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Hook-and-Loop laser fastening of an optical frequency comb to a strongly driven two-level quantum system

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ABSTRACT

In order to control the state of a two-level quantum system (e.g. the spin state of an ion qubit), optical frequency combs perform a two-photon stimulated Raman process through stimulated absorption from one comb tooth and stimulated emission into another comb tooth. If the two-level energy gap is an integer multiple of the repetition rate of the laser, resonant Rabi oscillations are excited. When these latter have a frequency close to the qubit's transition one, a strongly anharmonic phase-locked cycle may exist on the Bloch sphere, which generates a sub-harmonic series of very narrow, equally spaced, spectral lines. If the repetition rate of an optical frequency comb is appropriately tuned to these latter (up to the average carrier envelope frequency), a highly resonant dynamical regime of the two-level system should be reached where the Raman stimulated absorption and emission processes would occur for any pair of adjacent comb teeth.

By pointing out new spectral properties of the energy of a strongly-driven two-level quantum system:

$$E(t) = \langle \Psi(t)|\mathbf{H}(t)|\Psi(t)\rangle, \tag{1}$$

that is defined by its Hamiltonian:

$$\mathbf{H} = \mathcal{K}\sigma_x + \mathcal{E}(t)\sigma_z, \tag{2}$$

(where σ_x and σ_z are the two Pauli matrices) and by its two-component normalized spinor wave function $\Psi = \{\psi_a, \psi_b\}$, the present paper aims to contribute to the continuous feedback between basic science and technology [1] in the simplest paradigmatic case of a harmonic resonant drive:

$$\frac{\mathcal{E}(t)}{\mathcal{K}} = A \sin \Omega t. \tag{3}$$

The Larmor angular frequency $\Omega = 2\mathcal{K}/\hbar$ defines the splitting energy $2\mathcal{K}$ of the system. The $A \ll 1$ weakly-driven case is a well-known example of the use of the rotating wave approximation in order to display the harmonic Rabi oscillations of E between the two eigenstates $\pm\mathcal{K}$ of the undriven system. The periodicity of the driving field (3) allows the use of the Floquet theorem where the eigenstates now appear in the form of the product of a phase factor linear in time, defined by its constant Floquet quasienergy, and an oscillating amplitude – the normalized Floquet state – at the driving frequency Ω. In the undriven limit $A \to 0$, these states oscillate very rapidly at $\Omega \gg \Omega_R$ where $\Omega_R = \frac{1}{2}A\Omega$, which defines the quasienergy, is the angular Rabi frequency of the system. Therefore, within this high-frequency limit (or rotating wave approximation), they may be approximated as constant in time. Note that recently [2], an *exact* (i.e. *without* the use of rotating wave approximation) Hamiltonian description was performed in the $A \ll 1$ weak drive limit which led to the explanation – based on the quantum Zeno effect – of the abrupt energy flipping which has been observed in a continuously observed Rabi driven quantum system [3].

The present paper considers the $A \sim 1$ opposite case of a strongly driven two-level system where Ω_R is of order the splitting frequency Ω. Then obviously the rotating wave approximation is not valid. Indeed, neglecting the intrinsic time-dependence of the Floquet states is not possible because they may have a non-trivial time dependence [4]. In quantum information processing, for instance, the rotation of a quantum bit (qubit) is an important technical step in order to precisely control its quantum states. This rotation makes generally use of the Rabi flopping and has been performed in different physical systems such as nuclear magnetic resonance, ion traps, and systems made of Josephson junctions [5–7]. In general, a fast operation requires a high Rabi frequency which can easily be of the order of the transition frequency [8]. Similarly, in polariton chemistry, the onset of the ultrastrong coupling arises when the collective Rabi transition becomes of the order of the energy splitting of interest: $\Omega_R/\Omega > 10\%$. When this condition is fulfilled, deviations from the approximate Jaynes–Cummings and Tavis–Cummings light–matter coupling models become significant [9].

