Observation of Multiphoton Frequency Conversion in Superconducting Circuits

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(Dated: August 8, 2018)

Multiphoton up/down conversion in a transmon circuit, driven by a pair of microwaves tuned near and far off the qubit resonance, has been observed. The experimental realization of these high order non-linear processes is accomplished in the three-photon regime, when the transmon is coupled to weak bichromatic microwave fields with the same Rabi frequencies. A many-mode Floquet formalism, with longitudinal coupling, is used to simulate the quantum interferences in the absorption spectrum that manifest the multiphoton pumping processes in the transmon qubit. An intuitive graph theoretic approach is used to introduce effective Hamiltonians that elucidate main features of the Floquet results. The analytical solutions also illustrate how controllability is achievable for desired single- or multiphoton pumping processes in a wide frequency range.

Superconducting (SC) qubits are a leading contender to demonstrate the so-called quantum supremacy [1]. Achieving this task requires scaling up the number of qubits in the SC circuit. Since in many SC qubits circuits, microwaves couple the qubits and shuttle information [2], frequency conversion is vital for efficient interconnection among the qubits, and for hybrid quantum systems composed of SC qubits as components [3–7]. Microwave down-conversion has been demonstrated for three-level Λ qubits [8]. Up-conversion has also been reported for a three-Josephson-junction flux qubit coupled to a microwave drive [9, 10]. These conversions have, however, been achieved in circuit quantum electrodynamics systems, in the strong light-matter coupling regime [11]; and cannot be implemented in periodically driven systems.

Motivated by the rapidly growing interest in Floquet engineering [4, 12], and the ubiquitous use of microwave drives in quantum information processing [13], we implement a readily attainable scheme to up/down convert the frequency, when a SC qubit is periodically driven by bichromatic classical microwaves [14, 15]. We investigate the up/down frequency conversion, in a wide range, when multiphoton pumping between a pair of microwave fields is realized in a SC transmon qubit circuit [10]. Such non-linear response is usually driven by high intensity fields [17, 18]. Increasing the pump strength, also introduce undesirable coupling and interference that may limit the fidelity of the operations [19] [20]. In our approach, we observe nonlinear high order processes in the weak driving limit, i.e. when the Rabi frequency of the driving microwave field is much less than the qubit resonance frequency.

To establish the coupling scheme between the driving bichromatic microwave and the SC qubit, we take advantage of the intrinsic difference between the SC artificial atoms and the natural atomic systems. Unlike natural atoms, in which inversion symmetry only allows radiative coupling between opposite parity states, in SC artificial atoms, because the qubits can be inductively tuned, the transitions which break the symmetry are possible, i.e. non-dipole allowed transitions [21, 22]. This allows the coexistence of odd and even photon processes, via the longitudinal coupling. In fact, this is a non-conventional scheme, compared to the mostly commonly used trans-
verse coupling, in which the external field couples to qubit $\sigma_x$ degree of freedom [24]. The coexistence of different photon processes can induce additional phenomena which are not otherwise observed in systems that conform to symmetry selection rules [14, 24–26].

In this study, we consider a SC qubit with transition frequency $\omega_q = 2 \pi \times 8.8356$ GHz coupled to two microwave frequencies, $\omega_1$ and $\omega_2$ through longitudinal coupling. The assignment of indices to each of the microwaves is arbitrary. The time-dependent Hamiltonian, which describes the longitudinal (diagonal) coupling, ignoring relaxation and dissipation [14, 27],

$$\hat{H} = -\frac{1}{2} [\Delta \sigma_x + \delta(t) \sigma_z], \quad (1)$$

where $\Delta = 0.168$ MHz is the tunnel splitting, which represents the suppressed hopping between wells of the Josephson potential in the transmon qubit [25]. The Pauli matrices are $\sigma_{x,z}$. The bichromatic microwave field is introduced in the diagonal matrix elements, $\delta(t) = \omega_q + A_1 \cos(\omega t_1 + \phi_1) + A_2 \cos(\omega t_2 + \phi_2)$, with $A_1$ and $A_2$ the peak amplitudes of the fields, e.g. $A_1 = A_2 = 1.92$ MHz, in the weak limit, $(A_1, 2/\omega_q) \approx 2.2 \times 10^{-4}$. For simplicity, we set $\phi_1 = \phi_2 = 0$. Since we scan a large range of the frequencies near and far away from the qubit resonance, $\omega_1 + \omega_2$ is not necessarily time periodic. Therefore we apply the incommensurate frequencies condition in our calculations. This requires the implementation of the two-mode Floquet method [29, 30]. This methodology is introduced in the supplemental materials (SM) [31].

