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Non-Bloch decay of transient nutations in $S=\frac{1}{2}$ systems: An experimental investigation

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The decay of transient nutations has been experimentally investigated in $S=\frac{1}{2}$ spin systems at microwave frequency: E' centers in silica and $[AlO_4]^0$ centers in quartz have been studied. We have found that the damping is well described by a single exponential decay function, as expected from a T_1-T_2 model (Bloch model). However, the agreement is only qualitative. In fact the measured decay rate Γ is faster than expected and depends on the driving-field amplitude: it tends to the Bloch value $\Gamma_B=\frac{1}{2}T_2$ in the low-power limit and becomes faster and faster on increasing the input power. In all the cases examined the power dependence of the decay rate is fit well by a simple linear dependence of Γ on the induced Rabi frequency χ. The observed power dependence of Γ cannot be ascribed to the inhomogeneity of χ over the sample volume nor to the radiation damping, since both effects are negligible in our experiments. Other mechanisms, which can, in principle, yield a χ dependence of Γ, e.g., the direct interaction of the driving field with structural two-level systems or the spreading of the spin-field coupling constant, are not compatible with the experimental conditions. So, our results suggest that the homogeneous dephasing time of each isochromat contains an intrinsic term and a χ-dependent one. The latter may originate in a field-induced enhancement of the hyperfine or dipolar interaction; however, neither of these mechanisms completely fits the experimental features. The relationship with the decay properties of other coherent regimes is also discussed.

I. INTRODUCTION

The coherent transient regimes of the interaction between a radiation field and a system of two-level centers (atoms or spins) are given much attention in the current literature. One reason is that the kinetics of the system during these regimes yields information on the coherence-loss mechanisms in a time scale shorter than the relaxation time; this is a valuable complement to the information gained by exploring the steady state, where the relaxation interactions show up as averaged in time. Moreover, intense radiation is required to excite the coherent regimes; so the transient response can be used to test the validity of the dynamic equations of the active centers in the strong-field limit.

The transient regime considered here is the nutational response, which monitors the time evolution of a system, initially at thermal equilibrium, toward a new stationary state under the action of a very intense resonant field.[1,2] If the Rabi frequency χ, which measures the atom-field coupling strength, is higher than the dephasing rate $(1/T_2)$, the system response is oscillatory. In this case, the active centers are coherently driven up and down the two levels and the resulting oscillation of the population difference modulates the macroscopic response of the system. The oscillations (Rabi oscillations or transient nutations) are damped by two different mechanisms: the spreading of the precession frequencies and the interaction of each center with its fluctuating environment, which ultimately drives the system onto its steady state.

Transient nutations are a rather general effect of the atom-field interaction and have been observed in a variety of physical systems and in several bands of the electromagnetic (e.m.) spectrum: in nuclear and electron-spin systems,[1,3] in optical transitions of molecular and atomic systems,[4–6] and in multiphoton resonances.[7–9] The detection of this regime is frequently used for measuring the atom-field coupling, for testing the homogeneity of the driving field over the sample size, for disentangling overlapping spectra,[10] and also for investigating the correlation of atomic radiative properties with atomic states.[11]

Despite its generality and widespread use, poor attention has been paid to the decay properties of the nutational regime. On the one hand, their experimental study is not easy. In fact, because of the simultaneous presence of the strong driving field, the detection of transient nutations is often featured by a rather low signal-to-noise ratio, unsuitable for a reliable study of the decay. Moreover, the intrinsic damping of the oscillations is frequently masked by spurious effects as, e.g., the inhomogeneity of the driving field over the sample volume. On the other hand, the analysis of the experiments involving nutations is currently carried out using either the free-atom approximation[11,12] or the phenomenological relaxation times T_1 and T_2 (Bloch model).[3,6,10,13,14] However, the adequacy of the Bloch model to account for the system dynamics during the nutational regime is questionable. In fact, this regime occurs under the action of a strong field and in a time scale much shorter than T_2 and both conditions violate basic assumptions which the Bloch equations rely on.

We report here experimental results on the decay of the nutational regime in nearly ideal two-level electron-spin systems. By using nonlinear spectroscopy methods we can monitor the transient nutations with an accuracy high enough to study the decay properties in a relatively wide range of the driving-field intensity. We compare the experimental curves with the theoretical ones calculated for an ideal inhomogeneous system of free spins and by this procedure we show that the irreversible contribution to the damping is well described by a single exponential function. This behavior reproduces the predictions of a Bloch model, but only qualitatively. In fact, the decay rate appears to be faster than expected and to depend on the driving-field intensity. In particular, we find that the damping rate increases on increasing the input power level and this variation is well described by a surprisingly simple linear dependence of the decay rate on the Rabi frequency.

To our knowledge, experimental results on the decay properties of transient nutations have not been reported previously, except for a preliminary version of some of our results.[15] Anomalous decay of this regime was occasionally observed by Farrel, MacGillivray, and Standage,[6] but it was ascribed to propagating effects in the vapor and simultaneous pumping between hyperfine levels.