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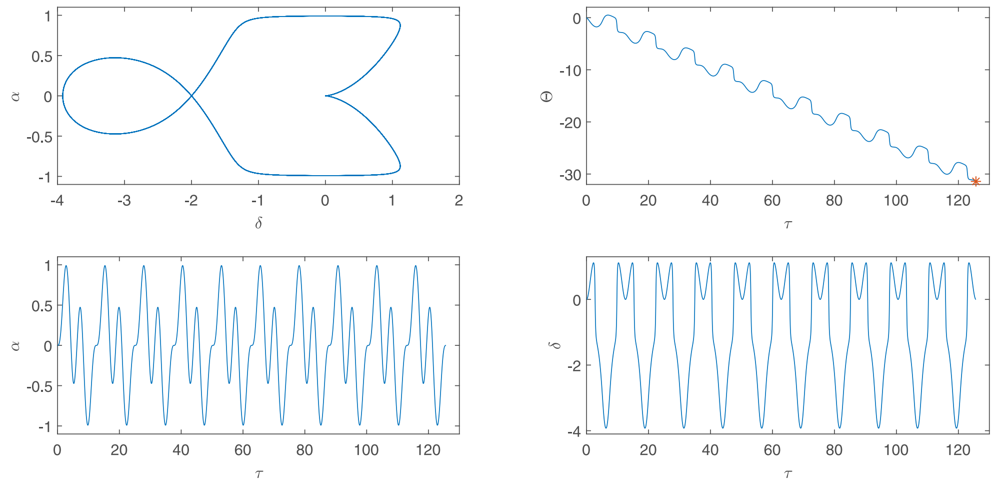

Fig. 1. $A = 1.008447$. Two upper plots: the HDS phase-locked cycle α vs δ (left) of period $T = 4\pi$ and the dynamics of the overall phase $\Theta(\tau)$ (right). The star at $\tau = 40\pi$ displays the final overall phase change $\Theta(40\pi) = -10\pi$. Two lower plots: the dynamics of the HDS variables $\alpha(\tau)$ (left) and $\delta(\tau)$ (right).

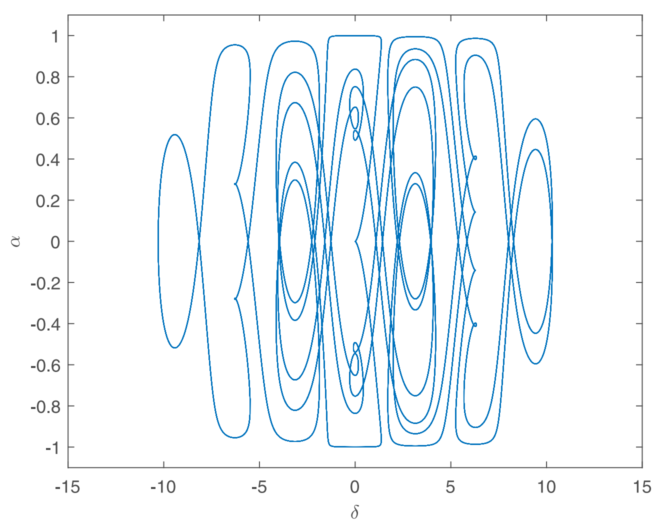

Fig. 2. The cyclic trajectory of period $T = 40\pi$ for $A = 1.1114430$.

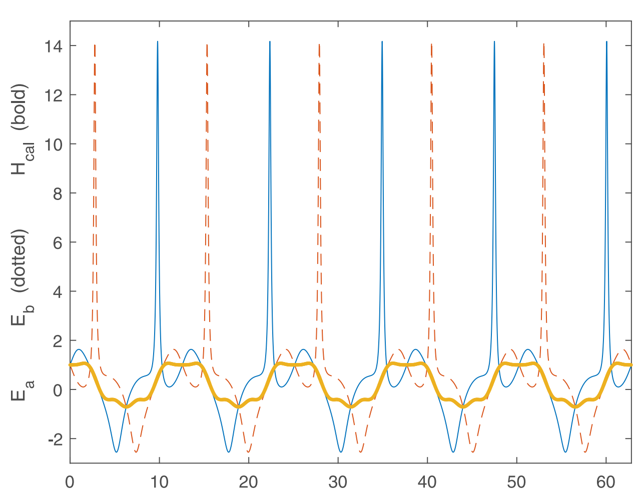

Fig. 3. The train of sharp energy pulses that emerge from the dynamics of $E_{a,b}$ defined by Eqs. (13)–(14) in units of $\mathcal{K}$ and corresponding to Fig. 1 for $A = 1.008447$. They are in marked contrast to the broad Rabi mean energy dynamics $\mathcal{H}(\tau)$ displayed in yellow bold. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

