Multilevel Interference Resonances in Strongly Driven Three-Level Systems

Jeroen Danon and Mark S. Rudner

Niels Bohr International Academy, and the Center for Quantum Devices, Niels Bohr Institute, University of Copenhagen, 2100 Copenhagen, Denmark

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We study multiphoton resonances in a strongly driven three-level quantum system, where one level is periodically swept through a pair of levels with constant energy separation E. Near the multiphoton resonance condition $n\hbar\omega = E$, where n is an integer, we find qualitatively different behavior for n even or odd. We explain this phenomenon in terms of families of interfering trajectories of the multilevel system. Remarkably, the behavior is insensitive to fluctuations of the energy of the driven level, and survives deep into the strong dephasing regime. The setup can be relevant for a variety of solid state and atomic or molecular systems. In particular, it provides a clear mechanism to explain recent puzzling experimental observations in strongly driven double quantum dots.

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The advent of intense microwave and laser sources has opened a range of new possibilities for investigating the strong-driving regime of both natural and artificial (solid-state) atoms and molecules [1–3]. In this regime, the amplitude of an applied ac driving field may greatly exceed both the driving field photon energy $\hbar\omega$ as well as the separation between energy levels of the system. While the dynamics of strongly driven two-level systems have been studied extensively [3–8], *multilevel* systems offer new avenues to explore. In particular, high-order multilevel and multiphoton processes [9,10] give rise to intriguing and potentially useful phenomena such as amplitude spectroscopy [11], population inversion [12–14], and microwave-induced cooling [15].

Recently, a new type of multiphoton resonance was discovered in experiments on spin-blockaded double quantum dots (DQDs) subjected to large-amplitude modulations of a nearby gate electrode [16,17]. The resonances show a striking asymmetry, with current *enhanced* when the electron Zeeman splitting matches an odd-integer multiple of the driving field photon energy, $E_Z = (2n+1)\hbar\omega$, and *suppressed* for even-integer resonances $E_Z = 2n\hbar\omega$. Such a dramatic even or odd effect, which is, furthermore, insensitive to changes in driving amplitude and dc offset over a wide range of values, is unknown in two-level systems, cf. [18]. Analytical [19,20] and numerical [21,22] investigations have accounted for the existence of multiphoton resonances, but crucially could not explain the even or odd asymmetry observed in [16,17] (though a Fano-like origin was speculated [19]).

Motivated by this puzzle, we look at the dynamics of a strongly driven multilevel system. Previously, such systems have been investigated in a variety of contexts. For example, driven transport through nanoscale conductors has been studied using Floquet theory [23,24]. The dynamics of superconducting qubits have been investigated via numerical simulations [12], or by viewing the dynamics in terms of isolated two-level crossings, omitting possible long-lived coherences [25]. Interferences between multilevel processes have also been studied, e.g., in ladder-type systems where multiphoton transitions can proceed directly or through a resonant intermediate state [26].

In this Letter, we focus on a three-level system and show that multilevel interferences between first and third order processes lead to multiphoton resonances with characteristics differing markedly from those of familiar two-level resonances. While the resonances require long-lived coherence between two of the levels, the phenomenon survives deep into the regime of strong dephasing of the third level. In this regime we derive analytic expressions for all interlevel transition rates. Finally, we explicitly connect our model to the experiments of Refs. [16,17] and show that it captures all relevant features of the data. The phenomena that we describe are, however, quite general, with potential relevance, e.g., for strongly driven spin qubits, superconducting qubits, and diamond NV centers.

To highlight the key qualitative differences between two-level and multilevel resonances, we first briefly review the phenomenology of multiphoton resonances in a two-level system. We consider a system with basis states $\{|1\rangle, |S\rangle\}$, its dynamics governed by the Hamiltonian

$$H_2(t) = \begin{pmatrix} 0 & q \\ q & -\varepsilon(t) \end{pmatrix}, \qquad \varepsilon(t) = \varepsilon_0 - A\cos\omega t. \quad (1)$$

Here we focus on the case of strong driving, $A \gg q$ and $A > |\varepsilon_0|$. Figures 1(a) and 1(b) show the instantaneous spectrum of this system, plotted versus detuning ε and time t.

