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Experimental study of Rabi oscillations induced by a phase-jump field

R. Boscaino and R. N. Mantegna
Istituto di Fisica, Università degli Studi, Via Archirafi 36, I-90123 Palermo, Italy
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The nutational regime induced by a stochastic field with phase telegraph noise is experimentally investigated in a two-level system at microwave frequency. Regular oscillations are observed in the average transient response of the system with an effective Rabi frequency whose value depends both on the jump amplitude and on the mean dwell time of the phase noise. Motional-narrowing, weak-damping, and strong-damping regimes are observed on varying the dwell time and the jump amplitude. The experimental results are in fair agreement with theoretical predictions.

In the interaction of a two-level system with a resonant field the Rabi frequency Ω measures the rate at which transitions are coherently induced between the atomic levels. Apparent manifestations of the Rabi frequency show up in intense fields or during transient regimes and include a.c. Stark splitting, spectral sidebands of fluorescent light, and transient nutations.

The theory of Rabi oscillations has been known for many years under the simplifying assumption of a two-level atom interacting with a monochromatic radiation.[1] More recently, the importance of the field fluctuations has been recognized and much theoretical work has been devoted to the examination of the effect of the statistical properties of the radiation on the atomic response. If one or more parameters of the driving field are fluctuating, the instantaneous Rabi frequency $\Omega(t)$ itself is a complex stochastic process; however, the average response of the system, namely its resonance fluorescence spectrum, or the dynamic splitting of its levels or, finally, the average nutational regime, may be characterized by an effective Rabi frequency Ω_R. In general, Ω_R is not given simply by the average of $\Omega(t)$; in fact, it depends on the interplay between the field statistics and the stochastic properties of the atomic system.

The effect of the field fluctuations on Ω_R has been studied theoretically for a variety of stochastic field models, e.g., the chaotic field,[2,3] the phase-diffusion field,[3-6] and several random-jump models.[7-11] It has been found that in the presence of field fluctuations the effective Rabi frequency Ω_R is different from the value Ω_0 induced by a monochromatic field carrying the same integral power. Moreover, fluctuations yield additional damping terms in the average evolution of the system. Apart from these general trends, the frequency and damping of the Rabi oscillations are affected by field fluctuations to an extent which depends on the particular stochastic model used to describe the field.

We have studied experimentally the effect of the field fluctuations on the average response of a two-level system by detecting its transient nutational regime. In this Rapid Communication, we report results on the effective Rabi frequency induced by a nonmonochromatic field whose phase includes a stochastic contribution $\phi(t)$: $\phi(t)$ switches between two allowed values, $-a$ and $+a$, at random times with a mean dwell time T. The effect of this kind of telegraph noise on the average response of the system has been examined theoretically by Eberly, Wodkiewicz, and Shore.[8] They have shown that the effective Rabi frequency Ω_R is practically the same as Ω_0 for $\Omega_0 T \gg 1$, namely at high-field intensity or for a long T; in this weak-damping regime the phase noise causes only additional dephasing. In the opposite limit, $\Omega_0 T \ll 1$ (motional-narrowing regime), Ω_R is expected to depend on the phase jump amplitude a as $\Omega_R = \Omega_0 |\cos(a)|$. Finally, within the intermediate range, oscillations disappear for $\Omega_0 T \approx 1$ (strong-damping regime). The experimental results we report here are consistent with the expected asymptotic behaviors and, moreover, they cover the intermediate range of $\Omega_0 T$. To our knowledge, measurements of the effective Rabi frequency in a nonmonochromatic field have not been reported previously.

Our experiments are carried out at microwave frequency in a $S = \frac{1}{2}$ electron-spin system. The system, initially at thermal equilibrium, is acted on by a pulse of microwave radiation whose phase is modulated by random telegraph noise (RTN) and we detect the transient response of the system ensuing the leading edge of the radiation pulse. By averaging over many pulses we observe regular oscillations at frequency Ω_R in the average transient response of the system. We report here results on the dependence of Ω_R on the parameters a and T of the RTN that modulates the field phase. A peculiarity of our experimental procedure is that the nutational regime of the spin system is induced by means of two-photon (TP) transitions and the system response that we consider is its second-harmonic (SH) emission.[12] In our experiments the two-level system is tuned to $\hbar\omega_0$ by the static field B_0, the microwave magnetic field has a mean frequency $\bar{\omega}=\frac{1}{2}\omega_0$ and we detect the radiation emitted by the system in a narrow spectral region centered at $2\bar{\omega}$. We point out that no resonant intermediate level is involved in the TP transitions considered here and a probe field is not required to detect the nonlinear response of the system. The experimental convenience of this procedure for investigating coherent transient regimes has been elucidated in previous papers[13,14] concerned with monochromatic excitation. Here we only remark that the Rabi frequency we are considering is that of the TP transition.