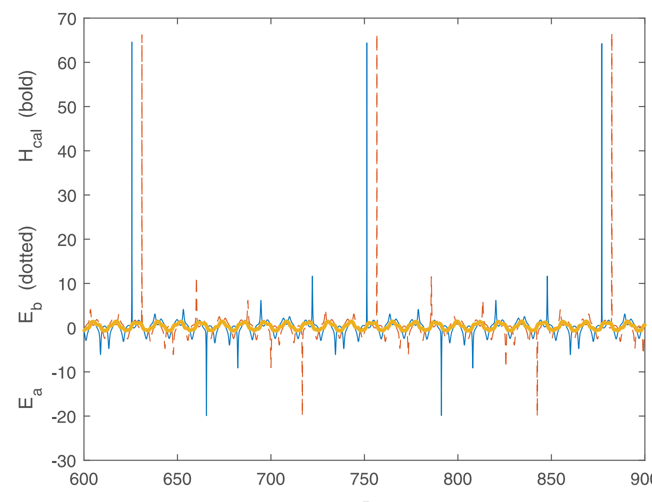

Fig. 4. The train of fast energy pulses $E_{a,b}$ defined by eqs (13)–(14) in units of $\mathcal{K}$ and corresponding to Fig. 2 for $A = 1.1114430$. Note the strong contrast with the quite smooth Rabi mean energy $\mathcal{H}$ of the system (in yellow bold). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

A two-level system may exhibit cyclic (or phase-locked) trajectories on its Bloch sphere [4]. We make use of the exact Hamiltonian description [11,12] in order to describe the resulting discrete energy spectra when $\Omega_R \sim \Omega$ —i.e. $A \sim 1$ in (3). These latter appear as frequency combs whose integer number of teeth separation $\nu_{rep} = 1/T$ – where T is the period of the corresponding cycle – spans the splitting frequency gap $2\mathcal{K}/h$ of the two-level system [10]. Therefore the parameter $q = 2\mathcal{K}/h\nu_{rep}$ is an integer and the resulting discrete multilevel energy spectrum defined by $h\nu_{rep}$ and by its harmonics actually replaces the original (unperturbed) two-level splitting $2\mathcal{K}$. "Combing this spectrum" [13] would then consist of tuning the train of short laser pulses emerging from an external mode-locked laser to the above frequency $\nu_{rep} = 1/T$ while the optical carrier envelope remains centered at $\nu_c \gg \nu_{rep}$. This procedure somehow resembles a "scratch" Velcro-type Hook-and-Loop fastening that would occur between these two discrete spectra by means of a two-photon stimulated Raman process [10]. Indeed, *any* pair of *adjacent* optical laser spectral teeth – the hook – could fasten at *any* pair of *adjacent* qubit frequency lines —the loop. A photon can be absorbed by one laser comb tooth and emitted into its next adjacent red one separated by the cyclic frequency ν_{rep} if the

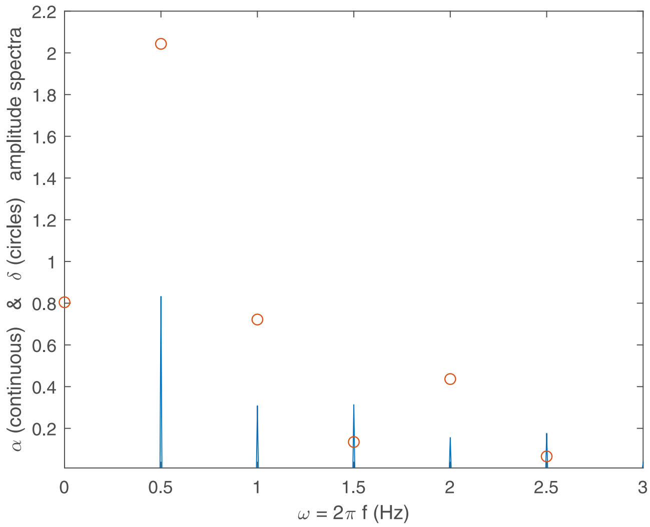

Fig. 5. The half-harmonic amplitude spectra of the conjugate coordinates α and δ corresponding to Fig. 1 for $A = 1.008447$.