In Sec. II we summarize the properties of the nutational regime of an idealized system of noninteracting spins with inhomogeneous spreading of the resonance frequencies. Experiments and results are described in Sec. III. A theoretical interpretation of the results is admittedly outside the scope of this paper and, in Sec. IV, we limit ourselves to a discussion of the possible origins of the observed non-Bloch behavior and on its relationship with the decay properties of other coherent regimes.

II. THE NUTATIONAL REGIME

To outline the experimental conditions and make clear the analysis procedure of the results, in this section we briefly recall the main features of the nutational regime in an inhomogeneous spin system. We consider a system of N two-level centers ($S=\frac{1}{2}$) in a static field $\mathbf{B}=B_0\hat{\mathbf{z}}$. In view of the experimental conditions, we assume an inhomogeneously broadened resonance line with a Gaussian profile and a standard deviation σ:

$$g(\epsilon)=(2\pi)^{-1/2}\sigma^{-1}\exp(-\epsilon^2/2\sigma^2)\ ,$$

where $\epsilon=\omega_0'-\omega_0$ measures the spectral distance of the generic spin (ω_0') from the mean frequency ω_0. As a first step, we assume that the relaxation interactions can be disregarded in the time scale of interest ($t \ll T_1, T_2$) and we will refer to this idealization as inhomogeneous system of free spins.

The system, initially at thermal equilibrium, is excited by a transverse field, $\mathbf{b}=2b_1\cos(\omega t)\hat{\mathbf{x}}$, abruptly turned on at $t=0$, and tuned to the line center ($\omega=\omega_0$). In this case ϵ measures both the spectral position of the packet and its detuning from the radiation.

In the usual rotating reference frame (RRF) the equations of motion of the generic spin are

$$\begin{aligned} \frac{du(\epsilon,t)}{dt}&=-\epsilon v(\epsilon,t)\ ,\\ \frac{dv(\epsilon,t)}{dt}&=\epsilon u(\epsilon,t)-\chi w(\epsilon,t)\ ,\\ \frac{dw(\epsilon,t)}{dt}&=\chi v(\epsilon,t)\ , \end{aligned} \tag{1}$$

where u, v, and w are the components of the generic packet magnetization in the RRF, $\chi=\gamma b_1$ is the Rabi frequency (the precession frequency of the resonant packet), and γ is the absolute value of the gyromagnetic ratio.

Within the rotating-wave approximation, Eqs. (1) have the well-known solutions[1-3]

$$\begin{aligned} u(\epsilon,t)&=-w_0(\epsilon\chi/\beta^2)[\cos(\beta t)-1]\ ,\\ v(\epsilon,t)&=-w_0(\chi/\beta)\sin(\beta t)\ ,\\ w(\epsilon,t)&=w_0[(\epsilon/\beta)^2+(\chi/\beta)^2\cos(\beta t)]\ , \end{aligned} \tag{2}$$

where $\beta=(\epsilon^2+\chi^2)^{1/2}$ is the precession frequency of the packet located at ϵ, and w_0 is the thermal equilibrium value of the longitudinal magnetization w.

One obtains the response (U,V,W) of the whole system in RRF by integrating the single packet solutions [Eqs. (2)] over the distribution $g(\epsilon)$. Due to the resonant excitation of the symmetric line, we get $U=0$. As for $V(t)$, for $\chi \ll \sigma$ (high-inhomogeneity limit), the solution can be approximated[1,2,5] as

$$V(t)=-M_0\chi t \tag{3a}$$

at short times ($t \ll \sigma^{-1}$), and

$$V(t)=-(\pi/2)^{1/2}(\chi/\sigma)M_0J_0(\chi t) \tag{3b}$$

at long times ($t \gg \sigma^{-1}$). Here $M_0=Nw_0$ and $J_0(x)$ denotes the zeroth-order Bessel function.[16] So, in the laboratory reference frame (LRF) the system response consists of a transverse magnetization

$$M_{x,y}(t)=2\,\mathrm{Re}[M_{x,y}(\omega,t)\exp(-i\omega t)]$$

oscillating at frequency $\omega=\omega_0$, with complex amplitudes $M_x(\omega,t)=-M_y(\omega,t)=-iV(t)$ modulated by the Bessel function $J_0(\chi t)$.

Equation (3b) shows that, even in the limit of free spins ($T_2=\infty$), the nutational response is damped by the static spreading of the resonance frequencies. This is a reversible damping,[12] to be distinguished from the irreversible decay related to the dephasing interactions which we are concerned with hereafter.

In a real system, the relaxation interactions cause an additional irreversible damping. If the relaxation rate is slow with respect to χ, we can assume the transient nutations to be described by $V'(t)$:

$$V'(t)=K(t)V(t) , \quad (4)$$

where $V(t)$ is the free-spin response [Eq. (3)] and $K(t)$ is a decay function representing the effect of the relaxation. The experiments described below aim at determining $K(t)$ by comparing the experimental curves $V'(t)$ of the transient nutations with the free-spin behavior $V(t)$.

