Non-Bloch Transients in Solids: Free Induction Decay and Transient Nutations

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(Received 19 June 1997)

Free induction decay and transient nutations are investigated in a system of two-level particles with an inhomogeneously broadened spectrum. The influence of the x (real) and y (imaginary) parts of the complex amplitude of the noise, induced by the driving field in the sample, is considered. It is shown that both noises affect T_1 and T_2 relaxation processes. When field-noise contributions to $1/T_2$ and $1/T_1$ relaxation rates are dominant and equal to each other, Bloch's saturation is changed to Redfield's saturation. This effect also accelerates transient nutation decay. Comparison with experimentally observed free induction decay and transient nutation in color centers in quartz is discussed. [S0031-9007(97)04230-0]

PACS numbers: 42.50.Md, 76.30.Mi, 76.90.+d

Conventional Bloch equations (CBE) are a fundamental starting point for describing the resonance effects of the atom-field interaction, including lasing, coherent spectroscopy, and pulse propagation in resonant dense media. Therefore the experimental indications of CBE failure in the description of coherent transients [1–6] and hole burning [7] in solids with inhomogeneous absorption spectra stimulated a widespread theoretical interest [8–20] (the list is not exhausted).

Indirect measurement of the hole width via free induction decay (FID) [1–5] and direct measurement by the pump-probe pulse train [7] revealed that the hole burnt in the inhomogeneous spectrum by a long pulse is much narrower than CBE predict. In fact, according to the CBE, the half width at half maximum (HWHM) of the hole equals $\sim\chi\sqrt{T_1/T_2}$ in the strong saturation ($\chi^2 T_1 T_2 \gg 1$) limit, where χ is the Rabi frequency, T_2 is the dephasing time, and T_1 is the relaxation time of the population difference. At variance, several kinds of experiments indicate a HWHM $\sim\chi$. As the ratio T_1/T_2 is usually very high for low-temperature solids, this huge difference was explained by several theories, hypothesizing that in the presence of a strong resonant field T_2 lengthens up to T_1 because of fast Rabi oscillations, which average out the random local field contribution to T_2. This matter continues to arouse interest in the study of nonlinear phenomena in solids with field-dependent dephasing (see, for example, the study [20] of nonlinear dephasing influence on two-wave mixing).

The theories mentioned above share a common frame in which the anomalous hole burning narrowing manifested by FID is ascribed to the dephasing suppression induced by the strong field. However, recent experiments on the transient nutations (TN) in magnetic resonance solid systems at low temperature [21] disprove these models. In fact, the direct measurement of the TN decay rate has shown that the dephasing rate increases, rather than decreases, with Rabi frequency. Therefore one can expect that there is some process giving the same contribution to the T_2 dephasing and to T_1 relaxation to produce the narrow hole.

In this Letter, we show that the anomalies experimentally observed in FID and TN decay are not conflicting with each other; on the contrary, they can have a common origin. We postulate the existence of x (in phase) and y (out of phase) noise components induced by the driving field in the sample. They are considered as uncorrelated and analyzed separately. We show that the two noise components influence to a different extent both T_1 and T_2 relaxation processes. The theoretical results are compared to experiments on FID [4] and TN [21].

In our model the dephasing acceleration is caused by the fluctuations induced by the driving field $\mathbf{H}_1(t) = \mathbf{H}_1 \exp(i\omega t)$ in the sample. We classify them in the following way. In Fig. 1 we present the diagram of fields

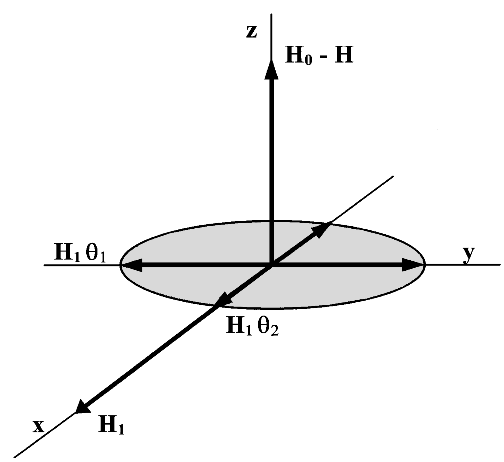

FIG. 1. Diagram of the fields interacting with the spin system in the rotating reference frame. The fluctuating fields $\mathbf{H}_1\theta_1$ and $\mathbf{H}_1\theta_2$ are shown by two-directed arrows. Coherent field $\mathbf{H}_1$ and constant field $\mathbf{H}_0 - \mathbf{H}$ are shown by single-directed arrows.