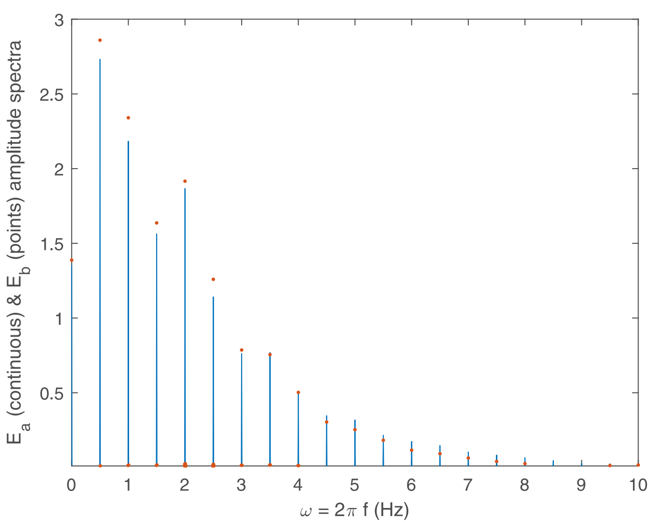

Fig. 6. The half-harmonic amplitude spectra of state energies $E_{a,b}$ displayed in Fig. 3 for $A = 1.008447$.

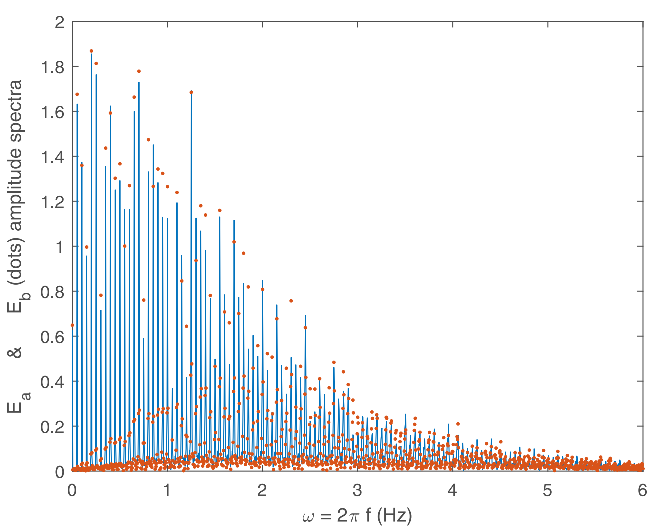

Fig. 7. The subharmonic amplitude spectra of state energies $E_{a,b}$ displayed in Fig. 4 for $A = 1.1114430$.

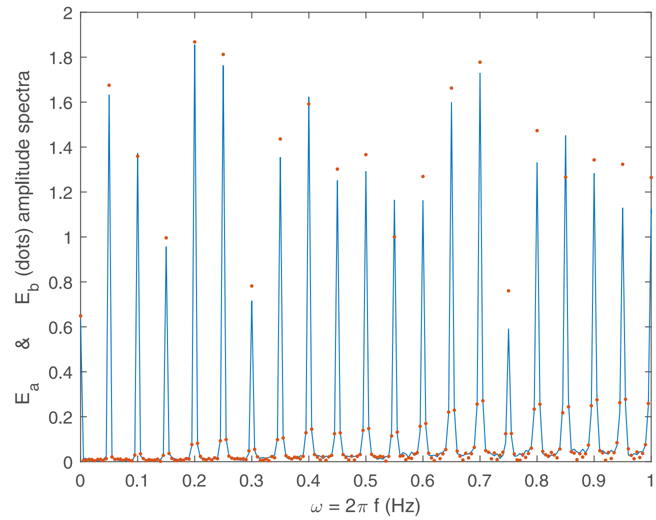

Fig. 8. The same as Fig. 7, but zoomed over the transition frequency range which is equal to unity in units of the Larmor angular frequency $\Omega = 2\mathcal{K}/\hbar$. Note that there are exactly $1/0.05 = 20$ spectral lines in the frequency gap of the two-level system: therefore, obviously like in Fig. 6, the resonant condition for the two-photon Raman stimulation of Rabi oscillations is fulfilled [10].

mode-locked laser is appropriately tuned to the qubit cycle [10]. Since several such "scratching" pairs of Raman-induced comb teeth do exist, they all coherently contribute to the Rabi stimulation of the same cyclic trajectory of period T [14].