As a particular case, we consider now the simple Bloch model, where the relaxation is accounted for by two rates, $\Gamma_1=1/T_1$ (longitudinal) and $\Gamma_2=1/T_2$ (transverse). The equations of motion of the individual packet assume the form

$$\frac{du}{dt}=-\epsilon v-\Gamma_2 u ,$$
$$\frac{dv}{dt}=\epsilon u-\chi w-\Gamma_2 v , \quad (5)$$
$$\frac{dw}{dt}=\chi v-\Gamma_1(w-w_0) .$$

The general solutions of Eqs. (5) are known[1,2,17,18] and can be cast in the form

$$z(t)=[A_1\cos(\lambda t)+A_2\sin(\lambda t)]\exp(-\lambda_1 t)$$
$$+A_3\exp(-\lambda_2 t)+A_4 , \quad (6)$$

where $z(t)$ is any of the components u, v, and w and A_i $(i=1,4)$, λ, λ_1, and λ_2 are rather complicated functions of χ, ϵ, Γ_1, and Γ_2. The exact solution of the transient response $Z(t)$ of the whole system requires to integrate Eq. (6) over $g(\epsilon)$, which cannot be carried out analytically. As will be discussed later, we have calculated numerically $V(t)$ in a few particular cases. Here we limit ourselves to consider an approximate solution, valid under the following conditions: (i) center excitation; (ii) $\Gamma_1 \ll \Gamma_2$; (iii) $\chi \gg \Gamma_1$, Γ_2. The extent to which these conditions are fulfilled in our experiments will be examined later. In these limits the solution $V_B(t)$ is approximately given by[1,2,5,17,19]

$$V_B(t)=-(\pi/2)^{1/2}(\chi/\sigma)M_0 J_0(\chi t)\exp(-\Gamma_2 t/2) . \quad (7)$$

The comparison with Eqs. (3b) and (4) shows that the decay function predicted by the Bloch model is a single exponential function $K_B(t)=\exp(-\Gamma_B t)$ with a rate $\Gamma_B=\Gamma_2/2$. This result will be used in the next section for discussing the experimental results.

III. EXPERIMENTAL RESULTS

A. Method

In a standard experiment for observing transient nutations, the system is irradiated by a step-modulated resonant field and one monitors the transient response of the system by revealing its oscillating polarization, whose amplitude is modulated at the Rabi frequency χ. In this organization, the response signal and the driving field have the same frequency ω and, from an experimental point of view, problems arise for separating them from each other. Balance techniques can be used to this aim; however, as a result, the Rabi oscillations can be observed with a reasonable signal-to-noise ratio only in a rather limited dynamic range, which prevents a reliable investigation of their decay.

To overcome these constraints a different organization was used in the experiments described here, in which nutations are excited by means of two-photon (TP) transitions and are monitored by revealing the system magnetization oscillating at the second harmonic (SH) frequency of the field. The advantage is that the large spectral distance between the signal and the field background reduces greatly the problems related to the detection. This configuration has been previously used successfully for time- and frequency-domain investigations of other coherent regimes.[20,21]

In our experiments, the driving field $\mathbf{b}_1$ is polarized at an angle $\phi=45°$ with respect to the static field $\mathbf{B}$ and its frequency ω is tuned to the TP resonance $\omega=\omega_0/2$, where ω_0 is the mean spin frequency. So, the upper and lower levels of each spin center are connected by TP transitions. As shown previously,[8,22] in a properly defined double rotating reference frame the time evolution of the generic packet is governed by the same equations as Eqs. (1), provided that ϵ and χ are given the meaning of detuning from the TP resonance condition, $\epsilon=\omega_0-2\omega$, and TP-Rabi frequency: $\chi_{TP}=-\gamma^2 b_1^2/\omega_0$.

Therefore, all the considerations reported in the previous section remain valid also for the TP excitation of the transient regime, and in particular the free-spin and the Bloch-model solutions. In the LRF the transient regime shows up as a transverse magnetization oscillating at 2ω whose amplitude is modulated by $V(t)$:

$$M_{x,y}(t)=2\,\mathrm{Re}[M_{x,y}(2\omega,t)\exp(-i2\omega t)] ,$$

where

$$M_x(2\omega,t)=-M_y(2\omega,t)=-iV(t)$$

with $V(t)$ given by Eqs. (3) (free spins) or by Eqs. (7) (Bloch model). Actually, in the experimental apparatus the oscillating macroscopic dipole radiates power in a narrow spectral band centered at 2ω; by detecting the emitted field we finally measure a signal $S(t)\propto|V'(t)|$.

