

# Generalized Bloch equations and dissipation of longitudinal magnetic-field energy

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For a spin- $\frac{1}{2}$  coupled to a Gaussian heat bath in the presence of an external magnetic field, we generalize the Bloch equations to the non-Markovian case. Using this generalization as the base, we show that the time-varying component of the magnetic field parallel to its constant part is absorbed by the spin system, especially at high temperatures of the heat bath. For the spectral density and dispersion of longitudinal spin fluctuations, we obtain expressions that are in good agreement with known results. [S1063-651X(97)04308-0]

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## I. INTRODUCTION

Dissipative dynamics of a spin- $\frac{1}{2}$  in the presence of an external magnetic field  $\vec{B}(t)$  is well known to be described by the Bloch equations [1–6] for the averaged Pauli matrices  $\vec{\sigma} = \{\sigma_x(t), \sigma_y(t), \sigma_z(t)\}$ :

$$\langle \dot{\sigma}_x(t) \rangle + \frac{1}{T_2} \langle \sigma_x(t) \rangle + \Delta \langle \sigma_y(t) \rangle = 2f_z(t) \langle \sigma_y(t) \rangle - 2f_y(t) \langle \sigma_z(t) \rangle, \quad (1)$$

$$\langle \dot{\sigma}_y(t) \rangle + \frac{1}{T_2} \langle \sigma_y(t) \rangle - \Delta \langle \sigma_x(t) \rangle = 2f_x(t) \langle \sigma_z(t) \rangle - 2f_z(t) \langle \sigma_x(t) \rangle,$$

$$\langle \dot{\sigma}_z(t) \rangle + \frac{\langle \sigma_z(t) \rangle - \sigma_z^0}{T_1} = 2f_y(t) \langle \sigma_x(t) \rangle - 2f_x(t) \langle \sigma_y(t) \rangle. \quad (2)$$

Here a magnetic field  $\vec{B}(t)$  is supposed to incorporate a  $z$ -directed constant part  $\vec{B}_0 = (0, 0, B_0)$  and a time-varying magnetic field  $\vec{B}_1(t)$ , so that  $\Delta = -2g\mu_0 B_0$ ,  $\vec{f}(t) = g\mu_0 \vec{B}_1(t)$ ,  $\mu_0 = e\hbar/2mc$  is the Bohr magneton, and  $g$  signifies the  $g$  factor of the spin particle with mass  $m$  ( $g=1$  for an electron). The longitudinal and transversal relaxation time are therewith denoted by  $T_1$  and  $T_2$ , respectively, while  $\sigma_z^0 = -\tanh(\Delta/2T)$  is the equilibrium  $z$  projection of the averaged spin matrix,  $T$  is a heat bath temperatures,  $\hbar=1$ , and  $k_B=1$ .

Among other things the Bloch equations (1) and (2) allow one to calculate the energy of an oscillating magnetic field  $\vec{B}_1(t)$  which is absorbed by the spin system in a unit of time and in a unit of volume [7]:

$$P(\omega) = \frac{\omega}{2\pi} \int_0^{2\pi/\omega} \left\langle \vec{B}_1(t) \cdot \frac{d\vec{\mu}(t)}{dt} \right\rangle dt, \quad (3)$$

where  $\omega$  is a frequency of magnetic-field oscillations,  $\vec{\mu}(t) = g\mu_0 \vec{\sigma}(t)$  is the magnetic moment of the spin particle,

and the brackets  $\langle \dots \rangle$  denote an ensemble average over the initial state of a heat bath in combination with the trace over spins.

If the magnetic field  $\vec{B}_1(t)$  rotates in a plane  $(x, y)$  which is normal to the direction of the constant magnetic field  $\vec{B}_0$ :  $\vec{B}_1(t) = (B_1 \cos(\omega t), B_1 \sin(\omega t), 0)$ , so the shape of the absorption line  $P(\omega)$ , Eq. (3), is determined by the imaginary parts of the transversal susceptibilities  $\Phi_{xx}(\omega)$  and  $\Phi_{yy}(\omega)$  [3,4]:

$$P_x(\omega) = \frac{1}{2} (g\mu_0)^2 B_1^2 \text{Im}\{\Phi_{xx}(\omega) + \Phi_{yy}(\omega)\}, \quad (4)$$

