Higher order geometric phase for qubits in a bichromatic field

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Abstract
The geometric phase in the dynamics of a spin qubit driven by transverse microwave (MW) and longitudinal radio frequency (RF) fields is studied. The phase acquired by the qubit during the full period of the ‘slow’ RF field manifests in the shift of Rabi frequency $\omega_1$ of a spin qubit in the MW field. We find out that, for a linearly polarized RF field, this shift does not vanish at the second and higher even orders in the adiabaticity parameter $\omega_{rf}/\omega_1$, where $\omega_{rf}$ is the RF frequency. As a result, the adiabatic (Berry) phases for the rotating and counter-rotating RF components compensate each other, and only the higher order geometric phase is observed. We experimentally identify that phase in the frequency shift of the Rabi oscillations detected by a time-resolved electron paramagnetic resonance.

(Some figures may appear in colour only in the online journal)

1. Introduction
Berry has shown [1] that during adiabatic and cyclic evolutions of Hamiltonian’s parameters, a non-degenerate eigenstate of a quantum system acquires a geometric phase in addition to its dynamic counterpart. The concept of the geometric phase was extended to non-cyclic and non-adiabatic (e.g., the Aharonov–Anandan phase [2]) evolutions, including degenerate states [2] and open systems with dissipation [3]. Geometric phases manifest in a wide variety of phenomena in quantum physics, optics and solid-state physics (see, e.g., [2, 4]). For example, the realizations of exotic (quantum [5], anomalous [6], optical [7] and phononic [8]) Hall effects as well as new propositions of logic gates for quantum computation [9] are closely related to the geometric phase. Various types of basic elements (qubits) for quantum computation have been presented in recent reviews [10, 11].

Energy level shifts of a quantum system due to off-resonant periodic perturbations, such as Bloch–Siegert, dynamic (ac) Zeeman and Stark shifts, are also directly related to the additional phase in the evolution of its stationary states. Indeed, slow periodic adiabatic perturbations with periods $\tau$ much longer than the characteristic time $\tau_r$ of the quantum system’s evolution give rise to energy shifts including those proportional to the first order in the adiabaticity parameter $\tau_r/\tau$ (see, e.g., [12]). This ensured that the quantum system accumulates the geometric phase [12]. In some cases [13], the energy level shift does not appear in the first order in $\tau_r/\tau$. As a result, the geometric phase can be acquired only at higher orders in $\tau_r/\tau$, i.e. when the adiabatic condition is violated.

The total phase $\phi$ acquired by the wavefunction of a quantum system in the adiabatically changing field can be expanded in a power series in a small parameter $\epsilon = \tau_r/\tau$:

$$\frac{d\phi}{d\tau} = \alpha_0 \epsilon^0 + \alpha_1 \epsilon^1 + \alpha_2 \epsilon^2 + \ldots$$  (1)

The dynamical phase of the wavefunction is given by the integral of the zeroth-order term over the time interval $(0, \tau)$. The integral of the first-order term depends on the geometry of the closed path in the space of slowly changing parameters and corresponds to the geometric (Berry) phase. Integrals of higher order terms yield corrections to the Berry phase and become zero in the exact adiabatic limit $\epsilon \rightarrow 0$. However, for real physical processes, adiabatic conditions are often not exactly fulfilled, i.e. the adiabaticity parameter has a small but finite magnitude. In these cases, higher order corrections in $\epsilon$ are needed (see [14] and references therein, and also [11]). Note that, in order to weaken the fulfillment of the exact adiabatic condition, Berry has also performed calculations taking into account the adiabatic correction terms (see p 494 in [2]). Examples of the higher order corrections to the geometric phase for simple systems can be found in the...
literature, e.g. in [15, 16]. The higher order corrections to the geometric phase [15] have been used to explain phase fluctuations observed in the superconducting circuit system [17]. Even for simple quantum systems such as spin qubits in a slowly changing electromagnetic field, the Berry phase, in its standard definition, exists only when the field is circularly polarized [18]. For the linearly polarized field, the first-order term in \( \epsilon \) in equation (1) that describes the standard Berry phase becomes zero, but the even higher order terms in the adiabaticity parameter differ from zero. Consequently, the geometric phase observed in that case can originate only from the higher order corrections. We call the higher order geometric phase as the part of the geometric phase given by the higher order corrections when the Berry phase is zero. The higher order geometric phase cannot be treated as the higher order corrections when the Berry phase is zero. We consider the ‘dressed’ states of the qubit at the precise resonance between the MW field and the qubit \( \omega_{\text{MW}} = \omega_0 \), also assuming that \( \omega_0 \gg \omega_1 \gg \omega_2 \). Since in our experiment \( \omega_1/\omega_{\text{MW}} \approx 10^{-4} \), the counter-rotating component of the MW field is neglected and the rotating-wave (RW) approximation is used for the interaction between the qubit and the MW field. For the RF field, the rotating and counter-rotating components should be taken into account because \( \omega_2 \) and \( \omega_2 \) can be comparable.

