Dressed State Nutation and Dynamic Stark Switching

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Using Raman heterodyne detected nuclear magnetic transition in diamond we have observed dressed state nutation for two situations: (i) pulsed probe + cw pump and (ii) cw probe + pulsed pump. The dressed state Rabi frequencies are measured for the first time and found to have a $\sin\theta$ and $\cos\theta$ dependence (θ is a function of pump intensity and detuning). A simple consideration based on the dressed state model gives a satisfactory account of the observed dependence. When a 90° phase shift is introduced, spin locking and strong oscillations are observed.

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A two-level atomic transition driven by a strong near-resonant field is a fundamental problem in the study of matter-radiation interaction [1]. Pump-probe spectroscopy has been used extensively for such a study, where the effects of a driving field on the two-level atomic transition are examined by a probe field. In the steady-state limit, both weak and strong probe field situations have been studied. Many interesting observations such as Autler-Townes doublets, optical Stark splitting, and gain without inversion have been reported, and a comprehensive understanding has been obtained [2,3]. Recently, time-dependent probe response has also been a subject of great interest [4–6], but its scope is much more limited than the cw case due to both theoretical and experimental difficulties. However, the observations made so far in the transient regime have already shown a phenomenological richness not found in the steady-state limit. For example, it has been shown that the transient Autler-Townes peaks display interesting temporal oscillations and that the transient probe spectra differ significantly for different initial atomic conditions [6]. In most earlier transient behavior studies, the situations were restricted to the weak probe limit, where the probe intensity is kept sufficiently low so that the interaction is linear and needs only be treated to first order. In this Letter we report the transient probe response for the case where both pump and probe are strong. The observations can be conveniently explained as dressed state nutation. The Rabi frequencies associated with dressed state transitions are measured experimentally, to our knowledge for the first time.

Nutation occurs when a strong near-resonant field is suddenly applied to a two-level atomic transition. It manifests the dynamic process occurring in matter-radiation interaction and, hence, has been studied extensively [1]. This Letter reports the observation of a new type of nutation by extending the common configuration of a two-level system interacting with a single pulsed field to that of a three-level system interacting simultaneously with a cw and a pulsed field. The experimental configuration is shown in Fig. 1(a), where a probe field of ω_2 with Rabi frequency χ_{gc} drives the $|g\rangle$-$|c\rangle$ transition and a pump field ω_1 with Rabi frequency χ_{gb} drives the $|g\rangle$-$|b\rangle$ transition. The nutation is observed in the $|g\rangle$-$|c\rangle$ transition for two situations, (i) cw pump + pulsed probe and (ii) pulsed pump + cw probe. Oscillations superimposed on the nutation signal are also observed. In the bare-atom state picture the observed nutation is a complicated process involving the coherences ρ_{gb} and ρ_{gc} being driven by the pump and probe fields at the $|g\rangle$-$|b\rangle$ and $|g\rangle$-$|c\rangle$ transitions, respectively. However, the dressed state approach [7] provides a simple physical picture in which the $|g\rangle$-$|b\rangle$ transition and the pump field ω_1 are treated as a global "atom+field" system. The nutation occurs when the probe field is suddenly applied resonant with transitions between the dressed states (eigenstates of the atom+field system). The nutation frequency is proportional to the dressed state transition dipole strength. For the cw pump + pulsed probe case, the dressed states are in the steady-state limit, and the observed nutation is referred to as dressed state nutation. For the pulsed

