Implementation of universal quantum gates by periodic two-step modulation in a weakly nonlinear qubit

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Reducing the leakage from qubit levels to other levels is of importance for quantum computation. Here, we develop a feasible method to effectively suppress this leakage and perform universal quantum gates in a weakly nonlinear qubit. The method is to exploit periodical two-step modulation of single control parameters in a artificial large detuning regime. It works well, regardless of whether the strength of control parameter is known or not. In particular, qubit leakage can also be suppressed in a counterintuitive parameter regime, where the control field drives the transition of qubit levels with large detuning, but resonance transition with leakage level. The results demonstrate that this method is robust against parameter perturbations, and the deformation of square waves does not accumulate the error of qubit operations but affects the gate time. In addition, this method provides us with a viable approach to measure the control parameters.

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I. INTRODUCTION

Accurate coherent qubit control (two-level system) is an essential technology in quantum computation [1,2]. Unfortunately, a qubit is difficult to be absolutely isolated from actual physical systems. This is because system-environment coupling or unwanted internal coupling exists in multilevel systems. These inevitable couplings give rise to qubit leakage. For the former case, many methods have been proposed to prevent decoherence-induced leakage, including dynamical decoupling [3–10], the construction of decoherence-free subspaces [11–17], etc. Certainly, different methods have their respective merits and application scope. For instance, the method of leakage elimination operators [18–20] (a type of dynamical decoupling) works well on eliminating leakage in multilevel systems but requires unbounded fast and strong pulses [21,22], which may be unattainable in experiments. To be compatible with distinct quantum systems and taking full advantage of the respective merits, different methods have been effectively combined in recent studies, such as incorporating the dynamical decoupling in decoherence-free subsystems [23–27] and adiabatic leakage elimination operators [28].

Here, we are interested in the latter case that leakage is induced by the internal coupling between multiple levels. Generally speaking, if the coupling strength of system fields is small enough, the influence of leakage can be dramatically ignored in anharmonic systems (i.e., the transition frequency of qubit levels slightly differs from the transition frequency of nonqubit levels) [29–34]. Nevertheless, this is a relatively contradictory choice, since strong system-field coupling strength is in favor of fast quantum operations [35]. Until now, substantial approaches have been introduced to solve this issue. One celebrated approach is called derivative removal by adiabatic gate (DRAG) [36–40], and it is a breakthrough in experiments where the DRAG shaping pulse has been exploited to improve single-qubit gates [41,42]. Nevertheless, this approach relies on employing several control parameters and pulse shaping techniques, increasing the complexity of manipulations. In particular, DRAG may be invalid for designing pulse shape if quantum systems show strong parameter perturbations. Another approach is called composite pulses [43–45], which is just a finite sequence of pulses with piecewise-constant values. The composite pulses have the merits of ultrahigh accuracy and robustness to parameter imperfections so that they have wide applications in quantum information processing. More recently, this approach has been employed in quantum dot charge quadrupole qubits to achieve arbitrary single-qubit rotations with high fidelity [46]. Particularly, when combined with quantum optimal control [47–53], the error of qubit operation can easily reach below the fault-tolerant threshold. The composite pulses are always composed of a mass of different constant values, and the time interval between different constant values is inequable. As a result, the pulse profile would be irregular. Besides, it might have the same problem as DRAG, namely, one should simultaneously modulate several physical parameters during qubit operations. In this work, based on inheriting the merits of composite pulses, we are highly focused on exploring simple and feasible pulses to realize robust universal quantum operations.

To our knowledge, the implementation of qubit operations is often based on resonance and near-resonance conditions

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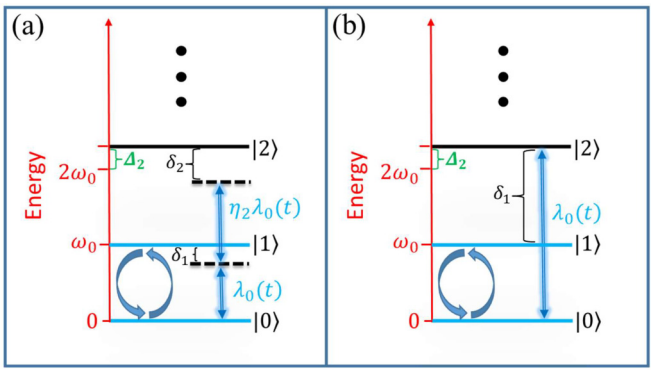

FIG. 1. The energy structure of the nonlinear oscillator coupled by a control field $\lambda_0(t)$, where the qubit is formed by $|0\rangle$ and $|1\rangle$. (a) The control field $\lambda_0(t)$ drives the transition $|0\rangle \leftrightarrow |1\rangle$ with detuning δ_1. (b) The control field $\lambda_0(t)$ resonantly drives the transition $|0\rangle \leftrightarrow |2\rangle$.

(in terms of the qubit-field coupling) but have been scarcely extended to large detuning conditions. This is due to the consensus that large detuning conditions cannot promote quantum state evolution, even freezing the population of qubit levels. In this work, by taking advantage of large detuning conditions, we develop a two-step modulation method to eliminate qubit leakage as well as reduce the number of control parameters and simplify pulse wave form. This method is used to regulate single control parameters with the shape of a periodical square wave rather than the analytically derived pulses [36]. Moreover, the two-step modulation method is still valid, even though the coupling strength of the control parameter is unknown. In addition, it is robust against parameter perturbation and wave-form deformation due to large detuning conditions. As a by-product, the two-step modulation method can be employed to experimentally measure unknown control parameters.

The rest of this paper is organized as follows. In Sec. II we introduce the physical model used in this work. In Sec. III we first present the protocol of two-step modulation in detail and provide an expression of the effective Hamiltonian of the system. Then we illustrate some examples of implementing qubit operations by strength modulation. In Sec. IV we phenomenologically give the typical expression of the evolution operator in two-step modulation. In Sec. V we discuss the leakage error, the validity of the rotation wave approximation (RWA), the physical mechanism, and the experimental feasibility of two-step modulation. In Sec. VI we demonstrate by numerical calculations that this method is robust against noise, including parameter perturbations, wave-form deformations, and decoherence. The conclusion is given in Sec. VII.

II. PHYSICAL MODEL

Consider the model that the d-level nonlinear oscillator is coupled by a control field, which can be used to describe superconducting qubits [54–63], nanomechanical oscillators [32], etc. As shown in Fig. 1(a), the qubit is composed of the two lowest levels $|0\rangle$ and $|1\rangle$ of the nonlinear oscillator. In the laboratory frame, the system Hamiltonian for a d-level structure reads ($\hbar = 1$)

$$\mathcal{H}(t) = \sum_{k=1}^{d-1}(k\omega_0 + \Delta_k)|k\rangle\langle k| + \lambda(t)\eta_k(|k\rangle\langle k-1| + |k-1\rangle\langle k|), \quad (1)$$

where ω_0 represents the transition frequency of nearest-neighbor levels. Specifically, the energy of the kth higher level is different from $k\omega_0$ by Δ_k, which is known as the anharmonicity, and we set $\Delta_1 = 0$. The form of the control field reads $\lambda(t) = \lambda_x(t)\cos(\omega_c t) + \lambda_y(t)\sin(\omega_c t)$, where ω_c is the control field frequency, and $\lambda_x(t)$ and $\lambda_y(t)$ represent two independent quadrature control parameters. η_k represents a dimensionless parameter weighting the relative coupling strength of the transition $|k-1\rangle \leftrightarrow |k\rangle$ and transition $|0\rangle \leftrightarrow |1\rangle$.

In the rotating frame defined by the unitary operator $U(t) = e^{-ik\omega_c t|k\rangle\langle k|}$, the Hamiltonian reads

$$\begin{aligned} H_0(t) &= U^\dagger(t)\mathcal{H}(t)U(t) - iU^\dagger(t)\dot{U}(t) \\ &= \sum_{k=1}^{d-1}\delta_k(t)|k\rangle\langle k| + \eta_k[\lambda_0(t) + \lambda_0^*(t)e^{-2i\omega_c t}]|k\rangle\langle k-1| \\ &\quad + \eta_k[\lambda_0^*(t) + \lambda_0(t)e^{2i\omega_c t}]|k-1\rangle\langle k|, \end{aligned} \quad (2)$$

where $\delta_k(t) = \Delta_k + k\delta_1(t)$, $\delta_1(t) = \omega_0 - \omega_c$ is the detuning of the qubit with respect to the control field, and $\lambda_0(t) = \frac{\lambda_x(t)}{2} + i\frac{\lambda_y(t)}{2}$. When $2\omega_c \gg |\lambda_0(t)|$, employing RWA, the Hamiltonian becomes

$$H_0(t) = \sum_{k=1}^{d-1}\delta_k(t)|k\rangle\langle k| + \eta_k\lambda_0(t)|k\rangle\langle k-1| + \text{H.c.} \quad (3)$$

In the following we study the dynamical evolution of the nonlinear oscillator by the Hamiltonian (3) in this frame.

In the ideal case, $\eta_k = 0$ ($k > 1$), the control field induces only the transition between qubit levels, and other higher levels cannot participate in this transition. As a result, the Hilbert space of the nonlinear oscillator can be projected into two irrelevant parts: the qubit subspace and the outside of the qubit subspace. Then the unitary operator of the nonlinear oscillator is expressed as $U_i = U_{\text{qb}} \oplus e^{i\phi}U_{\text{os}}$, where U_{qb} acts only on the qubit subspace, U_{os} acts only on the outside of the qubit subspace, and ϕ is an irrelevant relative phase. Therefore, universal qubit operations are easily achieved by modulating the control parameters. For instance, the Y gate $U_{\text{qb}} = \sigma_y$ is implemented by setting $\delta_1(t) = 0$, $\lambda_x(t) = 0$, and $\lambda_y(t) = \xi$, which satisfy $\int_0^{t_g}\xi dt = \frac{\pi}{2}$, where t_g denotes the gate time.

Nevertheless, in the general case, $\eta_k \neq 0$, the control field unavoidably induces the transition between qubit levels and higher levels. As a result, the undesired leakage into other higher levels leads to an error of qubit operations. To overcome this shortcoming, one can simultaneously regulate three independent control parameters [i.e., $\lambda_x(t)$, $\lambda_y(t)$, and the detuning $\delta_1(t)$] to effectively remove most of the leakage error [36]. At the same time, the shapes of three control parameters are always not familiar, since they are achieved from a set of differential equations. As a result, this would increase the difficulty of experimental operations. In the following, we

demonstrate that modulating only a single parameter [e.g., the detuning $\delta_1(t)$ or control parameter $\lambda_x(t)$] can also achieve the same goal, while other control parameters remain unchanged during the evolution process. Moreover, the modulation shape is extremely simple and common, i.e., the periodical square wave.

III. TWO-STEP MODULATION PROTOCOL

To begin with, we need to explicitly point out the control parameters in two-step modulation. In the weakly nonlinear qubit, the parameters $\{\lambda_x(t), \lambda_y(t), \delta_1(t)\}$ are individually adjustable. As a result, several adjustment manners can be performed in two-step modulation. For brevity, we enumerate two of their paradigms: (i) $\lambda_y = 0$, λ_x is fixed, and we periodically modulate $\delta_1(t)$, which we call frequency modulation [64]; (ii) $\lambda_y = 0$, δ_1 is fixed, and we periodically modulate $\lambda_x(t)$, which we call strength modulation. Note that other adjustment manners can be analyzed in a similar way, and we do not illustrate them here again.