Dynamics of the system with the Hamiltonian (2) is described by the Liouville equation for the density matrix \( \rho \):

\[ i\hbar \frac{\partial \rho}{\partial t} = [H, \rho] + i\Lambda \rho \]  

(3)

(furthermore, we assume \( \hbar = 1 \)). The superoperator \( \Lambda \) describing decay processes is defined by its action upon \( \rho \) as follows:

\[ \Lambda \rho = (\gamma_{12}/2)(2s^z\rho s^z - s^z s^z \rho - s^z s^z) \]
\[ + (\gamma_{12}/2)(2s^z s^z - s^z s^z \rho - s^z s^z) \]
\[ + (\eta/2)(2s^z s^z - \rho/2), \]

where \( \gamma_{12} \) and \( \gamma_{12} \) are the rates of the transitions from the excited state 2 of the qubit to its ground state 1 and vice versa, and \( \eta \) is the dephasing rate. After two canonical transformations \( \rho_2 = u_1^* u_1 \rho u_1 u_2^* \), \( u_1 = \exp(-i\omega_{\text{MW}} t s^z) \), \( u_2 = \exp(-i\pi s^x s^y) \), equation (3) is transformed into \( i\hbar \frac{\partial \rho_2}{\partial t} = [H_2, \rho_2] + i\Lambda \rho_2 \) if the conditions \( \gamma_{21}, \gamma_{12} \ll \omega_1 \) are fulfilled. Here

\[ H_2 = \omega_1 s^z - \frac{\omega_0}{2} (s^x e^{-i\omega_0 t} + \text{h.c.}) - \frac{\omega_2}{2} (s^y e^{i\omega_2 t} + \text{h.c.}), \]

(4)

where the second and third terms in the Hamiltonian (4) correspond to the RW and counter-rotating-wave (CRW) interactions between the RF field and the spin qubit, respectively, \( \Lambda \rho_2 \) has the same operator structure as \( \Lambda \rho \) and differs only in the relaxation parameters: \( \gamma_{21} \rightarrow \Gamma_{1} = (\gamma_{21} + \gamma_{12})/2, \gamma_{12} \rightarrow \Gamma_{1} = \Gamma_{1}, \eta \rightarrow \Gamma_{2} = \gamma_{21} + \gamma_{12} \).

The operator \( u_1 \) transforms the density matrix in the frame that rotates with the frequency \( \omega_{\text{MW}} \) around the z-axis of the laboratory frame, and \( u_2 \) tilts that frame in the spin space by the angle \( \pi/2 \) around the y-axis. Note that, when the condition \( \omega_{\text{RF}} \ll \omega_1 \) is fulfilled, the probabilities of the RW and CRW processes are close to each other in magnitude.

Using the transformation operator \( \rho_3 = \exp(-i\omega_{\text{RF}} t s^z) \), the Liouville equation can be transformed in the doubly rotating frame which rotates with the frequency \( \omega_{\text{RF}} \) around the z-axis of the tilted frame. In this frame in the Hamiltonian (4), the oscillations with the Rabi frequency \( \omega_0 \) occur. Since \( \omega_1 \gg \omega_{\text{RF}} \), the ratio \( \omega_{\text{RF}}/\omega_1 \) can be used as the adiabaticity parameter. Hence, the Hamiltonian (4) contains rapidly \( \exp(\pm i\omega_{\text{RF}} t) \) and slowly \( \exp(\pm i\omega_0 t) \) oscillating functions. The rapidly oscillating terms in the transformed Liouville equation can be eliminated by using the Krylov–Bogoliubov–Mitropolsky non-secular perturbation theory [22–24]. In the second order in
a small parameter $\omega_2/\omega_1$, we obtain, $i\partial(\rho_1)/\partial t = [H_{\text{eff}}, (\rho_1)] + i(\Lambda(\rho_1))$, 

$$H_{\text{eff}} = \frac{i}{\hbar} \left\{ \int_0^t \text{d}\tau \left[ H_3(\tau) - \langle H_3(\tau) \rangle, H_3(t) \right] \right\} = \delta \Omega^{(2)}(t) x^2,$$