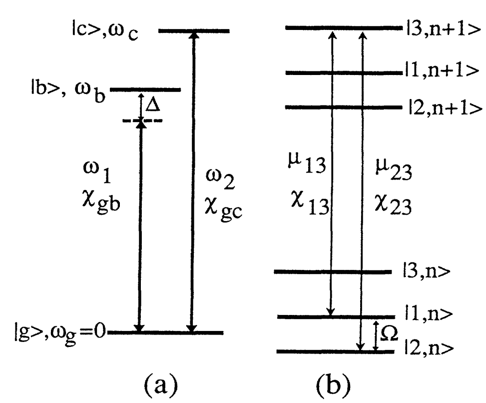

FIG. 1. Pump-probe spectroscopy in a three-level system for observing dressed state nutation and dynamic Stark switching. (a) In bare-atom state picture, the $|g\rangle$-$|b\rangle$ transition is driven by a pump field (frequency ω_1, Rabi intensity χ_{gb}, and detuning Δ), and the response of probe field (frequency ω_2, Rabi intensity χ_{gc}) at the $|g\rangle$-$|c\rangle$ transition is monitored. (b) In dressed state picture, nutation is observed at the $|3,n+1\rangle$-$|2,n\rangle$ and $|3,n+1\rangle$-$|1,n\rangle$ dressed state transitions.

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pump + cw probe field case, nutation occurs when the dressed state transition is brought into resonance with the cw probe field by the pump-field-induced dynamic Stark effect, thus, it is referred to as nutation by dynamic Stark switching in analogy to optical nutation experiments [8].

In the experimental configuration shown in Fig. 1(a), we consider the pump field ω_1 as the dressing field and the dressed states for the total three-level system + pump field are given by [7]

$$|1,n\rangle = \cos\theta|b,n\rangle + \sin\theta|g,n+1\rangle, \quad (1a)$$

$$|2,n\rangle = -\sin\theta|b,n\rangle + \cos\theta|g,n+1\rangle, \quad (1b)$$

$$|3,n\rangle = |c,n\rangle, \quad (1c)$$

where $\sin\theta = [(1-\Delta/\Omega)/2]^{1/2}$ and $\cos\theta = [(1+\Delta/\Omega)/2]^{1/2}$ are functions of the pump field Rabi frequency χ_{gb} and detuning Δ, $\Omega = [\Delta^2 + \chi_{gb}{}^2]^{1/2}$ is the generalized Rabi frequency. The energy level diagram of the resulting dressed states are shown in Fig. 1(b). The well-known Autler-Townes doublet [2,3] arises from the $|3,n+1\rangle$-$|1,n\rangle$ and $|3,n+1\rangle$-$|2,n\rangle$ dressed state transitions. The dressed state Rabi frequencies χ_{13} and χ_{23} are given by [7]

$$\chi_{13} = B_{\text{probe}}\mu_{13}/\hbar = B_{\text{probe}}\mu_{gc}\sin\theta/\hbar = \chi_{gc}\sin\theta, \quad (2a)$$

$$\chi_{23} = B_{\text{probe}}\mu_{23}/\hbar = B_{\text{probe}}\mu_{gc}\cos\theta/\hbar = \chi_{gc}\cos\theta, \quad (2b)$$

where B_{probe} is the magnetic field strength of the probe field. μ_{13} and μ_{23} are the dressed state transition dipole moments given by $\mu_{13} = \langle 3,n+1|\mu|1,n\rangle = \mu_{gc}\sin\theta$ and $\mu_{23} = \langle 3,n+1|\mu|2,n\rangle = \mu_{gc}\cos\theta$. Experimentally, χ_{13} and χ_{23} are determined from the nutation frequency. It is obvious from above discussions that when the Rabi intensities of the pump and probe fields χ_{gb} and χ_{gc} are held fixed, the dependence of the ratios of χ_{13}/χ_{gc} and χ_{23}/χ_{gc} on the pump detuning Δ is $\sin\theta$ and $\cos\theta$, respectively, and with a resonant pump field ($\Delta = 0$, $\theta = \pi/4$) the ratios are $\sin\theta = \cos\theta = 0.707$.