where the linear magnetic susceptibility  $\Phi_{ij}(\omega)$  ( $i, j = x, y, z$ ) is a Fourier transform of the averaged response function

$$\frac{\delta \langle \sigma_i(t) \rangle}{\delta f_j(t')} = (g\mu_0)^{-2} \frac{\delta \langle \mu_i(t) \rangle}{\delta (B_1)_j(t')} = \int \frac{d\omega}{2\pi} e^{-i\omega(t-t')} \Phi_{ij}(\omega) \quad (5)$$

taken at  $f_j=0$ . It follows from the Bloch equations (1) and (2) that absorption of the transversal magnetic field  $P_x(\omega)$  is described by the expression [3–6]

$$P_x(\omega) = 4(g\mu_0)^2 B_1^2 \tanh\left(\frac{\Delta}{2T}\right) \frac{\gamma_x \Delta \omega^2}{(\omega^2 - \Delta^2)^2 + 4\gamma_x^2 \omega^2} \quad (6)$$

if coupling of the spin to a heat bath is weak, so that  $\Delta \gg \gamma_x, \gamma_y, \gamma_z$ , where  $\gamma_x = \gamma_y = 1/T_2$  and  $\gamma_z = 1/T_1$ .

At the same time the most-used Bloch equations (1) and (2) predict a zero result for the longitudinal magnetic susceptibility  $\Phi_{zz}(\omega)$ :  $[\Phi_{zz}(\omega)]_{\text{Bloch}} = 0$ , and with it for the corresponding absorption spectrum

$$P_z(\omega) = \frac{1}{2} (g\mu_0)^2 B_1^2 \omega \text{Im}\Phi_{zz}(\omega); \quad (7)$$

that is, the energy of the time-varying magnetic field  $\vec{B}_1(t) = (0, 0, B_1 \cos(\omega t))$  is not bound to be absorbed by the spin system interacting with a heat bath:  $[P_z(\omega)]_{\text{Bloch}} = 0$ . It immediately follows from the fluctuation-dissipation theorem [8]

$$K_{zz}(\omega) = \text{Im}\Phi_{zz}(\omega) \coth(\omega/2T) \quad (8)$$

that longitudinal fluctuations of spin variables covered by the cumulant function

$$\langle \frac{1}{2} [\sigma_z(t), \sigma_z(t')]_+ \rangle = \int \frac{d\omega}{2\pi} e^{-i\omega(t-t')} K_{zz}(\omega) \quad (9)$$

are also nonexistent. But, in view of the fact that  $\sigma_z^2(t) = 1, (\sigma_z^0)^2 = \tanh^2(\Delta/2T)$ , the variance of  $\sigma_z$  fluctuations

$$\langle (\Delta\sigma_z)^2 \rangle = \langle \frac{1}{2} [\sigma_z(t), \sigma_z(t')]_+ \rangle = \int \frac{d\omega}{2\pi} K_{zz}(\omega), \quad (10)$$

can be significantly different from zero:

$$\langle (\Delta\sigma_z)^2 \rangle = \langle (\sigma_z)^2 \rangle - (\sigma_z^0)^2 = 1 - \tanh^2(\Delta/2T), \quad (11)$$

especially at high temperatures  $T \gg \Delta$ . Thus the conventional Bloch equations (1) and (2) lead to zero results for absorption of the longitudinal magnetic-field energy, and with it for the longitudinal fluctuations of spin variables, that cannot be accepted as reasonable in view of spin coupling to a heat bath.

The aim of the present paper is to obtain more correct expressions for the longitudinal absorption and fluctuation spectra of the spin system, and so to remedy the above-mentioned problem of the Bloch theory. The way to do this is through a non-Markovian generalization of the Bloch equations, taking into account memory effects in the interaction of the spin with the heat bath. A great deal of effort went into the derivation of non-Markovian kinetic equations [9–14] and into the investigation of non-Markovian effects [11–16]. The problem in question is also of special interest for understanding dissipative quantum tunneling in solids [17–22].