B. Experimental setup

The sample is located in a bimodal cavity, resonating at $\omega=2\pi$ 2.95 GHz in a partially coaxial model (pump mode) and at $\omega_2=2\pi$ 5.9 GHz in its TE_{102} rectangular mode (monitor mode). Both modes have relatively low quality factors ($Q\sim300$) to allow pulse operation. Fine tuning of the monitor mode to the harmonic condition $\omega_2=2\omega$ is achieved by inserting a quartz rod inside the cavity. The fields of the two modes are maxima and collinear at the sample position. The cavity is placed between the poles of an electromagnet that provides the static field B; care has been taken to ensure that the sample position falls in the middle of the region in which the spatial homogeneity of the static field is a maximum.

The output signal of a cw low-power (10 mW) generator tuned to ω is first pulse shaped by a fast modulator (transition time: 0.2 μs), driven by a pulse generator.

The duration t_p and the repetition frequency f_p of the pulses are so regulated that the obtained radiation can be considered as a single-shot step-modulated radiation: t_p, typically of 1 ms, is long enough to allow observing the whole transient regime and f_p, typically less than 1 Hz, is low enough to ensure complete thermal relaxation of the spin system between successive pulses. The microwave signal is then raised to the required power level (20 W) by a traveling-wave tube amplifier; finally, after filtering, it feeds the pump mode of the cavity. At the maximum available input power, the field amplitude at the sample site is 2.6 mT and induces a TP Rabi frequency $\chi=2\pi$ 0.3 MHz.

When the static field is adjusted to tune the spin system to the TP resonance ($\omega_0=2\omega$), the induced 2ω-oscillating magnetization of the sample radiates onto the monitor mode of the cavity. The microwave signal picked out from this mode is first amplified by a low-noise microwave amplifier (gain: 50 dB; bandwidth: 0.4 GHz; noise figure: 1.8 dB) and then revealed by a superheterodyne receiver. The intermediate frequency (IF) amplifier of the receiver has a bandwidth adjustable from 0.3 to 10 MHz. The receiver outputs a logarithmic video signal, calibrated with the accuracy of 0.2 dB, which is sent to a digital transient recorder (time resolution: 0.2 μs). Further improvement of the signal-to-noise ratio is required, especially at low input power levels, and is accomplished by averaging over a number of pulses, typically 512. The repetitive operation, the acquisition, the average and storage of the signal is controlled by a microcomputer system.

Due to the parasitic coupling between the two modes of the cavity, a small B-independent signal is usually present at the output of the cavity. In some measurements, especially at high power, its interference with the SH signal emitted by the sample causes distortion of the oscillation pattern and prevents the accurate measurement of the decay rate. To minimize these effects a bridge system is used at the output of the cavity, where the signal from the monitor mode is summed to a SH reference signal whose amplitude and phase are so regulated to reduce the field-independent signal. The stability of the bridge system is greatly improved by phase locking the cw low-power generator.

C. Samples

We report here the results obtained in three samples: samples no. 1 and no. 2 are quartz crystals containing $[AlO_4]^0$ centers with different concentrations, sample no. 3 is a glassy silica piece containing E' centers. Both kinds of centers are hole defects ($S=\frac{1}{2}$) and were created by room-temperature γ irradiation.

$[AlO_4]^0$ centers[23] are created nearby Al impurities in the quartz crystal. Their electron-spin resonance spectrum consists of six well-resolved lines separated by 0.6 mT due to the hyperfine interaction with the nuclear spin ($I=\frac{5}{2}$) of ^{27}Al nuclei. Each hyperfine line has an inhomogeneous width (peak-to-peak distance of the absorption derivative) of 0.018 mT, corresponding to $\sigma=2\pi$ 0.25 MHz. The measurements reported here were taken with the field **B** perpendicular to the **c** crystal axis: this orientation minimizes the overlapping with the lines of other defects; however, the field orientation plays no role in the problem addressed here. Unless otherwise specified, measurements refer to the highest field line.

E' centers[24] are created near an oxygen vacation. In a glassy matrix their resonance line is inhomogeneously broadened. In our sample we measured a width of 0.9 mT, corresponding to $\sigma=2\pi$ 1.25 MHz. Due to the high purity of the original piece of silica, no other absorption line was observed in this sample after γ irradiation.

Both kinds of sample were chosen for their relatively long relaxation times at low temperature ($T=4.2$ K). In Table I we list the values of T_1 and T_2 of our three samples, as measured by standard techniques (spin echo and saturation recovery) and in the same working conditions ($B=0.2$ T, $T=4.2$ K) as for nutation experiments. Spin concentrations indicated in Table I were estimated on the basis of the instantaneous diffusion[25] contribution to the echo decay.

D. Results

Typical experimental curves of the transient response to the stepwise excitation are shown in Fig. 1. These curves were taken in sample no. 1 at two different power levels: $\chi/2\pi=107$ kHz (a) and 21 kHz (b). In agreement with the expected behavior [Eqs. (3)], the curves exhibit a fast rise at the onset of the field followed by damped oscillations.