Let us briefly recall the exact Hamiltonian description of the – even strongly – driven two-level system [11,12]. It consists of using the non-autonomous dimensionless Hamiltonian dynamical system (HDS):

$$\dot{\alpha} = -\frac{\partial \mathcal{H}}{\partial \delta} = \sqrt{1-\alpha^2}\sin\delta \quad ; \quad \dot{\delta} = \frac{\partial \mathcal{H}}{\partial \alpha} = -\frac{\alpha}{\sqrt{1-\alpha^2}}\cos\delta + \frac{\mathcal{E}(t)}{\mathcal{K}}, \tag{4}$$

defined by its two conjugate canonical coordinates α, δ and by its corresponding Hamiltonian:

$$\mathcal{H} = \sqrt{1-\alpha^2}\cos\delta + \alpha\frac{\mathcal{E}(t)}{\mathcal{K}}. \tag{5}$$

The dot means the derivation with respect to the dimensionless time $\tau = \Omega t$ related to the Larmor frequency of the system $\Omega = 2\mathcal{K}/\hbar$. In order to fit with the two-state time-dependent Hamiltonian (2) and its spinor Schrödinger equation:

$$i\hbar\frac{\mathrm{d}\psi_a}{\mathrm{d}t} = \mathcal{E}(t)\psi_a(t) + \mathcal{K}\psi_b(t) \quad ; \quad i\hbar\frac{\mathrm{d}\psi_b}{\mathrm{d}t} = -\mathcal{E}(t)\psi_b(t) + \mathcal{K}\psi_a(t), \tag{6}$$

the two-component normalized spinor wave function $\Psi = \{\psi_a, \psi_b\}$ must be given by:

$$\psi_a(t) = \sqrt{\frac{1+\alpha(t)}{2}}\, e^{i\Theta(t)} \quad ; \quad \psi_b(t) = \sqrt{\frac{1-\alpha(t)}{2}}\, e^{i[\Theta(t)+\delta(t)]}. \tag{7}$$

The time-dependent overall phase $\Theta(t)$ is *not* a 3rd independent variable: it is slaved to the solution of HDS (4)–(5) by:

$$\dot{\Theta} = -\frac{1}{2}\left[\sqrt{\frac{1-\alpha}{1+\alpha}}\cos\delta + \frac{\mathcal{E}(t)}{\mathcal{K}}\right]. \tag{8}$$

It is worth noting that, contrary to intuitive opinions, the dynamics of the overall phase of a quantum state can indeed yield an observable physical effect. It causes for instance the famous 4π-symmetry of spinor wave functions that have been directly verified in both division-of-amplitude [15,16] and division-of-wave-front [17] neutron interferometry experiments; see also the upper right plot of Fig. 1 and related comments below. If the above system is regarded as a spin one-half, its angular positions θ and ϕ on the Bloch sphere are