The experimental setup consists of a SC transmon qubit coupled to a three-dimensional aluminum cavity as shown in Fig. 2(a). The length, width, and depth of the cavity are 15.5, 4.2, and 18.6 mm, respectively. The cavity is used for the quantum non-demolition measurement of qubit states. Three microwave drives are used: Qubit control waves, arbitrarily denoted as Mw1 and Mw2, are continuous while the readout wave (denoted as measure (black)), is triggered. (c) The Energy level and microwave drive diagrams represent the three-photon transition. The diagram shows the case of $\delta_1 > 0$ and $\delta_2 < 0$.

In Fig. 1(c), the first experimental photon pumping is observed at $\theta = 69.7^\circ$. The set $(\omega_1, \omega_2) = (2.9498, 2.9567)$ GHz at this point corresponds to photon numbers (5, 2), indicating a two-photon pumping process from Mw1 to Mw2. The next line in this spectrum indicates single photon pumping at $\theta = 78.2^\circ$, where $(\omega_1, \omega_2) = (2.9476, 2.9546)$ GHz. Here, three photons from Mw1 contributes to the qubit excitation, and one photon is pumped to Mw2 at 2.9546 GHz, i.e. (4, 1). The transition line at $\theta = 93.5^\circ$ represents the three-photon excitation by Mw1 only, i.e. (3, 0), at $\omega_1 = 2.9433$ GHz. The wide feature at $\theta = 119^\circ$ gives the contribution of two photons from Mw1 and one photon from Mw2, i.e. (2, 1), at $(\omega_1, \omega_2) = (2.9368, 2.9619)$ GHz. By symmetry, the other transition peaks in this plot can be labeled properly to determine the single- and multi-photon frequency conversions from Mw2 to Mw1 (see the labels in Fig. 1(b),(c)).

We extended the up- and down-conversions to frequen-
FIG. 3. Frequency conversion in 3-photon regime for the transition manifold (4,-1), away from the qubit resonance frequency. Each point is the contour map (top-view) of the transition probability peak corresponding to a (4,-1) conversion at the given frequencies. The up- and down-conversions are labeled by `+` and `-` signs on the plot, while the numbers indicate the conversion magnitude, $\Omega_{1,2} = \omega_2 - \omega_1$. For each conversion case, the theoretical result is shown as a blue circular contour plot, and the corresponding experimental peaks are indicated by red triangles. The breaks on the x- and y-axis indicate the near resonance region, discussed in Fig. (1).

FIG. 4. (a) Graph-theoretical illustration of the two-dimensional Floquet space. The three manifolds, namely (4,-1), (3,0), and (2,1), are indicated as the red, green and blue paths on the graph, respectively. The parameter space of this driven system, $\{\Delta, A_1, A_2\}$, is indicated as the weights of the graph edges. The transition spectra corresponding to these paths, obtained from Eqs. (2)-(4), are presented in (b). The full-Floquet result is given as black-dashed line for comparison purpose. (c) Comparison of the Floquet and analytical, Eq. (5), values for the shift observed in the (3,0) peak position as a function of ($A_2$). The Mw1 power is kept constant at $A_1 = 1.92$ MHz. The (3,0) peak is originally expected to be seen at $\omega_q/3 = 2.9452$ GHz (i.e. when $A_2 = 0$).