In this work we show that the generalized Bloch equations are a prime necessity in the treatment of the linear response function  $\langle \delta\sigma_z(t)/\delta f_z(t') \rangle_{f=0}$  as well as in the study of nonlinear memory phenomena associated with an influence of strong internal driving on the relaxation and decoherence processes [20,23,24]. The non-Markovian stochastic equations for a two-level system interacting with a Gaussian heat bath [14,15,25] have considerable utility for this purpose. In Sec. II we derive the non-Markovian equations for averaged spin variables, taking into account an influence of  $z$ -directed time-dependent magnetic field on spin relaxation. In Sec. III we calculate the linear response of the spin to the action of the longitudinal magnetic field, and find expressions for the absorption and fluctuation spectra. It should be noted that the generalized Bloch equations deduced in Sec. II are exact for a spin coupled to a Gaussian heat bath. However, we shall use a weak-damping limit for the calculation of the absorption and fluctuation spectra. Because of this, we shall restrict our consideration to the case of a super-Ohmic bath [17] with the spectral density  $J(\omega) \sim \omega^n, n > 1$ , or to the case of an Ohmic heat bath ( $n = 1$ ) provided that the system-environment coupling constant is small and the temperature  $T$  of the heat bath is modest. We also suppose that the temperature  $T$  is not too low:  $T \gg \hbar\gamma$ , with  $\gamma$  being a spin relaxation rate. It might be well to point out that a three-dimensional acoustic-phonon heat bath responsible for relaxation and fluctuations in solids is super-Ohmic with  $n = 3$  or  $5$  [17], and the same is true for a photon heat bath with  $n = 3$  [25,26].

## II. GENERALIZED BLOCH EQUATIONS

Starting with the microscopic Hamiltonian

$$H = \frac{\Delta}{2} \sigma_z - \vec{\sigma} \cdot \vec{Q}(t) - \sigma_z f_z(t) + H_B \quad (12)$$

for the spin subsystem coupled to a heat bath with variables  $\{Q_i(t)\} (i = x, y, z)$  and with the free Hamiltonian  $H_B$  in the presence of an external magnetic field  $\vec{B}(t) = \{0, 0, B_0 + B_1(t)\}$ , we obtain the Heisenberg equations

$$\dot{b}(t) = 2i\{Q_z(t)b(t) - Q_-(t)\sigma_z(t)\}, \quad (13)$$

$$\dot{\sigma}_z(t) = i\{Q_-(t)b^+(t) - Q_+(t)b(t)\} \quad (14)$$

for the spin operators

$$b(t) = \{\sigma_x(t) - i\sigma_y(t)\} \exp\left[i \int_0^t \Delta(\tau) d\tau\right], \quad (15)$$

$b^+(t)$ , and  $\sigma_z(t)$ .

Our main interest here is with absorption of the longitudinal magnetic field  $(\vec{B}_1)_z(t) = (g\mu_0)^{-1}f_z(t)$  only; therefore, the transversal projections of the time-varying magnetic field are taken as being zero:  $f_x(t) = f_y(t) = 0$ , while the longitudinal force  $f_z(t)$  is incorporated in the time-dependent splitting  $\Delta(t) = \Delta - 2f_z(t)$ . Here the functions  $Q_{\pm}$  are the new heat bath variables

$$Q_-(t) = \{Q_x(t) - iQ_y(t)\} \exp\left[i \int_0^t \Delta(\tau) d\tau\right] = [Q_+(t)]^+, \quad (16)$$

and the superscript  $(+)$  designates the Hermitian conjugation.

It was shown in Ref. [14] that the response of a Gaussian heat bath to the action of the dynamic subsystem is linear in spin variables  $\sigma_j(t) (j = x, y, z)$ ,

$$Q_i(t) = Q_i^{(0)}(t) + \int dt_1 \varphi_{ij}(t, t_1) \sigma_j(t_1), \quad (17)$$

where  $\{Q_i^{(0)}(t)\}$  are the unperturbed heat bath variables which are assumed to be Gaussian with zero mean values  $\langle Q_i^{(0)}(t) \rangle = 0$ , with a covariance

$$\begin{aligned} M_{ij}(t, t') &= \langle \frac{1}{2} [Q_i^{(0)}(t), Q_j^{(0)}(t')]_+ \rangle \\ &= \int \frac{d\omega}{2\pi} e^{-i\omega(t-t')} S_{ij}(\omega), \end{aligned} \quad (18)$$

and a response function

$$\begin{aligned} \varphi_{ij}(t, t') &= \langle i [Q_i^{(0)}(t), Q_j^{(0)}(t')]_- \rangle \theta(t - t') \\ &= \int \frac{d\omega}{2\pi} e^{-i\omega(t-t')} \chi_{ij}(\omega). \end{aligned} \quad (19)$$