Our procedure for generating a microwave field with RTN phase is straightforward.[15] The output signal of a low-power cw microwave source is passed through an electronic phase shifter driven by a RTN voltage $v(t)$. The microwave source is a cavity-stabilized klystron tuned to $\bar{\omega}=2\pi\times 2.95$ GHz with a residual bandwidth of less than 1 kHz and the phase shifter with a nominal transition time of 20 nsec. To obtain RTN voltages computer-generated sequences $\{z_i, i=1, 30000\}$ are preliminarily stored in the user memory of a waveform generator. Here z is a two-value (0,1) variable changing its state at $i=i_n$, with $n=1, N_j$. The original sequences $\{z_i\}$ are so built as to yield an exponential distribution $p(t_d)$ of the dwell time $t_d=t_n-t_{n-1}$: $p(t_d)=(1/T)\exp(-t_d/T)$ with a mean value T. A set of eight sequences has been used for the measurements described below, with different values of T, ranging from $T=1.78$–100 μsec. On external command, the selected sequence is retrieved and repeatedly converted to an analog voltage $v(t)$, with a conversion time step Δt (minimum 40 nsec). $v(t)$ jumps back and forth between 0 V and an adjustable amplitude V_R at times $t_n=i_n\Delta t$ $(n=1, N_j)$. For a given T, by adjusting V_R, we can vary the phase jump a from 5° to 85° with the accuracy of $\pm 2°$. We have verified that the spectrum of the obtained radiation consists of a δ-like contribution superimposed to a Lorentzian one, as expected;[8,16] the phase-jump amplitude a controls the relative relevance of the two contributions, whereas T determines the width of the Lorentzian part.

The radiation obtained as described above is then raised to the required power level (≈ 10 W) by a traveling-wave tube amplifier, and, finally, it feeds the pump mode of a bimodal cavity, where the sample is located. At the sample position the microwave magnetic field

$$\mathbf{b}(t)=\mathbf{b}_1\exp\{-i[\bar{\omega}t+\phi(t)]\}+\text{c.c.},$$

with a RTN phase $\phi(t)$, is directed at $\alpha=45°$ with respect to the static field $\mathbf{B}_0$, to maximize the TP transition probability of a $S=\frac{1}{2}$ spin.[13] When $\mathbf{B}_0$ is so adjusted as to fulfill the TP resonance condition $(2\bar{\omega}=\omega_0)$ a second-order transverse magnetization $m_\perp^{(2)}(t)$ oscillating at the mean frequency $2\bar{\omega}$ builds up in the sample:

$$m_\perp^{(2)}(t)=2\,\text{Re}[m_{\perp 2}(t)\exp(-2i\bar{\omega}t)]\,.$$

Another mode (detection mode) of the cavity, finely tuned to $2\bar{\omega}$, collects the radiation emitted by the dipole $m_\perp^{(2)}(t)$. A microwave signal, picked out from the detection mode, is firstly amplified and then detected by a superetherodyne receiver, which outputs a video signal $S(t)\propto|m_{\perp 2}(t)|$. We refer to our previous report[13] for a more detailed description of the experimental apparatus.

Actually, in order to excite the transient regime, the driving field $b(t)$ is pulse shaped, with a pulse duration long enough (typically 1 msec) to allow the detection of the whole transient regime and with a repetition frequency low enough (≈ 1 Hz) to ensure complete thermal relaxation of the spin system between successive pulses. Owing to the stochastic nature of the driving field $b(t)$, the output pulse $S(t)$ is a noisy signal; however, when $S(t)$ is averaged over a number of pulses (typically 1024), regular oscillations can be observed in the resulting curve $\langle S(t)\rangle$, where Ω_R can be measured directly.