A. Frequency modulation

In frequency modulation, the Hamiltonian of the system reads ($d = 3$)

$$H_0(t) = \delta_1(t)|1\rangle\langle 1| + [\Delta_2 + 2\delta_1(t)]|2\rangle\langle 2| + \lambda_x|1\rangle\langle 0| + \eta_2\lambda_x|2\rangle\langle 1| + \text{H.c.}, \tag{4}$$

where the detuning $\delta_1(t)$ is periodically modulated by a repeated two-step sequence,

$$\delta_1(t) = \begin{cases} \delta, & t \in [mT, mT+\tau], \\ \delta', & t \in (mT+\tau, (m+1)T]. \end{cases} \tag{5}$$

T is the square-wave period, $m \in \mathbb{N}$. That is, within a period, the detuning of system is δ in time interval $[0, \tau]$, and becomes δ' in time interval $(\tau, T]$. For concise representation, the system Hamiltonian is labeled by H_0 when the detuning is δ, while the system Hamiltonian is labeled by H_0' when the detuning is δ'. In addition, we mark $\tau' \equiv T - \tau$ hereafter.

In principle, the values of δ and δ' can be arbitrary but must be different from each other. Once $\delta = \delta'$, the time-dependent Hamiltonian (4) becomes static without any modulations. As a result, qubit operations cannot be achieved since the system dynamics cannot evolve in a large detuning regime ($|\delta| \gg \lambda_x$). Experimentally, it has two ways to adjust the detuning $\delta_1(t)$. The first way is to change the transition frequency of qubit levels. This situation can be realized by adding an external magnetic flux with square-wave shape [65–70]. The other way is to change the frequency of the control field. Namely, the frequency of the control field is modulated with a square-wave shape, which can be achieved by a square-wave generator.

There are two critical requirements for two-step modulation in the nonlinear oscillator. The first one is that the coupling between the qubit and control field is always in a large detuning regime during the whole evolution process, namely, $|\delta_1(t)| \gg |\lambda_x|$. The other one is that the time interval τ (τ') for the corresponding Hamiltonian H_0 (H_0') should satisfy

$$\tau = \frac{(2n+1)\pi}{E_1 - E_0}, \quad \tau' = \frac{(2n'+1)\pi}{E_1' - E_0'}, \quad n, n' \in \mathbb{N}, \tag{6}$$

where E_k and E_k' ($k = 0, 1$) are the two lowest eigenenergies of the Hamiltonian H_0 and Hamiltonian H_0', respectively. Therefore, the square-wave period reads

$$T = \tau + \tau' = \frac{(2n+1)\pi}{E_1 - E_0} + \frac{(2n'+1)\pi}{E_1' - E_0'}. \tag{7}$$

Obviously, under two-step modulation, the evolution operator of the system at arbitrary evolution time $t = t' + mT$ can be expressed as

$$U(t) = \mathbb{T} e^{-i\int_0^t H_0(t)dt} = \begin{cases} e^{-iH_0 t'}(e^{-iH_0'\tau'}e^{-iH_0\tau})^m, & t' \in [0, \tau], \\ e^{-iH_0'(t'-\tau)}e^{-iH_0\tau}(e^{-iH_0'\tau'}e^{-iH_0\tau})^m, & t' \in (\tau, T], \end{cases} \tag{8}$$

where $\mathbb{T}$ is the time-ordering operator. Note that the evolution operator (8) exactly describes the system dynamics, and we refer to the exact evolution or exact dynamics in the following.

To analyze the dynamical behaviors of the system, we need to give the specific expression of the time-independent effective Hamiltonian H_{eff}, which is defined by [71]

$$U(T) = e^{-iH_0'\tau'}e^{-iH_0\tau} \equiv e^{-iH_{\text{eff}}T}. \tag{9}$$

Compared to other periodic works [72,73], the remarkable difficulty is that we cannot employ the Baker-Campbell-Hausdorff expansion to achieve H_{eff}, since the modulation frequency $\omega = \frac{2\pi}{T}$ is the same (even small) magnitude to the energy gap of system. By making use of the second-order perturbation theory, the effective Hamiltonian of the nonlinear oscillator approximatively reads (see Appendix A for details)

$$H_{\text{eff}} = i\lambda_{\text{eff}}|0\rangle\langle 1| - i\lambda_{\text{eff}}|1\rangle\langle 0|, \tag{10}$$

where the constant $\lambda_{\text{eff}} = \frac{2\lambda_x|\delta'-\delta|}{T\delta\delta'}$.

It is easily found that the effective Hamiltonian (10) contains only the dynamical evolution of qubit levels $|0\rangle$ and $|1\rangle$, and the higher level $|2\rangle$ has been eliminated. Although the effective Hamiltonian (10) is derived from $d = 3$, it is also suitable for higher levels ($d > 3$) of the nonlinear oscillator. Moreover, one readily finds from Eq. (10) that the system dynamics now can be regarded as *Rabi-like* oscillation between qubit levels with "Rabi frequency" λ_{eff}. Therefore, the relevant quantum gates of the nonlinear oscillator can be implemented by choosing different evolution times. For instance, we can implement a Y gate at gate time $t_g = \frac{(1+4l)\pi}{2\lambda_{\text{eff}}}$ and an iSWAP gate at gate time $t_g = \frac{(1+8l)\pi}{4\lambda_{\text{eff}}}$, $l \in \mathbb{N}$, etc. On the other hand, if $\lambda_x = 0$ and λ_y are fixed, we can obtain the X gate at gate time $t_g = \frac{(1+4l)\pi}{2\lambda_{\text{eff}}}$. Remarkably, if the initial state of the system is $|0\rangle$, the final state of the system would be $|1\rangle$ at $t_g = \frac{(1+4l)\pi}{2\lambda_{\text{eff}}}$, which realizes population inversion between qubit levels. It is worth mentioning that t_g might not be exactly equal to mT in practice, but it does not affect main results, since we can interrupt the two-step sequence when the evolution time reaches t_g.

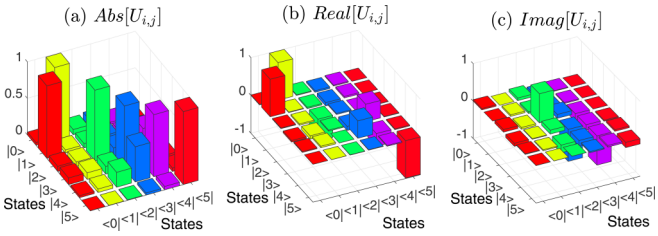

FIG. 2. The evolution operator $U(t_g) = \sum_{i,j=0}^{5} U_{i,j}|i\rangle\langle j|$ implemented by two-step modulation of the control parameter λ_x in a six-level nonlinear oscillator, where $\lambda_x/\lambda_0 = 1, \lambda_x'/\lambda_0 = -1, \lambda_y = 0$, $\Delta_2/\lambda_0 = 3, \delta_1/\lambda_0 = 3.6, \eta_k = \sqrt{k}, \Delta_{k+1} = \frac{k(k+1)}{2}\Delta_2, k = 1, \ldots, 4$.

B. Strength modulation

Alternatively, we can achieve relevant qubit operations in nonlinear oscillators by strength modulation, where $\lambda_y(t) = 0$, δ_1 is fixed, and $\lambda_x(t)$ is modulated by a two-step sequence:

$$\lambda_x(t) = \begin{cases} \lambda_x, & t \in [mT, mT + \tau], \\ \lambda_x', & t \in (mT + \tau, (m+1)T]. \end{cases} \tag{11}$$

Experimentally, strength modulation can be implemented by adjusting the phase (amplitude) of the control parameter λ_x with the phase (attenuation) modulator. And the phase (attenuation) modulators need to work under the square-wave shape.

Since the detailed derivations of the effective Hamiltonian in strength modulation are similar to those of frequency modulation, the expression is similar to Eq. (10):

$$H_{\text{eff}}' = i\lambda_{\text{eff}}'|0\rangle\langle 1| - i\lambda_{\text{eff}}'|1\rangle\langle 0|, \tag{12}$$

where the "Rabi frequency" $\lambda_{\text{eff}}' = \frac{2(\lambda_x - \lambda_x')}{T\delta}$. As an example, we plot in Fig. 2 the evolution operator $U(t_g)$ at gate time $t_g = 2.45/\lambda_0$, which is highly consistent with the ideal X gate. In this simulation, the levels of the nonlinear oscillator are chosen $d = 6$, and the results demonstrate that the subspace of qubit operation is almost isolated in the Hilbert space, indirectly proving the suppression of leakage from the qubit to other levels.

Next, we present a counterintuitive phenomenon in strength modulation, where the physical model is plotted in Fig. 1(b). To be specific, the control field $\lambda_0(t)$ still drives the transition $|0\rangle \leftrightarrow |1\rangle$ with large detuning δ_1. However, the detuning δ_1 exactly matches the transition frequency between $|1\rangle$ and $|2\rangle$ now. In other words, the control field $\lambda_0(t)$ resonantly drives the transition $|0\rangle \leftrightarrow |2\rangle$ at the same time. Thus, the Hamiltonian can be expressed as ($d = 3$)

$$H_0 = \delta_1|1\rangle\langle 1| + \lambda_0(t)|1\rangle\langle 0| + \lambda_0(t)|2\rangle\langle 0| + \text{H.c.} \tag{13}$$

It is a commonsense that Rabi oscillation occurs between $|0\rangle$ and $|2\rangle$ under static Hamiltonian (13) without two-step modulation. However, this Rabi oscillation is utterly helpless, even harmful for qubit operations, since the qubit basis is $\{|0\rangle, |1\rangle\}$ rather than $\{|0\rangle, |2\rangle\}$, and it would cause a large leakage from qubit subspace.

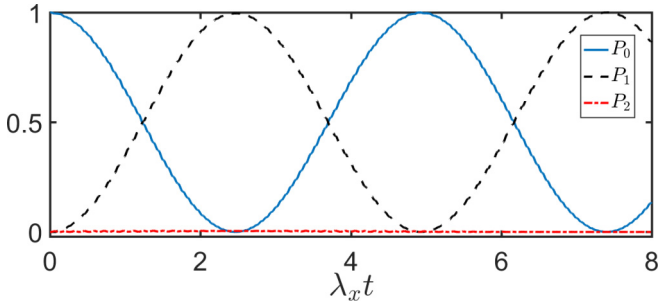

FIG. 3. The time evolution of population $P_l = |\langle l|\psi(t)\rangle|^2$ ($l = 0, 1, 2$) of each level under strength modulation of $\{\lambda_x, \lambda_x'\}$, where $\lambda_x' = -\lambda_x, \lambda_y = 0, \delta_1/\lambda_x = -48, \tau_1 = \frac{\pi}{E_1 - E_0}, \tau_1' = \frac{\pi}{E_1' - E_0'}$, and $|\psi(t)\rangle$ represents the system state at evolution time t.