Here  $\theta(t)$  is the unit Heaviside step function, and  $\chi_{ij}(\omega)$  and  $S_{ij}(\omega)$  are the susceptibility and the spectral density, respectively, of heat bath fluctuations. It is worth noting that the

independent-oscillator model of the heat bath [17,27] which is most extensively employed in dissipative quantum mechanics is also described by Gaussian statistics. In this case the imaginary part  $\text{Im}\chi(\omega)$  of the heat bath susceptibility is proportional to the spectral function  $J(\omega)$  used in works devoted to dissipative quantum tunneling [17,19,20]. For the isotropic heat bath we have  $\varphi_{ij}(t, t') = \delta_{ij}\varphi(t, t')$  and  $M_{ij}(t, t') = \delta_{ij}M(t, t')$ , so that the variables  $Q_{\pm}(t)$  and  $Q_z(t)$  may be written as

$$Q_z(t) = Q_z^{(0)}(t) + \int dt_1 \varphi(t, t_1) \sigma_z(t_1),$$

$$Q_{-}(t) = [Q_{+}(t)]^{+} = Q_{-}^{(0)}(t) + \int dt_1 \varphi_{-}(t, t_1) b(t_1), \quad (20)$$

with

$$\varphi_{-}(t, t_1) = [\varphi_{+}(t, t_1)]^{+} = \varphi(t, t_1) \exp\left[i \int_{t_1}^t d\tau \Delta(\tau)\right]. \quad (21)$$

The new spin and heat bath variables  $b(t)$ ,  $b^{+}(t)$ , and  $Q_{\pm}(t)$  allow us to take into consideration an effect of the arbitrary longitudinal magnetic field  $(\vec{B}_1)_z(t)$  on the relaxation process.

Substituting Eqs. (20) in the Heisenberg equations (13) and (14) followed by averaging over the initial state of the heat bath gives the generalized non-Markovian Bloch equations ( $\hbar = 1$ ):

$$\begin{aligned} \langle \dot{b}(t) \rangle + 2i \int dt_1 \{ \tilde{M}_{-}(t, t_1) \langle i[\sigma_z(t), b(t_1)]_{-} \rangle + \varphi_{-}(t, t_1) \\ \times \langle \frac{1}{2}[\sigma_z(t), b(t_1)]_{+} \rangle \} - 2i \int dt_1 \{ \tilde{M}(t, t_1) \\ \times \langle i[b(t), \sigma_z(t_1)]_{-} \rangle + \varphi(t, t_1) \langle \frac{1}{2}[b(t), \sigma_z(t_1)]_{+} \rangle \} = 0, \end{aligned} \quad (22)$$

$$\begin{aligned} \langle \dot{\sigma}_z(t) \rangle + i \int dt_1 \{ \tilde{M}_{+}(t, t_1) \langle i[b(t), b^{+}(t_1)]_{-} \rangle + \varphi_{+}(t, t_1) \\ \times \langle \frac{1}{2}[b(t), b^{+}(t_1)]_{+} \rangle \} - i \int dt_1 \{ \tilde{M}_{-}(t, t_1) \\ \times \langle i[b^{+}(t), b(t_1)]_{-} \rangle + \varphi_{-}(t, t_1) \langle \frac{1}{2}[b^{+}(t), b(t_1)]_{+} \rangle \} \\ = 0. \end{aligned} \quad (23)$$

The quantum Furutsu-Novikov theorem [14]

$$\langle \frac{1}{2}[Q_z^{(0)}(t), b(t)]_{+} \rangle = \int dt_1 \tilde{M}(t, t_1) \langle i[b(t), \sigma_z(t_1)]_{-} \rangle, \quad (24)$$

with  $\tilde{M}(t, t_1) = M(t, t_1) \theta(t - t_1)$ , and the function

$$M_{-}(t, t_1) = [M_{+}(t, t_1)]^{+} = M(t, t_1) \exp\left[i \int_{t_1}^t d\tau \Delta(\tau)\right], \quad (25)$$

are being used at the derivation of Eqs. (22) and (23).