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interacting with the two-level system in the rotating reference frame (RRF), where $\mathbf{H}_1$ is constant and aligned to the $\mathbf{x}$ axis, and $\mathbf{H}_0 - \mathbf{H}$ is the remnant of the constant field $\mathbf{H}_0$ (aligned along the $\mathbf{z}$ axis), which is reduced due to the rotation of the reference frame around the $\mathbf{z}$ axis with the frequency ω by the effective field $H = \hbar\omega/\gamma$; here γ is the gyromagnetic ratio. $\mathbf{H}_1\theta_1$ and $\mathbf{H}_1\theta_2$ are random fields induced by the driving field. The first is transverse to the $\mathbf{H}_1$ field and it is aligned along the $\mathbf{y}$ axis, so we name it "y noise." The second is aligned along the $\mathbf{x}$ axis and it is "x noise" in our classification. The probable origin of these fluctuations is the following.

(i) These fluctuations may be caused by amplitude and phase fluctuations of the input field. The first rises x noise. The second corresponds to the y noise when θ_1 is small. In general, these x and y components are, respectively, the real and imaginary parts of the complex amplitude of the noise. Their ratio determines noise squeezing of the driving field [22]. For example, when the y component is much smaller than the x component, we have a phase squeezed field. It should be noted that we consider the noise with the power exceeding the quantum-noise level.

(ii) The driving field excites a transverse magnetization in the sample. The spins neighboring the particular spin under consideration induce random fields with x and y components.

(iii) Multiple reflection of the field from the borders and imperfections in the dielectric sample may cause the fluctuations, too, in the optical band when the wavelength is shorter than the sample sizes.

Now one can consider the two-level system excited by the field with both random components, i.e.,

$$H_1(t) = H_1[1 + \theta_2(t) + i\theta_1(t)]\exp(i\omega t). \quad (1)$$

We admit that mean values of random variables $\theta_n(t)$ are zero, where $n = 1(2)$. As x and y noises are perpendicular to the H_0 field, they are nonadiabatic for Zeeman interaction and induce the transition between the spin levels split in the laboratory frame. As a result, they are expected to contribute to the relaxation of the z component of magnetization (T_1 relaxation). The y noise component is perpendicular to the coherent component H_1 of the field in Eq. (1) as well. Therefore, it induces transitions between the spin levels split by the H_1 field in the rotating frame and contributes to the relaxation of the x component of magnetization or T_{2x} relaxation. As x noise is adiabatic for Zeeman interaction of the spin system with the H_1 field in RRF, it gives no contribution to the relaxation of the x component of magnetization. However, being perpendicular to the y component of magnetization, it contributes to T_{2y} relaxation.

We consider x and y noises separately as we assume them uncorrelated. Moreover, we assume for both noises the Markovian uncorrelated jump model with $\theta_n(t)$ shifting instantly at successive times over a Poisson distribution. Then the stochastic method developed in [23] for solving a similar problem is applied (see also Refs. [15,16,19,24]). We choose this method as it has no restrictions on the parameter of the stochastic process, whereas the conventional method of kinetic equations derivation (see, for example, Ref. [25]) is valid within perturbation theory approach. In the stochastic method the partial density matrix $\rho(t,\theta_1)$ is introduced. The last obeys the equation of motion

$$\dot{\rho}(t,\theta_n) = -\frac{i}{\hbar}[\hat{\mathcal{H}}(\theta_n), \rho(t,\theta_n)] - \hat{R}\rho(t,\theta_n) - \frac{1}{\tau_c}\rho(t,\theta_n) + \frac{1}{\tau_c}\varphi(\theta_n)\int \rho(t,\theta_n')\,d\theta_n', \quad (2)$$

where $\hat{\mathcal{H}}(\theta_n)$ is the Hamiltonian of the spin system interacting with coherent and random fields, τ_c is the average time between two successive changes of random field, and $\varphi(\theta_n)$ is the stationary distribution of the random variable $\theta_n(t)$. One can normalize and reduce these equations as follows:

$$\dot{x}(\xi,\tau) = -(\hat{L}_0 + \xi\hat{L}_n)x(\xi,\tau) + \varphi(\xi)[\overline{x}(\tau) + \hat{\Lambda}], \quad (3)$$

where

$$x = \begin{bmatrix} u \\ v \\ w \end{bmatrix}; \qquad \hat{L}_0 = \begin{bmatrix} t_2 & z & 0 \\ -z & t_2 & -W \\ 0 & W & t_1 \end{bmatrix}; \qquad \hat{\Lambda} = w_{\mathrm{eq}}(t_1 - 1)\begin{bmatrix} 0 \\ 0 \\ 1 \end{bmatrix}; \qquad \overline{x}(\tau) = \int x(\xi,\tau)\,d\xi. \quad (4)$$

Variables u, v, and w are the following combinations of the elements of the partial density matrix: $u + iv = 2\rho_{12}(\theta_n)\exp(-i\omega t) = 2\sigma_{12}(\theta_n)$ and $w = \rho_{22}(\theta_n) - \rho_{11}(\theta_n)$, where index 1 (2) belongs to the ground (excited) state. We assume that random Rabi frequency is described by expression $\chi(t) = \chi_0[1 + i\theta_1(t)]$ or by $\chi(t) = \chi_0[1 + \theta_2(t)]$, depending on the noise type. Then two normalized parameters $W = \chi_0\tau_c$ and $\xi = \theta_n\chi_0\tau_c$ are introduced: $z = (\omega_0 - \omega)\tau_c$ is the normalized detuning from the resonant frequency ω_0; time τ in Eq. (3) is expressed in units of τ_c; $t_{1,2} = 1 + \tau_c/T_{1,2}$; $w_{\mathrm{eq}} = -\tanh(\hbar\omega_0/2kT)$ is the equilibrium population difference, where T is the thermal bath temperature. The operator $\hat{L}_n$ has the form

$$\hat{L}_1 = \begin{bmatrix} 0 & 0 & 1 \\ 0 & 0 & 0 \\ -1 & 0 & 0 \end{bmatrix} \quad \text{or} \quad \hat{L}_2 = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & -1 \\ 0 & 1 & 0 \end{bmatrix}, \quad (5)$$

depending on the kind of noise ($\hat{L}_1$ is valid for the y noise and $\hat{L}_2$ is defined for x noise). By applying the Laplace transformation [16,19], we get the solution of Eq. (3),

$$\overline{x}(p) = [1 - (1 - \overline{\hat{\mathcal{L}}})^{-1}]\frac{\hat{\Lambda}(t_1 + p\tau_c - 1)}{(1 - t_1)p}, \quad (6)$$

where p is the Laplace variable; $\overline{\hat{\mathcal{L}}} = \int \hat{\mathcal{L}}(\xi)\varphi(\xi)\,d\xi$ and $\hat{\mathcal{L}}(\xi) = (p\tau_c + \hat{L}_0 + \xi\hat{L}_n)^{-1}$.

The experimental TN signal in Ref. [21] was well fitted by the function

$$J_0(\chi_0 t)\exp(-\Gamma_{\mathrm{TN}} t), \tag{7}$$

and the decay rate Γ_{TN} was found to depend on the Rabi frequency as

$$\Gamma_{\mathrm{TN}} = \alpha + \beta\chi_0, \tag{8}$$

where α is close to the $(2T_2)^{-1}$. One can expect x and y noise contributions to the relaxation which are square dependent on Rabi frequency as the dispersion of the random process [25]. To fit the experimental linear dependence on the Rabi frequency we choose the random process with Lorentzian distribution

$$\varphi(\xi) = \frac{1}{\pi}\frac{a}{\xi^2 + a^2}, \tag{9}$$

as it has no defined dispersion and may provide non-square dependence (see, for example, Ref. [15]). Here $a = \theta_{n0}\chi_0\tau_c$ and θ_{n0} is the half-width of the distribution function. We consider the noise with very short correlation time conditioned by inequalities: $\tau_c \ll T_1, T_2, \chi_0^{-1}, \Delta^{-1}$. Moreover, we put the condition of $\theta_{n0} \ll 1$. Then we can confine our considerations to a time scale much longer than τ_c and restrict the analysis of the solution in Eq. (6) to small $p\tau_c$ ($|p\tau_c| \ll 1$). This approximation corresponds to disregarding the part of the solution that decays very rapidly with a rate $\sim 1/\tau_c$. In this condition, for example, the expression of the averaged $\overline{v}(p)$ component, influenced by y noise only, is reduced to

