*_\alpha\\
&\times[\hat{\mathcal{H}}'^{*}_\lambda(\vartheta - i\hbar\sigma), [\hat{\mathcal{H}}'_\lambda(i\hbar\sigma), \hat{M}_\alpha]]\} = \chi_\alpha\Omega^2_{\alpha\lambda} f_{\alpha\lambda}(\vartheta)
\end{aligned} \quad (40)$$

under the condition $f_{\alpha\lambda}(0) = 1$. Then

$$G^{(2)}_{\alpha\beta}(\tau) = -(-1)^\alpha\chi_\alpha\exp(-i\alpha\omega_0\tau)\,\psi_\alpha(\tau)\,\delta_{\alpha,-\beta}, \quad (41)$$

$$\psi_\alpha(\tau) = \sum_\lambda \Omega^2_{\alpha\lambda}\int_0^\tau(\tau - \vartheta)\exp(i\lambda\omega_0\vartheta) f_{\alpha\lambda}(\vartheta)\,d\vartheta. \quad (42)$$

The tensor $G_{\alpha\beta}(\tau)$ can now be written in the form

$$G_{\alpha\beta}(\tau) = (-1)^\alpha\chi_\alpha\exp(-i\alpha\omega_0\tau)(1 - \psi_\alpha)\,\delta_{\alpha,-\beta} - \frac{1}{kT}M^2_{0z}\delta_{\alpha 0}\delta_{\beta 0}. \quad (43)$$

As can be seen from (40) and (42), the function $\psi_\alpha(\tau)$ has the following properties:

$$\psi_\alpha(\tau) = \psi^*_\alpha(-\tau) = \psi^*_{-\alpha}(\tau). \quad (44)$$

4. If (43) is taken into account, Eq. (21) found above for the magnetization $\hat{M}_\alpha$ determines its variation with time in weak variable fields. The function $\psi_\alpha(\tau)$ can be obtained if the specific form of the correlation function $f_{\alpha\lambda}(t)$ is known. The function $f_{\alpha\lambda}(t)$ falls off rapidly with time. In the case that

$$\Omega_{\alpha\lambda}\tau_c \ll 1, \quad (45)$$

where τ_c is the time interval that characterizes the function $f_{\alpha\lambda}(t)$, then we can use for $\psi_\alpha(\tau)$ the following asymptotic expression

$$\psi_\alpha(\tau) = \tau\sum_\lambda \Omega^2_{\alpha\lambda}\int_0^\infty \exp(i\lambda\omega_0\vartheta) f_{\alpha\lambda}(\vartheta)\,d\vartheta, \quad \tau > 0. \quad (46)$$

The term neglected in the integral (42) is of order $(\Omega_{\alpha\lambda}\tau_c)^2$.

We can now write $\psi_\alpha(\tau)$ in the form

$$\psi_\alpha(\tau) = (1/T_\alpha)|\tau| + i\alpha\Delta\omega_0\tau, \quad (47)$$

where

$$\frac{1}{T_\alpha} = \sum_\lambda \Omega^2_{\alpha\lambda}\,\mathrm{Re}\int_0^\infty \exp(i\lambda\omega_0\vartheta) f_{\alpha\lambda}(\vartheta)\,d\vartheta, \quad (48)$$

$$\alpha\Delta\omega_0 = \sum_\lambda \Omega^2_{\alpha\lambda}\,\mathrm{Im}\int_0^\infty \exp(i\lambda\omega_0\vartheta) f_{\alpha\lambda}(\vartheta)\,d\vartheta. \quad (49)$$

Taking into account the fact that $\psi_\alpha(\tau)$ is small and the limiting condition $G(\infty) = 0$ we obtain in accordance with (21) and (43), up to terms of second-order perturbation theory:

$$M_\alpha = M_{0\alpha} + \chi_\alpha\int_0^\infty (i\alpha\omega_0 + \dot\psi_\alpha(\tau)) \times \exp(-i\alpha\omega_0\tau - \psi_\alpha(\tau))\,h_\alpha(t-\tau)\,d\tau. \quad (50)$$

The last expression completely determines the dependence of M_α on the time for a given $h(t)$.

