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Transient nutations decay: The effect of field-modified dipolar interaction

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The anomalous behavior of transient nutations is experimentally investigated in a set of two-level ($S=\frac{1}{2}$) spin systems differing only in spin concentration. Our results show that the non-Bloch power dependence of the decay rate of transient nutations is a concentration-dependent effect, which is more and more pronounced in more and more concentrated samples. The experimental results are interpreted in the framework of the recent theory by Shakhmuratov *et al.* [Phys. Rev. Lett. **79**, 2963 (1997)] and support the hypothesis that the anomalous decay of transient nutations in solids originates from radiation-induced changes of the dipolar field, rather than from residual fluctuations of the nominally coherent field. [S1050-2947(99)03905-0]

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I. INTRODUCTION

As is known, several aspects of the resonant interaction between a system of two-level centers (atoms or spins) and a resonant field cannot be explained thoroughly by conventional Bloch equations. In particular, discrepancies between experimental results and theoretical predictions have been reported in connection with free-induction decay (FID) [1–5], hole burning [6], transient nutations (TN) [7], and echo [8,9]. The failure of the Bloch equations is ascribed to the fact that they describe the relaxation processes in an oversimplified way and do not include the frequency fluctuations of the active centers and of the field source.

In a recent paper [10], Shakhmuratov *et al.* proposed a new set of modified Bloch equations (MBE's), capable of accounting for the decay properties of several coherent transients, and in particular FID and TN in low-temperature solids. The set of MBE's proposed in Ref. [10] relies on the assumption that each active center (atom or spin) experiences not only the coherent driving field but also a stochastic field, with the complex amplitude having both a real and imaginary component. The two components are supposed to depend linearly on the amplitude of the coherent field. For instance, for a magnetic resonance system, the hypothesized driving field on the single spin center has the form

$$H_1(t)=H_1[1+\theta_2(t)+i\theta_1(t)]\exp(-i\omega t), \qquad (1)$$

where $\theta_1(t)$ and $\theta_2(t)$ are random variables, uncorrelated from each other, with zero mean values. In that model, the simultaneous presence of the two random fields $H_1\theta_1$ and $H_1\theta_2$ causes the *shortening* of both relaxation times T_1 and T_2, which, in the presence of the strong coherent field, assumes values $T_{1\mathrm{eff}}$ and $T_{2\mathrm{eff}}$, shorter than their equilibrium values. This is the main element of this model with respect to all previous theories [11–24], where the anomalous decay of the coherent transients was ascribed to the *lengthening* of T_2 caused by the field-induced quenching of the dephasing interactions. This is also the reason why the MBE by Shakhmuratov *et al.* succeeded in explaining two apparently conflicting anomalous behaviors: the decay of TN, which was found experimentally to be much faster than predicted by the Bloch equations [7], and the damping of the FID signal which was found experimentally to be much slower than expected [1–6].

We are concerned here with the possible origin of the noise fields appearing in Eq. (1). This was left as an open problem in Ref. [10], where two possibilities were put forward: (i) the external driving field may modify, by some mechanism, the random local field acting on the individual spins and; (ii) the nominally coherent field is actually affected by a residual stochastic modulation. To gain insight into the physical nature of the hypothesized noise fields we have experimentally investigated the effect of varying the local dipolar field (by varying the concentration of active centers) while keeping the statistical properties of the external driving field unaltered. In particular, the experimental results reported below refer to ''non-Bloch'' power-dependence of the TN decay in a magnetic resonance system at microwave frequency.

II. EXPERIMENTAL RESULTS

We consider here a system of two-level ($S=\frac{1}{2}$) spins, whose resonance frequencies are inhomogeneously spread around ω_0 with a standard deviation σ. When the resonant field $H_1\exp(-i\omega_0 t)$is abruptly turned on, the ensuing transient response of the system is described by a transverse ω_0-oscillating magnetization whose amplitude is modulated at the Rabi frequency $\chi=\gamma H_1$ [25]. According to the conventional BE, the slowly varying amplitude *v(t)* of the ω_0-oscillating magnetization is proportional to

$$v(t)\propto K(t)J_0(t), \qquad (2)$$

where $J_0(t)$ is the zeroth order Bessel function and the decay function $K(t)$ is a single exponential law $K(t)=\exp(-\Gamma_B t)$ with the constant rate

$$\Gamma_B = 1/2T_2. \tag{3}$$

The above solution is valid for $T_1 \gg T_2$, in the underdamped regime ($\chi T_2 \gg 1$) and in the high-inhomogeneity limit ($\sigma T_2 \gg 1,\ \sigma \gg \chi$).