The situation is completely changed when we employ the following strength modulation:

$$\lambda_x(t) = \begin{cases} \lambda_x, & t \in [mT, mT + \tau], \\ -\lambda_x, & t \in (mT + \tau, (m+1)T]. \end{cases} \tag{14}$$

where $\lambda_y(t) = 0$, and 1.00,0.00,0.00 the time intervals τ and τ' also satisfy the condition: $\tau = \frac{(2n+1)\pi}{E_1 - E_0}$, $\tau' = \frac{(2n'+1)\pi}{E_1' - E_0'}$, $n, n' \in \mathbb{N}$. Figure 3 represents the time evolution of population $P_l = |\langle l|\psi(t)\rangle|^2$ ($l = 0, 1, 2$) of each levels under strength modulation, where $|\psi(t)\rangle$ represents the system state at time t. The results demonstrate that *Rabi-like* oscillation would occur between $|0\rangle$ and $|1\rangle$, regardless of the resonance condition between $|0\rangle$ and $|2\rangle$ in the original system. It is further shown in Fig. 3 that the populations of the leakage level $|2\rangle$ are nearly frozen during evolution process. Therefore, this counterintuitive phenomenon can offer us alternative preferences to realize universal qubit operations, since the undesired leakage is sharply suppressed.

IV. PHENOMENOLOGICAL DESCRIPTION OF TWO-STEP DYNAMICS

In two-step modulation, the dynamics described by the effective Hamiltonian (10) or (12) is fairly consistent with the exact dynamics of the system in a long timescale, but it is invalid in short timescales. In principle, it is easy to know the short-time dynamics of the system if we find out the explicit expression of evolution operator $U(t)$ in Eq. (8) at arbitrary time t. However, the exact solution is difficult to obtain in two-step modulation since the system no longer satisfies resonance conditions.

To better describe the exact dynamics for short timescales, taking frequency modulation as an example, the phenomenological estimation for the evolution operator of a qubit can be written as (see Appendix B for details)

$$U_{\text{qb}}^{\text{es}}(t) = \begin{pmatrix} |u_{11}^{\text{es}}(t)|e^{-i\Theta_{11}^{\text{es}}(t)} & |u_{12}^{\text{es}}(t)| \\ |u_{21}^{\text{es}}(t)|e^{-i\Theta_{21}^{\text{es}}(t)} & |u_{22}^{\text{es}}(t)|e^{-i\Theta_{22}^{\text{es}}(t)} \end{pmatrix}, \tag{15}$$

where the global phase is ignored and the expressions of amplitudes and phases read

$$|u_{11}^{\text{es}}(t)| = |u_{22}^{\text{es}}(t)| = \sqrt{|\cos^2(\lambda_{\text{eff}}t) - A(t)|},$$

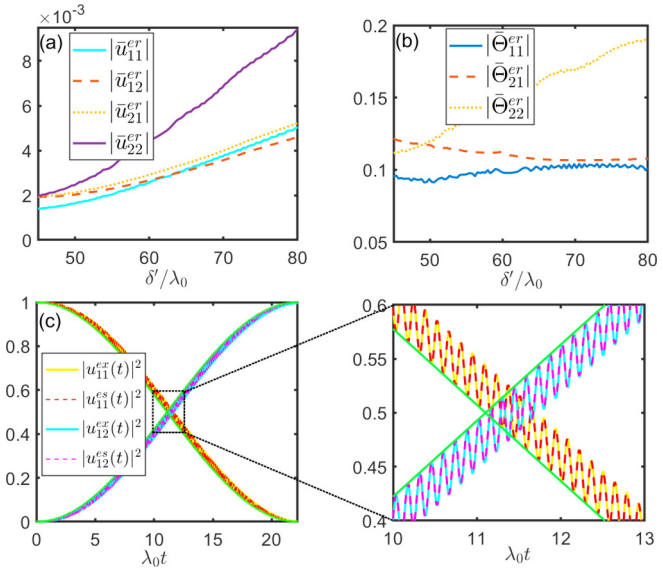

FIG. 4. The average error of (a) amplitudes and (b) phases as a function of δ' in frequency modulation, where $\lambda_x/\lambda_0 = 1$, $\Delta_2/\lambda_0 = 3$, $\eta_2 = \sqrt{2}$, $\delta/\lambda_0 = 40$. (c) The exact evolution and estimated evolution of amplitudes when $\delta' = 50$. In the inset, one can see more clearly that the estimated evolutions (dashed-red and dashed-pink lines) coincide well with the exact evolution (solid-yellow and solid-cyan lines).

$$|u_{12}^{\rm es}(t)| = |u_{21}^{\rm es}(t)| = \sqrt{|\sin^2(\lambda_{\rm eff}t) + A(t)|},$$

$$\Theta_{11}^{\rm es}(t) = -\left[\frac{1}{\sqrt{2}}A_m\left(\sin g(t) - \frac{1}{2}\right) + 1\right]\arctan\vartheta - \frac{\pi}{2},$$

$$\Theta_{22}^{\rm es}(t) = g(t) - \arctan\vartheta - \frac{\pi}{2},$$

$$\Theta_{21}^{\rm es}(t) = \pi + g(t), \quad A(t) = A_m \sin^2\left[\frac{g(t)}{2}\right]\sin(2\lambda_{\rm eff}t),$$

$$A_m = \frac{\lambda_x(\delta+\delta')}{\delta\delta'}, \quad \vartheta = \frac{\lambda_x^2}{\lambda_{\rm eff}^2}[2{\rm mod}(\lambda_{\rm eff}t/\pi) - \pi],$$

and the T-periodic function $g(t)$ reads

$$g(t) = \begin{cases} -\frac{\pi}{\tau}t, & t \in [mT, mT+\tau], \\ \frac{\pi}{\tau'}(T-t), & t \in (mT+\tau, (m+1)T]. \end{cases}$$

To quantify the deviation between the exact and estimated dynamical evolution, we define the average error within the gate time:

$$\bar{O}^{\rm er} = \frac{1}{t_g}\int_0^{t_g} |O^{\rm ex}(t) - O^{\rm es}(t)|dt, \tag{16}$$

where $O^{\rm ex}(t)$ and $O^{\rm es}(t)$ represent the time evolution of physical parameters governed by the exact evolution operator $U(t)$ [described by Eq. (8)] and the estimation evolution operator $U_{\rm qb}^{\rm es}(t)$ [described by Eq. (15)], respectively.

Figures 4(a) and 4(b) demonstrate the average error of amplitudes and phases as a function of detuning δ' at gate time $t_g = \frac{\pi}{2\lambda_{\rm eff}}$, respectively. The results show that the estimated expressions of amplitude and phase are pretty accurate to describe the exact evolution process since the value of average error is considerably small. As a concrete example, we plot in Fig. 4(c) the amplitudes as a function of evolution time when $\delta' = 50$, where the solid lines and dashed lines are described by the exact evolution operator $U(t)$ and the estimation evolution operator $U_{\rm qb}^{\rm es}(t)$, respectively. It is shown that the solid lines are in high agreement with the dashed lines. Hence, the exact dynamics $U(t)$ of the system in frequency modulation can be effectively described by the estimated expression $U_{\rm qb}^{\rm es}(t)$ at arbitrary time t. Furthermore, the green lines in Fig. 4(c) represent the time evolution of the system governed by the evolution operator $U_{\rm eff}(t) = e^{-iH_{\rm eff}t}$ with the "Rabi frequency" $\lambda_{\rm eff}$. We find that the system dynamics is well fitted by green lines in the long timescale, implicitly proving the validity of the effective Hamiltonian (10). On the other hand, one also observes in Figs. 4(a) and 4(b) that the values of $\bar{u}_{22}^{\rm er}$ and $\bar{\Theta}_{22}^{\rm er}$ are dramatically larger than other values when δ' is large. The reason is that we do not take into account leakage from the qubit to other levels in the expression $U_{\rm qb}^{\rm es}(t)$. When considering the leakage, the correction of amplitude $|u_{12}(t)|$ should be

$$|u_{12}^{\rm co}(t)| = |u_{21}^{\rm co}(t)| = \sqrt{1-2\xi^2}|u_{21}^{\rm es}(t)|, \tag{17}$$

where $\xi = |\frac{\frac{\eta_2\lambda_x}{\delta'+\Delta}(e^{-i\frac{\Delta\pi A_m}{\lambda_x}}+e^{-i\frac{\Delta\pi}{\delta}}) - \frac{\eta_2\lambda_x}{\delta+\Delta}(1+e^{-i\frac{\Delta\pi}{\delta}})}{1-e^{-i\frac{\Delta\pi A_m}{\lambda_x}}+i\lambda_{\rm eff}T}|$. Since qubit level $|1\rangle$ interacts with other levels during the evolution process, the correction of $|u_{22}(t)|$ is intricate and we do not discuss it here. Of course, to further reduce the leakage from qubit level $|1\rangle$, one can employ composite two-step modulation; see Appendix C for details.

V. DISCUSSIONS

A. Leakage error

Until now, we have demonstrated that two-step modulation has been successfully employed to implement universal quantum operations, including frequency modulation and strength modulation. It is worth mentioning that Eqs. (10) and (12) are effective expressions for periodical two-step modulation and they do not contain higher levels of the nonlinear oscillator, since we exploit the second-order perturbation theory. In fact, the exact evolution $U(t)$ of the nonlinear oscillator should include all levels under two-step modulation so that tiny leakage would always exist when we perform qubit operation. To quantify the leakage error of qubit operation, we adopt the Euclidean distance between the ideal qubit operation $U_{\rm qb}$ and the exact evolution $U(t_g)$, whose definition is [54,74]

$$F_{\rm error} = \tfrac{1}{4}||U_{\rm qb} - \mathcal{P}^\dagger U(t_g)\mathcal{P}||_2^2, \tag{18}$$

where $||O||_2^2 = {\rm trace}(O^\dagger O)$, and $\mathcal{P}$ is the projection operator on the qubit basis $\{|0\rangle, |1\rangle\}$. The exact evolution $U(t_g)$ is governed by Eq. (8) in two-step modulation.

As an illustration, we plot the relation between leakage error $F_{\rm error}$ and detuning δ' in Fig. 5 when achieving a Y gate at gate time $t_g = \frac{\pi}{2\lambda_{\rm eff}}$ by frequency modulation. The results demonstrate that the leakage error gradually decreases with the decrease of the difference between δ and δ', and frequency modulation is indeed effective in suppressing leakage error since the value of $F_{\rm error}$ is extremely small. It is worth mention-

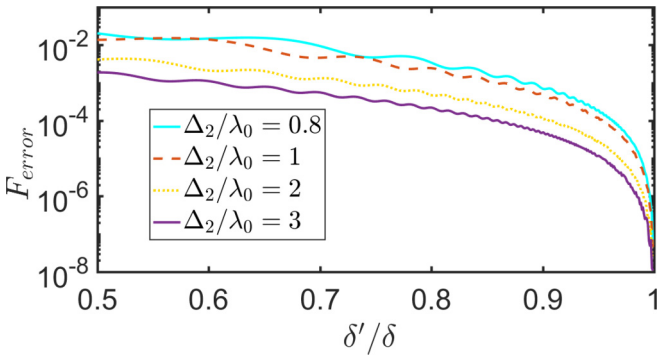

FIG. 5. Leakage error F_{error} vs detuning δ' in three-level nonlinear oscillator by frequency modulation where $\lambda_x/\lambda_0 = 1$, $\delta/\lambda_0 = 100$, $\eta_2 = \sqrt{2}$.

ing that the leakage error approaches zero when $\lim_{\delta' \to \delta} \frac{\delta'}{\delta} = 1$, namely, δ' infinitely approaches δ but $\delta' \neq \delta$. In fact, the decrease of leakage error is at the expense of long evolution times, which can be confirmed by the "Rabi frequency" λ_{eff} in Eq. (10). Therefore, there is a tradeoff between leakage error and evolution time in practice.