The relevant features of the driven system's dynamics can be understood heuristically in terms of families of interfering trajectories [Fig. 1(b)]. For strong driving, transitions

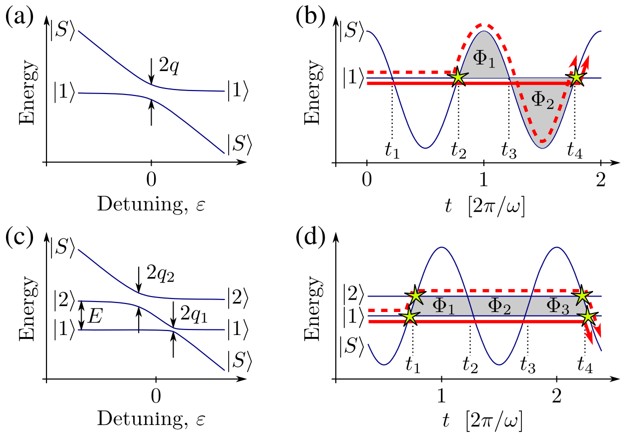

FIG. 1 (color online). Spectrum of (a),(b) the two-level Hamiltonian (1) and (c),(d) the three-level Hamiltonian (2). In (a),(c) the energy levels are plotted as a function of detuning and in (b),(d) as a function of time assuming strong driving (thin blue lines). We have set $A \gg q, q_{1,2}$. In (b),(d) we show interfering paths which bring the system from $|1\rangle$ to $|S\rangle$ (thick red lines).

take place at relatively well-defined points in time $\{t_p\}$ when the two levels are nearly degenerate. Two paths taking the system from state $|1\rangle$ to $|S\rangle$ are indicated by the dashed and solid red lines. In the illustration, the transitions occur at times t_2 and $t_4 = t_2 + T$, where $T = 2\pi/\omega$ is the driving period, and the interference phase corresponds to the difference of shaded areas shown, $\Phi = |\Phi_1| - |\Phi_2|$. When $\varepsilon_0 = n\omega$ (we set $\hbar = 1$), we have $\Phi = 2n\pi$, and for integer n the interference is constructive. In this case all paths featuring transitions at "even" times t_{2p} mutually interfere constructively, as do all paths with transitions at odd times t_{2p+1}. This provides a resonant response. Additional structure results from interferences between these two groups of trajectories, which are sensitive to the individual phases $\Phi_{1,2}$. For sinusoidal driving, this leads to the strength of the n-photon resonance line being modulated by the Bessel function $J_n(A/\omega)$ [5]. The intensities of the two-level multiphoton resonances are thus highly sensitive to the amplitude and frequency of driving, exhibiting sequences of peaks and nodes as A/ω is varied.

We now turn our attention to strong driving in a *multilevel* system. To clearly demonstrate the essential physics of multilevel resonances, we focus on the case of three levels. For simplicity, we assume that the driving field couples strongly to only one level, $|S\rangle$, while the energy separation between the other levels $|1\rangle$ and $|2\rangle$ is unaffected [see Figs. 1(c) and 1(d)]. The state $|S\rangle$ acts as a "shuttle," mediating population transfer between $|1\rangle$ and $|2\rangle$. This situation is described by the generic Hamiltonian

$$H_3(t) = \begin{pmatrix} E/2 & 0 & q_2 \\ 0 & -E/2 & q_1 \\ q_2 & q_1 & -\varepsilon(t) \end{pmatrix}, \qquad (2)$$

written in the basis $\{|2\rangle, |1\rangle, |S\rangle\}$. Here, E is the energy splitting between states $|1\rangle$ and $|2\rangle$, $q_{1,2}$ are the coupling matrix elements, and $\varepsilon(t) = \varepsilon_0 - A\cos\omega t$ as before.