It is now easy to obtain the differential equation that is satisfied by the components of the magnetization vector. On differentiating M_α with respect to time we obtain after simple transformations

$$\dot M_\alpha + [i\alpha(\omega_0 + \Delta\omega_0) + 1/T_\alpha](M_\alpha - M_{\alpha 0}) = -[i\alpha(\omega_0 + \Delta\omega_0) + 1/T_\alpha]\chi_\alpha h_\alpha(t). \quad (51)$$

By going over to the Cartesian coordinate system we obtain:

$$\begin{aligned} \dot M_x + (1/T_\perp)M_x &= (1/T_\perp)\chi_\perp h_x(t) + \gamma M_y H_0 - \gamma M_0 h_y(t) - \gamma(\chi_\perp H_0 - M_0)h_y(t), \\ \dot M_y + (1/T_\perp)M_y &= (1/T_\perp)\chi_\perp h_y(t) + \gamma M_0 h_x(t) - \gamma M_x H_0 + \gamma(\chi_\perp H_0 - M_0)h_x(t), \\ \dot M_z + (1/T_\parallel)(M_z - M_0) &= (1/T_\parallel)\chi_\parallel h_z(t), \end{aligned} \quad (52)$$

where

$$\gamma H_0 = \omega_0 + \Delta\omega_0, \quad (53)$$

and $T_\perp = T_{\pm 1}$ and $T_\parallel = T_0$ are the transverse and longitudinal relaxation times.

Equations (52) differ from the linearized Bloch equations (1) by their last terms, and also by the terms containing $\chi_\perp$ and $\chi_\parallel$.

In weak fields, $\hbar\omega_0 \ll kT$, as we have pointed out earlier, we have $\chi_\perp = \chi_\parallel = \chi$ independent of the field H_0. In this case $M_0 = \chi H_0$.

The quantity γ appearing in the equation differs from the gyromagnetic ratio for the free magnetic moment $\gamma_0 = g\mu_0/\hbar$ by the correction term (53)

$$\gamma = \gamma_0 + \Delta\omega_0/H_0, \quad (54)$$

where $\Delta\omega_0$ is determined by (49).

In the case $\omega_0\tau_c \ll 1$ we obtain from (49)

$$\Delta\omega_0/H_0 = \sum_\lambda \Omega^2_{1\lambda}\int_0^\infty \lambda\gamma_0\vartheta f_{1\lambda}(\vartheta)\,d\vartheta. \quad (55)$$

In the case under consideration this correction to γ is due certainly not to the spin-orbit interactions, which were not taken into account, but to dipole-dipole interactions. The magnitude of the correction depends in an essential manner on the nature of the thermal motion of the magnetic moments and on the exchange interaction. When $\omega_0\tau_c \gg 1$ the correction to the gyromagnetic ratio can be neglected.

The condition for the applicability of (52) is the inequality $\Omega_\alpha\tau_c \ll 1$.

In paramagnetic solutions τ_c is the correlation time which is determined by

$$\tau_c = (4\pi/3)a^3\eta/kT, \quad (56)$$

where a is the kinetic radius of the molecule. If we take into account that $\Omega_{\alpha\lambda} \sim \mu_0^2/d^3\hbar$, where d is the average distance between the magnetic moments, then

$$\Omega_{\alpha\lambda}\tau_c \sim (4\pi\mu_0^2\eta/3\hbar kT)(a/d)^3. \quad (57)$$

For aqueous solutions of paramagnetic salts at room temperatures $\Omega_{\alpha\lambda}\tau_c \sim (a/d)^3 < 1$ and rapidly decreases upon dilution.

For ferromagnetic substances at temperatures below the Curie temperature we always have $\Omega_{\alpha\lambda}\tau_c \ll 1$.

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