B. Validity of RWA

As mentioned in Sec. II, the main results are valid by making use of the RWA. Thus, it is important to know the regime in which the RWA is well satisfied. Note that the system dynamics would emerge with Rabi oscillation if we employ the effective Hamiltonian (10) or (12). However, the system dynamics is not strictly Rabi oscillation in two-step modulation (a detailed explanation is given in Sec. IV). Thus we would use Hamiltonian (3) rather than the effective Hamiltonian (10) or (12) to explore the validity of the RWA. For brevity, we label the time evolution of the system state as $|\psi^O(t)\rangle$ when employing the original Hamiltonian (1) and label the time evolution of the system state as $|\psi^R(t)\rangle$ when employing Hamiltonian (3) under the RWA. Then, the following definition can be exploited to study the parameter range of safely employing the RWA,

$$P_l^D = \max\left\{\left|P_l^O(t) - P_l^R(t)\right|\right\},\ l = 0, 1, \tag{19}$$

where $P_l^O(t) = |\langle l|\psi^O(t)\rangle|^2$ ($P_l^R(t) = |\langle l|\psi^R(t)\rangle|^2$) represents the population evolution of level $|l\rangle$ without (with) RWA, and P_l^D represents the maximum deviation between two dynamics during time evolution. Remarkably, a high value of P_l^D means that RWA is no longer suitable, and the RWA would be more valid when P_l^D is lower.

Figure 6 demonstrates the maximum deviation P_l^D as a function of the transition frequency ω_0 and the control field frequency ω_c in strength modulation. As expected, the RWA is quite effective in most of the region when the control field frequency is much larger than the coupling strength ($\omega_c \gg \lambda_0$) or the system satisfies a large detuning condition ($|\omega_o - \omega_c| \gg \lambda_0$), since the value of P_l^D is as low as 0.03. Furthermore, it can be observed that we cannot employ the RWA when the control field frequency is low ($\omega_c/\lambda_0 \simeq 1$) or the system is in the near-resonance regime ($|\omega_o - \omega_c| \simeq 0$).

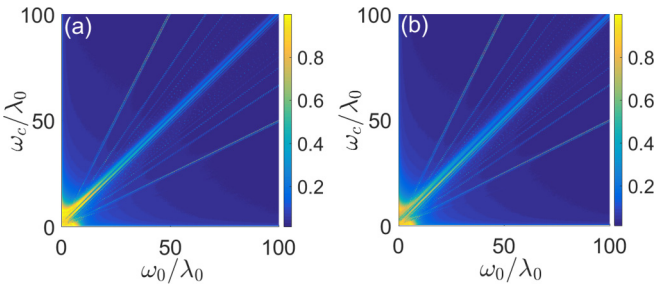

FIG. 6. (a) The maximum deviation P_0^D vs the transition frequency ω_0 and the control field frequency ω_c. (b) The maximum deviation P_1^D vs the transition frequency ω_0 and the control field frequency ω_c. The initial state is $|0\rangle$ in strength modulation, and the other parameters are $\lambda_x/\lambda_0 = 1$, $\lambda_x'/\lambda_0 = 1.2$, $\lambda_y = 0$, $\Delta_2/\lambda_0 = 3$, and $\eta_2 = \sqrt{2}$.

In addition, the RWA is also invalid when $\omega_c/\omega_0 = 2$ or $\omega_0/\omega_c = 2$, since the detuning $\delta_1 = -\omega_0$ or $\delta_1 = \omega_c$ in this case.

C. Physical mechanism

At first glance, it seems inconceivable to perform qubit operations by two-step modulation in a large detuning regime. In fact, the physical mechanism of two-step modulation can be comprehended from two perspectives. First, the system should satisfy a *frequency match*. The frequency ω of two-step modulation exactly compensates the detuning between qubit levels. This is distinctly confirmed in strength modulation, where the detuning between qubit levels is δ_1 and the frequency ω of two-step modulation reads

$$\omega = \frac{2\pi}{T} = \frac{2\pi}{\tau + \tau'} = \frac{2\pi}{\frac{(2n+1)\pi}{E_1 - E_0} + \frac{(2n'+1)\pi}{E_1' - E_0'}} \simeq \frac{\delta_1}{\mathfrak{n}+1}. \tag{20}$$

Here $\mathfrak{n} = n + n' \in \mathbb{N}$, and we call it a "$(\mathfrak{n}+1)$-photon resonance" condition because the detuning δ_1 is $\mathfrak{n}+1$ times the frequency ω (since this process is different from traditional n-photon resonance, we use this terminology with quotation marks). It is easily found from Eq. (20) that different time intervals τ and τ' determine different values of $\mathfrak{n}$, namely, they determine how many "photons" are required to compensate the detuning between qubit levels. When the system satisfies the frequency match condition, it is possible that Rabi oscillation emerges between qubit levels in two-step modulation. However, not all square waves with the same frequency ω can be used to perform universal qubit operations. Different square-wave duty ratios affect the dynamical phase during the evolution process, and the system should satisfy *phase match*. For each static Hamiltonian H_0 and H_0', the dynamical phase difference between qubit levels $|1\rangle$ and $|0\rangle$ must be odd times of π, i.e.,

$$\begin{aligned} \Theta_1 &= (E_1 - E_0)\tau = (2n+1)\pi,\quad n \in \mathbb{N}, \\ \Theta_1' &= (E_1' - E_0')\tau' = (2n'+1)\pi,\quad n' \in \mathbb{N}. \end{aligned} \tag{21}$$

In Fig. 7(a), we plot the maximum population $P_1^{\max}$ of qubit level $|1\rangle$ during the evolution process versus different frequencies $\omega(=\frac{2\pi}{T})$ and duty ratios ζ in strength modulation, where $\zeta T = \tau$ represents the time interval that the system

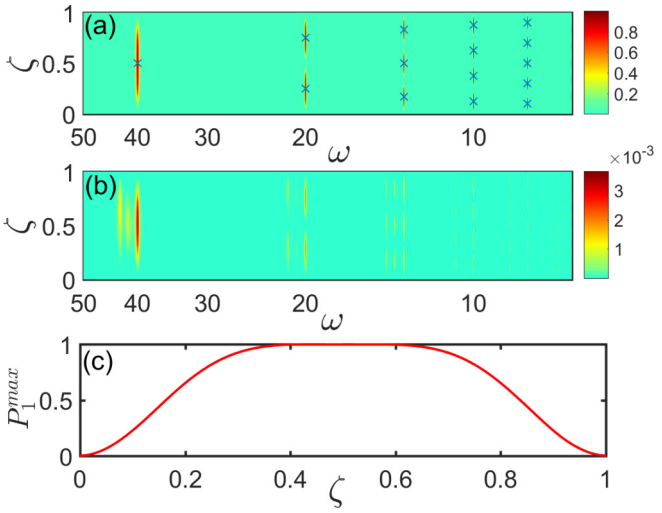

FIG. 7. (a) The maximum population $P_1^{\max}$ of qubit level $|1\rangle$ and (b) the maximum population $P_2^{\max}$ of leakage level $|2\rangle$ during whole evolution process under strength modulation of $\{\lambda_x, \lambda_x'\}$, where the initial state is in $|0\rangle$, and $\lambda_x' = 0.8\lambda_x$, $\lambda_y = 0$, $\delta_1/\lambda_x = 40$, $\Delta_2/\lambda_x = 3$, $\eta_2 = \sqrt{2}$, $\tau = \frac{\pi}{E_1 - E_0}$, $\tau' = \frac{\pi}{E_1' - E_0'}$. (c) The maximum population $P_1^{\max}$ of qubit level $|1\rangle$ as a function of duty ratio ζ when $\omega_c = 40$ under strength modulation. $P_1^{\max} \simeq 1$ means that there exist Rabi oscillations between qubit levels meanwhile sharply suppressing the leakage, and vice versa.

is governed by the static Hamiltonian H_0 and $(1-\zeta)T = \tau'$ represents the time interval that the system is governed by the static Hamiltonian H_0'. Among all cases of strength modulation, we can observe that only those frequencies that satisfy the frequency match condition can realize the transition $|0\rangle \leftrightarrow |1\rangle$ of qubit levels, which is denoted by dark-red vertical lines in Fig. 7(a), and the $(\mathfrak{n}+1)$th-row dark-red vertical line (look from left to right) corresponds to the "$(\mathfrak{n}+1)$-photon resonance" condition. Further inspection reveals that the $(\mathfrak{n}+1)$th-row dark-red vertical line is divided into $(\mathfrak{n}+1)$ segments. This is because there are $(\mathfrak{n}+1)$ possibilities to constitute the frequency ω of two-step modulation, i.e., the value of n can be chosen from 0 to $\mathfrak{n}$ according to the expression $\mathfrak{n} = n + n' \in \mathbb{N}$ in Eq. (20). Furthermore, only when the system satisfies the phase match condition can the transition between qubit levels be achieved very well. This is confirmed by X marks in Fig. 7(a), where the positions of the X's (i.e., frequencies ω and duty ratios ζ) are calculated by Eqs. (20) and (21).

On the other hand, Fig. 7(a) shows that the two-step modulation we design is robust against the mismatch caused by time interval τ, since there is a platform where the maximum population of qubit level $|1\rangle$ is high. This can be also verified in Fig. 7(c), the two-dimensional graph when $\omega = 40$ in Fig. 7(a). It implicitly reveals that the conditions satisfying both frequency match and phase match are optimum for qubit operations, since the value of $P_1^{\max}$ is maximum at the position of the X marks, the center of each dark-red vertical line in Fig. 7(a). In order to prove whether two-step modulation can effectively suppress the leakage, we plot in Fig. 7(b) the maximum population $P_2^{\max}$ of leakage level $|2\rangle$ versus different frequencies ω and duty ratios ζ. The results indicate that the maximum of $P_2^{\max}$ is on the order of 10^{-3}, verifying that the populations of leakage levels are indeed sharply suppressed when we perform qubit operations by two-step modulation.

D. Experimental feasibility

Next we discuss the feasibility of the proposed method. Experimentally, the nonlinear qubit can be found in the transmon-type Cooper-pair box (CPB) circuit [69,70,75]. In a CPB circuit, due to the presence of weak anharmonicity in the transmon regime, the energy spacings of adjacent levels are different from each other. Thus, the lowest two levels of the transmon system can be employed to act as a qubit and the remainder are leakage levels. For a typical transmon qubit [76–78], the transition frequency of the qubit is $\omega_0/2\pi = 5$ GHz, and the anharmonicity is $\Delta_2/2\pi = -200$ MHz. When adopting frequency modulation in Fig. 1(a), we exploit a frequency modulator with a square wave to generate $\delta/2\pi = -1.2$ GHz and $\delta'/2\pi = -2.4$ GHz. Thus, the period is $T = 0.625$ ns, the control parameter is $\lambda_x/2\pi = 66.7$ MHz, and the gate time of achieving the Y gate is $t_g = 18.22$ ns. When adopting strength modulation, we control only the applied voltage to keep $\lambda_x/2\pi = 66.7$ MHz and $\lambda_x'/2\pi = -66.7$ MHz. Thus, the period is $T = 3.90$ ns, the detuning is $\delta_1/2\pi = -240$ MHz, and the gate time of achieving the X gate is $t_g = 5.85$ ns. In addition, if we adopt strength modulation for the physical model in Fig. 1(b), we need to control the applied voltage to keep $\lambda_x/2\pi = 100$ MHz and $\lambda_x'/2\pi = -100$ MHz, and the detuning $\delta_1/2\pi = 4.8$ GHz. Then the period of two-step modulation is $T = 0.208$ ns, and the gate time of achieving the Y gate reads $t_g = 3.92$ ns.