We note that the characteristic time τ_R of the initial fast rise is lower than the time resolution of our experimental setup (0.2 μs). This circumstance makes unreliable the evaluation of the amplitude of the first maximum, especially at high-power levels. Moreover, we recall that the signal $S(t)$ measured by our receiver (and plotted in Fig. 1) is proportional to the absolute value of the system response $V'(t)$, so that the distance between successive maxima in the curves of Fig. 1 is half a period:

TABLE I. Relevant quantities measured in our three samples. T_1 and T_2 are measured at $B=0.2$ T and $T=4.2$ K. Γ_B is the decay rate expected from the Bloch model. α and β are the best-fitting parameters of the dependence of Γ on χ.

Sample no.	c (10^{16} cm^{-3})	T_1 (ms)	T_2 (μs)	$\Gamma_B/2\pi$ (kHz)	$\alpha/2\pi$ (kHz)	β (10^{-2})
1	4.0±0.4	5.0±0.8	120±15	0.66±0.6	0.78±0.08	2.4±0.1
2	0.9±0.1	45±5	625±75	0.13±0.01	0.12±0.01	2.1±0.1
3	4.0±0.4	200±30	87±15	0.91±0.1	0.72±0.08	3.3±0.2

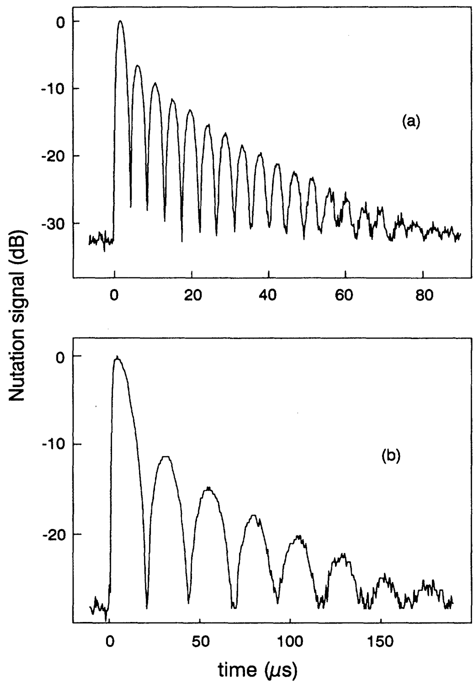

FIG. 1. Experimental curves of the nutational response $[S(t)\propto|V'(t)|]$ as measured in sample no. 1 at two different input power levels: $\chi=2\pi\times107$ kHz (a) and $\chi=2\pi$ 21 kHz (b). The driving field is switched on at $t=0$. Each curve is normalized to its maximum. The bandwidth of the receiver was (a) 10 MHz for the high-power curve and (b) 0.3 MHz for the low-power curve.

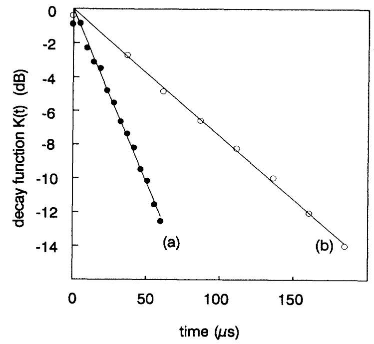

FIG. 2. Decay function $K(t)$. Points are the experimental values, as determined from the curves of Fig. 1 by points (at the maxima): $\chi=2\pi$ 107 kHz (●), $\chi=2\pi$ 21 kHz (○). The lines plot the single exponential functions $K(t)=\exp(-\Gamma t)$, the best fit the experimental points, excluding the initial ones (see text). We get $\Gamma=2\pi\times3.7$ kHz (a) and $\Gamma=2\pi\times1.37$ kHz (b).

π/χ. Finally, we point out that the bandwidth of the receiver is not fixed; rather, for each power level, it is regulated to maximize the signal-to-noise ratio without altering the oscillation pattern, namely, it is adjusted to a greater value when taking high-power curves, as (a), than for low-power curves, as (b). This is the reason why the absolute noise level appears to be different in the two curves of Fig. 1. In any case, the steady-state response falls below the noise level.

The number of observable maxima depends on the input power level, being nearly 20 at the maximum available power level ($\chi/2\pi=0.3$ MHz). For our investigation of the decay properties, we considered only those curves where the intensity of at least five maxima could be measured. This requirement fixed a lower limit for the explored range of χ: $\chi/2\pi=5$ kHz.