As previously reported [7], at variance to this prediction, the decay of TN in impurity-ion solids at low temperature was found to proceed at a higher and power-dependent rate Γ. The experimental data of Γ were well fitted by a linear dependence on the Rabi frequency χ:

$$\Gamma = \alpha + \beta\chi, \tag{4}$$

where α is equal to Γ_B within the experimental uncertainties and β is a parameter of the order of 10^{-2}.

In the MBE proposed by Shakhmuratov *et al.* [10], owing to the presence of the random components of the driving field, the TN decay rate Γ_S is calculated to be

$$\Gamma_S = \frac{1}{2T_2} + \left(\frac{1}{2}\theta_{10} + \theta_{20}\right)\chi, \tag{5}$$

which relates the phenomenological parameter β to the noise field

$$\beta = \frac{1}{2}\theta_{10} + \theta_{20}, \tag{6}$$

where θ_{10} and θ_{20} are the halfwidths of the distributions of the random variables θ_1 and θ_2. In the experiments described below we investigate the dependence of β on the spin concentration c, which, in the framework of this model, images the concentration dependence of the random fields $H_1\theta_1$ and $H_2\theta_2$.

The experiments described below were carried out in samples of high-purity glassy silica (a-SiO_2) containing E' centers [26]. E' centers ($S=\frac{1}{2}$) can be considered as a model spin system particularly suitable for this kind of experiment because of their narrow ESR line and their unusually long relaxation times ($T_2 \cong 100\ \mu s$, $T_1 \cong 1$ s at $T=4.2$ K). Moreover, we note that the inhomogeneous resonance line of E' centers is concentration independent in the range of c considered here and, for our purposes, it can be approximated by a Gaussian line with a standard deviation $\sigma/2\pi \cong 1$MHz so the conditions $\sigma T_2 \gg 1$ and $\sigma \gg \chi$ are well satisfied in our experimental conditions. E' centers can be generated by exposing the silica glass to γ rays and their concentration can be varied by varying the accumulated dose [27]. Here we report the results obtained in three samples whose spin concentrations and relaxation times are summarized in Table I. The relaxation time T_2 and the concentration c were determined in each sample by detecting the echo decay and by analyzing the decay data by the instantaneous diffusion model [28]; the relaxation time T_1 was measured by the standard saturation recovery method.

The experimental apparatus and procedure for exciting and detecting the nutational regime were reported in a previous paper [7], to which we refer for a detailed description. Here we limit ourselves to a brief outline of the aspects relevant to the present work. We used the two-photon-excitation-second-harmonic-detection scheme: the spin system is tuned at the frequency ω_0 ($\omega_0/2\pi = 5.95$ GHz) by the external magnetic static field and irradiated by an intense field at the frequency $\omega = \omega_0/2$, which couples the ground and excited states of each center by two-photon (TP) transitions. The nutational regime is monitored by revealing the radiation emitted by the spin system at the frequency ω_0, namely, at the second harmonic (SH) of the driving field.

TABLE I. Characteristic parameters (spin concentration c and transverse and longitudinal relaxation times) in our three samples. The last column reports the best fit values of the parameter β, as obtained from the data in Fig. 3.

Sample no.	Concentration (10^{17} spins cm^{-3})	T_2 (10^{-4} s)	T_1 (s)	β (10^{-2})
1	0.75±0.2	3.6±0.3	0.9	4.8±0.5
2	1.6±0.5	1.6±0.5	0.7	6.1±0.5
3	2.4±0.8	0.75±0.07	1.2	10.6±0.5

In view of the discussion below, we characterize the spectral properties of the exciting field in more detail. In the experimental setup the source is a low-power (10 mW) cw synthesized microwave generator. Its output signal is raised to the required power level (20 W) by a traveling-wave-tube amplifier (TWTA), whose output feeds the fundamental mode of the bimodal cavity where the sample is located. The TP-effective driving field is $H_{1\mathrm{eff}} = H_1^2/H_0$ [7], where H_0 is the static field and H_1 is the ω-oscillating field on the sample. In order to get a reliable representation of the spectral properties of the effective driving field, the output of the TWTA was sent to a quadratic device (a diode) whose SH response was examined by a microwave spectrum analyzer (SA). A typical result is reported in Fig. 1. As shown, the spectrum consists of an intense coherent part (the central peak) superimposed to a broad structure. The spectrum was

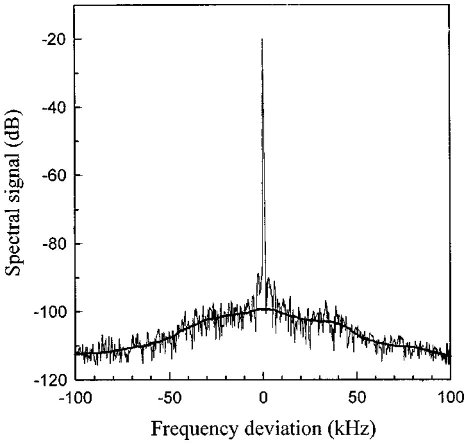

FIG. 1. Spectral density of the TP-effective field driving the spin system. The spectrum was measured by a microwave spectrum analyzer tuned to the center frequency 5.95 GHz and operating with its minimum resolution of 10 Hz. The solid line smooths the noisy components which has a peak spectral density of −100 dB/10 Hz and a spectral full width of nearly 50 kHz. The center peak is narrower than the resolution bandwidth and we can measure only its integrated power (−20 dB).