The measurements reported here were taken in a system of $[\text{AlO}_4]^0$ centers in quartz,[17] obtained by γ irradiating a synthetic single crystal in a ^{60}Co source at room temperature. $[\text{AlO}_4]^0$ centers have spin $S=\frac{1}{2}$. Owing to the hyperfine interaction with ^{27}Al nuclear spins, the ESR spectrum of these centers is split in six well-resolved lines, each one having an inhomogeneous width of nearly 0.15 mT. This system has been chosen for the present measurements owing to its relatively long relaxation times. In our sample, we measured $T_1=55$ msec and $T_2=0.65$ msec on the resonance line considered here and in our experimental conditions: $T=4.2$ K and $B_0=0.21$ T.

In Fig. 1 we report the experimental values of Ω_R measured at various jump amplitudes a of the field phase, for $\Omega_0 T=0.17$. At the input power level used for these measurements, the Rabi frequency Ω_0 in the absence of phase fluctuations was $\Omega_0=9.5\times 10^4$ rad sec^{-1}. When the phase was modulated by a RTN with a mean dwell time $T=1.78$ μsec, we found that on increasing a from 5° to 30° the oscillations of $\langle S(t)\rangle$ occurred at a frequency Ω_R less than Ω_0 and more and more quickly damped. No oscillations could be observed in the window $30°\lesssim a\lesssim 60°$ which corresponds to an overdamping regime of the average response. Oscillations appeared again for $a\gtrsim 60°$ at a frequency Ω_R progressively increasing toward Ω_0. In Fig. 2 we report the experimental values of Ω_R (normalized to Ω_0) measured at various values of the mean dwell time T for a fixed (nominal) value of the phase-jump amplitude $a=20°$. As shown, experimental values of Ω_R/Ω_0 close to one are found for large values of $\Omega_0 T$ (weak-damping limit), whereas they tend to 0.78 ± 0.03 in the opposite motional-narrowing limit.

The response of a two-level system to a RTN phase field has been investigated theoretically by Eberly *et al.*[8] for the one-photon resonance of a system with $T_2=2T_1$ within the rotating-wave approximation. As shown there, the equation of motion of the average population difference $\langle n(t)\rangle$ has a finite algebraic solution for this kind

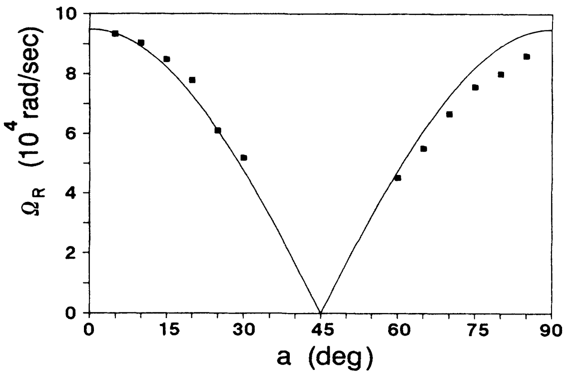

FIG. 1. Effective Rabi frequency Ω_R vs the phase-jump amplitude a. Experimental data were taken for $\Omega_0=9.5\times10^4$ rad sec^{-1} and $T=1.78$ μsec. The solid curve plots the theoretical dependence as calculated by solving Eq. (1) for $\Omega_0 T=0.17$, $T_1/T_2=85$, and $T_2/T=365$.

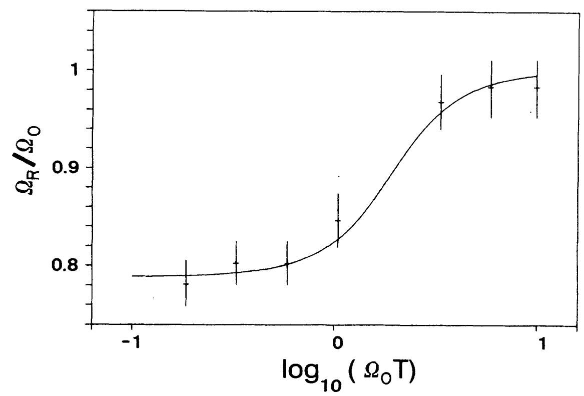

FIG. 2. Effective Rabi frequency Ω_R (normalized to Ω_0) vs $\Omega_0 T$. Measurements were taken for $\Omega_0=1.05\times10^5$ rad sec^{-1} and $a=20°$ (nominal). The solid curve is the theoretical dependence as obtained by solving Eq. (1) for $a=19°$ and $T_1/T_2=85$.