The matrix can be extracted from the original Floquet matrix by eliminating the intermediate dressed states, within the adiabatic approximation (introduced in Ref. [3]),

$$\hat{H}_F^{(3,0)} = \begin{bmatrix} E^{(3,0)}_g & g_{e f f}^{(3,0)} \\ g_{e f f} & E_{[0,0,e]} \end{bmatrix},$$

$$g_{e f f}^{(3,0)} = -\frac{4A_1^2\Delta}{E_{[3,0,g]}^3},$$

$$\hat{H}_F^{(2,1)} = \begin{bmatrix} E^{(2,1)}_g & g_{e f f}^{(2,1)} \\ g_{e f f} & E_{[0,0,e]} \end{bmatrix},$$

$$g_{e f f}^{(2,1)} = -\frac{10A_1^2A_2\Delta}{E_{[2,1,g]}^3}.$$
\[ H_{\text{eff}}^{(4,-1)} = \begin{bmatrix} E_{[4,-1,g]}^{(4,-1)} \\ g_{\text{eff}}^{(4,-1)} \\ E_{[0,0,e]}^{(4,-1)} \end{bmatrix}, \]
\[ g_{\text{eff}}^{(4,-1)} = -\frac{20 A_1^4 A_2 \Delta}{E_{[4,-1,g]}^5}. \]

where \( E_{[3,0,g]} = -\frac{\omega_1^2}{2} + 3\omega_1 \), and \( E_{[0,0,e]}^{[2,1,g]} = -\frac{\omega_1^2}{2} - 2\omega_1 + \omega_2 \), and \( E_{[4,-1,g]} = -\frac{\omega_1^2}{2} + 4\omega_1 - \omega_2 \).

The graph theoretic representation is helpful to derive these effective Hamiltonians. An example is presented in Fig. 4(a). There are three manifolds in this figure, (4,-1), (3,0), and (2,1). The red path in Fig. 4(a) indicates the (4-1) transition; the green and blue paths correspond to the (3,0) and (2,1) transitions, respectively. As can be seen in Eqs. (4,4), for each case, the diagonal entries of the corresponding \( 2 \times 2 \) Hamiltonian are the energies of the initial and final states of each graph path; and the off-diagonal coupling elements are proportional to the product of the weights of the edges involved in each path. These weights construct the parameter space of the driven system, \( \{ \Delta, A_1, A_2 \} \). The transition probabilities presented in Fig. 4(b) are obtained by solving the eigenvalue problem in Eqs. (2)-(4). The prominent peaks in the full Floquet spectrum are reproduced, but shifted, with the reduced graph-theoretic Hamiltonian approach.

The spectral peaks shift from the bare states due to photon dressing \([33]\). Within the rotating-wave approximation, such resonance shift can be expressed analytically in the form,
\[ \xi_n = \frac{A_1^2}{2(\omega_1 + \omega_2)} \left( \frac{1}{n + 1} + \frac{(A_1/A_2)^2}{n + 1} \right). \]

where \( n \) indicates the number of photons involved in the transition. This effect is presented in Fig. 4(c) for the shift observed in the position of the (3,0) transition peak.

We now address the origin of the 3-photon transition suppression shown in Fig. 5(a,b). The arrows indicate the transition suppression in the (0,3) manifold. The effect is also observed in the other 3-photon manifolds, e.g. (-1,4), (-2,5), (-3,6), etc.. The transmission blockade of more than two photons has been previously reported \([34,35]\). The suppression of the transition probability can also be due to the coherent superposition of high-order Fourier harmonics closing the dynamical gap between the Floquet states \([36]\).

To explain the mechanism of such suppression for the (0,3) manifold, we reduced the size of the original Floquet matrix, Eq. (S18), by allowing only the absorption and emission processes up to three photons for each microwave mode. The Fourier index set for each mode is now reduced to \( n_i = 0, \pm 1, \pm 2, \pm 3 \). The bare state is denoted by index 0, and the dressed states are given by photon number \( \pm 1, \pm 2, \pm 3 \). The truncated matrix has now the size of \( 2 \times 7 \times 7 = 98 \). Since we have removed all of the higher multiphoton contributions from the Floquet matrix, the only available paths on the reduced 2D Floquet space involve only four states, see Fig. (S1).

As presented in the inset contour plot in Fig. 5 the suppression effect can be achieved by tuning the driving frequencies in the vicinity of a crossing between the Floquet states. The 3-photon resonance suppression can be explained by population trapping on the graph vertices connecting the \( |0,3,g> \) state to the \( |0,0,e> \) state, e.g. Fig. (S1). An intuitive analogy is the traffic congestion at an intersection along a route that connects two points on a map. As the solid red curve in Fig. 5 indicates, such population trapping occurs in the \( |0,1> \) state. The occurrence of a maximum in the \( |0,1> \) population manifests itself as the suppression of the (0,3) transition probability (solid blue line).