A preliminary set of measurements was devoted to determine the functional shape of the decay function. To this aim, we considered only the maxima of the signal: for each experimental curve, the values S_i $(i=1,N)$ of the maxima were compared to the extremes $|J_0|_i$ (absolute value of maxima and minima) of the zeroth-order Bessel function. The obtained values of $K_i=S_i/|J_0|_i$ $(i=1,N)$ were arranged in plots of K vs t, as in Fig. 2, which refers to the curves of Fig. 1. Data in Fig. 2 indicate that the decay function $K(t)$ is well described by a single exponential, at least for $t>\pi/\chi$. As discussed above, the departure of the first maximum from the straight line is possibly related to the limited time resolution of our receiver system. This behavior is typical in the sense that a single exponential decay was observed in all the examined cases. Departure from this behavior was observed only when large-size samples were used and was ascribed to the inhomogeneity of χ over the sample volume; this aspect will be discussed later.

We note that a single exponential form of $K(t)$ is consistent with the Bloch model [Eq. (7)]. However, the experimental rate is faster than expected, and it especially depends on the input power, at variance to the predictions of this model: in Fig. 2, we measure a decay rate $\Gamma/2\pi=3.7$ kHz for $\chi/2\pi=107$ kHz (a) and $\Gamma/2\pi=1.3$ kHz for $\chi/2\pi=21$ kHz (b), to be contrasted to the power-independent Bloch value

$$\Gamma_B=1/2T_2=2\pi(0.66\pm0.06)\text{ kHz} .$$

In a successive set of measurements a somewhat more refined analysis of the experimental curves was performed, in which an exponential $K(t)$ was assumed and all the points of the experimental curves (not only the maxima) were involved. In this method the function

$$S(t)=C\{[J_0(\chi t)\exp(-\Gamma t)+A]^2+B^2\}^{1/2} \qquad (8)$$

was used to fit the experimental curves. In this form of $S(t)$ the leading term is

$$C|J_0(\chi t)\exp(-\Gamma t)| ,$$

which is the signal emitted by the nutating spin system, as given by Eqs. (3b) and (4), for an exponential decay function $K(t)=\exp(-\Gamma t)$; the parameter C incorporates the conversion factor of the receiver. $S(t)$ in Eq. (8) also includes two minor terms, A and B, which represent the effect of the spurious signal originating from the parasitic coupling between the cavity modes. As discussed above, the major part of this small signal is balanced out by the bridge system and we get free of the residual part by including it in Eq. (8), as an unknown term: A is the amplitude of its component in phase with the nutational signal,

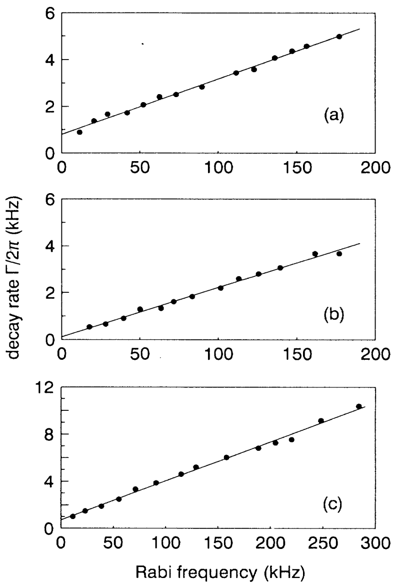

FIG. 3. Power dependence of the decay rate in sample no. 1 (a), no. 2 (b), and no. 3 (c). The input power is scaled as Rabi frequency. Both Γ and χ are expressed in frequency units. Dots are experimental points; full curves are the best-fit straight lines: $\Gamma=\alpha+\beta\chi$. The best-fit values of α and β for the three samples are listed in Table I.

B the out-of-phase one. For each experimental curve, taken at a given input power level, we determined the parameters A, B, C, χ, and Γ by a standard least-squares procedure. Note that, even if the Rabi frequency is an externally imposed condition, its value χ was determined by fitting to make the analysis independent of the unreliable evaluation of the microwave field strength at the sample position.

By this procedure, we obtained for each sample a set of experimental values of Γ at different input power levels. Data are reported in Fig. 3, where Γ is plotted versus χ, both in frequency units. As shown, we found that in our three samples and in the investigated range of χ (from 5 to 300 kHz), the decay rate of the nutational regime increases on increasing the driving-field intensity.

This dependence is well described as a simple linear variation of Γ with χ:

$$\Gamma=\alpha+\beta\chi \ . \tag{9}$$

The quantities α and β depend on the sample and their values, as obtained by the fitting, are listed in Table I. α has the physical meaning of the low-power extrapolation of the experimental decay rate. A remarkable aspect of our results is that in all the samples in this limit the decay rate of the nutational regime recovers the Bloch behavior $[\Gamma\rightarrow\Gamma_B=(2T_2)^{-1}$, for $\chi\rightarrow 0]$, within the experimental uncertainties. This is shown in Table I, where the values of the Bloch-model rate Γ_B are compared to the values of α determined experimentally.

A final remark concerns the crystalline samples (no. 1 and no. 2), where, as mentioned earlier, data reported

TABLE II. Values of the parameter β obtained as described in the text for the six hyperfine lines of sample no. 2 and in sample no. 3.

