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ORIGINAL PAPER

Bloch-Siegert shift in the Rabi oscillations on the "dressed" electron spin states

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Abstract. Using electron paramagnetic resonance (EPR) at Rabi frequencies, we study the Bloch-Siegert effect in the coherent dynamics of electron spin states driven by a bichromatic radiation consisting of a transverse microwave (MW) and longitudinal radio frequency (RF) fields. The Rabi oscillations on the electron spin states "dressed" by the MW field are observed at the double resonance, when the MW frequency equals the Larmor frequency of the spin system and the frequency ω_{rf} of the RF field is close to the Rabi frequency ω_1 in the MW field. The dispersive ($|\omega_{rf} - \omega_1| \sim \omega_{rf}, \omega_1$) and near-resonant ($\omega_{rf} \approx \omega_1$) regimes are investigated. We demonstrate that the Bloch-Siegert shift increases with increasing RF amplitude and at the fixed RF amplitude it has the largest values in the "low-frequency" ($\omega_{rf} < \omega_1$) range of the dispersive regime. Experimental results were obtained in the time-resolved EPR for E_1' centres in quartz.

Key words: electron paramagnetic resonance (EPR) • Bloch-Siegert shift • Rabi oscillation • bichromatic field • dressed state • spin qubit

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Introduction

The coherent Rabi oscillations are one of the fundamental effects of the resonant interaction between an electromagnetic radiation and a quantum system. This phenomenon has been studied for a wide range of physical systems, including nuclear and electron spins in nuclear magnetic resonance (NMR) [14] and electron paramagnetic resonance (EPR) [5], atoms in optical cavities, quantum dots, flux, and charge qubits in superconducting systems [9]. In the usual description of the resonant interaction, the linearly polarized electromagnetic field is decomposed into two mutually counterrotating circularly polarized fields. In the weak-driving limit, only the corotating component interacts efficiently with the quantum system. At resonance, this rotating-wave approximation (RWA) yields the Rabi frequency that is defined by the amplitude of the electromagnetic field. When the electromagnetic field is strong, the interaction with the counter-rotating component of the electromagnetic field needs to be taken into account. This interaction leads to the Bloch-Siegert shift in the resonance frequency ω_0 of a quantum system [1, 2, 4]. The Bloch-Siegert effect becomes significant in the ultrastrong coupling regime of field-qubit interaction, when the driving is so strong that the Rabi frequency ω_1 approaches the Larmor frequency ω_0 ($\omega_1/\omega_0 \geq 0.1$).

This regime has been studied in the continuous-wave double (optical-magnetic) resonance [3]. Recently, the quantum Bloch-Siegert effect has been observed in an LC resonator very strongly coupled to a flux superconducting qubit [6]. On the other hand, the Bloch-Siegert effect can be measured in the coherent dynamics of a two-level electron spin system (electron spin qubit) using the Rabi oscillations in the time-resolved experiments. The ultrastrong coupling regime can be realized in the pulsed double resonance with the bichromatic field. In such EPR experiments, transverse microwave (MW) and longitudinal radio frequency (RF) fields [8] are applied. When the MW frequency ω_{mw} is equal to the Larmor frequency ω_0 of an electron spin qubit and the RF ω_{rf} is close to the Rabi frequency ω_1 in the MW field, the so-called Rabi (nutational) resonance is observed [7, 10, 11]. In this case the Rabi oscillations between the spin states dressed by the MW field can be excited by pulses of a linearly polarized RF field. Since it is possible to access the regime where the amplitude of the RF field is comparable with ω_1 the Rabi oscillations on the dressed spin states enable investigations of the Bloch-Siegert effect in the coherent dynamics of the qubit [11–13].

In this paper, we study the Bloch-Siegert effect in the Rabi oscillations of spin qubits driven by the bichromatic (MW and RF) fields. The peculiarities of this effect in the dispersive ($|\omega_{rf} - \omega_1| \sim \omega_{rf}, \omega_1$) and in the near-resonant ($\omega_{rf} \approx \omega_1$) regimes are analyzed. Experimental results obtained in the time-resolved EPR for E_1' centers in quartz are presented.

Theoretical predictions

Based on the theoretical description [11–13], we consider spin qubits in a static magnetic field directed along the z-axis of the laboratory frame and driven by bichromatic field consisting of a transverse MW and a longitudinal RF fields. The RWA is used for the interaction between the qubit and the MW field, because $\omega_1 << \omega_0, \omega_{mw}$ and the Bloch-Siegert effect can arise only due to the counter-rotating component of the RF field. We use the non-RWA for the description of the RF field-qubit interaction.