The exact microscopic equations (22) and (23) can be put in a more simple form assuming a weak coupling of the spin subsystem to the heat bath. We suppose that the damping rate  $\gamma$  of spin variables is much less than the frequency  $\Delta$ :  $\gamma \ll \Delta$ , so that we have approximately  $b(t) \approx b(t_1)$ ,  $\sigma_z(t) \approx \sigma_z(t_1)$  and

$$[b^{+}(t), b(t_1)]_{-} = 4\sigma_z(t_1); [b(t), b^{+}(t_1)]_{+} = 4,$$

$$[\sigma_z(t), b(t_1)]_{-} = 2b(t_1); [\sigma_z(t), b(t_1)]_{+} = 0.$$

It is known [17] that such underdamped behavior of the two-level system takes place for a super-Ohmic heat bath or for an Ohmic heat bath if the system-environment coupling is sufficiently weak and the heat bath temperature is not too high. The investigation of the spin coupling to a sub-Ohmic heat bath is much more cumbersome [17,21] and is not performed here. Thus in the weak-damping limit the following non-Markovian equations may be obtained from the generalized Bloch equations (22) and (23):

$$\langle \dot{b}(t) \rangle + \int dt_1 G(t, t_1) \langle b(t_1) \rangle = 0, \quad (26)$$

$$\langle \dot{\sigma}_z(t) \rangle + \int dt_1 G_z(t, t_1) \langle \sigma_z(t_1) \rangle + \alpha_z(t) = 0. \quad (27)$$

These equations incorporate an effect of the magnetic field  $f_z(t)$  on the relaxation process, as indicated by the dependence of the functions  $G(t, t_1)$ ,  $G_z(t, t_1)$ , and  $\alpha_z(t)$  on the time-varying splitting  $\Delta(\tau) = \Delta - 2f_z(\tau)$ :

$$G(t, t_1) = 4\tilde{M}(t, t_1) \left[ 1 + \exp\left(i \int_{t_1}^t d\tau \Delta(\tau)\right) \right],$$

$$G_z(t, t_1) = 8\tilde{M}(t, t_1) \cos\left(\int_{t_1}^t d\tau \Delta(\tau)\right), \quad (28)$$

$$\alpha_z(t) = 4 \int dt_1 \varphi(t, t_1) \sin\left(\int_{t_1}^t d\tau \Delta(\tau)\right).$$

In the absence of a time-dependent magnetic field [ $f_z(t) = 0, \Delta(\tau) = \Delta$ ] we obtain the well-known results [1–6,17] describing the relaxation of longitudinal and transversal spin variables:

$$\langle \sigma_z(t) \rangle = e^{-\gamma_z t} \langle \sigma_z(0) \rangle + [1 - e^{-\gamma_z t}] \sigma_z^0,$$

$$\langle \sigma_{tr}(t) \rangle = e^{-\gamma_x t - i\Delta t} \langle \sigma_{tr}(0) \rangle, \quad (29)$$

where  $\sigma_{tr} = \sigma_x - i\sigma_y$ ;  $\sigma_z^0 = -\tanh(\Delta/2T)$  is the equilibrium  $z$  projection of the spin;  $\gamma_x = 2[S(0) + S(\Delta)]$  and  $\gamma_z = 4S(\Delta)$  are the transversal and longitudinal damping rates, respectively; and  $S(\omega)$  is the spectral density of heat bath fluctuations (18) which is proportional to the imaginary part  $\text{Im}\chi(\omega)$  of the heat bath susceptibility (19) [8]:  $S(\omega) = \text{Im}\chi(\omega) \coth(\omega/2T)$ . As expected [17], for super-Ohmic and Ohmic heat baths, there are conditions which correspond to the underdamped behavior of the spin system and justify the weak-damping limit ( $\gamma/\Delta \ll 1$ ).

### III. ABSORPTION OF THE MAGNETIC FIELD AND LONGITUDINAL SPIN FLUCTUATIONS

We now proceed to a calculation of the linear-response function  $\langle \delta\sigma_z(t)/\delta f_z(t') \rangle$  determining the absorption  $P_z(\omega)$ , Eq. (7), of the longitudinal magnetic field. As for the standard Bloch equations (1) and (2), the longitudinal magnetic field  $f_z(t)$  does not appear in the generalized equation (27) in an explicit form. However, by virtue of time nonlocality the functions  $G_z(t, t_1)$ ,  $G(t, t_1)$ , and  $\alpha_z(t)$  are functionals of the magnetic field  $f_z(\tau)$ . That is why the equation for the linear longitudinal response function, resulting from the non-Markovian equation (27), has a nonzero right-hand side:

$$\begin{aligned} \frac{d}{dt} \left\langle \frac{\delta\sigma_z(t)}{\delta f_z(t')} \right\rangle + \int dt_1 G_z(t, t_1) \left\langle \frac{\delta\sigma_z(t_1)}{\delta f_z(t')} \right\rangle \\ = \frac{\delta\alpha_z(t)}{\delta f_z(t')} - \sigma_z^0 \int dt_1 \frac{\delta G_z(t, t_1)}{\delta f_z(t')} \\ = 8\theta(t-t') \int_{-\infty}^{t'} dt_1 L_z(t-t_1), \end{aligned} \quad (30)$$

with

$$L_z(\tau) = \varphi(\tau) \cos(\Delta\tau) - 2\tilde{M}(\tau) \sigma_z^0 \sin(\Delta\tau). \quad (31)$$

Here we take into account the relation

$$\begin{aligned} \frac{\delta}{\delta f_z(t')} \int_{t_1}^t d\tau [\Delta - 2f_z(\tau)] = -2 \int_{t_1}^t d\tau \delta(\tau - t') \\ = -2\theta(t-t')\theta(t'-t_1), \end{aligned} \quad (32)$$

and on differentiation with respect to  $f_z(t')$  set  $f_z=0$ . For longitudinal susceptibility  $\Phi_{zz}(\omega)$  one obtains, from Eqs. (5) and (30),

$$\Phi_{zz}(\omega) = \frac{8}{i\omega} \frac{\Lambda_z(\omega) - \Lambda_z(0)}{-i\omega + \Gamma_z(\omega)}, \quad (33)$$

where  $\Lambda_z(\omega)$ ,  $\Gamma_z(\omega)$ , and  $\tilde{S}(\omega)$  are the Fourier transforms of the functions  $L_z(\tau)$  [Eq. (31)],  $G_z(\tau)$  [Eq. (28)], and  $\tilde{M}(\tau) = M(\tau)\theta(\tau)$ , respectively:

$$\begin{aligned} \Lambda_z(\omega) = \frac{1}{2} [\chi(\omega + \Delta) + \chi(\omega - \Delta)] \\ + i\sigma_z^0 [\tilde{S}(\omega + \Delta) - \tilde{S}(\omega - \Delta)], \end{aligned} \quad (34)$$

$$\Gamma_z(\omega) = 4[\tilde{S}(\omega + \Delta) + \tilde{S}(\omega - \Delta)]. \quad (35)$$

The longitudinal magnetic-field absorption  $P_z(\omega)$  is determined by the imaginary part of the susceptibility  $\Phi_{zz}(\omega)$ , Eq. (33):

$$P_z(\omega) = 4(g\mu_0)^2 B_1^2 \frac{\omega \text{Im}\Lambda_z(\omega)}{\omega^2 + [\text{Re}\Gamma_z(\omega)]^2}. \quad (36)$$

The functions  $\text{Re}\Gamma_z(\omega)$  and  $\text{Im}\Lambda_z(\omega)$  can be written, in view of the relation  $\text{Re}\tilde{S}(\omega) = (1/2)S(\omega)$  and the Callen-Welton fluctuation-dissipation theorem (FDT) [8], as

$$\begin{aligned} \text{Re}\Gamma_z(\omega) = 2\text{Im} \left[ \chi(\omega - \Delta) \coth\left(\frac{\omega - \Delta}{2T}\right) \right. \\ \left. + \chi(\omega + \Delta) \coth\left(\frac{\omega + \Delta}{2T}\right) \right], \end{aligned} \quad (37)$$

$$\begin{aligned} \text{Im}\Lambda_z(\omega) = \frac{1}{2} \text{Im}\chi(\omega - \Delta) \left[ 1 + \tanh\left(\frac{\Delta}{2T}\right) \coth\left(\frac{\omega - \Delta}{2T}\right) \right] \\ + \frac{1}{2} \text{Im}\chi(\omega + \Delta) \left[ 1 - \tanh\left(\frac{\Delta}{2T}\right) \coth\left(\frac{\omega + \Delta}{2T}\right) \right]. \end{aligned} \quad (38)$$

According to the FDT and Eq. (33), the spectral density of the fluctuations of the longitudinal spin variable  $\sigma_z(t)$  will look like

$$K_{zz}(\omega) = 8 \coth\left(\frac{\omega}{2T}\right) \frac{\text{Im}\Lambda_z(\omega)}{\omega^2 + [\text{Re}\Gamma_z(\omega)]^2}. \quad (39)$$

It should be emphasized that the calculations of the spectral density  $K_{zz}(\omega)$  performed by means of stochastic equations and fluctuation sources furnish the same result.