$$\overline{v}(p) = \frac{w_{\mathrm{eq}}\chi_0(T_2^{-1} + p)(T_1^{-1} - \theta_{10}\chi_0 + p)}{p[(T_1^{-1} + p)\Delta^2 + (T_2^{-1} + p)\chi_0^2 + (T_1^{-1} + p)(T_2^{-1} + p)(T_2^{-1} - \theta_{10}\chi_0 + p)]}. \tag{10}$$

One can easily reconstruct from Eq. (10) and similar equations for the other components of the density matrix, averaged on the noise jumps, the modified Bloch equations

$$\begin{aligned} \frac{d\overline{u}}{dt} &= -\Delta\overline{v} - \Gamma_u\overline{u}, \\ \frac{d\overline{v}}{dt} &= \Delta\overline{u} + \chi_0\overline{w} - \Gamma_v\overline{v}, \\ \frac{d\overline{w}}{dt} &= -\chi_0\overline{v} - \Gamma_w\overline{w} + \frac{w_{\mathrm{eq}}}{T_1}, \end{aligned} \tag{11}$$

where $\Gamma_u = 1/T_2 + \theta_{10}\chi_0$, $\Gamma_v = 1/T_2$, and $\Gamma_w = 1/T_1 + \theta_{10}\chi_0$.

As admitted above, the x and y noises are uncorrelated, so their effects are additive. By applying the same procedure as before for the x noise, one can derive the same Eqs. (11), with redefined decay rates

$$\begin{aligned} &\Gamma_u = T_2^{-1} + \theta_{10}\chi_0; \qquad \Gamma_v = T_2^{-1} + \theta_{20}\chi_0; \\ &\Gamma_w = T_1^{-1} + (\theta_{10} + \theta_{20})\chi_0, \end{aligned} \tag{12}$$

which take into account both x and y noises.

Equations (11), with the decay rates in Eqs. (12), give the following expression for the FID rate:

$$\Gamma_{\mathrm{FID}} = \frac{1}{T_2} + \sqrt{\Gamma_u\Gamma_v + \chi_0^2\frac{\Gamma_u}{\Gamma_w}}. \tag{13}$$

As will be shown below, Eq. (13) is capable of accounting for the power dependence of Γ_{FID} experimentally measured in magnetic resonance systems [4]. Here we point out that in the high-power limit when θ_{10} and θ_{20} are larger than $(\chi_0 T_2)^{-1}$ and $(\chi_0 T_1)^{-1}$, the decay rate Eq. (13) is approximated by

$$\Gamma_{\mathrm{FID}} = \frac{1}{T_2} + \chi_0\sqrt{\frac{\theta_{10}}{\theta_{10} + \theta_{20}}}. \tag{14}$$

When θ_{10} and θ_{20} values are equal we get the asymptote

$$\Gamma_{\mathrm{FID}} = \frac{1}{T_2} + \frac{\chi_0}{\sqrt{2}}, \tag{15}$$

which represents the Redfield limit [1]. Any noise squeezing or deviation of ratio θ_{10}/θ_{20} from one changes the FID rate dependence on Rabi frequency in the high-power limit. Thus one can use the hole burning or FID to detect the noise squeezing.

According to Eqs. (11), the TN signal [Eq. (7)] is well approximated by the zeroth order Bessel function, exponentially damped with a rate

$$\Gamma_{\mathrm{TN}} = \tfrac{1}{2}(\Gamma_w + \Gamma_v), \tag{16}$$

which, for $T_1 \gg T_2$ (this inequality is typical for solids at low temperature), reduces to

$$\Gamma_{\mathrm{TN}} = \frac{1}{2T_2} + \left(\frac{\theta_{10}}{2} + \theta_{20}\right)\chi_0. \tag{17}$$