The three-level system used in our experiment arises from nuclear magnetic transitions within the ground state hyperfine levels of the nitrogen-vanancy (N-V) center in diamond. The magnetic transitions are detected by a sensitive optical-radio-frequency double resonance method: the Raman heterodyne technique [9]. The details of the NMR three-level system and the experimental arrangement have been given previously [3]. Briefly, the N-V center ground state is an electronic spin triplet, and the hyperfine splitting arises from the nitrogen nuclear spin of $I = 1$. The specific transitions involved are the hyperfine levels associated with the $S_Z = 0$ electronic spin level subjected to an axial static magnetic field of 1050 ± 10 G. The $I_Z = |0\rangle$-$|1\rangle$ transition [corresponding to the $|g\rangle$-$|b\rangle$ transition in Fig. 1(a)] at 4.7 MHz is driven by a pump field, and the nutation is observed in $I_Z = |0\rangle$-$|-1\rangle$ transition [corresponding to the $|g\rangle$-$|c\rangle$ transition in Fig. 1(a)] at 5.4 MHz which is coupled by a probe field.

Figure 2 shows the nutation signal observed using a pulsed probe field with [Fig. 2(b)] and without [Fig. 2(a)] a cw pump field. The cw pump field is applied resonant with the $|g\rangle$-$|b\rangle$ transition ($\omega_{gb} = 4.7$ MHz) and has a Rabi frequency $\chi_{gb} = 60$ kHz. In Fig. 2(a) the pulsed probe field is resonant with the $|g\rangle$-$|c\rangle$ transition ($\omega_{gc} = 5.4$ MHz), while in Fig. 2(b) it is resonant with the $|3,n+1\rangle$-$|1,n\rangle$ transition whose frequency is $\omega_{gc} - \chi_{gb}/2$. The two traces were taken using the same probe field intensity. Similar nutation is also observed when the pulsed probe field is resonant with the $|3,n+1\rangle$-$|2,n\rangle$ transition at $\omega_{gc} + \chi_{gb}/2$. The nutation shown in Fig. 2(a) is the normal nutation observed in bare-atom state with a Rabi nutation frequency of $\chi_{gc} = 9.17$ kHz, whereas the nutation shown in Fig. 2(b) is in the dressed state with a Rabi nutation frequency of $\chi_{13} = 6.37$ kHz (for the $|3,n+1\rangle$-$|2,n\rangle$ transition the observed Rabi nutation frequency is $\chi_{23} = 6.49$ kHz). The ratio between dressed state and bare-atom state Rabi nutation frequencies χ_{13}/χ_{gc} (χ_{23}/χ_{gc}) is 0.69 (0.71). This is in excellent agreement with the theoretical prediction of 0.707 for resonant pumping. There is also a temporal oscillation corresponding to the 60 kHz pump Rabi frequency superimposed on the nutation shown in Fig. 2(b), which is not resolved due to low time resolution. When the pump Rabi intensity is reduced to $\chi_{gb} = 30$ kHz, the observed dressed state nutation in the $|3,n+1\rangle$-$|2,n\rangle$ transition [see the inset in Fig. 2(b)] clearly reveals the

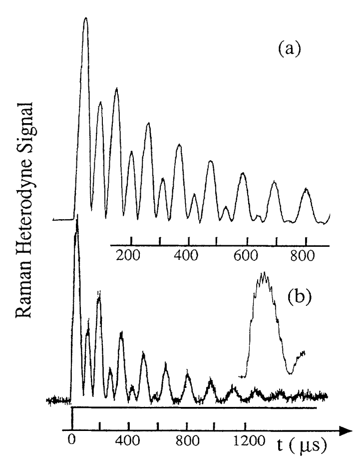

FIG. 2. (a) Bare-atom state nutation, the pulsed probe field is resonant with the $|g\rangle$-$|c\rangle$ transition. (b) Dressed state nutation, the cw pump field of Rabi frequency 60 kHz is resonant with the $|g\rangle$-$|b\rangle$ transition. The pulsed probe field is resonant with the $|3,n+1\rangle$-$|1,n\rangle$ transition. Similar nutation is also observed when the pulsed probe field is resonant with the $|3,n+1\rangle$-$|2,n\rangle$ transition. The inset shows the oscillation superimposed on the nutation when the pump Rabi frequency is reduced to 30 kHz, the oscillation frequency is measured to be 30.8 kHz.

temporal oscillation of approximately 30.8 kHz. This is in good agreement with the 30 kHz splitting measured by steady-state Autler-Townes spectrum in the frequency domain. The temporal oscillation can be viewed as arising from the quantum beats between the dressed state doublets [4–6].