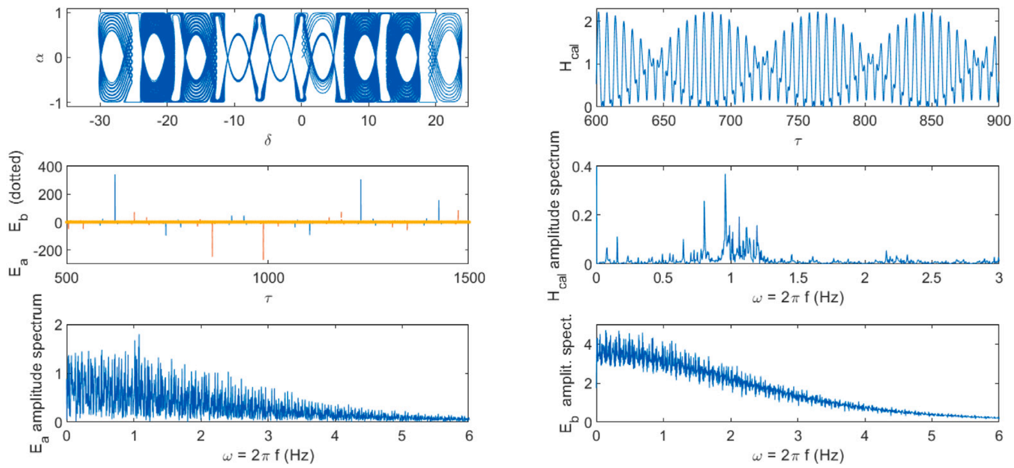

Fig. 9. A=2 . *Upper row*: $\{\alpha \text{ vs } \delta\}$ aperiodic Hamiltonian trajectory for $0 < \tau < 500\pi = 1571$ (*left*) and the time series of the mean Rabi energy $\mathcal{H}(\tau)$ (*right*). *Middle row*: the time series of the state energies $E_a(\tau)$ (blue) and $E_b(\tau)$ (dotted, red) while the thick yellow line represents at scale the mean Rabi energy $\mathcal{H}(\tau)$ (*left*) ; the amplitude spectrum of this latter (*right*). *Lower row*: the amplitude spectrum of the state energies $E_a(\tau)$ (*left*) and $E_b(\tau)$ (*right*): compare with Figs. 7 and 8. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

straightforward, according to:

$$\alpha = \cos\theta \quad ; \quad \phi = \Theta + \frac{\delta}{2}. \tag{9}$$

Then:

$$\psi_a(t) = e^{i\phi(t)} \cos\frac{\theta(t)}{2} e^{-i\delta(t)/2} \quad ; \quad \psi_b(t) = e^{i\phi(t)} \sin\frac{\theta(t)}{2} e^{+i\delta(t)/2}. \tag{10}$$

We point out the existence of phase-locked cyclic trajectories in the $\{\alpha \,\text{vs}\, \delta\}$ HDS phase space —or equivalently on the Bloch sphere (9)–(10)— for definite values of the drive amplitude A. Figs. 1 display the properties of the half-harmonic cycle of period $T = 4\pi$ (and hence of frequency $1/2$) obtained for $A = 1.008447$. The two upper plots display the phase-locked cycle α vs δ and the dynamics of the overall phase $\Theta(\tau)$. The star at $\tau = 40\pi$ refers to the final overall phase change $\Theta(40\pi) = -10\pi$. This means that the overall phase change is $-\pi$ after each 4π cycle period. Therefore the qubit needs *two* HDS periods T in order to get its overall phase Θ unchanged. This is reminiscent of the well-known 4π-symmetry of spinor wave functions [15–17] that has also been emphasized in the case of weakly driven resonant Rabi systems [11]. The two lower plots in Figs. 1 respectively display the periodic dynamics of the two HDS conjugate canonical coordinates $\alpha(\tau)$ and $\delta(\tau)$. Similarly, the spectacular very-long-period cycle with $T = 40\pi = 125.66$ in our reduced units is illustrated by Fig. 2. It is defined by the external drive (3) with $A = 1.1114430$.

The energy of the system is given by:

$$\langle \Psi | \mathbf{H} | \Psi \rangle = E = \mathcal{K}\mathcal{H}. \tag{11}$$

As expected, the classical-like HDS Hamiltonian $\mathcal{K}\mathcal{H}$ equals the quantum expectation value of the qubit energy defined by its Hamiltonian $\mathbf{H}$. We now define its *time-dependent state energies* $E_{a,b}$. In agreement with eqs (6), (7) and (11), we have the following exact alternative definition of the energy of the system:

$$E = |\psi_a|^2 E_a + |\psi_b|^2 E_b, \tag{12}$$

where the two state energies $E_{a,b}$ read either:

$$E_a = \frac{\mathcal{K}\mathcal{H} + \mathcal{E}}{1+\alpha} \quad ; \quad E_b = \frac{\mathcal{K}\mathcal{H} - \mathcal{E}}{1-\alpha}, \tag{13}$$

or:

$$E_a = -\hbar\frac{\mathrm{d}\Theta}{\mathrm{d}t} = -2\mathcal{K}\dot{\Theta} \quad ; \quad E_b = -\hbar\frac{\mathrm{d}(\Theta+\delta)}{\mathrm{d}t} = -2\mathcal{K}(\dot{\Theta}+\dot{\delta}). \tag{14}$$

