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MULTIPHOTON TRANSIENT EPR NUTATIONS IN A DOUBLY RESONANT BICHROMATIC FIELD

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We demonstrate theoretically and experimentally that multiplication of spin qubits arises at double resonance in a bichromatic field when the frequency of the radio-frequency field is close to that of the in the microwave field, provided its frequency equals the Larmor frequency of the initial qubit. The effect is investigated beyond the rotating wave approximation using transient nutations in the pulse EPR of *E*′ centers in crystalline quartz. It is show that the operational multiphoton transitions of qubits dressed by the bichromatic field can be selected by the choice of both the rotating frame and the radio-frequency phase.

INTRODUCTION

It is known (Belas, 2006) that the resonant interaction between electromagnetic radiation and two-level quantum system (qubit) induces Rabi oscillations, which are the basis for quantum operations. The Rabi frequency ω_R is defined by the amplitude of the electromagnetic field and usually is much smaller than the energy difference ω_0 (in frequency units) between the qubit's states. The "dressing" of qubit by the electromagnetic field splits each level into two giving rise to two new qubits with energy difference ω_R. The spectrum of the multilevel "qubit + field" system consists of three lines at the frequencies ω_0 and $\omega_0 \pm \omega_R$. The second low-frequency electromagnetic field with the frequency close to the Rabi frequency ω_R could induce an additional Rabi oscillation on dressed states of new qubits. These qubits are attracting interest because their coherence time is longer than that of the initial qubit (Jeschke, 1999; Fedoruk, 2004; Fedaruk, 2006; Greenberg, 2007; Saiko & Fedoruk, 2008). The results of studies of qubits dressed by bichromatic radiation formed by fields with strongly different frequencies are important for a wide range of physical objects, including, among others, nuclear and electron spins, double-well quantum dots, flux and charge qubits in superconducting systems. In NMR (Redfield, 1955; Hatanaka, Sugiyama, & Tabuchi, 2003) and EPR (Jeschke, 1999; Fedoruk, 2004; Saiko & Fedoruk, 2008; Fedoruk, Saiko, Markevich & Poklonskaya, 2009) such investigations are used in the development of line-narrowing methods. In EPR above mentioned multiphoton transitions on dressed states are observed at double resonance when the frequency ω of one (transverse microwave) field is equal to the Larmor frequency ω_0 of the spin system and the frequency ω_{rf} of the other (longitudinal radio-frequency) field is close to the Rabi frequency in a microwave field.

Another multiphoton transitions are induced in a two-level spin system dressed by the strong rf field. In this case a bichromatic field excites not only the single-photon transitions at ω, but also a number of multiphoton transitions at frequencies $\omega + k\omega_{rf}$, where k is a integer referring to the number of of rf photons absorbed (positive k) or emitted (negative k) (Kälin, Gromov & Schweiger, 2004; Fedin, Kälin, Gromov & Schweiger, 2003). Transient nutation EPR spectroscopy was used to observe directly such multiphoton transitions and to measure the effective field (Rabi frequency) for such transitions (Saiko, Fedoruk & Markevich, 2006; Saiko, Fedoruk & Markevich, 2007; Fedoruk & Poklonskaya 2009). The effective field amplitude was examined in the case of a relatively strong microwave field, when the derivation of the effective Hamiltonian cannot be reduced to first-order perturbation theory in ω_R/ω_{rf} (Saiko *et al.*, 2007).

In this report, we study the dynamics of spin qubits at double resonance in a bichromatic field using transient EPR nutations of *E*′ centers in crystalline quartz. The peculiarities of Rabi oscillations are investigated in the rotating frame beyond the rotating wave approximation.

THEORETICAL BACKGROUND

Let an electron spin qubit be in three fields: a microwave (mw) one directed along the x axis of the laboratory frame, a radio-frequency (rf) one directed along the z axis, and a static magnetic one also directed along the z axis. The Hamiltonian of the qubit in these fields can be written as follows:

$$H = H_0 + H_\perp(t) + H_\parallel(t). \quad (1)$$

Here $H_0 = \omega_0 s^z$ is the Hamiltonian of the Zeeman energy of a spin in the static magnetic field B_0, where $\omega_0 = \gamma B_0$, and γ is the electron gyromagnetic ratio. Moreover, $H_\perp(t) = 2\omega_1 \cos(\omega t + \psi) s^x$ and $H_\parallel(t) = 2\omega_2 \cos(\omega_{rf} t + \psi) s^z$ are the Hamiltonians of the spin interaction with linearly polarized mw and rf fields, respectively. B_1 and B_2, ω and ω_{rf}, and φ and ψ denote the respective amplitudes, frequencies, and phases of the mw and rf fields. Finally, $\omega_1 = \gamma B_1$ and $\omega_2 = \gamma B_2$ stand for the Rabi frequencies, whereas $s^{x,y,z}$ are the components of the spin operator.

The mw phase $\varphi = 0$ and the counter-rotating component of the mw field is neglected. We also assume that the exact resonance condition is fulfilled $\omega_0 - \omega = 0$, and that ω_1, $\omega_{rf} >> \omega_2$.