Two-level resonances between $|S\rangle$ and $|1\rangle$ or $|2\rangle$, analogous to those described above, can occur whenever the corresponding static detuning $\varepsilon_0 \pm E/2$ matches the n-photon energy $n\omega$. Such resonances do not present qualitatively new physics.

More interestingly, we investigate the existence of resonances associated with the energy splitting E. Such resonances must occur via the strongly modulated level $|S\rangle$, thereby constituting a true multilevel phenomenon.

How could such resonances arise? In the spirit of the discussion above, in Fig. 1(d) we illustrate a characteristic pair of interfering trajectories, from $|1\rangle$ to $|S\rangle$. For large driving amplitude $A \gg \varepsilon_0$, E, the interference phase is given by $\Phi_1 + \Phi_2 + \Phi_3 = E(t_4 - t_1) = 3E(T/2)$. Importantly, this phase is controlled only by the splitting E and the driving half-period $T/2 = \pi/\omega$, and *not* by the driving amplitude or waveform. Many such paths exist, where the last two transitions take place approximately at the same time $t_{p>1}$. These paths interfere constructively when $\Phi_1 = \pi E/\omega = 2\pi n$, suggesting the existence of resonances at $E = 2n\omega$, i.e., at *even* multiples of ω. Similar considerations for transitions from $|1\rangle$ to $|2\rangle$ reveal a series of processes depending on the *full* driving period T, predicting additional resonances at *all* multiples of the photon energy, $E = n\omega$. We thus expect to find resonances with very different behavior when E is an even or an odd multiple of ω. Further, in sharp distinction with the two-level case discussed above, the interference phase Φ_1 and thus the resonances are only weakly sensitive to the amplitude A and detuning ε_0.

We now begin our detailed analysis, which is based on a perturbative treatment in terms of the small parameters $q_{1,2}^2/(A\omega)$ that characterize the strong driving limit. To most clearly exhibit the effect, and to allow us to arrive at analytic results, we focus on a regime of strong dephasing where coherences between $|1\rangle$ and $|S\rangle$ and between $|2\rangle$ and $|S\rangle$ are rapidly lost, on a time scale shorter than the driving period. In contrast, we allow coherences between $|1\rangle$ and $|2\rangle$ to be long-lived on this time scale. The dephasing is modeled by Gaussian white-noise fluctuations on each of the unperturbed energy levels via

$$\delta H_3(t) = \sum_\alpha \xi_\alpha(t)|\alpha\rangle\langle\alpha|, \quad \alpha \in \{1, 2, S\}, \qquad (3)$$

with $\overline{\xi_\alpha(t)\xi_\beta(t')} = \Gamma_\alpha \delta(t - t')\delta_{\alpha\beta}$, where the overbar indicates averaging over noise realizations. Within this model we calculate the rates of interlevel transitions, working up to fourth order in the couplings $q_{1,2}$.

Strong dephasing is particularly relevant for the experiments in Refs. [16,17], where the level corresponding to $|S\rangle$ exhibits strong lifetime broadening due to coupling to a nearby reservoir (see discussion below). The multilevel resonances survive deep into the strong-dephasing regime, where the quasi-two-level resonances at $\varepsilon_0 \pm E/2 = n\omega$ are completely washed out.

The first analytical step is to transform to a modified interaction picture via $|\psi_R(t)\rangle = e^{iR(t)}|\psi(t)\rangle$, with $R(t) = \sum_\alpha \phi_\alpha(t)|\alpha\rangle\langle\alpha|$. The phases ϕ_α are given by $\phi_\alpha(t) = -\int_0^t d\tau \tilde{\varepsilon}_\alpha(\tau)$, with $\tilde{\varepsilon}_{1,2}(\tau) = \mp\frac{1}{2}E + \xi_{1,2}(\tau)$ and $\tilde{\varepsilon}_S(\tau) = \varepsilon(\tau) + \xi_S(\tau)$. States in this interaction picture evolve according to $i(d/dt)|\psi_R\rangle = \tilde{H}_3(t)|\psi_R\rangle$, with

$$\tilde{H}_3(t) = q_1 e^{i\phi_{S1}(t)}|S\rangle\langle 1| + q_2 e^{i\phi_{S2}(t)}|S\rangle\langle 2| + \text{H.c.}, \quad (4)$$

where $\phi_{\alpha\beta}(t) \equiv \phi_\alpha(t) - \phi_\beta(t)$.