By these means the generalized Bloch equations (22) and (23) point to the occurrence of absorption of the longitudinal magnetic-field energy, Eq. (36), by the spin- $\frac{1}{2}$  coupled to a heat bath, and with it to the existence of equilibrium fluctuations of longitudinal spin variables. In view of the relation

$$[1 - \coth(A)\tanh(B)]\coth(A-B) = \coth(A) - \tanh(B),$$

the function  $\text{Im}\Lambda_z(\omega)\coth(\omega/2T)$  may be rearranged to give

$$\begin{aligned} \text{Im}\Lambda_z(\omega)\coth\left(\frac{\omega}{2T}\right) = \frac{1}{2} \text{Im}\chi(\omega - \Delta) \left[ \coth\left(\frac{\omega - \Delta}{2T}\right) \right. \\ \left. + \tanh\left(\frac{\Delta}{2T}\right) \right] + \frac{1}{2} \text{Im}\chi(\omega + \Delta) \\ \times \left[ \coth\left(\frac{\omega + \Delta}{2T}\right) - \tanh\left(\frac{\Delta}{2T}\right) \right]. \end{aligned} \quad (40)$$

Substituting this function into Eqs. (39) and (10) yields the desired result (11) for the variance of longitudinal fluctuations  $\langle (\Delta\sigma_z)^2 \rangle$

$$\begin{aligned} \langle (\Delta\sigma_z)^2 \rangle = \left[ 1 - \tanh^2\left(\frac{\Delta}{2T}\right) \right] \int_{-\infty}^{+\infty} \frac{d\omega}{\pi} \frac{\gamma_z}{\omega^2 + \gamma_z^2} \\ = 1 - \tanh^2\left(\frac{\Delta}{2T}\right), \end{aligned} \quad (41)$$

provided the damping rate  $\gamma_z = \text{Re}\Gamma_z(\omega=0) = 4\text{Im}\chi(\Delta)\coth(\Delta/2T)$  is far less than the frequency  $\Delta$  and the temperature  $T$ :  $\gamma_z \ll \Delta, T$ . Consequently, the generalized non-Markovian equations (22) and (23) produce better results for the longitudinal spin fluctuations than the conventional Bloch equations (1) and (2) do. It is also worth noting that for an Ohmic heat bath with  $\text{Im}\chi(\omega) = \lambda\omega$  ( $\lambda = \text{const}$ ) the spectral density  $K_{zz}(\omega)$ , Eq. (39), has the Lorentzian shape  $K_{zz}(\omega) = 2\gamma_z/(\omega^2 + \gamma_z^2)$  at high temperatures

( $T \gg \hbar \omega, \hbar \Delta$ ) when  $\text{Im}\Lambda_z(\omega) = \lambda \omega, \text{Re}\Gamma_z(\omega) = \gamma_z = 8\lambda T$ . The maximum value of the longitudinal absorption, Eq. (36), is therewith much less than the maximum of transversal absorption (6):  $P_z(\gamma_z)/P_x(\Delta) \approx \gamma_z \gamma_x / \Delta^2 \approx (8\lambda T)^2 / \Delta^2 \ll 1$ , and yet this ratio increases as the splitting  $\Delta$  decreases.