Note that since the system dynamics of two-step modulation can be regarded as Rabi-like oscillation between qubit levels, different quantum gates can be implemented by choosing different gate times while the operation sequence is the same, namely, the two-step sequence. Particularly, the gate time is controllable, because it is the moment when we remove the two-step sequence. Thus there are variety ways of modulating control parameters to realize our scheme in the transmon qubit.

VI. ROBUSTNESS AGAINST NOISES

A. The effect of parameter perturbation

The universal way of qubit operations is to make the coupling between qubit levels in the resonance or near-resonance regime [79–81]. In these regimes, one of the main characteristics is that the error of qubit operations would dramatically increase with the existing strong perturbations in the control parameter. However, this situation can be significantly improved in two-step modulation, benefiting from highly detuning the qubit [$|\delta_1(t)| \gg \lambda_0$].

Suppose that the nonlinear oscillator exists with perturbation in control parameter λ_0, and the expression of $\lambda_0^{\rm per}$ reads

$$\lambda_0^{\rm per} = (1+\varepsilon)\lambda_0, \tag{22}$$

where ε quantifies the strength of unknown perturbation, and λ_0 is the exact value to calculate the time interval $\tau^{(\prime)}$ in frequency modulation. Figure 8(a) depicts the leakage error $F_{\rm error}$ versus the perturbation ε when we achieve the Y gate

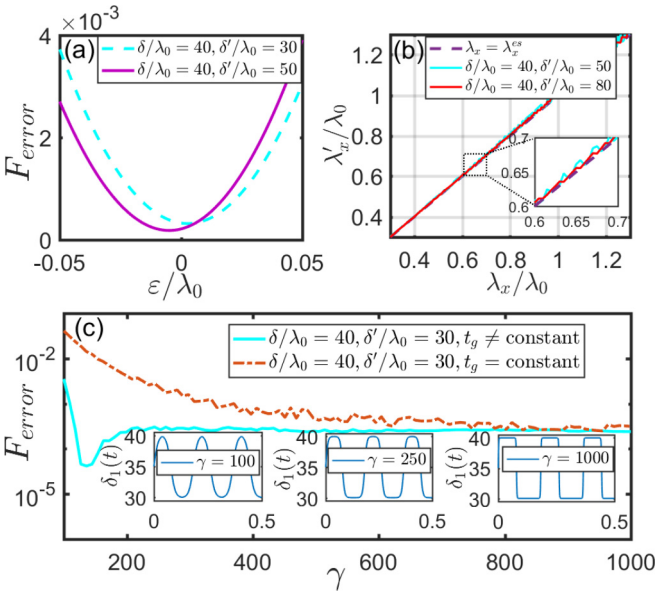

FIG. 8. (a) The leakage error F_{error} vs the perturbation ε in frequency modulation, where $\lambda_x/\lambda_0 = 1$, $\Delta_2/\lambda_0 = 3$, $\eta_2/\lambda_0 = \sqrt{2}$. (b) The unknown exact value λ_x vs the estimated value λ_x^{est} under different frequency modulations, where the purple-dashed line satisfies the equation $\lambda_x = \lambda_x^{\text{est}}$, and the other parameters are $\lambda_x/\lambda_0 = 1$, $\Delta_2/\lambda_0 = 3$, $\eta_2/\lambda_0 = \sqrt{2}$. (c) The leakage error F_{error} vs the hardness γ in frequency modulation, where the dashed-dot orange line denotes the value of F_{error} at the same gate time, the solid-cyan line denotes the minimum value of F_{error} during the modulation process, and the inset denotes the concrete shape of square waves with different γ. The other parameters are $\lambda_x/\lambda_0 = 1$, $\Delta_2/\lambda_0 = 3$, $\eta_2/\lambda_0 = \sqrt{2}$.

by different frequency modulations. The results demonstrate that F_{error} is still below 4×10^{-3}, even when $\varepsilon/\lambda_0 = \pm 5\%$, verifying that it makes little difference in the presence of perturbation. Hence, this modulation method is robust against the perturbation of control parameter λ_0.

What is more, one can employ this robustness property to measure the unknown coupling strength of control parameters by frequency modulation. In the large detuning regime $[|\delta_1(t)| \gg \lambda_0]$, we directly choose $\tau = \frac{\pi}{\delta}$ and $\tau' = \frac{\pi}{\delta'}$, since the coupling strength of the control parameters hardly affects the energy gap of the system. Due to the robust performance of time intervals τ and τ', which is shown in Fig. 7, frequency modulation is still valid and the leakage from the qubit to other levels is sharply suppressed, even though the coupling strength of the control parameter is unknown. Then, according to Eq. (10), the populations of qubit levels would emerge with *Rabi-like* oscillation, and the oscillation period $\mathcal{T}_R$ can be readily acquired by means of experimental techniques. Hence the unknown coupling strength of the control parameter λ_x can be estimated as

$$\lambda_x^{\text{est}} = \frac{\pi^2(\delta' + \delta)}{2\mathcal{T}_R(\delta' - \delta)}. \tag{23}$$

The advantage of this modulation is that we can measure the coupling strength repeatedly by choosing different values of detunings. In Fig. 8(b) we plot the unknown exact value λ_x versus the estimation value λ_x^{est}. One easily observes that all lines basically coincide with each other, and the estimated expression of λ_x^{est} would be more accurate if the exact value of λ_x were smaller. Thus, Eq. (23) is a validity expression to estimate the unknown coupling strength of control parameter λ_x in frequency modulation.

B. The effect of wave-form deformation

In a realistic modulation process, perfect square waves may not be readily obtained. Here we study this effect on the leakage error of qubit operations by an approximate square waves, whose expression reads

$$\delta_1(t) = \begin{cases} \delta' + \frac{\delta - \delta'}{1 + e^{-\gamma \text{mod}(t/T)}}, & \text{mod}(t/T) < \frac{\tau}{2}, \\ \delta' + \frac{\delta - \delta'}{1 + e^{\gamma[\text{mod}(t/T) - \tau]}}, & \frac{\tau}{2} \leqslant \text{mod}(t/T) \leqslant T - \frac{\tau'}{2}, \\ \delta' + \frac{\delta - \delta'}{1 + e^{-\gamma[\text{mod}(t/T) - T]}}, & \text{mod}(t/T) > T - \frac{\tau'}{2}. \end{cases} \tag{24}$$

The dimensionless parameter γ characterizes the hardness of square waves. That is, the shape of Eq. (24) approaches square waves when γ is large, and it becomes perfect square wave if $\gamma \to \infty$. Figure 8(c) represents the leakage error F_{error} as a function of γ in frequency modulation, and the results demonstrate that even when γ is small (e.g., $\gamma = 100$), which means the square wave is in heavy deformation, the leakage error would not be distinctly enlarged. In particular, the leakage error stays extremely low with heavy deformation by suitably selecting the gate time, as shown by the solid-cyan line in Fig. 8(c). Therefore, this modulation is also robust against the deformation of the square wave.

C. The effect of decoherence

So far we have not considered the influence of decoherence of qubit operations. In the presence of decoherence, the Lindblad master equation can be expressed as

$$\begin{aligned} \dot{\rho} = &-i[H_0(t), \rho] + \sum_{k=1,2} \frac{\gamma_k}{2}(2\sigma_{k-1,k}\rho\sigma_{k,k-1} \\ &- \sigma_{k,k-1}\sigma_{k-1,k}\rho - \rho\sigma_{k,k-1}\sigma_{k-1,k}) \\ &+ \frac{\Gamma_k}{2}(2\sigma_{k,k}\rho\sigma_{k,k} - \sigma_{k,k}\rho - \rho\sigma_{k,k}), \end{aligned} \tag{25}$$

where $\sigma_{n',m'} = |n'\rangle\langle m'|$, and γ_k and Γ_k denote the dissipation and dephasing rate of level $|k\rangle$, respectively. Note that the master equation (25) describes the evolution of density matrix rather than evolution operator; thus we set the initial state of the system as $|0\rangle$. When we choose the gate time $t_g = \frac{\pi}{2\lambda_{\text{eff}}}$, the final state would be $|1\rangle$. Figures 9(a) and 9(b) depict the influence of decoherence on the population P_1 of qubit level $|1\rangle$ in strength modulation. It can be easily found that the effect of decoherence induced by qubit level $|1\rangle$ is much more serious than that induced by leakage level $|2\rangle$, because the population of leakage level $|2\rangle$ is dramatically suppressed during the modulation process. In addition, the population P_1 can also reach a high value, even when both the dissipation rate γ_1 and the dephasing rate Γ_1 are large, which benefits from the short evolution time of the modulation process.

VII. CONCLUSION

In conclusion, we have demonstrated that universal quantum gates can be achieved by two-step modulation, even

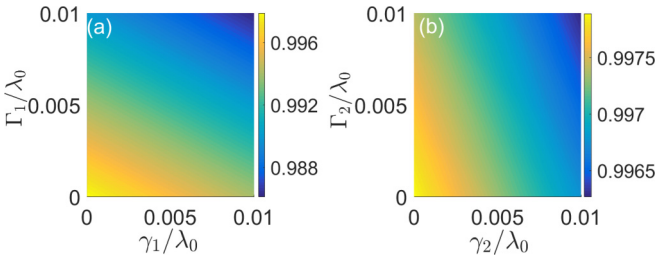

FIG. 9. (a) The population P_1 vs the dissipation rate γ_1 and the dephasing rate Γ_1 of qubit level $|1\rangle$ by strength modulation, where $\lambda_x/\lambda_0 = 1$, $\lambda_x'/\lambda_0 = -1$, $\lambda_y = 0$, $\Delta_2/\lambda_0 = 3$, $\delta_1/\lambda_0 = 3.6$, $\eta_2 = \sqrt{2}$, $d = 3$. (b) The population P_2 vs the dissipation rate γ_2 and the dephasing rate Γ_2 of leakage level $|2\rangle$ by strength modulation, where the parameters are the same as inset (a).

though the anharmonicity of the nonlinear qubit is very weak and the coupling strength is unknown. We have deduced the expression of the effective Hamiltonian by second-order perturbation theory and shown that the exact dynamical behaviors of two-step modulation can be well described by the effective Hamiltonian in a long timescale and the phenomenological evolution operator in whole process. The modulation shape is extremely common, namely, square wave. Particularly, the deformation of the square wave would not substantially increase the error of qubit operations but change the gate time. The purpose of artificially adding multiple large detunings is to sharply suppress the leakage from qubit to other levels and make qubit operations robust against the perturbation of control parameters. Thus, the nonlinear oscillator can be validly reduced to a two-level system, i.e., qubit.