Figure 3 shows the dressed state Rabi nutation frequencies χ_{13} and χ_{23} as a function of pump detuning Δ with fixed pump and probe intensities: $\chi_{gb} = 80$ kHz and $\chi_{gc} = 8.83$ kHz. The theoretical calculation of Eq. (2a) [(2b)] is plotted in Fig. 3 for comparison, where the only parameter used is $\chi_{gc} = 8.83$ kHz, determined from experiment. It is seen that the theoretical calculations are in excellent agreement with the experimental observations. It is well known that the dressed state transition dipole moments can be deduced from Eq. (2) [7]; however, our result provides, to our knowledge, its first experimental proof.

Figure 4 shows the nutation observed using a pulsed pump and a cw probe field. The pump field is resonant with the $|g\rangle$-$|b\rangle$ transition and the cw probe field resonant with the $|3, n+1\rangle$-$|1, n\rangle$ transition, whose position is determined from the steady-state Autler-Townes spectrum. Similar nutation is also observed when the $|3, n+1\rangle$-$|2, n\rangle$ transition is probed. There is also an oscillation superimposed on the nutation signal which corresponds to the Autler-Townes splitting. Nutation in the optical frequency has been observed using the Stark switching method [8]. In this technique a cw probe laser is used and a pulsed dc electrical field is applied by which the two-level atomic transition is suddenly brought into resonance with the cw laser field by the static Stark effect. In our experiments, the transition is suddenly brought into resonance with the cw probe field by the dynamic Stark effect using a pulsed pump field. In this sense we refer to our method as nutation by dynamic Stark switching. The observed temporal oscillation also have a close analog: Raman beats in the optical region [10], where again a pulsed dc electrical field is used to Stark split a degenerate level, and Raman beats occur at the frequency of the degenerate level Stark splitting due to the coherence prepared before the dc Stark pulse. There are, however, fundamental differences between these two methods and hence the observations. In the dc Stark switching method, the atomic levels are shifted by the static Stark effect, but the eigenstates are still the bare-atom eigenstates. Thus the Rabi nutation frequency corresponds to the bare-atom state Rabi frequency. On the other hand, in the dynamic Stark switching method the atomic states being shifted are no longer bare-atom states but the superposition of bare-atom states, thus the Rabi nutation frequency is no longer the bare-atom state Rabi frequency. The Raman beats observed in the optical region [10] rely on the coherence prepared during the period when the levels are degenerate, while in our case the coherence exists as an intrinsic property of the dressed state.

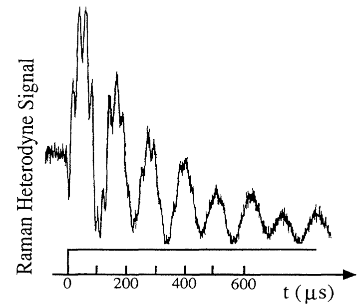

FIG. 4. Nutation by dynamic Stark switching. The cw probe field is resonant with the $|3, n+1\rangle$-$|1, n\rangle$ transition. Nutation is observed when the pump field of Rabi frequency 39 kHz resonant with the $|g\rangle$-$|b\rangle$ transition is switched on. Oscillation is also observed. Similar nutation is observed when the $|3, n+1\rangle$-$|2, n\rangle$ transition is probed.