(5)

where $\rho_1 = \omega_1^2 \rho_2 u_1$, the symbol $\langle \ldots \rangle$ denotes time averaging over rapid oscillations of the type $\exp(\pm i \omega_0 t)$ given by $\langle O(t) \rangle = \frac{1}{2\pi} \int_0^{2\pi} O(t) \text{d}t$ and the upper limit $t$ of the indefinite integral indicates the variable on which the result of the integration depends and square brackets denote the commutation operation. The averaging does not affect the form of the superoperator, $\langle \Lambda' \rangle = \Lambda'$. The frequency shift in the Rabi oscillations is given by

$$\delta \Omega^{(2)}(t) = \frac{1}{2 \omega_1^2} \left( \frac{1}{\omega_1 - \omega_{\text{rf}}} + \frac{1}{\omega_1 + \omega_{\text{rf}}} \right) (1 + \cos 2 \omega_{\text{rf}} t).$$

(6)

Since, in the doubly rotating frame, the phase $\phi^{(2)}$ acquired by the qubits equals $\phi^{(2)}(t) = \int \delta \Omega^{(2)}(t) \text{d}t$, using equation (1) and expanding $\delta \Omega^{(2)}$ (6) in a power series of $\omega_{\text{rf}}/\omega_1$ yield $\delta \Omega^{(2)} = \delta \Omega^{(2)}_{\text{rf}} + \delta \Omega^{(2)}_{\text{rf}}$ where $\delta \Omega^{(2)}_{\text{rf}} = (\omega_2^2/\omega_1^2)(1 + \cos 2 \omega_{\text{rf}} t)$ is the dynamical Zeeman shift, and $\delta \Omega^{(2)}_{\text{rf}} = (\omega_1^2 / \omega_1^2) (1 + \cos 2 \omega_{\text{rf}} t)$ is the shift corresponding to the geometric phase. The latter shift is quadratically dependent on the adiabaticity parameter $\omega_{\text{rf}}/\omega_1$ (even higher order terms are neglected).

After a full cycle $\tau = 2 \pi / \omega_{\text{rf}}$ of the ‘slow’ RF field’s evolution, the qubit accumulates the dynamic phase $\phi^{(2)}_{\text{rf}} = 2 \pi \omega_{\text{rf}}^2 / \omega_1 \omega_{\text{rf}}$ and the geometric phase $\phi^{(2)}_{\text{g}} = 2 \pi \omega_{\text{rf}} / \omega_1 \omega_{\text{rf}}$. The geometric phase is the first-order term in the adiabaticity parameter and tends to zero at the adiabatic limit of $\omega_{\text{rf}}/\omega_1 \rightarrow 0$. Consequently, we obtain $\delta \Omega^{(2)} = (\phi^{(2)}_{\text{g}} \omega_{\text{rf}} / 2 \pi) (1 + \cos 2 \omega_{\text{rf}} t)$.

The terms $(\omega_1 - \omega_{\text{rf}})^{-1}$ and $(\omega_1 + \omega_{\text{rf}})^{-1}$ in equation (6) are due to the RW and CRW components of the RF field in the Hamiltonian $H_3$ (4), respectively. The ‘interference’ between these components is described by the term containing $\cos 2 \omega_{\text{rf}} t$ in the shift $\delta \Omega^{(2)}$. The ‘interference’ contribution is eliminated by averaging over the period $2 \pi / \omega_{\text{rf}}$, and we obtain $\delta \Omega^{(2)} = \omega_2^2 / \omega_1 + \phi^{(2)}_{\text{g}} / \omega_1 / 2 \pi$. The higher order (at least the fourth-order) corrections in $\omega_{\text{rf}}/\omega_1$ allow us to improve the theoretical description. The application of the non-secular perturbation theory [22] gives the fourth-order correction to the shift of the Rabi frequency in the form $\Delta \Omega^{(4)} = -\omega_2^4 / 4 \omega_1^3 - 3 \omega_2^2 \omega_{\text{rf}}^2 / 2 \omega_1^3$.