This two-step modulation method would have wide applications in quantum physics, since the system is no longer limited to resonance conditions and the values of control parameters are controllable and adjustable. Note that the advantages of two-step modulation originate from the large detuning between qubit levels and are suitable for a general three-level system rather than a specific system. While this work mainly focuses on universal control of qubits and measuring the coupling strength of control parameters, it can also find applications in other systems, such as the atomic system. In a word, the periodic two-step modulation method would offer us a significant step forward in the development of qubits, which is feasible for quantum information processing.

ACKNOWLEDGMENTS

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APPENDIX A: GENERAL FORM FOR THE EFFECTIVE HAMILTONIAN IN TWO-STEP MODULATION: DERIVATION OF EQS. (10) AND (12)

In this Appendix, we first demonstrate that the Baker-Campbell-Hausdorff formula cannot be employed to calculate the time-independent effective Hamiltonian in this physical model. For simplicity, we only consider a special case of frequency modulation, where the detuning $\delta_1(t)$ is periodically adjusted by the following repeated two-step sequence,

$$\delta_1(t) = \begin{cases} \delta_1, & t \in \left[mT, mT + \frac{T}{2}\right], \\ -\delta_1, & t \in \left(mT + \frac{T}{2}, (m+1)T\right]. \end{cases} \tag{A1}$$

As a result, the effective Hamiltonian satisfies the following equation:

$$e^{-iH_{\text{eff}}T} = e^{-iH_0'\frac{T}{2}} e^{-iH_0\frac{T}{2}}. \tag{A2}$$

By using the Baker-Campbell-Hausdorff formula,

$$e^A e^B = \exp\left(A + B + \tfrac{1}{2}[A,B] + \tfrac{1}{12}[A-B,[A,B]] + \cdots\right), \tag{A3}$$

we can yield the expression of the effective Hamiltonian

$$\begin{aligned} H_{\text{eff}} &= H_{\text{eff},0} + H_{eff,1} + O(T^2), \\ H_{\text{eff},0} &= \lambda_x|1\rangle\langle 0| + \eta_2\lambda_x|2\rangle\langle 1|, \\ H_{\text{eff},1} &= -i\frac{T}{4}[\lambda_x\delta_1|0\rangle\langle 1| + \eta_2\lambda_x(\delta_1-\delta_2)|2\rangle\langle 1|] + \text{H.c.}, \end{aligned} \tag{A4}$$

where $H_{\text{eff},0}$ and $H_{\text{eff},1}$ represent the zero-order term and first-order term of the effective Hamiltonian. As shown in Eq. (7) of the main text, the square-wave period is approximately equal to

$$\begin{aligned} T = \tau + \tau' &= \frac{(2n+1)\pi}{E_1 - E_0} + \frac{(2n'+1)\pi}{E_1' - E_0'} \\ &\approx \frac{(2n+1)\pi}{\delta_1} + \frac{(2n'+1)\pi}{\delta_1}. \end{aligned} \tag{A5}$$

When we choose $n = n' = 0$, the period reads $T \approx \frac{2\pi}{\delta_1}$. Then we substitute the period T into the first-order term of the effective Hamiltonian $H_{\text{eff},1}$, and we find that

$$\begin{aligned} H_{\text{eff},1} &= i\frac{T}{4}[\lambda_x\delta_1|0\rangle\langle 1| + \eta_2\lambda_x(\delta_1-\delta_2)|2\rangle\langle 1|] \\ &\approx -i\frac{\pi}{2}\left[\lambda_x|0\rangle\langle 1| + \eta_2\lambda_x\left(1 - \frac{\delta_2}{\delta_1}\right)|2\rangle\langle 1|\right]. \end{aligned} \tag{A6}$$

Remarkably, the first-order term $H_{\text{eff},1}$ has the same magnitude as the zero-order term $H_{\text{eff},0}$. Therefore the effective Hamiltonian (A4) is diverging, and it is invalid for employing the Baker-Campbell-Hausdorff formula in this system.

In order to obtain the effective Hamiltonian of the nonlinear oscillator, one can first calculate the evolution operator $U(T) = e^{-iH_0'\tau'}e^{-iH_0\tau}$ within one period. Then by inversely solving the equation $U(T) = e^{-iH_{\text{eff}}T}$, we naturally achieve the effective Hamiltonian H_{eff}. In the following, we introduce this method in detail.

Since the Hamiltonian of the two-step modulation can be regarded as two static Hamiltonians H_0 and H_0', we only need to study one of the evolution operators (e.g., $e^{-iH_0\tau}$) and the form of another evolution operator would be similar. At first, one should achieve the eigenstates and eigenvalues of the Hamiltonian H_0 in Eq. (3) of the main text in order to obtain the analytical expression of the evolution operator $e^{-iH_0\tau}$.

If the system satisfies the two-photon resonance condition, i.e., $\delta_2 = 0$, the analytical expressions for the eigenstates and eigenvalues of the Hamiltonian H_0 are extremely simple. Nevertheless, in this work, we do not require this rigorous condition, i.e., δ_2 can be arbitrary but nonzero. As a result, the expressions of eigenstates become intricate, and they are quite useless for us. Thus we solve it by standard perturbation theory.

When $\lambda_0 \ll \delta_1$ (i.e., the large detuning condition), according to the second-order perturbation theory, the eigenvalues E_k and eigenstates $|E_k\rangle$ $(k = 0, 1, 2)$ of the Hamiltonian H_0 approximatively read $(d = 3)$

$$\begin{aligned} E_0 &\simeq -\delta_1 a_1^2, & |E_0\rangle &\simeq |0\rangle - a_1|1\rangle, \\ E_1 &\simeq \delta_1 + \delta_1 a_1^2 - (\delta_2 - \delta_1)a_2^2, & |E_1\rangle &\simeq a_1|0\rangle + |1\rangle - a_2|2\rangle, \\ E_2 &\simeq \delta_2 + (\delta_2 - \delta_1)a_2^2, & |E_2\rangle &\simeq a_2|1\rangle + |2\rangle, \end{aligned} \tag{A7}$$

where $a_1 = \frac{\lambda_0}{\delta_1} \ll 1$, $a_2 = \frac{\eta_2\lambda_0}{\delta_1+\Delta} \ll 1$, and the coefficient of eigenstates is not normalization. As a result, the evolution operator of the system becomes

$$\begin{aligned} U(t) = e^{-iH_0t} &= \sum_{k=0}^{2} e^{-iE_kt}|E_k\rangle\langle E_k| \\ &= e^{-iE_0t}(|0\rangle - a_1|1\rangle)(\langle 0| - a_1\langle 1|) + e^{-iE_1t}(a_1|0\rangle + |1\rangle - a_2|2\rangle)(a_1\langle 0| + \langle 1| - a_2\langle 2|) + e^{-iE_2t}(a_2|1\rangle + |2\rangle)(a_2\langle 1| + \langle 2|) \\ &\simeq e^{-iE_0t}[(|0\rangle\langle 0| - a_1|1\rangle\langle 0| - a_1|0\rangle\langle 1|) + e^{-i(E_1-E_0)t}(|1\rangle\langle 1| + a_1|1\rangle\langle 0| - a_2|1\rangle\langle 2| + a_1|0\rangle\langle 1| - a_2|2\rangle\langle 1|) \\ &\quad + e^{-i(E_2-E_0)t}(|2\rangle\langle 2| + a_2|1\rangle\langle 2| + a_2|2\rangle\langle 1|)] \\ &= e^{-iE_0t}[|0\rangle\langle 0| + e^{-i\Theta_1}|1\rangle\langle 1| + e^{-i\Theta_2}|2\rangle\langle 2| + a_1(e^{-i\Theta_1} - 1)(|1\rangle\langle 0| + |0\rangle\langle 1|) + a_2(e^{-i\Theta_2} - e^{-i\Theta_1})(|1\rangle\langle 2| + |2\rangle\langle 1|)], \end{aligned}$$

where we ignore higher-order terms ($\sim a_1^2, a_2^2, a_1a_2$), and set $\Theta_1 = (E_1 - E_0)t$ and $\Theta_2 = (E_2 - E_0)t$. It is instructive to adopt the matrix form of the evolution operator in the basis $\{|0\rangle, |1\rangle, |2\rangle\}$, which reads

$$U(t) = e^{-iH_0t} = \begin{pmatrix} 1 & a_1(e^{-i\Theta_1} - 1) & 0 \\ a_1(e^{-i\Theta_1} - 1) & e^{-i\Theta_1} & a_2(e^{-i\Theta_2} - e^{-i\Theta_1}) \\ 0 & a_2(e^{-i\Theta_2} - e^{-i\Theta_1}) & e^{-i\Theta_2} \end{pmatrix}, \tag{A8}$$

where we ignore the global phase e^{-iE_0t}. Note that Eq. (A8) of the evolution operator $U(t)$ has general form under the static Hamiltonian H_0, and it is a starting point for two-step modulation.

Next, we need to determine the time intervals τ and τ' in two-step modulation, where the dynamical phase difference between qubit levels $|0\rangle$ and $|1\rangle$ of the system should be odd times of π, i.e., $\Theta_1 = (E_1 - E_0)\tau = (2n + 1)\pi$, $\Theta_1' = (E_1' - E_0')\tau = (2n' + 1)\pi$, $n, n' \in \mathbb{N}$. To be specific, when the time evolution of the system under static Hamiltonian H_0 satisfies $\Theta_1 = (2n + 1)\pi$, we immediately switch to the static Hamiltonian H_0', and when the time evolution of the system under the static Hamiltonian H_0' satisfies $\Theta_1' = (2n' + 1)\pi$, we immediately switch back to the static Hamiltonian H_0.

Note that there are several kinds of physical parameters that can become variable in two-step modulation, such as the quadrature control parameters $\lambda_x(t)$, $\lambda_y(t)$ or the detuning $\delta_1(t)$. Here, we enumerate two paradigms of them. One is that the quadrature control parameters are fixed (i.e., $\lambda_y = 0$ and λ_x is a constant) and the detuning is variable [cf. Eq. (5) in the main text], which we call frequency modulation. The other is that we set the detuning δ_1 to be fixed and the control parameter λ_x to be variable [cf. Eq. (11) in the main text, $\lambda_y = 0$], which is called strength modulation. For simplicity, we give the detailed derivative procedure of frequency modulation. According to Eq. (A8), in two-step modulation, the evolution operator within a period T reads

$$\begin{aligned} U(T) = U(\tau')U(\tau) = e^{-iH_0'\tau'}e^{-iH_0\tau} &= \begin{pmatrix} 1 & 2a_1 & 0 \\ 2a_1 & -1 & a_2(e^{-i\Theta_2} + 1) \\ 0 & a_2(e^{-i\Theta_2} + 1) & e^{-i\Theta_2} \end{pmatrix}\begin{pmatrix} 1 & 2a_1' & 0 \\ 2a_1' & -1 & a_2'(e^{-i\Theta_2'} + 1) \\ 0 & a_2'(e^{-i\Theta_2'} + 1) & e^{-i\Theta_2'} \end{pmatrix} \\ &= \begin{pmatrix} 1 & b_1 & 0 \\ -b_1 & 1 & b_2 \\ 0 & b_3 & b_4 \end{pmatrix}, \end{aligned} \tag{A9}$$

where $b_1 = 2(a_1 - a_1')$, $b_2 = a_2e^{-i\Theta_2'}(1 + e^{-i\Theta_2}) - a_2'(1 + e^{-i\Theta_2'})$, $b_3 = -a_2(1 + e^{-i\Theta_2}) + a_2'e^{-i\Theta_2}(1 + e^{-i\Theta_2'})$, $b_4 = e^{-i(\Theta_2+\Theta_2')}$, and we also ignore higher-order terms. It is worth mentioning again that the above approximations are valid on the large detuning condition ($\lambda_0 \ll |\delta_1|$). Since the diagonal terms $U_{11} = U_{22} \neq U_{33}$, the off-diagonal terms $\pm b_1$ are major contributions for the eigenstates of $U(T)$, and we then ignore the off-diagonal terms b_2 and b_3 (the

leakage caused by b_2 and b_3 can be canceled by composite two-step modulation, as shown in Appendix C). In addition, due to $b_1 \ll 1$, the equations $1 = \cos(b_1)$ and $b_1 = \sin(b_1)$ hold approximately. Therefore, the system dynamics can be regarded as Rabi-like oscillation in qubit levels, without stimulating other levels. On the other hand, suppose that the effective Hamiltonian of the nonlinear oscillator has the following form:

$$H_{\text{eff}} = i\lambda_{\text{eff}}|0\rangle\langle 1| - i\lambda_{\text{eff}}|1\rangle\langle 0|, \tag{A10}$$

where the unknown coefficient λ_{eff} is required to be solved. Hence, the effective evolution operator in the basis $\{|0\rangle, |1\rangle\}$ reads

$$U_{\text{eff}}(t) = e^{-iH_{\text{eff}}t} = \begin{pmatrix} \cos(\lambda_{\text{eff}}t) & \sin(\lambda_{\text{eff}}t) \\ -\sin(\lambda_{\text{eff}}t) & \cos(\lambda_{\text{eff}}t) \end{pmatrix}. \tag{A11}$$