The transition rate between states $|\alpha\rangle$ and $|\beta\rangle$ is calculated as the time derivative of the transition probability,

$$W_{\alpha\to\beta} = \frac{d}{dt}\overline{|\langle\beta|U(t)|\alpha\rangle|^2}, \quad (5)$$

where $U(t)$ evolves the system between times 0 and t. We expand the time-evolution operator in powers of $q_{1,2}$ as $U(t) = 1 + U^{(1)}(t) + U^{(2)}(t) + \cdots$, with

$$U^{(m)}(t) = (-i)^m \int_0^t dt_1 \cdots \int_0^{t_{m-1}} dt_m \tilde{H}_3(t_1)\cdots\tilde{H}_3(t_m).$$

Working up to third order in $q_{1,2}$ gives access to the transition *rates* up to fourth order in the couplings.

For illustration, we now evaluate $W_{1\to S}$ to lowest (second) order; other rates are obtained similarly. We write

$$\begin{aligned} W^{(2)}_{1\to S} &= \frac{d}{dt}\overline{|\langle S|U^{(1)}(t)|1\rangle|^2} \\ &= q_1^2 \frac{d}{dt}\int_0^t dt_1 \int_0^t dt_2 \overline{e^{i[\phi_{S1}(t_1) - \phi_{S1}(t_2)]}}, \end{aligned} \quad (6)$$

and use $\overline{e^{i\int d\tau \xi(\tau)}} = e^{-(1/2)\int d\tau \int d\tau' \overline{\xi(\tau)\xi(\tau')}}$. For white noise the correlator in the exponent is $\propto \delta(\tau - \tau')$; Gaussian noise with other spectral densities can be considered similarly. The result is simplified for $\Gamma_S \gg \omega, \Gamma_{1,2}$, giving

$$W^{(2)}_{1\to S} = \frac{q_1^2 \Gamma_S}{(\frac{1}{2}E - \varepsilon_0 + A\cos\omega t)^2 + \frac{1}{4}\Gamma_S^2}. \quad (7)$$

Moving to the strong driving limit $A \gg \Gamma_S$ and assuming $A > |\varepsilon_0 - \frac{1}{2}E|$, the transition rate displays sharp bursts, well separated in time, occurring whenever the levels cross, i.e., when $A\cos\omega t \approx \varepsilon_0 - \frac{1}{2}E$. Averaging these bursts over one period yields $W^{(2)}_{1\to S} \approx 2q_1^2/[A^2 - (\frac{1}{2}E - \varepsilon_0)^2]^{1/2}$. Similarly, we find $W^{(2)}_{2\to S} \approx 2q_2^2/[A^2 - (\frac{1}{2}E + \varepsilon_0)^2]^{1/2}$ in the same limit, and identical rates for the reverse processes $W^{(2)}_{S\to 1}$ and $W^{(2)}_{S\to 2}$.

Multilevel interference resonances first arise at fourth order, $W^{(4)}_{\alpha\to\beta} = (d/dt)\{2\text{Re}\overline{\langle\alpha|U^{\dagger(3)}(t)|\beta\rangle\langle\beta|U^{(1)}(t)|\alpha\rangle} + \overline{|\langle\beta|U^{(2)}(t)|\alpha\rangle|^2}\}$. Because of the form of $\tilde{H}_3$, the rates $W^{(4)}_{1,2\leftrightarrow S}$ only involve the first term, while the rates $W^{(4)}_{1\leftrightarrow 2}$ involve only the last. Proceeding along similar lines as above, we assume $\Gamma_S \gg \omega, \Gamma_{1,2}$ and work in the strong-driving limit $A \gg \Gamma_S$. Note that in our regime of interest, $n \gtrsim 1$, this also ensures that $\Gamma_S \ll A\omega/E$.