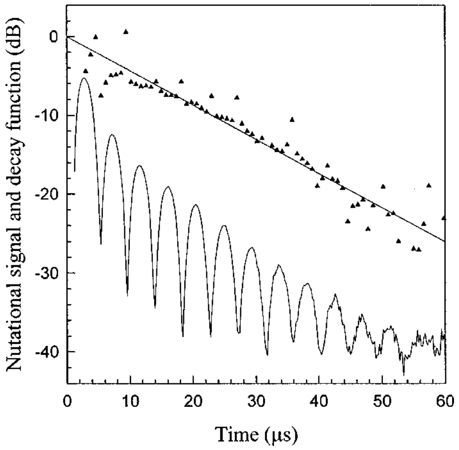

FIG. 2. Typical nutational response as measured in our sample No. 2 for $\chi/2\pi = 113$ kHz. By comparing the experimental curve to the function $|J_0(\chi t)|$ we determine by points (▲) the experimental decay function *K(t),* shown in the upper part of the figure and normalized to initial value. The solid line plots the exponential function $y = \exp(-\Gamma t)$ that best fits the experimental point, $\Gamma/2\pi = 6.8 \pm 0.5$ kHz.

detected by using the minimum available spectral bandwidth of the SA (10 Hz), which is surely low enough to ensure a faithful representation of the broad structure. The solid regular line smooths the noise component, which has a spectral full width of nearly 50 kHz. Taking into account the resolution bandwidth of the SA, its integral power content is $P_{\text{noise}} = 5 \times 10^{-7}$ mW. On the other hand the detection of the central peak is limited by the finite bandwidth of the SA, so that its maximum value (10^{-2} mW) represents the power area of the coherent part. The power ratio between the coherent component and the noisy one is of the order of 43 dB. We verified that this ratio is nearly independent on the power level. This evaluation is relevant to the discussion below.

All the measurements were carried out at the liquid helium temperature $T = 4.2$ K and with the spin system tuned to the exact resonance at the line center. In Fig. 2 we report a typical TN signal as detected in sample No. 2 with a Rabi frequency $\chi/2\pi = 113$ kHz. As discussed in Ref. [7], these experimental TN curves are well described by the damped Bessel functions $K(t)|J_0(\chi t)|$. In the upper part of Fig. 2 we report the experimental decay function *K(t),* as obtained by directly comparing the experimental curve to the function $|J_0(\chi t)|$, with the appropriate value of χ. As shown, the decay function *K(t)* is well described by a single exponential $K(t) = \exp(-\Gamma t)$, over a dynamic range of more than 20 dB.

We measured, for each value of χ, the value of the corresponding decay rate Γ. The results are reported in Fig. 3 for the three samples considered here. The dependence of the decay rate Γ on the Rabi frequency χ manifests the departure of the actual TN regime from the Bloch picture and is well described by the linear law given by Eq. (4) in all the investigated samples, with the values of the parameter β listed in Table I. As shown, β increases on increasing the concentration *c*.

III. DISCUSSION

First we discuss the possibility that the non-Bloch behavior of the TN decay can be ascribed to the residual bandwidth of the exciting field. As shown in Fig. 1, the field used in our experiments consists of a sharp peak (coherent part) that collects most of the incoming power (nearly 99.995%), superimposed to a much broader and less intense structure, which collects the remaining 0.005% of the power. Both values (low power and narrow bandwidth) are in conflict with the model considered in Ref. [10]. First, the small relative power of the noise ruins the advantage of the Lorentzian distribution of the noise amplitude, which provides the linear dependence of the calculated decay rate on the distribution halfwidths and ultimately on the Rabi frequency. In the framework of that model such low values of the noisy power requires us to cut the wings of the noise amplitude distribution at values much less than the halfwidths $\theta_{10}H_1$ and $\theta_{20}H_2$; in this limit Eq. (5) is no longer valid and the dependence of the decay rate on the Rabi frequency χ is no longer linear. Second, according to the model by Shakhmuratov *et al.* [10], when the Rabi frequency exceeds the halfwidth of the noise spectrum (nearly 25 kHz in our experiments) the expected effect of the noise on the dephasing rate decrease due to the ‘‘motional narrowing’’ effect, which is not revealed in our experiments. So, we can conclude that the residual noisy modulation of the driving field is much less than required for explaining the non-Bloch decay of TN at least in magnetic resonance systems.