When both the second-order and the fourth-order corrections are considered, the shift becomes

$$\delta \Omega = \delta \Omega^{(2)} + \delta \Omega^{(4)} = \omega_2^2 / \omega_1^2 \left( 1 - \frac{\omega_2^2}{4 \omega_1^2} \right) + \frac{\omega_2^2 \omega_{\text{rf}}^2}{\omega_1^2} \left( 1 - \frac{3 \omega_2^2}{2 \omega_1^2} \right),$$

(7)

and the geometric phase is

$$\phi_{\text{g}} = \phi^{(2)}_{\text{g}} + \phi^{(4)}_{\text{g}} = \frac{2 \pi \omega_2^2}{\omega_1^2} \left( 1 - \frac{3 \omega_2^2}{2 \omega_1^2} \right) \omega_{\text{rf}} / \omega_1.$$  

(8)

The results of the non-secular perturbation theory lead to the important conclusion. Because the ‘interference’ of the RW and CRW terms in the frequency shift and in the geometric phase is averaged to zero over the period $2 \pi / \omega_{\text{rf}}$, the contributions of these terms in the Hamiltonian (4) can be calculated independently in all orders in $\omega_2/\omega_1$. Therefore, it is possible to consider the RW ($H_{\text{RW}}^2$) and CRW ($H_{\text{CRW}}^2$) parts in the Hamiltonian (4) separately. Each of these parts is exactly diagonalized: $H_{\text{RW}}^2 = f(\omega_{\text{rf}}) x^2$, $H_{\text{CRW}}^2 = f(\omega_{\text{rf}}) x^2$, where $f(\omega_{\text{rf}}) = (\omega_1 - \omega_{\text{rf}}^2 + \omega_2^2)^{1/2}$. Taking into account in the same way the contributions of the RW and CRW components of the RF field, we calculate the shift of the Rabi frequency up to the second order in $\omega_{\text{rf}}/\omega_1$. In the arbitrary order in $\omega_{\text{rf}}/\omega_1$ we have

$$\delta \Omega = 2 f(0) - \omega_1^2 f^{-3}(0) \omega_{\text{rf}}^2.$$  

(9)

Equation (9) is ‘exact’ in comparison with equation (7) because it is correct for all ratios of $\omega_2/\omega_1$. The geometric phase corresponding to the frequency shift (9) is

$$\phi_{\text{g}} = \frac{2 \pi \omega_2^2}{\omega_1^2} \left( 1 - \frac{3 \omega_2^2}{2 \omega_1^2} \right) \omega_{\text{rf}} / \omega_1.$$  

(10)

Hence, equations (9) and (10) are generalizations of equations (7) and (8) calculated by using the perturbation theory for the case of the arbitrary ratio of $\omega_2/\omega_1$.

Figure 1 shows the qubit dynamics on the Bloch sphere in the frame rotating with the frequency $\omega_{\text{MW}}$ and tilted by the angle $\pi/2$ around the $y$-axis in the spin space. The vectors $\omega_{\text{rf}}$ and $-\omega_{\text{rf}}$ represent the rotating and counter-rotating components of the RF field. At the exact resonance ($\omega_{\text{MW}} = \omega_1$) in the RW approximation for the MW field, the vector of the qubit magnetization $\mu$ can precess about the vectors $\omega_+\text{ and } \omega_-$ in the plane perpendicular to those vectors.
given by \( \omega_- = f(\omega_0) \) and \( \omega_+ = f(-\omega_0) \), respectively. The geometric phase acquired by the qubit state after a full cycle of the evolution, which is described by the Hamiltonian \( H_{\text{RF}}^\text{SRW}(H_{\text{CRW}}^\text{SRW}) \), equals \( +\Theta_{C-}/2 \) (\(-\Theta_{C+}/2 \)), where \( \Theta_{C-} (\Theta_{C+}) \) is the solid angle enclosed by the loop traversed by \( \omega_- (\omega_+) \). The solid angles are given by \( \Theta_{C-} = 2\pi[1 - \cos(\theta + \theta')] \) and \( \Theta_{C+} = 2\pi[1 - \cos(\theta - \theta')] \), where \( \cos(\theta + \theta') = (\omega_1 - \omega_0)/(\omega_1 + \omega_0) f^{-1}(\omega_0) \) and \( \cos(\theta - \theta') = (\omega_1 + \omega_0) f^{-1}(\omega_0) \). Expanding \( \Theta_{C-} \) and \( \Theta_{C+} \) in the power series of \( \omega_0/\omega_1 \) up to the first-order term, we take into account the higher order effect and obtain \( \Theta_{C-} - \Theta_{C+} = 4\pi\omega_0^2\omega_0 f^{-3}(0) = \Theta'_{C-} \).

The difference \( \Theta'_{C-} \) has the finite value at the first order in the adiabaticity parameter \( \omega_0/\omega_1 \) and becomes zero at ideal adiabaticity. In addition to the MW and RF amplitudes, the solid angle \( \Theta'_{C-} \) depends on the RF frequency. According to equation (10) the higher order geometric phase is \( \varphi_{E'0} = \Theta'_{C-}/2 \). Hence, the higher order geometric phase is determined by the half of the solid angle obtained by the difference between the solid angles corresponding to the RW and CRW components of the RF field.