We now can perform two of the three time integrals in the rates, yielding

$$\begin{aligned} W^{(4)}_{1,2\leftrightarrow S} = \int_0^t d\tau & \frac{2q_1^2 q_2^2 \Gamma_S e^{-(\Gamma'\omega/2\pi)(t-\tau)}}{(\frac{1}{2}E - \varepsilon_0 + A\cos\omega t)^2 + \frac{1}{4}\Gamma_S^2} \\ & \times \text{Im}\left\{\frac{e^{-iE(t-\tau)}}{\frac{1}{2}E - \varepsilon_0 + A\cos\omega\tau + \frac{i}{2}\Gamma_S}\right\}, \end{aligned} \quad (8)$$

with the dimensionless dephasing rate $\Gamma' = (\Gamma_1 + \Gamma_2)\pi/\omega$, and a similar expression for $W^{(4)}_{1\leftrightarrow 2}$. We can find analytic approximations for the integrals in two important cases, valid for times $t \gg \Gamma_{1,2}^{-1}$. First, at zero detuning, $\varepsilon_0 = 0$, we find $W^{(4)}_{1,2\leftrightarrow S} \approx -g_0 \bar{W}$ and $W^{(4)}_{1\leftrightarrow 2} \approx (\frac{1}{2}g_0 + h_0)\bar{W}$, with $\bar{W} = 2\pi q_1^2 q_2^2/(A^2\omega)$ and

$$\begin{aligned} g_0 &= \frac{2\cos(n\pi)\sinh(\frac{1}{2}\Gamma') + e^{\Gamma'} - \cos 2n\pi}{\cosh\Gamma' - \cos 2n\pi}, \\ h_0 &= \frac{\sin^2(\frac{1}{2}n\pi)\sinh(\Gamma')}{\cosh\Gamma' - \cos 2n\pi}, \end{aligned}$$

using the (continuum-valued) photon number $n = E/\omega$. Second, for arbitrary detuning but *integer* n, we find $W^{(4)}_{1,2\leftrightarrow S} \approx -g_i \bar{W}$ and $W^{(4)}_{1\leftrightarrow 2} \approx (\frac{1}{2}g_i + h_i)\bar{W}$, with

$$\begin{aligned} g_i &= \frac{\cos(nd_-)[\sinh(\frac{\Gamma' d_-}{2\pi}) + \sinh(\frac{\Gamma' d_+}{2\pi})] + e^{\Gamma'} - 1}{(\cosh\Gamma' - 1)(1 - \delta^2)}, \\ h_i &= \frac{\sin^2(\frac{1}{2}nd_+)\coth(\frac{1}{2}\Gamma')}{1 - \delta^2}, \end{aligned}$$

where $d_\pm = \pi \pm 2\sin^{-1}\delta$ and $\delta = \varepsilon_0/A$.

In Fig. 2 we plot the rates as a function of n for $\delta = 0$ (a),(c) and as a function of δ for $n = 1, 2, 3$ (b),(d), in all plots setting $\Gamma'/\pi = 0.3$. The rates display resonant features at integer n. Moreover, the resonances for even and odd n are qualitatively different, as anticipated above. The negative sign of $W^{(4)}_{1\to S}$ indicates that this fourth-order contribution provides a suppression of the large (second-order) background transition rate $W^{(2)}_{1\to S}$. As long as $\Gamma_1 + \Gamma_2 > W^{(2)}_{1\to S}$ the total rate $W^{(2)}_{1\to S} + W^{(4)}_{1\to S}$ is positive. For $W^{(2)}_{1\to S} > \Gamma_1 + \Gamma_2$, lifetime broadening of $|1\rangle$ and $|2\rangle$ due to driving-induced transitions to $|S\rangle$ becomes dominant. To capture this effect, higher terms must be included.