The above result is in agreement with experiments on TN, whose decay rate was well fitted by the linear law in Eq. (8). The comparison with theoretical law allows us to relate the phenomenological parameter β to the normalized noise amplitudes θ_{10} and θ_{20}, as

$$\beta = \tfrac{1}{2}\theta_{10} + \theta_{20}. \tag{18}$$

To make the comparison quantitative between theoretical and experimental results, we report in Figs. 2 and 3 the experimental data of the χ_0 dependence of Γ_{TN} and of Γ_{FID}, respectively, as measured in the sample of $[\mathrm{AlO}_4]^0$ centers in quartz (Ref. [4] and sample No. 1 of Ref. [21]). The χ_0 dependence of Γ_{TN} in Fig. 2 is well fitted by the linear law Eq. (17) with slope $\beta = 0.025 \pm 0.002$, which gives the condition Eq. (18) on noise amplitudes. The full line in Fig. 3 plots the curve in Eq. (13) with the constraint in Eq. (18) calculated for nonsqueezed noise ($\theta_{10} = \theta_{20}$) to reach the Redfield limit. As shown, the agreement is satisfying to a large extent, except for the very-low power

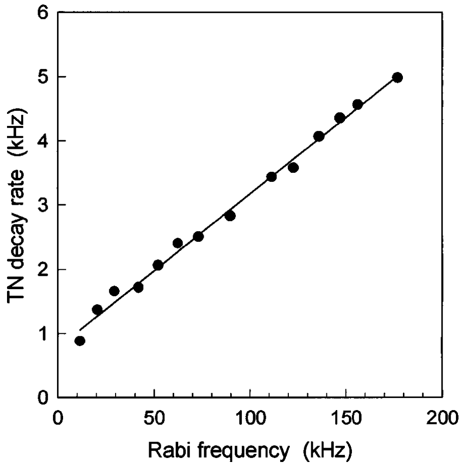

FIG. 2. Transient nutations decay dependence on Rabi frequency. Solid line is theoretical plot of Eq. (17) and black circles are experimental points from Ref. [21].

region. If we cut the wings of distribution $\varphi(\theta_n)$ starting from the values $\pm 1/\theta_{n0}$, then the contribution $\theta_{n0}\chi_0$ to the relaxation rates is changed as

$$\theta_{n0}\chi_0 \frac{2}{\pi} \arctan\left(\frac{\chi_0}{\chi_c}\right), \tag{19}$$

where $\chi_c = \theta_{n0}/\tau_c$. Assuming $(2\pi\tau_c)^{-1}$ is equal to 0.5 MHz, one can get a better fit at low power. At the same time, there is no appreciable effect of wings cut on linear dependence of $\Gamma_{\rm TN}$ on Rabi frequency shown in Fig. 2. Apart from the improved agreement between theory and experimental results, we get finite power of the noise by wings cut. Thus broad-band noise has much smaller intensity than the coherent driving field within its narrow spectrum.

In conclusion, we have shown that the consideration of *x* and *y* noise in the spin-system interaction is capable of explaining the "non-Bloch" behavior of both TN decay and FID in agreement. Admittedly, the origin of the noise remains an open problem not addressed here. Experimental and theoretical work is in progress.

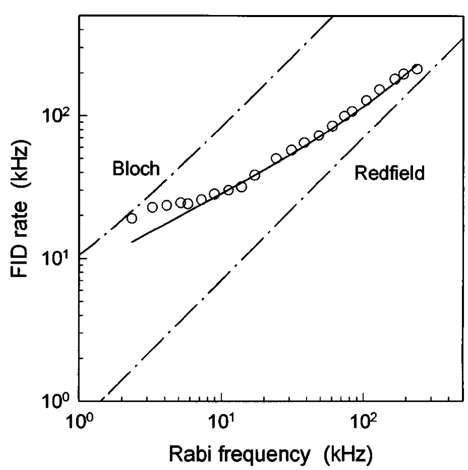

FIG. 3. FID rate vs Rabi frequency. The curve line is the plot of Eq. (13). Circles are experimental points from Ref. [4]. Bloch and Redfield limits are indicated by dash-dotted lines.

The authors express their thanks to R. Boscaino for valuable comments and discussions and for current interest to the research. R. N. Shakhmuratov expresses his thanks to the program on Physics of Quantum and Wave Processes; Trend: Fundamental Spectroscopy (Project No. 4.8). Partial support by Italian Ministry for Scientific Research is also acknowledged.

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