It is notable that under these conditions the spectral density of heat bath fluctuations  $S(\omega) = \text{Im}\chi(\omega) \coth(\omega/2T)$  does not depend on the frequency,  $S(\omega) = 2\lambda T$ , so that the correlation function (18) is  $M(t, t') = 2\lambda T \delta(t - t')$ . This limit corresponds to the Markovian approximation. The spectral density  $\Xi_{zz}(\omega)$  of fluctuating forces responsible for the fluctuations of longitudinal spin variables is known to be determined by the numerator of the function  $K_{zz}(\omega)$ , Eq. (39):  $\Xi_{zz} = 8\text{Im}\Lambda_z(\omega) \coth(\omega/2T)$ . As one would expect, for a Gaussian Ohmic heat bath the spectral density  $\Xi_{zz}(\omega)$  is independent of the frequency at high temperatures:  $\Xi_{zz}(\omega) = 16\lambda T = 2\gamma_z$ , and the corresponding fluctuating forces become  $\delta$  correlated. However, now we have obtained the correct expression (41) for the variance of longitudinal fluctuations, whereas the Bloch equations (1) and (2) using the Markovian correlation function  $M(t, t') = 2\lambda T \delta(t - t')$  from the outset predict a zero result. Dissipation of longitudinal magnetic-field energy and longitudinal spin fluctuations also exist in the case of the high-temperature Ohmic heat bath, when the Markovian approximation nominally takes place. However, a transition to the Markovian limit in the generalized Bloch equations (22) and (23) never must be performed until the linear or nonlinear response functions of interest are calculated.

It should be mentioned that the results beyond the Markovian limit can be also obtained by means of the instanton technique in the noninteracting-blip approximation (NIBA) [17 (Chap. IV), 19, 20]. As may be seen from Eqs. (4.21b) and (4.22b) of Ref. [17], the NIBA is true for a classical Ohmic heat bath when the Markovian description is correct. Nonetheless, the results deduced in Refs. [17–19] point to the fact that the breakdown of the Markovian approximation does not necessarily signify the breakdown of the NIBA.

At low temperatures ( $\Delta \gg T \gg \gamma$ ) longitudinal absorption and longitudinal fluctuations covered by Eqs. (36) and (39) are significantly diminished because the function  $\text{Im}\Lambda_z(\omega)$ , Eq. (38), is nil over the range  $-\Delta < \omega < \Delta$ ,

$$\text{Im}\Lambda_z(\omega) = \text{Im}\chi(\omega - \Delta)\theta(\omega - \Delta) + \text{Im}\chi(\omega + \Delta)\theta(-\omega - \Delta), \quad (42)$$

with  $\theta(\omega > 0) = 1$ , and  $\theta(\omega < 0) = 0$ . In this case absorption of the longitudinal magnetic-field energy  $P_z(\omega)$ , Eq. (36), as

well as the spectral density  $K_{zz}(\omega)$ , Eq. (39), begin from the (positive) frequency  $\omega = \Delta$  that is far in excess of the absorption line width  $\text{Re}\Gamma_z(\omega)$ , Eq. (37). That is the reason why a contribution of the spectrum  $K_{zz}(\omega)$  to the variance of longitudinal fluctuations  $\langle (\Delta\sigma_z)^2 \rangle$ , Eq. (10), is negligibly small, so that the longitudinal projection of the spin has the fixed value  $\sigma_z = \sigma_z^0$  at  $T = 0$ .

#### IV. CONCLUSIONS

We have analyzed the relaxation of a spin- $\frac{1}{2}$  interacting with a Gaussian heat bath in the presence of an external magnetic field. The magnetic field involves a  $z$ -directed constant part and a time-varying component which is also parallel to the  $z$  axis. With the microscopic method, we derived the generalized non-Markovian Bloch equations which allow for an effect of time-dependent magnetic field on the relaxation process. The longitudinal susceptibility of the spin system calculated by means of these equations in the weak-damping limit is nonvanishing, suggesting that a dissipation of longitudinal magnetic-field energy exists. The absorption spectrum peaks in the low-frequency region at high temperature, whereas for low temperature the absorption of longitudinal magnetic-field energy takes place on frequencies in excess of the splitting frequency  $\Delta$ . The energy of the longitudinal magnetic field is rather weakly absorbed by the spin system in comparison with the dissipation of the transversal magnetic-field energy. However, at sufficiently elevated temperatures the spectral density and the dispersion of longitudinal spin fluctuations compare well with those for fluctuations of transversal spin variables. The fluctuations of the longitudinal spin projection are “squeezed” as the temperature decreases, so that the  $z$  projection of the spin has a fixed value at  $T = 0$ . We have emphasized that application of the conventional Bloch equations leads to zero results for the dissipation of the longitudinal magnetic-field energy, and for fluctuations of longitudinal spin variables at an arbitrary temperature of the heat bath. Only when the non-Markovian equations are used can more correct results be obtained which are in good agreement with Eq. (11) for the dispersion  $\langle (\Delta\sigma_z)^2 \rangle$ .

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