In this work, nonlinear multiphoton up/down frequency conversion in a SC transmon circuit is realized and reproduced with a full Floquet technique with longitudinal coupling. Multiphoton spectra over a wide microwave range, show photon pumping from one microwave field to another. To reveal the underlying physics in the single- and multiphoton pumping mechanisms, ef-
fective Hamiltonians are derived with a graph-theoretic approach, to model the transitions. The suppression of pumping at particular resonant frequencies are shown to be due to population trapping in dressed qubit states of the qubit. The proposed experimental design and the theoretical approach can be extended to tunable qubit circuits or solid state semiconductors, which offer controllability over the energy gaps. Such feasibly tunable multiphoton conversion is of interest in quantum transducer design and fabrication for future quantum networks, and to maintain the interconnectivity of remote SC quantum machines.

ACKNOWLEDGMENTS

HRS and HZJ acknowledge the NSF support through a grant for ITAMP at Harvard University. S.H. is supported in part by NSF (PHY-1314861). This work was partially supported by the NKRDP of China (Grant No.2016YFA0301801), NSFC (11474154), PAPD, and Dengfeng Project B of Nanjing University.

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[31] (A) Experimental details. (B) The derivation of the two-mode Floquet matrix from the Hamiltonian in Eq.(1) is provided in the supplemental materials. (C) The 2-by-2 effective Hamiltonians for different transition manifolds are extracted from the full Floquet matrix by adiabatic elimination of the intermediate dressed states.

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SUPPLEMENTAL MATERIAL

A. Experimental details

The transmon is made via electron beam lithography and double-angle evaporation, in which a single Al/AlO\(_x\)/Al Josephson junction is capacitively shunted by two Al pads on a high-resistance Si substrate. The fundamental TE101 mode of the aluminum cavity is \(\omega_{\text{cav}} = 10.673 \text{ GHz}\). The device is located in an Oxford Triton 400 dilution refrigerator below 10 mK with magnetic shielding. Three microwave drives are used: Qubit control waves are continuous while the readout wave is triggered. The measure wave and the microwaves are combined by two power splitters at room temperature before being sent to the dilution refrigerator. The microwave lines to the cavity are heavily attenuated at each stage of the dilution refrigerator and sent to the cavity through low-pass filters with a cutoff frequency of 12 GHz. The output signal from the cavity is passed through cryogenic circulators and a high-electron-mobility transistor amplifier located in the dilution refrigerator and further amplified at room temperature. It is then mixed down and digitized by a data acquisition card. In a sampling period, the measure wave is turned on at \(t = 70 \text{ ns}\) for \(2.1 \mu\text{s}\) and the data acquisition card starts to record at \(t = 270 \text{ ns}\), with a repetition rate of 10 kHz. The states of the transmon are measured with a Jaynes-Cummings readout. The measure wave is applied at the bare cavity frequency.

B. Two-mode Floquet formalism with longitudinal coupling scheme

We consider the interaction of the qubit (modeled as a two-level system) with two microwaves. The relaxation terms have not been included in the interaction Hamiltonian. As the presented results demonstrate, this approximation does not affect the observation of the multiphoton pumping qualitatively. The assignment of MW1 and Mw2 for the two microwave fields are arbitrary. Within the two-mode semiclassical Floquet approach the system will be treated quantum mechanically whereas the fields classically. The time evolution of the wave function is determined by the time-dependent Schrödinger equations (in atomic units, \(\hbar = m = e = 1\)),

\[
\frac{i}{\partial t} \Psi = \hat{H} \Psi, \tag{S.1}
\]

where

\[
\hat{H} = \hat{H}_0 - \frac{1}{2} \sum_i \xi_i(t) \sigma_z. \quad i = p, c \tag{S.2}
\]

