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Spin Echo at the Rabi Frequency in Solids

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We report on the observation of spin echo signals at the generalized Rabi frequency of a strongly driven two level system. The results are obtained for a radio-frequency transition of the nitrogen-vacancy center in diamond. A physical picture of the experiment is given in terms of the dressed-state basis, and the echo decay results are analyzed semiclassically and found to be in excellent agreement with our transient solution to the generalized Bloch equations. [S0031-9007(96)02144-8]

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A coherently driven two level system (TLS) may be conveniently discussed in the coupled “field + matter” dressed-state basis [1]. The energy levels of the dressed state system form a ladder of doublets that are parameterized by n , the number of photons in the field. Within each doublet the energy spacing is $\hbar\Omega$ ($\Omega \equiv$ generalized Rabi frequency), and each doublet is separated from its neighboring pairs by the energy associated with a single photon in the field. To date, most spectroscopic studies of a driven TLS have been concerned with either the transitions between different doublets in the ladder or those to a third uncoupled level. However, when a TLS has nonzero diagonal dipole moment matrix elements, the transition between the levels of a single dressed-state doublet, i.e., at the generalized Rabi frequency of the driving field, is allowed [2]. Recently, this transition has been observed by driving an optical transition in a ⁸⁷Rb atomic beam [3]. As expected, its frequency lies in the radio-frequency (rf) spectral region determined by the achievable driving intensity. In contrast with this optical experiment, the present measurements were undertaken by strongly driving an electron spin resonance transition such that both the driven transition and the Rabi transition lie in the rf spectral region. In this Letter, we report the first observation of the Rabi transition in a solid and, more importantly, the first report of a coherent transient measurement, namely, the two-pulse echo [4] (referred to as a *spin* echo by adopting the normal convention), in such a transition. Spin echo experimental results at the generalized Rabi frequency are presented and analyzed in terms of the relaxation coefficients associated with the undriven system.

A brief overview of the dressed-state formalism is presented first. Consider the energy level scheme of Fig. 1(a), where a TLS with resonance frequency ω_0 is driven by a pump field of frequency ω_1 , detuning $\delta = \omega_0 - \omega_1$, and Rabi frequency χ . Let $|g\rangle$ and $|e\rangle$ represent the ground and excited states, respectively, of the TLS which are eigenstates of the Hamiltonian \mathcal{H}_{TLS} . In the remainder of this Letter these will be referred to as the “bare” states of the system. The Hamiltonian of the

combined system is $\mathcal{H} = \mathcal{H}_{\text{TLS}} + \mathcal{H}_F + \mathcal{H}_I$, where \mathcal{H}_F is the Hamiltonian for the field and \mathcal{H}_I is the TLS-field interaction Hamiltonian. Using the rotating wave approximation, the eigenstates of \mathcal{H} are given by

$$|\bar{e}, \bar{n} - 1\rangle = \cos \theta |e, n - 1\rangle + \sin \theta |g, n\rangle,$$

$$|\bar{g}, \bar{n}\rangle = \cos \theta |g, n\rangle - \sin \theta |e, n - 1\rangle, \quad (1)$$

where $\cos \theta = \frac{1}{\sqrt{2}}\sqrt{1 + \delta/\Omega}$, $\sin \theta = \frac{1}{\sqrt{2}}\sqrt{1 - \delta/\Omega}$, and the overscore implies the dressed-state representation. These eigenstates form the ladder of doublets separated by the pump field frequency, ω_1 . The frequency separation between the states $|\bar{g}, \bar{n}\rangle$ and $|\bar{e}, \bar{n} - 1\rangle$ is $\Omega = \sqrt{\delta^2 + \chi^2}$ [see Fig. 1(b)].

The TLS studied here consists of the $m_s = 0$ and $m_s = -1$ spin substates of the ³A orbital ground state of the nitrogen-vacancy (N-V) center in diamond [5]. In this case, the prerequisite for a net transition moment at Ω is that the diagonal matrix elements, $\langle g, n | \hat{\mu} \cdot \hat{x} | g, n \rangle$ and $\langle e, n - 1 | \hat{\mu} \cdot \hat{x} | e, n - 1 \rangle$ where \hat{x} is the polarization direction of the field and $\hat{\mu}$ is the magnetic dipole moment, have different values. To achieve this situation, we mix “pure” electron spin states ($m_s = -1$ and $m_s = 0$) by a Zeeman-field-induced level anticrossing effect.