of stochastic field. The theoretical treatment by Eberly *et al.* can be extended in a straightforward way to our case of TP nutations; in fact, the time evolution of the two-level system near the TP resonance can be described by a set of Bloch-like equations provided that the Rabi frequency Ω and the detuning Δ from the resonance are properly redefined.[18] In particular, for our case of a $S=\frac{1}{2}$ spin system, the appropriate expression of the Rabi frequency Ω_0 is $\Omega_0=\gamma^2 b_1^2/\omega_0$, in the absence of field fluctuations. By the same procedure as in Ref. 8, we get the following equation for the average population difference $\langle n(t)\rangle$ for the case of exact TP resonance ($\Delta=0$) induced by a RTN phase field:

$$\langle \dddot{n}\rangle+(\Gamma_1+2\Gamma_2+2\Gamma)\langle\ddot{n}\rangle+[\Omega_0^2+2\Gamma_1\Gamma_2+2(\Gamma_1+\Gamma_2)\Gamma+\Gamma_2^2]\langle\dot{n}\rangle + \{\Omega_0^2\Gamma_2+2\Omega_0^2\Gamma[\cos(2a)]^2+\Gamma_1\Gamma_2(2\Gamma+\Gamma_2)\}\langle n\rangle+\Gamma_1\Gamma_2(2\Gamma+\Gamma_2)=0 \quad (1)$$

with $\Gamma=1/T$, $\Gamma_1=1/T_1$, $\Gamma_2=1/T_2$. Note that Eq. (1) reduces to Eq. (3.4) of Ref. 8 by posing $\Gamma_1=2\Gamma_2=A$ and replacing $2a$ with a. The latter substitution is the only consequence of exciting the nutational regime by means of TP transitions.

Equation (1) can be solved exactly by standard procedure in terms of the roots of a cubic polynomial to get the effective Rabi frequency Ω_R. In particular, it can be easily shown that Ω_R tends to Ω_0 and to $\Omega_0|\cos(2a)|$ in the limits $\Omega_0 T\gg1$ and $\Omega_0 T\ll1$, respectively. The full curves reported in Figs. 1 and 2 plot the theoretical dependence of Ω_R on a and T, as calculated from Eq. (1) by inserting the appropriate values of Γ, Γ_1, Γ_2, and Ω_0. In Fig. 1 the theoretical curve is practically indistinguishable from the asymptotic expression $\Omega_R=\Omega_0|\cos(2a)|$, as expected for $\Omega_0 T=0.17$. In Fig. 2 best fitting of the experimental data is obtained for $a=19°$. The agreement with the experimental data is quite good in both figures, with the only exception of the data at $a\gtrsim70°$ in Fig. 1, which appear to be less than expected. In our opinion, this departure has instrumental origins. In fact, as a approaches 90°, the δ-like contribution in the input power spectrum is reduced and most of the input power is carried by the Lorentzian part;[8,16] in these conditions, the filter action of the cavity pumpmode may cut a part of the integral power. Moreover, it is possible that for large values of a, spurious amplitude modulation of the input field may occur due to the variation of the insertion loss of the phase shifter when working at full scale. Both effects tend to reduce the integral power acting on the sample, thus lessening the effective Rabi frequency.

A final remark concerns the detected signal $\langle S(t)\rangle$. We have been concerned here with the frequency of the oscillations appearing in $\langle S(t)\rangle$. However, we have observed that $\langle S(t)\rangle$ contains a time-dependent nonoscillating component as well, which depends in a complicated way on a, T, and Ω_0. It is worth noting that $\langle S(t)\rangle$ involves the average of a nonlinear function, $|m_{\perp}^2(t)|$, of the density matrix elements. So, the theoretical consideration of its detailed time dependence cannot be carried out merely on the basis of Eq. (1), which indeed regards the average population difference. Rather, numerical simulation is required to account for the nonoscillating component of $\langle S(t)\rangle$. Work in this direction is in progress.

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