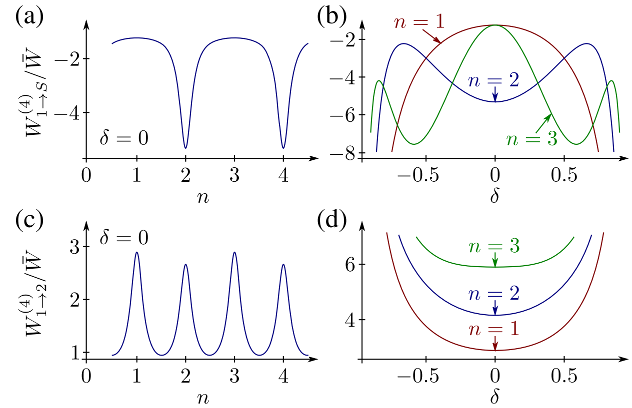

FIG. 2 (color online). The rate $W^{(4)}_{1\to S}$ at (a) zero detuning as a function of $n = E/\omega$, and (b) for integer $n = 1, 2, 3$ as a function of δ. (c),(d) The same for the rate $W^{(4)}_{1\to 2}$. In all plots we used $\Gamma'/\pi = 0.3$. In (d) the curves are offset in steps of $\frac{3}{2}$.

So far, our results are general for strongly driven three-level systems. The different behavior at even and odd n is predicted to be a generic feature of resonances at $E = n\omega$; to arrive at analytic results we added the assumption of strong fluctuations of the driven level. We now connect our results to the experiments of Refs. [16,17], in which current through spin-blockaded DQDs was measured in the presence of strong ac driving.

In the two-electron regime, the low-energy electronic subspace of the DQD is spanned by five states: a "(1,1)" spin singlet and a spin triplet with a single electron in each dot, and a "(0,2)" spin singlet with double occupancy of the right dot (the left dot being empty). In spin blockade, current flow is mediated by the (0,2) singlet state, which is the only state with direct coupling to the drain lead. Coupling between the blocked spin triplet and the singlet levels occurs via spin-orbit, hyperfine, and/or inhomogeneous Zeeman coupling. Away from singlet-triplet degeneracy points, ac driving can provide the energy necessary to stimulate triplet-singlet transitions [27,28]. When the driving frequency and level splittings are in resonance, such coupling is expected to lift the blockade and produce an *enhancement* of current. The striking even or odd effect observed in Refs. [16,17] thus clearly does not fit in this simple picture.

As we will now show, the multilevel multiphoton resonances described above account for all of the main features of the experimental data. To make the connection explicit, state $|S\rangle$ in our model represents the (0,2) singlet state of the DQD, while $|1\rangle$ represents the triplet state T_+, with both electron spins pointing up, and $|2\rangle$ represents a particular superposition of the (1,1) singlet and T_0 states, which is determined by Zeeman energy inhomogeneities in the DQD [29].

Using all contributions to the transition rates up to fourth order, we construct a master equation for the time-dependent level occupation probabilities $\{p_\alpha\}$,

$$\dot{p}_1 = p_2\left(W_{2\to 1} + \frac{1}{2}W_{2\to S}\right) - p_1\left(\frac{1}{2}W_{1\to S} + W_{1\to 2}\right), \tag{9}$$

where $p_2 = 1 - p_1$. To eliminate p_S, we assumed that the decay of $|S\rangle$ and the consecutive reloading of $|1\rangle$ or $|2\rangle$ (with equal probabilities) happens instantaneously on the time scale of the dynamics of $p_{1,2}$. We solve Eq. (9) for the steady-state values $p^{(\mathrm{eq})}_{1,2}$, which give the steady-state current $I/e = p^{(\mathrm{eq})}_1 W_{1\to S} + p^{(\mathrm{eq})}_2 W_{2\to S}$.