We obtained the equation for the absorption signal observed in this case in the laboratory frame (LF) (Saiko & Fedaruk, 2010). It follows from this equation that the resonant interaction between the mw field and the qubit creates its dressed states and two new qubits with energy splitting equal to the Rabi frequency ω_1.

The rf field with the frequency ω_{rf}, which is close to the Rabi frequency ω_1 of the new qubits, "dresses" these qubits, giving rise to four qubits with the energy splitting ε. Allowed multiphoton transitions between states of these qubits afford nine spectral lines observed in the laboratory frame. Here $\varepsilon = \left[(\omega_1 - \omega_{rf} + \Delta_{BS})^2 + \omega_2^2\right]^{1/2}$ is the frequency of the Rabi oscillations between the spin states dressed simultaneously by the mw and rf field, and $\Delta_{BS} \approx \omega_2^2 / 4\omega_{rf}$ is the Bloch–Siegert-like frequency shift. This shift of the dressed-state frequency is due to effect of the counter-rotating (antiresonance) component of the rf field.

There is the possibility of selecting the observed transitions of four qubits by employing the rotating frame. In the singly rotating frame (SRF), which rotates with frequency ω around the z axis of the laboratory frame, the absorption signal can by written as (Saiko & Fedaruk, 2010)

$$\begin{aligned} \upsilon_{SRF}(t) = (1/8)[&2\sin^2\xi\left(\sin\omega_{rf} t + \sin(\omega_{rf} t + 2\psi)\right) + \\ &+ (1+\cos\xi)^2 \sin(\omega_{rf} + \varepsilon)t - \\ &- \left(1-\cos^2\xi\right)^2 \sin\left((\omega_{rf} + \varepsilon)t + 2\psi\right) + \\ &+ (1-\cos\xi)^2 \sin(\omega_{rf} - \varepsilon)t - \\ &- \left(1-\cos^2\xi\right)^2 \sin\left((\omega_{rf} - \varepsilon)t + 2\psi\right), \end{aligned} \quad (2)$$

where $\sin\xi = -\omega_2/\varepsilon$, $\cos\xi = (\omega_1 - \omega_{rf} + \Delta_{BS})/\varepsilon$.

For the random rf phase, the absorption signal has three comparable oscillating components with frequencies ω_1 and $\omega_1 \pm (\omega_2^2 + \Delta_{BS}^2)^{1/2}$. For the rf phase $\psi = 0$, the sidebands are smaller than those at the random rf phase by the factor $\Delta_{BS} / (\omega_2^2 + \Delta_{BS}^2)^{1/2}$. When we use $\psi = \pi/2$, the component with frequency ω_1 vanishes and the sidebands are comparable to those at the random rf phase.

Note that in the strong-field regime the counter-rotating component of the rf field gives rise to the Bloch–Siegert effect, and the high-frequency sideband is always more intensive than the low-frequency one.

Upon the rotating wave approximation ($\Delta_{BS} \to 0$), it follows from Eq. (2) that for $\psi = 0$ only the component with frequency ω_1 remains. At the same time, for both $\psi = \pi/2$ and the random rf phase, the intensities of the sidebands are equal. The equalization of sidebands can be used to indicate the validity of the rotating wave approximation. On the contrary, their asymmetry reveals the effect of the counter-rotating component of the rf field.

In the doubly rotating frame (DRF), in which the Hamiltonian is diagonalized to the form $H_{diag} = \varepsilon s^z$, the absorption signal can be written as follows:

$$\upsilon_{DRF}(t) = (\cos\xi \cos\psi \sin\varepsilon t - \sin\psi \cos\varepsilon t)/2. \quad (3)$$

According to Eq. (3), the absorption signal in the doubly rotating frame is caused by the transitions between spin states dressed simultaneously by the mw and rf fields. At the exact resonance ($\omega_1 = \omega_{rf}$), the signal for $\psi = 0$ is smaller than the signal for $\psi = \pi/2$ by the factor $\Delta_{BS} / (\omega_2^2 + \Delta_{BS}^2)^{1/2}$. If $\Delta_{BS} \to 0$, the signal for $\psi = 0$ disappears. In this case, for $\psi = \pi/2$, the absorption signal oscillates with the Rabi frequency ω_2.

RESULTS AND DISCUSSION

The predicted effects are confirmed by observations of the transient nutations (Rabi oscillations) in the pulse EPR of *E*′ centers in crystalline quartz. We used the experimental method and the sample described in (Saiko & Fedoruk, 2008). The experiments were carried out at room temperature. The duration, amplitude, and repetition period of the magnetic-field pulses were equal to 10 μs, 0.12 mT, and 1.25 ms, respectively. To improve the signal-to-noise ratio, the digital summation (up to 10^3) of the nutation signals obtained during each pulse was used.