Note that these state energies $E_{a,b}$ are *not* Floquet's characteristic exponents: indeed, if they were so, their sum should vanish [18], which is obviously not the case as shown by Eqs. (13) and (14). They are respectively displayed (in units of $\mathcal{K}$) by Figs. 3 and 4 for the cycles shown in Figs. 1 and 2. The striking feature of these time series concerns the very narrow high-amplitude energy pulses that are periodically emitted at frequency $\nu_{rep} = 1/T$ by the two-level cyclic system. They are in marked contrast to the broad Rabi mean energy dynamics $\mathcal{H}(\tau)$ displayed in yellow bold. These sharp energy pulses resemble the train of short laser pulses that emerge from a mode-locked laser and generate a frequency comb. Hence the basic idea of this paper: proposing a two-photon stimulated Raman coupling between these two trains of ultrashort energy pulses, using an ad-hoc external mode-locked optical laser.

The corresponding amplitude spectra for the two phase-locked trajectories displayed by Figs. 1 and 2 are respectively given by Fig. 5 for the HDS conjugate coordinates $\alpha(\tau)$, $\delta(\tau)$, Fig. 6 for the state energies $E_{a,b}(\tau)$ and Fig. 7 for the very-long-period state energies $E_{a,b}(\tau)$. The respective fundamental angular frequency $\omega_{rep} = 2\pi\nu_{rep}$ is given by $2\pi/T = 2\pi/4\pi = 1/2$ for spectra 5 and 6, while it amounts to $2\pi/T = 2\pi/40\pi = 0.05$ for spectra 7, as evidenced in Fig. 8 when zoomed over the transition gap $0 < \omega < 1$. Since the teeth from the above frequency spectra all come from the same cyclic trajectory, they are automatically phase-coherent with each other. Fig. 8 shows that the 20th harmonic of the repetition rate ν_{rep} is equal to the qubit splitting frequency $2\mathcal{K}/h$, implying that the parameter $q = 2\mathcal{K}/h\nu_{rep}$ be an integer [10]. Indeed we have $q = 1/0.05 = 20$. This also means that the period T of the $A \sim 1$ strongly driven HDS cycle is a multiple of the elementary 2π orbit period of the $A = 0$ conservative case [11,12]. This remarkable property clearly appears in the two above examples illustrated by Figs. 1 and 2 where we respectively have $T = 4\pi$ and $T = 40\pi$.

Finally, we display a non-cyclic trajectory in order to emphasize the basic differences with the above cyclic ones: see Fig. 9. We choose the $A = 2$ high-amplitude drive which would yield the angular frequency 1 of the harmonic Rabi oscillation in units of the Larmor frequency $\Omega = 2\mathcal{K}/\hbar$ if the rotating wave approximation were valid. Indeed the Rabi angular frequency would then be equal to $\Omega_R = \frac{1}{2}A\Omega = \Omega$. This latter survives as the highest peak in the mean energy spectrum of $\mathcal{H}(\tau)$ (middle row: right), however at a slightly lower frequency, namely, 0.96. The HDS trajectory being aperiodic, there are obviously no discrete nor equally-spaced comb-like spectra like in the above previous cases.

As a conclusion, we wish to point out that the existence of phase-locked cycles in the strongly driven Hamiltonian dynamics generates sharp and regular pulse-train spectra for the time-dependent state energies $E_{a,b}$ of the original two-level system. These latter are in sharp contrast with their smooth mean value $\mathcal{H}$ defining the anharmonic Rabi flipping of the system. They could consequently be excited by an appropriately-tuned optical frequency comb emerging from an external mode-locked laser. This would provide a highly resonant dynamical regime where a two-photon Raman transition process would occur for *all pairs* of *adjacent* comb teeth. As a result, these two-photon stimulated excitations would coherently sum up in order to secure the phase-locked cycle of the strongly driven system.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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