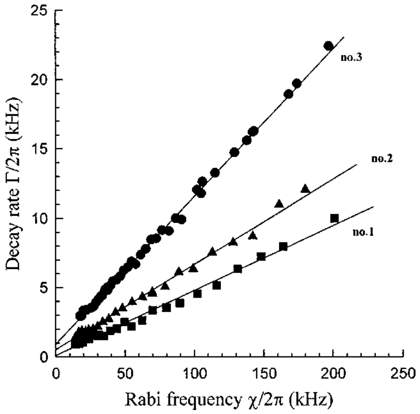

FIG. 3. Experimental dependence of the TN decay rate Γ on the Rabi frequency χ in our three samples: No. 1 (■), No. 2 (▲), and No. 3 (●). In all the samples, the power dependence of Γ is well fitted by a linear law (solid lines): $\Gamma = \alpha + \beta\chi$. The best fit values of β are listed in Table I.

On the other hand, the results reported in Fig. 3 indicate that the non-Bloch behavior of TN is a concentration-dependent effect, which is more and more pronounced in more and more concentrated samples, as reported in Table I. This experimental fact suggests to ascribe the anomalous decay to the modifications of the dipolar field induced by the driving radiation. Hereby we outline a qualitative scheme of a possible mechanism.

Let us consider the generic spin S_i and represent its dipolar interaction with its neighboring spins S_j by a local magnetic field $H_{Li} = \Sigma_j H_{ij}$, where H_{ij} is the field generated at the site S_i by the spin S_j. The spin flips of the sources S_j are responsible for the random time dependence of $H_{Li}(t)$. At equilibrium, spin transitions of S_j are induced by the thermal bath and $H_{ij}(t)$, which collects the effects of many uncorrelated transitions, is well approximated by a white noise. This circumstance is on the basis of the applicability of the conventional BE in near-equilibrium conditions. To be concrete, from the unperturbed values of T_2, of the order of 10^{-4} s, we estimate that the mean square value of H_L ($H_L \cong 1/\gamma T_2$) is a few mG in our samples.

In the presence of the intense resonant field H_1 the spins S_j undergo forced precessions at the frequency ω_0, which induce new spectral components in $H_{Li}(t)$. For instance, the spin S_j with resonance frequency $\omega_{0j} = \omega_0 + \delta_j$, has transverse components oscillating at the field frequency ω_0 but is amplitude modulated at the precession frequency $\beta_j = (\chi^2 + \delta_j^2)^{1/2}$. Because the local field $H_{ij}(t)$ depends on the state of the spin S_j, the forced precession of the latter induces new spectral components into $H_{Li}(t)$. The local field $H_{Li}(t)$ at the site S_i is the superposition of many contributions $H_{ij}(t)$, each one modulated at a different frequency β_j, so, ultimately, it appears noisy. Moreover, as the precession phase of the individual spin S_j with respect to H_1 depends on its detuning δ_j, the driving field-induced dipolar field H_{ij} has both in-phase and in-quadrature components.

So, in the rotating reference frame, the field $H_{Li}(t)$ appears as a noisy field, with properties quite similar to those hypothesized for the stochastic field in the theory by Shakhmuratov *et al.* [10]. In fact, it has both *x*- and *y*-noisy components. Moreover, the amplitude of each component is expected to maintain a linear dependence on χ, since each contribution $H_{ij}(t)$ arises from the H_1-induced component of S_j. Finally, due to its dipolar nature, $H_{Li}(t)$ is expected to increase with increasing concentration *c*, in agreement with the experimental results reported in this paper.

IV. CONCLUSION

In conclusion, our experimental data show that the anomalous decay of the TN is a concentration-dependent effect. In the framework of the MBE recently proposed by Shakhmuratov *et al.* [10] this fact supports the hypothesis that the stochastic field responsible for the anomalous decay originates from the dipolar interaction. On the other hand, the residual noise of the nominally coherent exciting radiation plays no relevant role in determining the TN decay properties. The latter statement may be peculiar of magnetic resonance systems, excited by highly coherent radiation sources; for optical resonances excited by laser sources, additional contributions to the decay of coherent transients may come from the intrinsic noise of the source. A further test of the validity of the new MBE proposed by Shakhmuratov *et al.* [10] can be obtained by investigating the concentration dependence of the anomalous FID emission. Experimental work is in progress.

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