According to the definition $U(T) \equiv e^{-iH_{\text{eff}}T}$ [71], we would obtain the equation $b_1 = \lambda_{\text{eff}}T$. As a result, we readily achieve the constant $\lambda_{\text{eff}} = \frac{2\lambda_0|\delta'-\delta|}{T\delta\delta'}$. Note that the derivative procedure of strength modulation is similar to that of frequency modulation, and the effective Rabi frequency is $\lambda'_{\text{eff}} = \frac{2(\lambda_x-\lambda'_x)}{T\delta}$.

APPENDIX B: PHENOMENOLOGICAL DESCRIPTION OF THE EVOLUTION OPERATOR IN TWO-STEP MODULATION

In this Appendix, we explain how to achieve the estimation expression of amplitudes in the evolution operator (15) in the main text. According to the effective Hamiltonian in Eq. (10) in the main text, the time evolutions of amplitudes are mainly dominated by the trigonometric functions: $\cos(\lambda_{\text{eff}}t)$ or $\sin(\lambda_{\text{eff}}t)$. On this basis, there should exist a time-dependent term $A(t)$ to describe rapid oscillation during two-step modulation. By making use of the normalization condition, we can immediately give the following estimation expressions: $|u^{\text{es}}_{11}(t)| = |u^{\text{es}}_{22}(t)| = \sqrt{|\cos^2(\lambda_{\text{eff}}t) - A(t)|}$ and $|u^{\text{es}}_{12}(t)| = |u^{\text{es}}_{21}(t)| = \sqrt{|\sin^2(\lambda_{\text{eff}}t) + A(t)|}$, where $A(t)$ is unknown. To determine the estimation expression of $A(t)$, we must be confronted with the following facts:

(i) The amplitude of $A(t)$ should be closely connected to the detunings $\delta^{(\prime)}$ of two-step modulation. That is, the amplitude of $A(t)$ is smaller when the detunings $\delta^{(\prime)}$ are larger, which is reflected in the term A_m.

(ii) The period of the rapidly oscillating term $A(t)$ should coincide exactly with the period of two-step modulation, which is reflected in the term $\sin^2[\frac{g(t)}{2}]$, where $g(t)$ represents a periodic function with the period T.

(iii) By carefully observing the evolution process of two-step modulation, we find that the amplitude of rapid oscillation is not always invariable and approaches zero in the vicinity of $\lambda_{\text{eff}}t = m\pi$, $m \in \mathbb{N}$. Thus, the value of $A(t)$ is also determined by the effective "Rabi frequency," which is reflected in the term $\sin(2\lambda_{\text{eff}}t)$.

According to the above facts and combining with numerical simulations, the estimation expression of $A(t)$ is given by $A(t) = A_m \sin^2[\frac{g(t)}{2}]\sin(2\lambda_{\text{eff}}t)$, where $A_m = \frac{\lambda_x(\delta+\delta')}{\delta\delta'}$ and $g(t) = \{^{-\frac{\pi}{\tau}t,}_{\frac{\pi}{\tau'}(T-t),} \; ^{t\in[mT,mT+\tau]}_{t\in(mT+\tau,(m+1)T]}$. The numerical calculations in Fig. 4(a) of the main text show that the estimation expression of amplitudes is describes the actual evolution process well, since the value of average error is considerably small, on the order of 10^{-3}. Then we briefly explain the estimation expression of phases in the evolution operator. For the off-diagonal term $\Theta^{\text{es}}_{21}(t)$, one can easily find that the value linearly increases with the evolution time when ignoring the period T, which is reflected in the term $g(t) = \{^{-\frac{\pi}{\tau}t,}_{\frac{\pi}{\tau'}(T-t),} \; ^{t\in[mT,mT+\tau]}_{t\in(mT+\tau,(m+1)T]}$. However, there exists a phase transition from 0 (π) to π (0) in the vicinity of the evolution time $t = m\pi/\lambda_{\text{eff}}, m \in \mathbb{N}$, which is described by the term $\arctan\vartheta$. So the estimation expression of $\Theta^{\text{es}}_{22}(t)$ reads $\Theta^{\text{es}}_{22}(t) = g(t) - \arctan\vartheta - \frac{\pi}{2}$. Combining with numerical simulations, the estimation expression of $\Theta^{\text{es}}_{11}(t)$ is given by $\Theta^{\text{es}}_{11}(t) = -[\frac{1}{\sqrt{2}}A_m(\sin g(t) - \frac{1}{2}) + 1]\arctan\vartheta - \frac{\pi}{2}$. Note that the variable range of the actual value of phases is large in the vicinity of evolution time $t = m\pi/\lambda_{\text{eff}}, m \in \mathbb{N}$, leading to the generation of a large error for the estimation expressions. As a result, the average errors of phases are accordingly larger than those of amplitudes.

Since $U(T)$ is a unitary matrix, there exists a unitary transformation $V = \begin{pmatrix} V_{11} & V_{12} & V_{13} \\ V_{21} & V_{22} & V_{23} \\ V_{31} & V_{32} & V_{33} \end{pmatrix}$ to make it diagonalization, i.e.,

$$VU(T)V^\dagger = \begin{pmatrix} e^{i(b_1+\epsilon_1)} & 0 & 0 \\ 0 & e^{-i(b_1+\epsilon_2)} & 0 \\ 0 & 0 & e^{-i(\Theta_2+\Theta'_2+\epsilon_3)} \end{pmatrix}, \tag{B1}$$

where $\epsilon_k \ll b_k \in \mathbb{R}$ is a small quantity, $k = 1, 2, 3$. For the expression of Eq. (15) in the main text, we do not take into account the leakage from the qubit to other levels, i.e., $V_{31} = V_{13} = V_{32} = V_{23} = 0$. When considering the leakage, the correction of amplitude $|u_{12}(t)|$ approximately reads $|u^{\text{co}}_{12}(t)| \simeq |u^{\text{co}}_{21}(t)| = \sqrt{1-2\xi^2}|u^{\text{es}}_{21}(t)|$, where $\xi = |\frac{V_{31}}{V_{21}}|$ can be determined by the secular equation of the unitary matrix $U(T)$.

APPENDIX C: FURTHER LEAKAGE REMOVAL BY COMPOSITE TWO-STEP MODULATION

In the above section, in order to deduce the effective Hamiltonian (A10), we have ignored the leakage caused by the off-diagonal terms b_2 and b_3. Generally speaking, the off-diagonal terms b_2 and b_3 are the main source of qubit leakage in two-step modulation. Next we demonstrate that the leakage caused by the off-diagonal terms b_2 and b_3 can be canceled by composite two-step modulation.

At first, we need to proof that the phase θ of the control parameter λ_0 only affects the phase of off-diagonal terms of the evolution operator in two-step modulation. Suppose that the Hamiltonian of the nonlinear oscillator reads

$$H_0 = \begin{pmatrix} 0 & \lambda_0 e^{i\theta} & 0 \\ \lambda_0 e^{-i\theta} & \delta_1 & \eta_2\lambda_0 e^{i\theta} \\ 0 & \eta_2\lambda_0 e^{-i\theta} & \delta_2 \end{pmatrix}. \tag{C1}$$

It is easy to calculate that the eigenvalues E_k $(k = 0, 1, 2)$ of H_0 are roots of the following cubic equation:

$$E_k^3 - (\delta_1 + \delta_2)E_k^2 + \left(\delta_1\delta_2 - \lambda_0^2 - \eta_2^2\lambda_0^2\right)E_k + \delta_2\lambda_0^2 = 0. \tag{C2}$$

One can find that the eigenvalues E_k do not show any connection to the phase θ of the control parameter λ_0. Furthermore, we can also give the expression of eigenstates $|E_k\rangle$, i.e.,

$$\begin{aligned}|E_k\rangle = N_k[&(E_k - \delta_2)\lambda_0 e^{i\theta}|1\rangle + E_k(E_k - \delta_2)|2\rangle \\ &+ E_k\eta_2\lambda_0 e^{-i\theta}|3\rangle],\end{aligned} \tag{C3}$$

where N_k is the normalization constant. As a result, the evolution operator by static Hamiltonian H_0 reads

$$\begin{aligned}U(\tau) = e^{-iH_0\tau} &= \sum_{k=0}^{2} e^{-iE_k\tau}|E_k\rangle\langle E_k| \\ &= \begin{pmatrix} S_{11}e^{i\theta} & S_{12}e^{i\theta} & S_{13}e^{i\theta} \\ S_{21} & S_{22} & S_{23} \\ S_{31}e^{-i\theta} & S_{32}e^{-i\theta} & S_{33}e^{-i\theta}\end{pmatrix}\begin{pmatrix}1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & e^{-i\Theta_2}\end{pmatrix}\begin{pmatrix} S_{11}e^{-i\theta} & S_{12} & S_{13}e^{i\theta} \\ S_{21}e^{-i\theta} & S_{22} & S_{23}e^{i\theta} \\ S_{31}e^{-i\theta} & S_{32} & S_{33}e^{i\theta}\end{pmatrix} \\ &= \begin{pmatrix} \mathbb{S}_1 & \mathbb{S}_4e^{i\theta} & \mathbb{S}_5e^{i2\theta} \\ \mathbb{S}_4e^{-i\theta} & \mathbb{S}_2 & \mathbb{S}_6e^{i\theta} \\ \mathbb{S}_5e^{-i2\theta} & \mathbb{S}_6e^{-i\theta} & \mathbb{S}_3\end{pmatrix},\end{aligned} \tag{C4}$$

where

$$\begin{aligned}&S_{1k} = N_k(E_k - \delta_2)\lambda_0, \quad S_{2k} = N_kE_k(E_k - \delta_2), \quad S_{3k} = N_kE_k\eta_2\lambda_0, \\ &\mathbb{S}_1 = S_{11}^2 - S_{12}^2 + e^{-i\Theta_2}S_{13}^2, \ \mathbb{S}_4 = S_{11}S_{21} - S_{12}S_{22} + e^{-i\Theta_2}S_{13}S_{23}, \\ &\mathbb{S}_2 = S_{21}^2 - S_{22}^2 + e^{-i\Theta_2}S_{23}^2, \ \mathbb{S}_5 = S_{11}S_{31} - S_{12}S_{32} + e^{-i\Theta_2}S_{13}S_{33}, \\ &\mathbb{S}_3 = S_{31}^2 - S_{32}^2 + e^{-i\Theta_2}S_{33}^2, \ \mathbb{S}_6 = S_{21}S_{31} - S_{22}S_{32} + e^{-i\Theta_2}S_{23}S_{33}.\end{aligned}$$