Line center (mT)	β
Sample no. 2	
206.0	0.025±0.002
206.7	0.025±0.002
207.4	0.027±0.003
208.0	0.029±0.003
208.6	0.027±0.003
209.2	0.021±0.002
Sample no. 3	
208.0	0.033±0.003

above refer to the highest-field line of the hyperfine sextuplet. In both samples, the other hyperfine lines have the same value of T_2 and their nutation decay rate exhibits a similar linear behavior as a function of χ, even if with a slightly different value of β. In Table II we list the experimental values of β, as obtained in the six hyperfine lines of sample no. 2; as shown, the significance of the variation of β is uncertain as it is on the limits of the experimental error.

IV. DISCUSSION

Before attempting to explain the above results, we wish to comment on the possibility that the observed power dependence of the decay rate originates in nonideal experimental conditions or unsuitable analysis.

First, we have not taken into account the damping of the magnetization caused by the emission of radiation. As known,[26] the radiation damping is characterized by a rate $\Gamma_R=(2\pi M_0\gamma Q\eta)$, where Q is the quality factor of the cavity and η is the filling factor ($\eta=V_{\text{sample}}/V_{\text{cavity}}$). In our conditions $Q\sim 300$, $\eta\sim 2\times 10^{-4}$, and γM_0 is $2\times 10^3\ \text{s}^{-1}$ in samples no. 1 and no. 3 and $5\times 10^2\ \text{s}^{-1}$ in sample no. 2. So, in our experiments $\Gamma_R/2\pi$ is 120 Hz in samples no. 1 and 3 and 30 Hz in sample no. 2. These values are overestimated, as we used the value of the whole magnetization for M_0 whereas only a fraction of the order of χ/σ, directly involved in the emission process, should be used. In any event, even the overestimated rates are much slower than the experimental ones, so that this effect can be safely discarded from our considerations.

Second, the residual inhomogeneity of the field over the sample may induce spreading of χ and damp the oscillations in a χ-dependent way. To explore the effect of the spatial inhomogeneity of χ, measurements were carried out in a series of samples of different sizes, all obtained from the same original piece of irradiated silica as the sample no. 3. We found that in large samples the inhomogeneity of b_1 does speed up the damping of the oscillatory transient in a χ-dependent way; however, the first evidence of the inhomogeneity of χ is a departure of the decay function from the single exponential law. In smaller and smaller samples, $K(t)$ progressively recovers its linear shape in the log-lin plot of Fig. 2 and this condition can be taken in turn as evidence that the spreading of χ no longer affects the decay. All the samples used for the measurements described in the previous section have sizes that fall well inside these limits.

Finally, our analysis of the decay curves is based on the high-inhomogeneity approximation: $\chi \ll \sigma$. In our experimental conditions, the ratio χ/σ ranges from 0.02 to 0.8 in samples no. 1 and no. 2, and from 0.004 to 0.16 in sample no. 3. One may wonder if the only partial fulfillment of the high inhomogeneity condition may endanger the determination of $K(t)$. To explore this point, we have numerically evaluated the transient response of the system, by using the exact analytical solutions [Eq. (6)] of the Bloch equations[17,18] integrated over the distribution $g(\epsilon)$ for various values of the ratio χ/σ. We found that the determination of $K(t)$ is unaffected by the value of χ/σ up to $\chi/\sigma \sim 2$.

The above results and considerations indicate unambiguously that the intrinsic decay rate of the nutational regime has two contributions, $\Gamma = \alpha + \beta\chi$, the former is consistent with the relaxation rate measured by the spin-echo decay, whereas the latter depends on the field amplitude and is not accounted for by a Bloch model. In this regard we recall that the inadequacy of the Bloch equations to account for the dynamics of dilute solid systems has been considered recently in connection with other transient regimes and, in particular, the free-induction decay (FID). In those experiments, both in optical resonances[27] and in spin systems,[20] the decay was found to be much slower than expected at high power. We remark that the situation is opposite in regard to the nutational regime, whose decay rate is found to be faster than expected and to increase on increasing the field strength. A remarkable conceptual difference between these two regimes is that the FID emission manifests the properties of the saturated state reached by the system during the preparative stage, whereas the nutational regime is a true coherent regime. Theoretical treatments of the models proposed to explain the anomalous FID[28] can therefore make use of time averages for calculating the steady-state properties and this circumstance prevents their application to the nutational regime.