To compare with the data in Fig. 2(d) of Ref. [17], we set $\delta = 0$ and assume that $\omega, E \sim 1$–$10\ \mu$eV. We set $q_1^2/A = 0.05\ \mu$eV and $q_2^2/A = 0.5\ \mu$eV, i.e., $q_2^2/q_1^2 = 10$ [30], and choose $\Gamma_{1,2} = 1\ \mu$eV. In Fig. 3(a) we plot the resulting steady-state current, normalized to I_{bg}, the off-resonant "background" current (i.e., the current due to "direct" second-order transitions $W^{(2)}_{1,2\to S}$ associated with repeated sweeps through the S-T_+ level crossing; in the experiment $I_{\mathrm{bg}} \sim 15$ pA). The model reproduces all important features of the data: a resonant response of current along all n-photon lines, alternating between enhancement for odd n and suppression for even n [31]. At even n, the negative contributions $W^{(4)}_{1,2\to S}$ suppress escape from $|1\rangle$ and $|2\rangle$ to $|S\rangle$, resulting in a reduction of current relative to the background. The rate $W^{(4)}_{1\leftrightarrow 2}$ is largest for odd n, where it efficiently mixes $|1\rangle$ and $|2\rangle$ and thus enhances the escape rate out of the most strongly blocked state, $|1\rangle$, thereby increasing the total current. Including a second unpolarized (1,1) level [29], split from $|1\rangle$ by E', would yield another fan of current peaks and dips at $E' = n\omega$, reproducing the "doubled" fan of Fig. 2(d) of Ref. [17].

We finally investigate the detuning dependence of the current, which in the experiment showed a strikingly slow

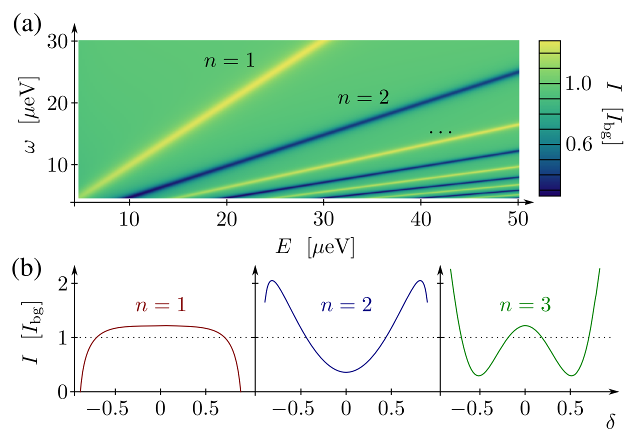

FIG. 3 (color online). Calculated current through a driven double quantum dot in spin blockade, normalized to the background current I_{bg}. (a) The current at $\delta = 0$ as a function of ω and E. (b) Slow modulation of the resonances: the current as a function of δ for fixed $n = 1, 2, 3$. In all plots we used $q_1^2/A = 0.05\ \mu$eV, $q_2^2/A = 0.5\ \mu$eV, and $\Gamma_{1,2} = 1\ \mu$eV.

modulation (on the scale of $\varepsilon_0 \sim A$) with distinct characteristic shapes for each of the resonances, see Fig. 3(b) of [17]. In Fig. 3(b) we plot the current as a function of δ at fixed $n = 1, 2, 3$, using the same parameters as for Fig. 3(a). The detuning dependence of I agrees well with the experimental observations. Here it arises from the weak dependence of the interference phases Φ_n on ε_0, as explained above.

To summarize, we investigated multiphoton resonances in a strongly driven three-level quantum system. We identified new resonant responses which crucially depend on the multilevel structure of the system. We further revealed how these resonances provide a mechanism to explain recent puzzling experimental observations in strongly driven double quantum dots. Interestingly, the behavior survives deep into the regime of strong dephasing on one of the levels. Detailed explorations of the fully coherent regime, the role of decoherence, and connections to other physical systems are interesting directions for further study.

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