\(\hat{H}_0\) is the unperturbed Hamiltonian of the two-level system, with eigenstates \([g>, |e>]\) and eigenvalues \((E_g, E_e)\),

\[
\hat{H}_0 = -\frac{1}{2}(\omega_q \sigma_z + \Delta \sigma_x), \tag{S.3}
\]

with \(\omega_q\) being the qubit resonance frequency, \(\Delta\) the tunnel splitting, and \(\sigma_{x,z}\) the Pauli matrices. The external field, \(\xi_i(t)\) is given by

\[
\xi_i(t) = A_i \cos(\omega_i t + \phi_i), \tag{S.4}
\]

with \(A_i\) and \(\omega_i\) being the peak amplitude, and the frequency of the microwave, respectively. According to the generalized Floquet theory, Eq. (S.1) has a solution that can be written as

\[
\Psi(t) = \exp(-iqt)\Phi(t). \tag{S.5}
\]

where \(\Phi(t)\) is periodic in time and \(q\) is called the quasienergy. Substituting Eq. (S.5) into Eq. (S.1), we obtain an eigenvalue equation for the quasienergy,

\[
\left[\hat{H}(t) - i \frac{\partial}{\partial t}\right] \Phi(t) = q\Phi(t). \tag{S.6}
\]

Since we are scanning a wide range of the two microwave frequencies, we consider the case of incommensurate frequencies, where the combined \(\omega_1\) and \(\omega_2\) frequencies is not necessarily periodic in time. According to the many-mode Floquet theorem, one can transfer this time-dependent problem into an equivalent time-independent infinite
dimensional Floquet matrix eigenvalue problem, so that the temporal part of the Hamiltonian $H(t)$ and the quasienergy eigenfunction $\Phi(t)$ can be expanded with the double-Fourier components of any arbitrary set of the fundamental frequencies $(\omega_1, \omega_2)$,

$$
\Phi(t) = \sum_{m_1, m_2} \Phi^{[m_1, m_2]} \exp[-i(m_1 \omega_1 t + m_2 \omega_2 t)],
$$

(S.7)

$$
\hat{H}(t) = \sum_{m_1, m_2} H^{[m_1, m_2]} \exp[-i(m_1 \omega_1 t + m_2 \omega_2 t)].
$$

(S.8)

where the time-independent coefficients $\Phi^{[m_1, m_2]}$ and $H^{[m_1, m_2]}$ are spanned by any orthonormal basis set. We employ the Floquet-state nomenclature,

$$
|\alpha, \{m_1, m_2\} \rangle = |\alpha> \otimes |\{m_1, m_2\} >.
$$

(S.9)

where $g$ and $e$ are system indices, representing the ground and excited states of the two-level system, respectively. $m_1$ and $m_2$ are the Fourier indices that run from $-\infty$ to $+\infty$. Substituting Eqs.(S.7), (S.8) into Eq.(S.6) and employing Eq.(S.9), gives the following time-independent two-mode Floquet matrix eigenvalue equation.

$$
\sum_{e} \sum_{\{k\}} <g; \{m_1, m_2\}|\hat{H}_F|e; \{k\}> <e; \{k\}|q_{\mu; \{n\}} > = q_{\mu; \{n\}} <g; \{m_1, m_2\}|q_{\mu; \{n\}} >.
$$

(S.10)

where $q_{\mu\nu}$ is the quasienergy eigenvalue and $|q_{\mu\nu}>$ is the corresponding eigenvector. The time-independent two-mode Floquet matrix, $\hat{H}_F$, is defined in terms of Eq.(S.9). More explicitly, $\hat{H}_F$ can be written, in matrix form as,

$$
<g; \{m_1\}|\hat{H}_F|e; \{k\}> = \hat{H}_F^{[m-e]} + \sum_i m_i \omega_i \delta_{\mu\nu} \delta_{\{m_i\}; \{k\}}. \quad i = 1, 2
$$

(S.11)

where $\delta_{\{i,j\}}$ is the Kronecker delta function. For the effective Hamiltonian of the superconducting transmon qubit, with longitudinal coupling,

$$
\hat{H} = -\frac{1}{2} \begin{bmatrix}
\delta(t) & \Delta \\
\Delta & \delta(t)
\end{bmatrix}.
$$

(S.12)

the structure of the Floquet matrix, which is Hermitian, is illustrated below,

$$
C = -\frac{1}{2} \begin{bmatrix}
\omega_q & \Delta \\
\Delta & -\omega_q
\end{bmatrix},
$$