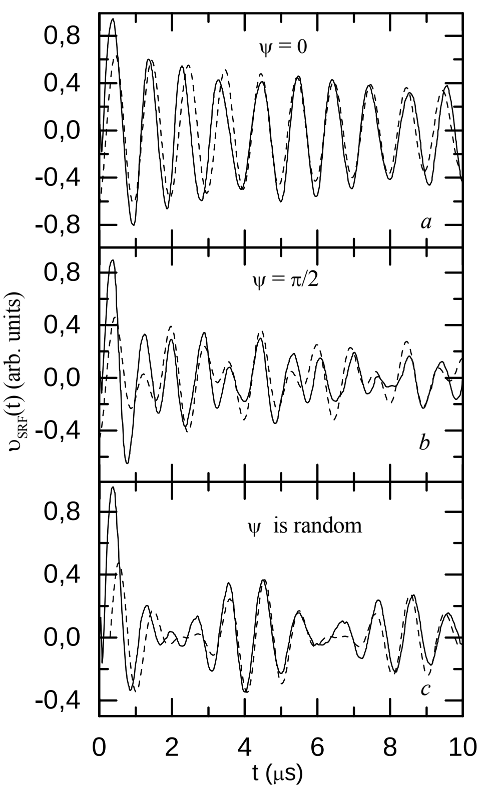

Fig. 1. Time evolution of the absorption EPR signals in the singly rotating frame at $\omega = \omega_0$, $\omega_1 = \omega_{rf} = 2\pi$ 1.0 MHz, $\omega_2 = 2\pi$ 0.24 MHz. The solid and dashed lines correspond to the experiment and theory, respectively.

Fig. 1 shows the time evolution of absorption EPR signals observed at a doubly resonant bichromatic field in the singly rotating frames. The signals were obtained for the following parameters of the bichromatic field: $\omega = \omega_0$, $\omega_1 = \omega_{rf} = 2\pi\, 1.0$ MHz, $\omega_2 = 2\pi\, 0.24$ MHz, for the rf phase $\psi = 0$ (Fig. 1a), $\psi = \pi/2$ (Fig. 1b) and the random rf phase (Fig. 1c). The signal shown in Fig. 1c is the result of the averaging of nutation signals at the uniform distribution of random phases of the rf field over the interval from 0 to 2π. The Fourier spectra of obtained signals are presented in Fig. 2.

The fitting of the observed signals by Eq. (2) shown by the dashed lines in Figs. 1 and 2 demonstrates a good agreement between the theory and experiment and confirms that the observed signal has three oscillating components with frequencies ω_1 and $\omega_1 \pm \left(\omega_2^2 + \Delta_{BS}^2\right)^{1/2}$. The predicted dependence of these components on the rf phase is also observed. The approximation of the damping of the signals was done using in Eq. (2) the exponential decay function with T = 14 μs.

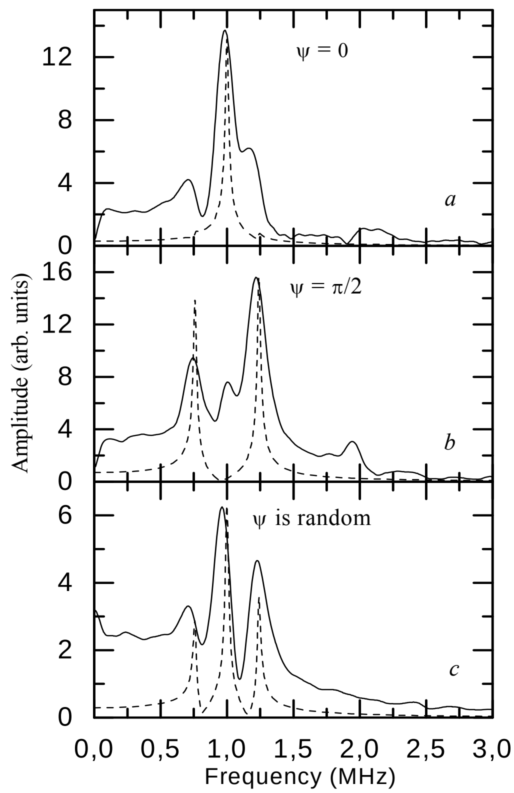

Fig.2. Fourier spectra of the absorption signals shown in Fig. 1. The solid and dashed lines correspond to the experiment and theory, respectively.

It is seen in the signal shown in Fig. 2 that the violation of the rotating-wave approximation is manifested in the asymmetry of the amplitudes of signals at the frequencies $\omega_1 \pm \left(\omega_2^2 + \Delta_{BS}^2\right)^{1/2}$.

Same difference between observed and calculated signals is due to inhomogeneous broadening of EPR line of our sample, which was not taken into account in the theoretical description. In the Fourier spectra, the broadening of observed lines in comparison with the calculated those is due to the limited time interval of the observation of the nutation signals.

CONCLUSIONS

We have shown theoretically and experimentally that multiplication of spin qubits arises at double resonance in a bichromatic field ($\omega = \omega_0$ and $\omega_1 = \omega_{rf}$). We demonstrate that the operational multiphoton transitions of dressed qubits can be selected by the choice of both the rotating frame and the rf phase. The experimental results obtained in the two-level EPR system in the ratating frame show that the theory correctly describes the dynamics of spin qubits dressed by the bichromatic field. The effects predicted beyond the rotating wave approximation are also confirmed by the observation of transient nutations.

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