Dispersive regime

In the dispersive regime, when the conditions $\omega_2 << |\omega_{rf} - \omega_1| \sim \omega_{rf}, \omega_1$ are fulfilled, the Bloch-Siegert effect results in the shift in the frequency ω_1 of the absorption signal of the Rabi oscillations observed in the frame rotating around the z-axis of the laboratory frame with the frequency ω_{mw}. [13]. The Bloch-Siegert effect is accompanied by the dynamical (ac) Zeeman effect. According to [13], under our experimental conditions ($\omega_{mw} = \omega_0$), the shift in the Rabi frequency is

$$(1) \qquad \delta\Omega = \frac{1}{2}\omega_2^2\left(\frac{1}{\omega_1 - \omega_{rf}} + \frac{1}{\omega_1 + \omega_{rf}}\right) \equiv \delta\Omega_Z + \delta\Omega_{BS}$$

where $\omega_1 = \gamma B_1$, $\omega_2 = \gamma B_2$, γ is the gyromagnetic ratio, B_1 and B_2 are the respective amplitudes of the MW and RF fields, $\delta\Omega_Z$ and $\delta\Omega_{BS}$ are the frequency shifts caused by the ac Zeeman and the Bloch-Siegert effects, respectively. The observed Rabi frequency is given by $\Omega_R^{eff} = \omega_1 + \delta\Omega$.

Near-resonant regime

In the near-resonant regime, when ω_{rf} is close to ω_1, we also consider the exact resonance in the MW field ($\omega_{mw} = \omega_0$) assuming that $\omega_1, \omega_{rf} >> \omega_2$. In the rotating frame the absorption signal of the Rabi oscillations consists of three components oscillating at frequencies ω_{rf} and $\omega_{rf} \pm \varepsilon$ [12]. Here

$$(2) \qquad \varepsilon = \left[(\omega_1 - \omega_{rf} + \Delta_{BS})^2 + \omega_2^2\right]^{1/2}$$

where $\Delta_{BS} = \omega_2^2/2(\omega_1 + \omega_{rf})$ is the Bloch-Siegert shift. In this case the Bloch-Siegert effect causes the shift in the frequency ε.

Experimental results and discussion

The Rabi oscillations between the qubit states were detected in the time-resolved EPR for E_1' centres in crystalline quartz. The width of the EPR line of the E_1' centres is $\Delta B_{pp} = 16$ μT, and the longitudinal and transverse relaxation times are $T_1 = 0.2$ ms and $T_2 = 3.5$ μs, respectively. The experimental method was described in [11]. The experiments were carried out at room temperature. The duration, amplitude, and repetition period of

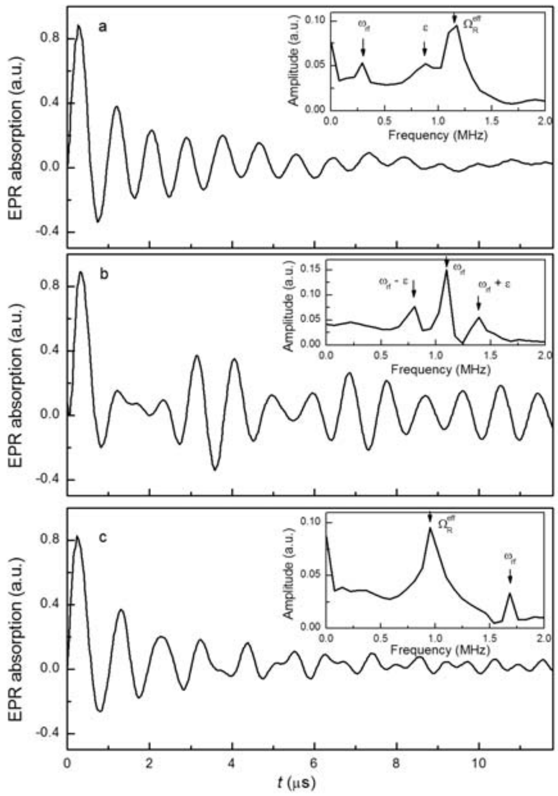

Fig. 1. Absorption signals of the Rabi oscillations for the fixed amplitudes of the MW and RF fields ($\omega_1/2\pi = 1.005$ MHz, $\omega_2/2\pi = 0.320$ MHz) at the three different radio frequencies $\omega_{rf}/2\pi = 0.300$ MHz (a), $\omega_{rf}/2\pi = 1.100$ MHz (b), and $\omega_{rf}/2\pi = 1.700$ MHz (c). The insets show the Fourier transformed signals.

the magnetic-field pulses were equal to 12 μs, 0.12 mT, and 1.25 ms, respectively. To improve the signal-to-noise ratio, the digital summation (up to 10^3 times) of the Rabi oscillations signals was used. The phase of the RF field was randomly changed in the range of $(0, 2\pi)$.