We have not yet found a satisfactory explanation of the observed power dependence of the decay rate and limit ourselves here to discussing a few possible mechanisms. A possible cause is the inhomogeneous spreading of the spin-field coupling constant γ, due to locally different microscopic environments. This may induce a spreading $\Delta\chi$ of the value of the Rabi frequency, $\Delta\chi/\chi = \Delta\gamma/\gamma$ ($=2\Delta\gamma/\gamma$, for TP excitation), which, in turn, may yield a contribution $\Gamma' = \beta'\chi$ to the decay rate, with $\beta' = \Delta\gamma/\gamma$. This mechanism yields a linear χ dependence of Γ, in agreement with the experiments, but problems arise on a quantitative ground. In fact, the γ spreading should also affect the relationship between the static field B and the resonance frequency ω_0, thus contributing to the line broadening. Even if one assumes that the whole inhomogeneous width σ originates from this γ spreading, one gets an upper limit $\Delta\gamma/\gamma$ of 4×10^{-5} for samples no. 1 and no. 2, and 2×10^{-4} for sample no. 3, which are two orders of magnitude less than the measured values of β.

Therefore, we are led to consider the anomalous decay as an homogeneous effect, in the sense that it should be related to a power-dependent dephasing rate of the individual spin. To account for the observed χ dependence of Γ we need to hypothesize an effective width of the homogeneous packet line $\sigma_h = (1/T_2) + 2\beta\chi$. Now we examine the possible origins of this homogeneous power-dependent extra broadening.

(i) *Structural-two-level systems.* As is known, at low temperature, the microwave field interacts strongly and in a short time scale with the structural two-level systems (TLS's), which are peculiar of amorphous materials, and, in particular, it can affect their population distribution.[29] On the other hand, the TLS's are known to play a role in the dephasing dynamics of spin systems.[30] So, one is tempted to relate the observed anomalous decay of the nutational regime to the direct interaction of the microwave field with the TLS's and to their dephasing action on the spin precession. However, this mechanism should work only in the amorphous sample no. 3, and it should be much less effective or completely absent in the crystalline samples no. 1 and no. 2. On the contrary, the measured values of β are of the same order in all three samples (Table I).

(ii) *Nuclear spin flipping.* The rapid precession of the electron spins induces flipping of the neighboring nuclear spins, which are, in turn, sources of dephasing for the electron spins. This kind of local-field feedback is known to affect the saturation dynamics of systems of electron spins surrounded by nuclear spins and in particular it causes spectral diffusion.[31] Should this mechanism be responsible for the observed χ dependence of Γ, one should expect the constant β to depend on the value of the nuclear spin $\langle I_z \rangle$ and then on the particular hyperfine line considered. On the contrary, in samples no. 1 and no. 2 we measured that β varies very little from line to line, near the limits of the experimental uncertainties (Table II). Moreover, this mechanism should be less effective in sample no. 3, where hyperfine interactions play only a minor role.[24]

(iii) *Enhanced dipolar interaction.* In the systems considered here the spin centers have a random spatial distribution not correlated to their spectral position within the resonance line. The nearest neighbors of each spin have different resonance frequencies and detuning from the radiation. During the nutational regime, each spin will be acted on by the local dipolar field created by the neighboring spins, which precess each one at a different frequency. Due to the broad spectral composition, the superposition of these fields at the spin site loses its coherent features and has the same effect as a random field with a short correlation time. The resulting local field, whose amplitude and correlation time are expected to depend on χ, may yield a χ-dependent contribution to the coherence loss rate. In this interpretation the effect of the field is not only to induce the precession of the spins but also to change the nearly static dipolar interaction in a time-dependent one. A similar mechanism is effective in the echo decay kinetics and its effect, known as instantaneous diffusion (ID),[25] is to add a term to the damping rate. The tentative interpretation is partially supported by the effectiveness of ID mechanisms in our three samples, as proven by the experimental study of their echo decay kinetics.[21] On the other hand, it is challenged by two circumstances. First, according to this mechanism of field-induced enhancement of the dipolar interaction, the parameter β should depend on the spin concentration, whereas, as shown in Table I, the experimental values of β in our samples are not correlated to their concentrations. Second, we note that the mechanism considered here enhances the dipolar interaction among the spins falling within a narrow band $(-\chi, +\chi)$ around the line center. This kind of band-limited fast spectral diffusion has been theoretically considered in connection with the saturation of inhomogeneous lines in Ref. 32, where it has been calculated that it contributes a term proportional to χ^2 to the homogeneous width. So, if this theory can be extended to cover the transient case, it will yield a damping rate proportional to χ^2 rather than to χ.

In conclusion, we have reported experimental results on the anomalous decay of the transient nutation, which seems to escape a satisfactory explanation. We have tentatively considered a few mechanisms trying to relate the power dependence of Γ to a field-induced variation of the interaction of a spin center with its fluctuating environment, but none of them seems to account for the experimental behavior. So, the interpretation of the observed effects remains an open problem. A theoretical treatment has to take into proper account that this regime occurs in a time scale shorter than the relaxation time T_2 and that the spin-field coupling is stronger than the dephasing interaction. Both peculiarities may invalidate the very concept of spin packet and require us to treat the relaxation interactions as acting on the coupled spin-field states. Moreover, in our opinion, deeper insight on this effect can be gained by further experimental work, especially the investigation of the decay properties in a true homogeneous system and in a different band of the e.m. spectrum, e.g., in optics.

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