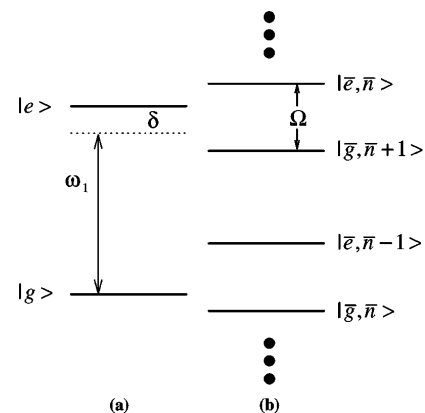


FIG. 1. Energy levels of the driven TLS in the (a) bare-state and (b) dressed-state representation.

Through this, $|g\rangle$ and $|e\rangle$ are admixtures of the $m_s = 0$ and $m_s = -1$ spin substates. In zero static field the $m_s = \pm 1$ states are degenerate and are separated in frequency by 2.88 GHz from the $m_s = 0$ state. In an externally applied field of $H_z = 1028$ G along the N-V axis ([111] direction), the $m_s = -1$ and $m_s = 0$ levels anticross due to a small misalignment of the center's axis from the static field direction. In the experiments reported here we worked with a field of approximately $H_z = 1000$ G, and the crystal was aligned to within $\pm 0.5^\circ$. Using these values in the spin Hamiltonian formalism appropriate for this center, the Rabi transition for the driven system has a strength of

$$\langle \bar{g}, \bar{n} | \mu_x | \bar{e}, \bar{n} - 1 \rangle = A \mu_{ge} \sin 2\theta = A \mu_{ge} \left(\frac{\chi}{\Omega} \right), \quad (2)$$

with the mixing parameter $A \approx 0.44$ which is strongly dependent on the exact values of misalignment and static field.

A typical weak-probe spectrum of our driven system for the case of $\omega_0/2\pi = 101.8$ MHz, $\omega_1/2\pi = 80.3$ MHz, and $\chi/2\pi = 7.6$ MHz is represented by the solid line in Fig. 2(a). The three-peaked structure of the transitions is due to the hyperfine interactions and all the above numbers are with respect to the central feature. It should be

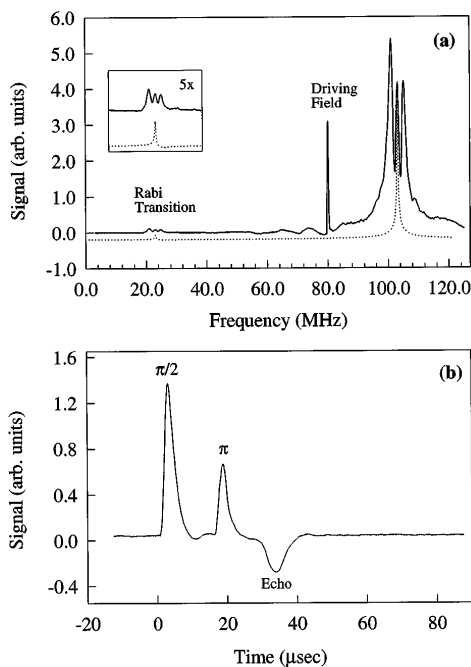


FIG. 2. (a) Solid line: weak-probe spectrum for the driven system proportional to the amplitude of the Raman heterodyne signal. The main feature at ~ 103 MHz is the "light-shifted" transition while the weaker at ~ 23 MHz is the transition at the generalized Rabi frequency, Ω . The spike at ~ 80 MHz represents the frequency of the driving field. Dotted line: results of our numerical calculation based on the SBE. The homogeneous linewidth was artificially increased in the calculation for clarity. (b) Phase-sensitive spin echo at the generalized Rabi frequency. The pulse lengths were on the order of 10^{-7} s with a separation of $16 \mu\text{s}$.

noted that the experimental value for χ is calculated using the measured values of Ω and δ , and includes the first order Bloch-Seigert shift, $\chi^2/4\omega_0$, of the resonance. Figure 2(b) shows an illustrative example of the detected spin echo which was obtained by applying a $\pi/2 - \tau - \pi$ pulse sequence at the frequency Ω . It is important to note that both the pulses *and* the echo were at the generalized Rabi frequency [6]. Decay curves were constructed from the echo amplitudes obtained as a function of τ and fit to an exponentially decaying function, thus yielding the irreversible coherence relaxation rate constant Γ_n for the Rabi transition. We then determined Γ_n as a function of the θ in the experiment, i.e., by either changing the detuning or the amplitude of the rf field at ω_1 . The results are shown in Fig. 3. Clearly, the relaxation rate for the Rabi transition is found to have a functional dependence on θ . It should be noted that, for the spin echo data shown, the detuning did not exceed 10 MHz. Also, data could not be acquired for the range $32^\circ < \theta < 45^\circ$ because, with the small detunings needed to achieve these values, the hyperfine levels were not distinctly spaced in the Rabi transition. Consequently, an echo typical of a *single* hyperfine transition could not be realized. In the range $0^\circ < \theta < 10^\circ$, data could not be acquired because the transition strength at Ω is greatly reduced [see Eq. (2)]. The data was recorded using the Raman heterodyne detection technique, details of which have been presented elsewhere [7].