We also find that the values of $\mathbb{S}_l (l = 1, ..., 6)$ are not affected by the phase θ of the control parameter λ_0. Furthermore, the evolution operator by two-step modulation reads

$$\begin{aligned}U(T) = U(\tau_1')U(\tau_1) = e^{-iH_0'\tau'}e^{-iH_0\tau} &= \begin{pmatrix} \mathbb{S}_1' & \mathbb{S}_4'e^{i\theta} & \mathbb{S}_5'e^{i2\theta} \\ \mathbb{S}_4'e^{-i\theta} & \mathbb{S}_2' & \mathbb{S}_6'e^{i\theta} \\ \mathbb{S}_5'e^{-i2\theta} & \mathbb{S}_6'e^{-i\theta} & \mathbb{S}_3'\end{pmatrix}\begin{pmatrix} \mathbb{S}_1 & \mathbb{S}_4e^{i\theta} & \mathbb{S}_5e^{i2\theta} \\ \mathbb{S}_4e^{-i\theta} & \mathbb{S}_2 & \mathbb{S}_6e^{i\theta} \\ \mathbb{S}_5e^{-i2\theta} & \mathbb{S}_6e^{-i\theta} & \mathbb{S}_3\end{pmatrix} \\ &= \begin{pmatrix} \mathcal{S}_{11} & \mathcal{S}_{12}e^{i\theta} & \mathcal{S}_{13}e^{i2\theta} \\ \mathcal{S}_{21}e^{-i\theta} & \mathcal{S}_{22} & \mathcal{S}_{23}e^{i\theta} \\ \mathcal{S}_{31}e^{-i2\theta} & \mathcal{S}_{32}e^{-i\theta} & \mathcal{S}_{33}\end{pmatrix},\end{aligned} \tag{C5}$$

where

$$\begin{aligned}&\mathcal{S}_{11} = \mathbb{S}_1\mathbb{S}_1' + \mathbb{S}_4\mathbb{S}_4' + \mathbb{S}_5\mathbb{S}_5', \quad \mathcal{S}_{12} = \mathbb{S}_4\mathbb{S}_2' + \mathbb{S}_1\mathbb{S}_4' + \mathbb{S}_5\mathbb{S}_6', \quad \mathcal{S}_{13} = \mathbb{S}_5\mathbb{S}_3' + \mathbb{S}_1\mathbb{S}_5' + \mathbb{S}_4\mathbb{S}_6', \\ &\mathcal{S}_{21} = \mathbb{S}_4\mathbb{S}_1' + \mathbb{S}_2\mathbb{S}_4' + \mathbb{S}_6\mathbb{S}_5', \quad \mathcal{S}_{22} = \mathbb{S}_2\mathbb{S}_2' + \mathbb{S}_4\mathbb{S}_4' + \mathbb{S}_6\mathbb{S}_6', \quad \mathcal{S}_{23} = \mathbb{S}_6\mathbb{S}_3' + \mathbb{S}_4\mathbb{S}_5' + \mathbb{S}_2\mathbb{S}_6', \\ &\mathcal{S}_{31} = \mathbb{S}_5\mathbb{S}_1' + \mathbb{S}_6\mathbb{S}_4' + \mathbb{S}_3\mathbb{S}_5', \quad \mathcal{S}_{32} = \mathbb{S}_6\mathbb{S}_2' + \mathbb{S}_5\mathbb{S}_4' + \mathbb{S}_3\mathbb{S}_6', \quad \mathcal{S}_{33} = \mathbb{S}_3\mathbb{S}_3' + \mathbb{S}_5\mathbb{S}_5' + \mathbb{S}_6\mathbb{S}_6'.\end{aligned}$$

Again, we find that the values of $\mathcal{S}_{mn}(m, n = 1, 2, 3)$ are not affected by phase θ of the control parameter λ_0. In other words, phase θ of the control parameter λ_0 affects only the phase of off-diagonal terms of the evolution operator in two-step modulation. Note that Eq. (C5) is the exact expression of the evolution operator in two-step modulation, and Eq. (A9) is the approximate expression when $\theta = 0$ and $\lambda_0 \ll \delta_1$.

To cancel the leakage caused by the off-diagonal terms $\mathcal{S}_{23}(\simeq b_2)$ and $\mathcal{S}_{32}(\simeq b_3)$ in Eq. (C5), we need to introduce another evolution operator of two-step modulation, which has a phase θ on the control parameter λ_0 while other parameters remain unchanged. Hence, the period of composite two-step modulation is $2T$ now, and the evolution operator of the system reads

$$\mathcal{U}(2T) = U(T)U'(T) = \begin{pmatrix} \mathcal{S}_{11} & \mathcal{S}_{12} & \mathcal{S}_{13} \\ \mathcal{S}_{21} & \mathcal{S}_{22} & \mathcal{S}_{23} \\ \mathcal{S}_{31} & \mathcal{S}_{32} & \mathcal{S}_{33}\end{pmatrix}\begin{pmatrix} \mathcal{S}_{11} & \mathcal{S}_{12}e^{i\theta} & \mathcal{S}_{13}e^{i2\theta} \\ \mathcal{S}_{21}e^{-i\theta} & \mathcal{S}_{22} & \mathcal{S}_{23}e^{i\theta} \\ \mathcal{S}_{31}e^{-i2\theta} & \mathcal{S}_{32}e^{-i\theta} & \mathcal{S}_{33}\end{pmatrix} = \begin{pmatrix} \mathcal{S}_{11}' & \mathcal{S}_{12}' & \mathcal{S}_{13}' \\ \mathcal{S}_{21}' & \mathcal{S}_{22}' & \mathcal{S}_{23}' \\ \mathcal{S}_{31}' & \mathcal{S}_{32}' & \mathcal{S}_{33}'\end{pmatrix}, \tag{C6}$$

where

$$\mathcal{S}_{12}' = \mathcal{S}_{22}\mathcal{S}_{12} + e^{i\theta}\mathcal{S}_{11}\mathcal{S}_{12} + e^{-i\theta}\mathcal{S}_{13}\mathcal{S}_{32}, \ \mathcal{S}_{21}' = \mathcal{S}_{11}\mathcal{S}_{21} + e^{-i\theta}\mathcal{S}_{21}\mathcal{S}_{22} + e^{-i2\theta}\mathcal{S}_{23}\mathcal{S}_{31},$$

$$\mathcal{S}'_{13} = \mathcal{S}_{13}\mathcal{S}_{33} + e^{i\theta}\mathcal{S}_{12}\mathcal{S}_{23} + e^{i2\theta}\mathcal{S}_{11}\mathcal{S}_{13}, \quad \mathcal{S}'_{31} = \mathcal{S}_{11}\mathcal{S}_{31} + e^{-i\theta}\mathcal{S}_{21}\mathcal{S}_{32} + e^{-i2\theta}\mathcal{S}_{31}\mathcal{S}_{33},$$
$$\mathcal{S}'_{23} = \mathcal{S}_{23}\mathcal{S}_{33} + e^{i\theta}\mathcal{S}_{22}\mathcal{S}_{23} + e^{i2\theta}\mathcal{S}_{13}\mathcal{S}_{21}, \quad \mathcal{S}'_{32} = \mathcal{S}_{22}\mathcal{S}_{32} + e^{i\theta}\mathcal{S}_{12}\mathcal{S}_{31} + e^{-i\theta}\mathcal{S}_{32}\mathcal{S}_{33},$$
$$\mathcal{S}'_{11} = \mathcal{S}^2_{11} + e^{-i\theta}\mathcal{S}_{12}\mathcal{S}_{21} + e^{-i2\theta}\mathcal{S}_{13}\mathcal{S}_{31}, \quad \mathcal{S}'_{22} = \mathcal{S}^2_{22} + e^{i\theta}\mathcal{S}_{12}\mathcal{S}_{21} + e^{-i\theta}\mathcal{S}_{23}\mathcal{S}_{32},$$
$$\mathcal{S}'_{33} = \mathcal{S}^2_{33} + e^{i\theta}\mathcal{S}_{23}\mathcal{S}_{32} + e^{i2\theta}\mathcal{S}_{13}\mathcal{S}_{31}.$$

In the large detuning condition, it has the relation $|\mathcal{S}_{13}|, |\mathcal{S}_{31}| \ll |\mathcal{S}_{12}|, |\mathcal{S}_{12}|, |\mathcal{S}_{23}|, |\mathcal{S}_{32}| \ll |\mathcal{S}_{11}|, |\mathcal{S}_{22}|, |\mathcal{S}_{33}|$. Thus we can safely ignore the off-diagonal terms $\mathcal{S}'_{13}$ and $\mathcal{S}'_{31}$. To eliminate the off-diagonal terms $\mathcal{S}'_{23}$ and $\mathcal{S}'_{33}$, one should satisfy the following equations:

$$\mathcal{S}_{23}(\mathcal{S}_{33} + e^{i\theta}\mathcal{S}_{22}) + e^{i2\theta}\mathcal{S}_{13}\mathcal{S}_{21} = 0,$$
$$\mathcal{S}_{32}(\mathcal{S}_{33} + e^{i\theta}\mathcal{S}_{22}) + e^{i2\theta}\mathcal{S}_{31}\mathcal{S}_{12} = 0. \tag{C7}$$

Due to $|\mathcal{S}_{13}| \simeq |\mathcal{S}_{31}| \simeq 0$, we choose the phase θ only to satisfy the equation $(\mathcal{S}_{33} + e^{i\theta}\mathcal{S}_{22}) = 0$. On the other hand, $|\mathcal{S}_{22}| \simeq |\mathcal{S}_{33}| \simeq 1$, and the equation can be further reduced as $|\mathcal{S}_{33}|(1 + e^{i(\theta+\vartheta')}) = 0$, where ϑ' denotes the phase difference between $\mathcal{S}_{33}$ and $\mathcal{S}_{22}$. Hence, when $\theta = \pi - \vartheta'$, we can cancel the leakage caused by the off-diagonal terms $\mathcal{S}'_{23}$ and $\mathcal{S}'_{32}$ in the evolution operator in Eq. (C6). Note that the most convenient way to find this specific θ is to employ numerical calculations. That is, we reversely calculate the amplitudes of $\mathcal{S}'_{23}$ and $\mathcal{S}'_{32}$ for all $\theta \in [0, 2\pi]$. Then we naturally achieve this specific θ corresponding to the minimum value of $|\mathcal{S}'_{23}|$ and $|\mathcal{S}'_{32}|$.

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