(S.13)

$$
X = -\frac{1}{4} \begin{bmatrix}
A_1 & 0 \\
0 & -A_1
\end{bmatrix},
$$

(S.14)

$$
Y = -\frac{1}{4} \begin{bmatrix}
A_2 & 0 \\
0 & -A_2
\end{bmatrix},
$$

(S.15)

$$
A = \begin{bmatrix}
\ddots & \\
C - 2\omega_1 I & X & 0 & 0 & 0 \\
X & C - \omega_1 I & X & 0 & 0 \\
0 & X & C & X & 0 \\
0 & 0 & X & C + \omega_1 I & X \\
0 & 0 & 0 & X & C + 2\omega_1 I \\
\ddots & 
\end{bmatrix},
$$

(S.16)
\[ B = \begin{bmatrix}
Y & 0 & 0 & 0 \\
0 & Y & 0 & 0 \\
0 & 0 & Y & 0 \\
0 & 0 & 0 & Y
\end{bmatrix}, \quad (S.17) \]

\[ \hat{H}_F = \begin{bmatrix}
\vdots & & & \\
A - 2\omega_2 I & B & 0 & 0 & 0 \\
B & A - \omega_2 I & B & 0 & 0 \\
0 & B & A & B & 0 \\
0 & 0 & B & A + \omega_2 I & B \\
0 & 0 & 0 & B & A + 2\omega_2 I \\
\vdots & & & & 
\end{bmatrix} \quad (S.18) \]

The multiply periodic structure of \( \hat{H}_F \) holds the following important periodic relationships for its eigenvalues, and eigenvectors,

\[ q_{\mu;\{n+k\}} = q_{\mu;\{n\}} + \sum_i k_i \omega_i, \quad i = 1, 2 \quad (S.19) \]

and

\[ < g;\{m_i + k\}|q_{\mu;\{n+k\}}|e;\{m\}> = < g;\{m_i\}|q_{\mu;\{n\}}|e;\{0\}>. \quad (S.20) \]

Eigenvalues of the Floquet matrix in Eq.(S.18) are numerically solved by truncating the number of Floquet blocks. The converged results presented in this paper are generated by the inclusion of 21 blocks (adsorption and emission up to 10 photons for each mode). After solving the eigenvalue problem of the Floquet matrix, the time-averaged transition probability between \( |g> \) and \( |e> \) can be computed as,

\[ \mathcal{P}_{g\rightarrow e} = \sum_{\mu} \sum_{\{m\}} \sum_{\{n\}} |< e;\{m\}|q_{\mu;\{n\}}> \cdot < q_{\mu;\{n\}}|g;\{0\}>|^2. \quad (S.21) \]

Eq.(S.21) is corresponding to the probability of finding the qubit system in the excited state in the experiment. The transition probability plots and contour plots in Figs.(1), (3) are obtained by applying this equation.

There has been no approximation made in Eq.(S.18) to solve the time-dependent Hamiltonian of Eq.(S.12). Therefore this matrix can be applied for all parameter regimes. Indeed, all the theoretical results presented in Fig.(1), and Fig.(3) are obtained by solving the eigenvalue problem of this two-mode Floquet matrix, and calculating the time-averaged transition probabilities between \( |g> \) and \( |e> \) using Eq.(S.21). Although truncated, the size of the Floquet matrix that one needs to actually solve to get each data points in a contour plot, like the one presented in Fig.1(b), can increase dramatically for the higher-order processes. In this study, to obtain convergence in the three-photon transition regime, we truncated the matrix at \( 2 \times 21 \times 21 = 882 \), which includes all the transition channels for the qubit with \( \pm 10 \) photons processes.

C. Effective Hamiltonian in multiphoton region

Figure (S1) shows the four possible paths from \( |3,0,g> \) to \( |0,0,e> \), which are responsible for the \( (3,0) \) peak reported in Fig.4(b).