The density matrix in the laboratory frame is given by \( \rho(t) = u_i u_j^* \rho(0) u_j u_i^* \exp\left(-i\int_0^t \delta\Omega \, dt\right) \exp(i\int_0^t \delta\Omega(t') \, dt') \), where \( \rho(0) = 1/2 \cdot \sigma_z \) provided that the qubit is found in the ground state at the initial moment. By using \( \rho(t) \), we obtain the absorption signal \( V(t) \) of the qubit in the rotating frame:

\[
V(t) = \text{tr}\{\rho(t)\sigma_z\} = \frac{1}{2i} \left[\left(\{1, 1\}(\rho(t)[2] - (2, 1)\rho(t)[1]) \right) - \frac{1}{2} \exp(-\Gamma_\perp t) \sin\left(\omega_1 t + \int_0^t \delta\Omega \, dt\right)\right],
\]

where \([1, 1]\) and \([2, 1]\) are the ket-vectors of the ground and excited states of the qubit, \( \Gamma_\perp = (\Gamma_\perp + \Gamma_\parallel + \Gamma_\parallel)/2 \). The renormalized relaxation rates \( \Gamma_\perp \) can be expressed in terms of the longitudinal relaxation times \( T_1 \) and transverse relaxation times \( T_2 \) in the laboratory frame: \( \Gamma_{\perp} = 1/2T_2 + 1/2T_1 \).

If the system of qubits has a distribution of its own frequencies \( \omega_0^i \) (where the index \( i \) numbers the qubit), the absorption signal of the system \( V_E \) can be found by summing up signals of every qubit with an appropriate weight function [25]. Assuming that \( \Gamma_\perp \) and \( \delta\Omega \) are the same for all qubits and that the width of the weight function is much larger than \( \omega_1 \), we obtain

\[
V_E \sim \omega_1 \exp(-\Gamma_\perp t) \left[ J_0(\omega_0 t) \cos\left(\frac{1}{2} \int_0^t \delta\Omega \, dt\right) - N_0(\omega_0 t) \sin\left(\frac{1}{2} \int_0^t \delta\Omega \, dt\right) \right],
\]

where \( J_0(x) \) and \( N_0(x) \) are the zero-order Bessel function of the first and second kind, respectively.

### 3. Experimental results and discussion

Rabi oscillations were formed by the so-called Zeeman pulse technique [26]. The continuous MW and RF fields were used. The resonant interaction with the spin system was abruptly established with a pulse of a longitudinal magnetic field. Pulses of a magnetic field with amplitude \( \Delta B = B - B_0 = 0.12 \text{ mT} \) and duration of 10 \( \mu \text{s} \) were used. The experiments were performed at room temperature using an X-band homemade EPR spectrometer. Multichannel digital summation was used to improve the signal-to-noise ratio. Due to the long relaxation times and the small EPR line width, the \( E_1 \) centres \((S = 1/2) \) in neutron-irradiated quartz were used in our experiments. The EPR spectrum of the \( E_1 \) centres consists of an isolated line with the width of \( \Delta B_{pp} = 16 \mu\text{T} \) at the magnetic field direction along the crystal optical axis. The values of \( \omega_1 \) and \( \omega_2 \) were directly measured with the precision of about 1 kHz at \( \omega_{\text{max}} = \omega_0 \) by using the frequencies of the Rabi oscillations in the MW field \((\omega_0 = 0) \) and in the bichromatic field at \( \omega_{\text{RF}} = \omega_1 \).
The solid angle was varied by changing the frequency geometric phase on the solid angle for the data presented in \( \phi \). The experimental values of the agreement with the theoretical predictions. In our experiment, only the higher order geometric phase is observed. Moreover, the observable dephasing is due to the conventional relaxation time \( T_2 \), and the geometric dephasing is eliminated by the stability of the bichromatic field.

4. Summary

We have shown both theoretically and experimentally that, in the evolution of a spin qubit driven by the bichromatic (MW and RF) field, the geometric phase during the full period of the ‘slow’ RF field appears as a shift of the Rabi frequency of the qubit in the MW field. We have demonstrated that for the linearly polarized RF field, only the higher order geometric phase is observed. This phase can be realized in a variety of quantum systems, thus opening new possibilities for their geometric control, and our results can be useful in any practical applications beyond ideal adiabaticity.

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