To our knowledge, the relaxation properties of the transition at the generalized Rabi frequency in solids have not been treated theoretically [8]. This is not surprising considering the lack of motivating experimental results as well as the computational difficulties arising from the inclusion of diagonal dipole matrix elements in the theory. In order

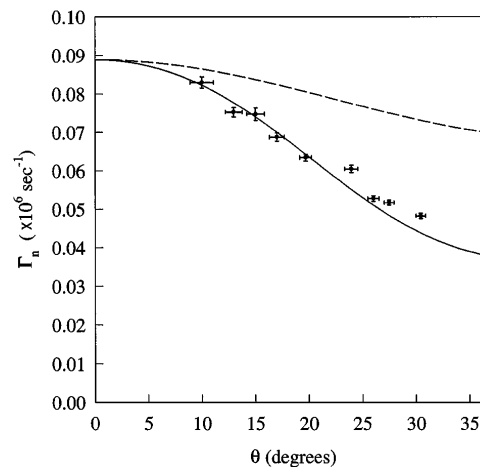


FIG. 3. Experimental values (circles with error bars) for the decay of the spin echo amplitudes in the Rabi transition as a function of θ . The solid line is a plot of Eq. (7) which was derived using the strongly driven GBE. The broken line is a plot of the linewidth dependence at Ω of our numerical calculations based on the SBE. The experimental bare-state values of $T_1 = 26 \pm 5 \mu\text{s}$ and $T_2 = 11 \pm 0.25 \mu\text{s}$ were used in the calculations.

to provide general insight into the analysis of our experiment, we have first performed numerical calculations based on a model in which the TLS has a nonzero dipole moment in the direction of a *classical* driving field and where the bare-state relaxation rates are treated phenomenologically as in the standard Bloch equations (SBE) [9]. A semi-classical approach is used because, although the dressed-state basis provides an excellent physical picture of the Rabi transition, the relaxation parameters have their simplest form in the bare-state basis. Within our model, we obtained, using Floquet's theorem [10], the steady state solutions to the density matrix equations of motion of the driven system probed by a weak field. The usual continued fraction solution [11] of the infinite set of coupled coefficient equations relating the various frequency components, however, could not be obtained because the dipole moment in the field direction lowers the symmetry of the equations. Keeping terms up to third order in the strong field and first order in the weak field, the response at the probe field frequency is obtained. An example calculation is shown as the dotted line in Fig. 2(a).

In the same manner, weak-probe absorption spectra were calculated using the experimental parameters associated with each of the data points in Fig. 3. The linewidths of the resonant responses at Ω and $\omega_1 \pm \Omega$ for each point were then compared, and found to be identical to within numerical error. Subsequently, in order to quantify their effect on the linewidths, the diagonal dipole moments were set to zero and the calculations repeated. There was found to be no measurable difference of the linewidths at $\omega_1 \pm \Omega$ compared to the calculations in which the diagonal matrix elements were nonzero. The functional dependence of the linewidths on θ using this model is represented by the broken line in Fig. 3.

It is clear from the figure that, although the above model correctly predicts the position of the resonant response at Ω , it provides a poor description of the observed homogeneous linewidth. Furthermore, the result that the linewidths are narrower than predicted theoretically is reminiscent of the breakdown of the SBE for systems under the influence of high power fields, first demonstrated in magnetic resonance by Redfield in 1955 [12] and at optical frequencies by Devoe and Brewer in 1983 [13]. These observations prompted a number of authors to separately develop the so-called generalized Bloch equations (GBE) [14]. For these equations, instead of using the low power phenomenological relaxation rates as in the SBE, the TLS is considered to be coupled to an unobserved reservoir [15] such that the total Hamiltonian of the system gains a term of the form [16]

$$\mathcal{H}_{sr} = \varepsilon\{\Lambda \otimes S_+ + \Lambda^\dagger \otimes S_- + \Delta \otimes S_z\}, \quad (3)$$

where ε is a dimensionless parameter related to the interaction strength, $S_+ = |e\rangle\langle g|$ and $S_- = |g\rangle\langle e|$ are the raising and lowering operator for the TLS, $S_z = \frac{1}{2}(|e\rangle\langle e| - |g\rangle\langle g|)$, and Λ and Δ are the off-diagonal and diagonal reservoir operators, respectively.