Figure 1 shows the absorption signals of the Rabi oscillations of the E_1' centres observed for the fixed amplitudes of the MW and RF fields at three different radio frequencies. Two of them correspond to the low- and high-frequency ranges of the dispersive regime and the third one (Fig. 1b) corresponds to the near-resonant regime. The insets are the Fourier transformations of the detected signals. The observed signals demonstrate the dependence of frequency of the Rabi oscillations on the detuning $(\omega_1 - \omega_{rf})$ from the Rabi resonance. The frequencies Ω_R^{eff}, ε and $\omega_{rf} \pm \varepsilon$ are identified in the observed Rabi oscillations. These frequencies are in very good agreement with our analytical model. In accordance with theoretical predictions, the differences between the frequencies of the observed Rabi oscillations and the frequency of the excited RF field are larger in the dispersive regime than those in the near-resonant regime. The oscillations at the frequency ω_{rf} are also identified in the observed signals. In the near-resonant regime these oscillations are predicted [12]. According to [13], there are no such oscillations in the dispersive regime. They can be observed because the non-homogeneous line width is larger than the RF amplitude.

Figure 2 depicts the dependence of the measured frequencies of the Rabi oscillations on the RF. For the used RF amplitude $\omega_2/\omega_1 = 0.32$, and the ultrastrong coupling regime of the RF field-qubit interaction is realized. In that case the Bloch-Siegert effect becomes perceptible in the frequency shift of the Rabi oscillations between the dressed spin states. The frequencies of the Rabi oscillations Ω_R^{eff} and $\omega_{rf} \pm \varepsilon$ were obtained with the precision not worse than 5 kHz. The measured frequencies of the Rabi oscillations agree well with theoretical predictions, Eqs. (1) and (2). In order to illustrate the contribution of the Bloch-Siegert shift, the dependences of Ω_R^{eff} and $\omega_{rf} \pm \varepsilon$ assuming the RWA are indicated by the dashed lines. The observed frequencies of the Rabi oscillations demonstrate that in the dispersive regime the Bloch-Siegert shift is larger than this effect in the near-resonant regime. The shift is the most significant in the "low-frequency" $(\omega_{rf} < \omega_1)$ range of the dispersive regime and increases with increasing detuning from the Rabi resonance. In accordance with Eqs. (1) and (2), the contribution of the Bloch-Siegert effect is smaller in the "high-frequency" $(\omega_{rf} > \omega_1)$ range. Theoretical predictions of Ω_R^{eff} give good results only in the finite RF range, i.e. for $\omega_{rf}/2\pi < 0.7$ MHz and $\omega_{rf}/2\pi > 1.4$ MHz. Equation (2) yields a quite good agreement for the dependencies of $\omega_{rf} \pm \varepsilon$ with the experimental values in the RF range which is even wider than determined by the near-resonant regime.

It is known that in the dispersive regime the Bloch--Siegert effect is accompanied by the ac Zeeman effect [13]. In the near-resonant regime only the Bloch-Siegert shift occurs, but this effect is smaller than in the dispersive regime. Therefore, it is better to illustrate the contribution of the Bloch-Siegert shift in the near-resonant regime using the dependence of the frequency ε of the Rabi oscillations between the dressed spin states on the detuning $\omega_1 - \omega_{rf}$. Such a dependence for the two RF amplitudes is depicted in Fig. 3. The dotted lines represent the theoretical dependences given by Eq. (2) assuming the RWA. In this case ε does not depend on the sign of the detuning and for the fixed RF amplitude is presented by a single line. According to Eq. (2), due to the Bloch-Siegert effect ε depends asymmetrically on the sign of the detuning resulting in two lines for each RF amplitude. Figure 3 shows that at $\omega_2/2\pi = 0.120$ MHz the contribution of the Bloch-Siegert effect is almost unobservable. On the other hand, at $\omega_2/2\pi = 0.320$ MHz the contribution of the Bloch-Siegert shift is noticeable and is most significant for $\omega_{rf} < \omega_1$ and increases with increasing detuning.

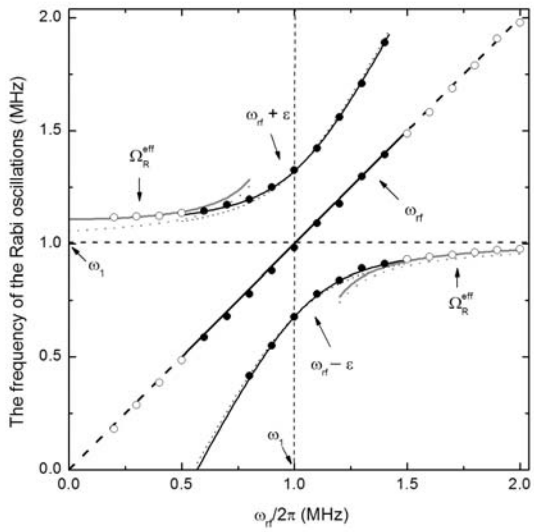

Fig. 2. The frequency of the Rabi oscillations vs. the radio frequency at $\omega_1/2\pi = 1.005$ MHz and $\omega_2/2\pi = 0.320$ MHz. The gray and black solid lines represent theoretical calculations of Ω_R^{eff} and $\omega_{rf} \pm \varepsilon$, respectively. The dotted lines show theoretical values of Ω_R^{eff} and $\omega_{rf} \pm \varepsilon$ in the RWA. The white and black circles are the measured frequency in the dispersive and near-resonant regimes, respectively.