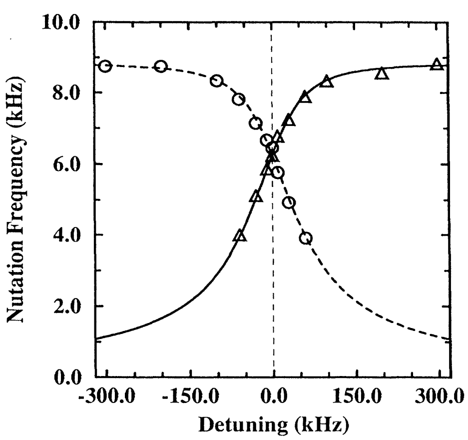

FIG. 3. Dressed state Rabi nutation frequencies χ_{13} (circle) and χ_{23} (triangle) as a function of the cw pump field detuning Δ. The cw pump field intensity is held constant with a resonant Rabi frequency of 80 kHz. The dashed (solid) trace is a theoretical plot of Eq. (2a) [(2b)] with $\chi_{gc} = 8.83$ kHz and $\chi_{gb} = 80$ kHz determined from experiment.

With constant phase pulsed fields the nutations observed are similar for the two situations. However, when a 90° phase shift of the pulsed field is introduced after the pulsed field has been switched on for $\pi/2$ period, the observed transient behavior differs dramatically depending on which field is pulsed (as shown in Fig. 5). For the pulsed probe field spin locking [11] is observed [Fig. 5(a)] for the $|3, n+1\rangle$-$|1, n\rangle$ transition. The nutation is prohibited by the locking field, and the signal exhibits exponential decay with a time constant of approximately 340 μsec. The oscillation superimposed on spin-locking signal has a frequency of 39 kHz, which is in good agreement with the measured Autler-Townes splitting of 39 kHz. Figure 5(b) shows the "anomalous" strong oscillations when the pump field is pulsed and the

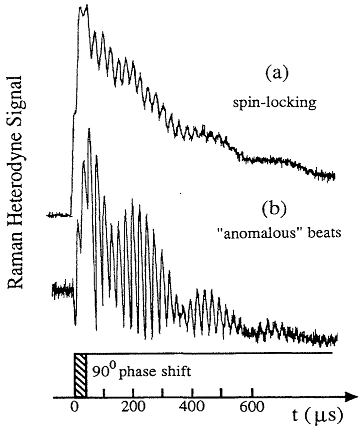

FIG. 5. Spin-locking (a) and "anomalous" strong oscillations (b). In (a) the probe field is pulsed, and its phase is shifted by 90° after a $\pi/2$ period with a cw pump field applied resonantly at the $|g\rangle$-$|b\rangle$ transition. In (b) the resonant pump field is pulsed, and its phase is shifted by 90° after a $\pi/2$ period with a cw probe field resonant with the $|3, n+1\rangle$-$|1, n\rangle$ transition.

cw probe field is resonant with the $|3, n+1\rangle$-$|1, n\rangle$ transition. The nutation is totally concealed by the strong oscillations which again correspond to the measured Autler-Townes splitting of 39 kHz. The effects of a 90° phase shift can be schematically illustrated using the vector model [1]. With a resonant, constant-phase, pulsed probe field, the Bloch vector is perpendicular to and continually precesses about the field vector which gives rise to the nutation signal observed. When a 90° phase shift is produced at $t = \pi/2$, the field vector is rotated so that it is parallel (or antiparallel) to the Bloch vector, and the precession of the Bloch vector is suppressed [6,11]. In the case of pulsed pump field, the effect of the 90° phase shift is to prepare the atomic system in different initial conditions [6]. Our results demonstrate that the transient process can be influenced strongly by the initial condition of the dressed state and that such a dependence for strong probe fields differs from that for weak probe reported in Ref. [6].

In summary, we report on the first experimental investigation of nutation in dressed state in a three-level system pump-probe configuration. The dressed state Rabi frequencies can be readily determined by nutation measurements, and the experimental results reported are in good agreement with dressed state theory. Spin locking in dressed state is also reported. The anomalous strong oscillation demonstrate that the transient behavior can be influenced dramatically by the initial conditions of the dressed state. Full understanding of this strong oscillation challenges further experimental and theoretical studies which are in progress.

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