The reservoir operators of Eq. (3) can be treated either stochastically or quantum mechanically to arrive at the relaxation parameters of the TLS. Consider, for a moment, the field-free case. For weak TLS-reservoir coupling ($\varepsilon \ll 1$) [17], Eq. (3) can be treated as a perturbation. To second order in ε the off-diagonal reservoir operators completely determine the population relaxation rate, $1/T_1$, of the TLS, whereas all three terms contribute to the coherence relaxation rate, $1/T_2$, such that

$$\frac{1}{T_2} = \frac{1}{2T_1} + \frac{1}{T_2'}, \quad (4)$$

where $1/T_2'$ is fully determined by Δ [18]. These are the standard relaxation parameters which change considerably and become field dependent for the strongly driven system.

Geva, Kosloff, and Skinner [16] have recently presented a detailed derivation of the GBE in the case of a TLS weakly coupled to a reservoir and subject to a monochromatic field of arbitrary intensity. In the limit that $1/\tau_c^\Delta \ll \Omega \ll \omega_0$, where τ_c^Δ is the correlation time of the reservoir operator Δ , the GBE have the form

$$\frac{d}{dt} \begin{pmatrix} \mathbf{S}_x \\ \mathbf{S}_y \\ \mathbf{S}_z \end{pmatrix} = \begin{pmatrix} -\Gamma & -\delta & \Gamma_{xz} \\ \delta & -\Gamma & -\chi \\ 0 & \chi & -\Gamma_z \end{pmatrix} \begin{pmatrix} \mathbf{S}_x \\ \mathbf{S}_y \\ \mathbf{S}_z \end{pmatrix} - \begin{pmatrix} 0 \\ 0 \\ \gamma_z \end{pmatrix}, \quad (5)$$

where

$$\begin{aligned} \Gamma &= \frac{1}{2T_1} + \cos^2(2\theta) \frac{1}{T_2'}, \\ \Gamma_z &= \frac{1}{T_1}, \\ \Gamma_{xz} &= \sin(2\theta) \cos(2\theta) \frac{1}{T_2'}, \\ \gamma_z &= -\frac{S_z^{eq}}{T_1}, \end{aligned}$$

and S_z^{eq} is the equilibrium value of S_z in the absence of the driving field. An analytical solution such as this is not possible if the TLS has nonzero diagonal dipole moment matrix elements. Therefore, consistent with the results of our numerical calculations with the SBE that showed no measurable change in the linewidth with such an inclusion, we will use Eq. (5) in our analysis.

Analogous to spin echo experiments in an undriven system, the $\pi/2 - \tau - \pi$ pulse sequence at Ω produces an echo after a time 2τ . However, during the time in which the pulses are off, the system is still strongly driven by the cw field. Therefore, in order to deduce the decay rate of the echo signals, the strongly driven GBE must be solved in the transient regime. In 1949, Torrey [19] presented the transient solution to the driven SBE using the Laplace transform method for solving coupled differential equations. The well known result of his work

was that the decay of the transient nutation signal of the strongly driven TLS is given by

$$\frac{1}{T_2} - \frac{1}{2} \left(\frac{1}{T_2} - \frac{1}{T_1} \right) \sin^2(2\theta). \quad (6)$$

Alternatively, the SBE may be solved by transforming to the semiclassical dressed-state basis [20] in which, ignoring nonsecular contributions, the same functional dependence is obtained for the linewidths of the resulting nonstationary states. As expected, Eq. (6) accurately models the θ dependence of the linewidths we obtained with the numerical calculations based on the SBE discussed above and shown as the broken line in Fig. 3.

We have derived the transient solutions to the GBE of Eq. (5) in the limit $\chi \gg \Gamma, \Gamma_z$ using the Laplace transform method. The inclusion of the nonsecular term, Γ_{xz} , in the equations is the main difference between our derivation and that of Torrey. The decay of the dominant term of both S_x and S_y and, hence, the echo signal in our experiments is found to be given by

$$\Gamma_n = \Gamma - \frac{1}{2}(\Gamma - \Gamma_z) \sin^2(2\theta) + \frac{1}{4}\Gamma_{xz} \sin(4\theta). \quad (7)$$

This equation is plotted as the solid line in Fig. 3 and is seen to be in excellent agreement with the experimental results.

Using our analysis, it can be seen that the echo at Ω has the same decay rate as would be predicted for rotary echo experiments. Therefore, it represents an alternative method for determining the homogeneous decay rate of the strongly driven transition with the added advantage that the detection takes place at a frequency far from the strong driving field.

In summary, we have presented the first observation of a coherent transient measurement at the generalized Rabi frequency. We have used the spin echo technique to arrive at the transient decay rate, Γ_n , of the driven system as a function of the dressed-state coupling parameter, θ , and found it to be in excellent agreement with the theoretical predictions of the GBE in the strong field limit.

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