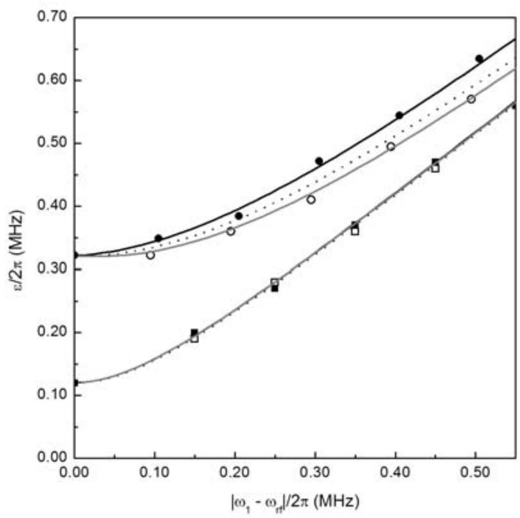

Fig. 3. The dependence of the frequency ε of the Rabi oscillations between the dressed spin states on the detuning $|\omega_1 - \omega_{rf}|$ for the two RF amplitudes. The circles and squares represent the experimental data for $\omega_2/2\pi = 0.320$ MHz and 0.120 MHz, respectively; for $\omega_{rf} < \omega_1$ the black symbols and for $\omega_{rf} > \omega_1$ the white those. Solid (dotted) lines are calculated using the non-RWA (RWA).

Conclusions

The Bloch-Siegert effect results in the frequency shift of the Rabi oscillations on the dressed electron spin states observed in the time-resolved EPR in the bichromatic (microwave and longitudinal radio frequency) field. Changing the radio frequency allows us to measure the Rabi resonance in the dispersive and resonance regimes. In both regimes the Bloch-Siegert effect gives the contribution to the frequency of the Rabi oscillations. We demonstrate that the Bloch-Siegert shift increases with increasing RF amplitude and at the fixed RF amplitude it has the largest values in the "low-frequency" ($\omega_{rf} < \omega_1$) range of the dispersive regime. The obtained results demonstrate the failure of the rotating-wave approximation in the description of the coherent dynamics of electron spin qubits, but can be useful for quantum computation operating in the ultrastrong coupling regime of field-qubit interaction.

References

1. Abragam A (1961) The principles of nuclear magnetism. Clarendon Press, Oxford
2. Allen C, Eberly JH (1975) Optical resonance and two--level atoms. Wiley, New York
3. Arimondo E, Moruzzi G (1973) Bloch-Siegert shift in optically oriented ^{199}Hg vapour. J Phys B 6:2382–2389
4. Bloch F, Siegert A (1940) Magnetic resonance for nonrotating fields. Phys Rev 57:522–527
5. Fedoruk GG (2002) Transient nutation EPR spectroscopy of condensed media. J Appl Spectrosc 69:161–182
6. Forn-Diaz P, Lisenfeld J, Marcos D *et al*. (2010) Observation of the Bloch-Siegert shift in a qubit-oscillator system in the ultrastrong coupling regime. Phys Rev Lett 105:237001
7. Jeschke G (1999) Coherent superposition of dressed spin states and pulse dressed electron spin resonance. Chem Phys Lett 301:524–530
8. Kälin M, Fedin M, Gromov I, Schweiger A (2006) Multiple-photon transitions in EPR spectroscopy. Lect Notes Phys 684:143–183
9. Le Bellac M (2006) A short introduction to quantum information and quantum computation. Cambridge University Press, Cambridge
10. Redfield AG (1955) Nuclear magnetic resonance saturation and rotary saturation in solids. Phys Rev 98:1787–1809
11. Saiko AP, Fedaruk GG (2008) Effect of the Bloch-Siegert shift on the frequency responses of Rabi oscillations in the case of nutation resonance. JETP Lett 87:128–132
12. Saiko AP, Fedaruk R (2010) Multiplication of qubits in a doubly resonant bichromatic field. JETP Lett 91:681–685
13. Saiko AP, Fedaruk R, Kolasa A, Markevich SA (2012) Dissipative dynamics of qubits driven by a bichromatic field in the dispersive regime. Phys Scr 85:045301
14. Torrey HC (1949) Transient nutations in nuclear magnetic resonance. Phys Rev 76:1059–1068