The truncated Floquet Hamiltonian for this manifold can be written as,

\[ \hat{H}_F = \begin{bmatrix}
-\frac{\omega_2}{2} + 3\omega_1 & \Delta & 0 & 0 & 0 \\
\Delta & \frac{\omega_2}{2} + 3\omega_1 & A_1 & 0 & 0 \\
0 & A_1 & \frac{\omega_2}{2} + 2\omega_1 & A_1 & 0 \\
0 & 0 & A_1 & \frac{\omega_2}{2} + \omega_1 & A_1 \\
0 & 0 & 0 & A_1 & \frac{\omega_2}{2}
\end{bmatrix}. \quad (S.22) \]
For any of these paths, one can write the set of five Schrödinger equations for the evolution of the dressed states involved, with their corresponding amplitudes, $c_1$-$c_5$. For instance, the path presented in the top-left panel in Fig. (S1), would be formulated as,

\[
\begin{align*}
    i\dot{c}_1 &= \left(-\frac{\omega q}{2} + 3\omega_1\right)c_1 + \Delta c_2, \\
    i\dot{c}_2 &= \left(\frac{\omega q}{2} + 3\omega_1\right)c_2 + A_1 c_3, \\
    i\dot{c}_3 &= \left(\frac{\omega q}{2} + 2\omega_1\right)c_3 + A_1 c_4, \\
    i\dot{c}_4 &= \left(\frac{\omega q}{2} + \omega_1\right)c_4 + A_1 c_5, \\
    i\dot{c}_5 &= \left(\frac{\omega q}{2}\right)c_5.
\end{align*}
\]

(S.23)

Under adiabatic approximation, we can assume that the population of the three intermediate states, do not change significantly, i.e. $c_2 = c_3 = c_4 = 0$. This gives,

\[
\begin{align*}
    c_2 &= -\frac{A_1 c_3}{\frac{\omega q}{2} + 3\omega_1}, \\
    c_3 &= -\frac{A_1 c_4}{\frac{\omega q}{2} + 2\omega_1}, \\
    c_4 &= -\frac{A_1 c_5}{\frac{\omega q}{2} + \omega_1}.
\end{align*}
\]

(S.24)

The effective coupling rate between the initial, $|3,0,g\rangle$, and final, $|0,0,e\rangle$, states, through this particular path, can be then obtained by plugging in these coefficients into the $c_1$ and $c_5$ rate equations in Eq. (S.23),

\[
g_{eff}^{\text{path}(1)} = -\frac{A_1^3 \Delta}{E_{[3,0,e]} E_{[2,0,e]} E_{[1,0,e]}}.
\]

(S.25)

where $E_{[3,0,e]} = \frac{\omega q}{2} + 3\omega_1$, $E_{[2,0,e]} = \frac{\omega q}{2} + 2\omega_1$, and $E_{[1,0,e]} = \frac{\omega q}{2} + \omega_1$. To obtain the total effective coupling rate, one should sum over the coupling strengths through other three possible pathways; meaning that,

\[
g_{eff}^{\text{total}} = g_{eff}^{\text{path}(1)} + g_{eff}^{\text{path}(2)} + g_{eff}^{\text{path}(3)} + g_{eff}^{\text{path}(4)}.
\]

(S.26)
FIG. S2. Graph-theoretical illustration of the ten possible paths on the 2D Floquet subspace, representing the (2,1) transition manifold. The weight of the edges are given as the Mw1 power, $A_1$, Mw2 power, $A_2$, and the tunnel splitting, $\Delta$.

where,

\[
\begin{align*}
 g_{\text{path}(2)\ eff} &= -\frac{A_1^3 \Delta}{E_{[2,0]g}E_{[2,0]e}E_{[1,0]e}}, \\
 g_{\text{path}(3)\ eff} &= -\frac{A_1^3 \Delta}{E_{[2,0]g}E_{[1,0]g}E_{[1,0]e}}, \\
 g_{\text{path}(2)\ eff} &= -\frac{A_1^3 \Delta}{E_{[2,0]g}E_{[1,0]g}E_{[0,0]g}},
\end{align*}
\] (S.27)

with $E_{[3,0]g} = -\frac{\omega q}{2} + 2\omega_1$, $E_{[1,0]g} = -\frac{\omega q}{2} + \omega_1$, and $E_{[0,0]g} = -\frac{\omega q}{2}$.

Eq. (S.27) can be simplified by writing all the energies in terms of $E_{[3,0]g}$ as (see Fig. (S1))

\[
\begin{align*}
 E_{[3,0]e} &= E_{[3,0]g} + \Delta, \\
 E_{[2,0]e} &= E_{[3,0]g} + \Delta - A_1, \\
 E_{[1,0]e} &= E_{[3,0]g} + \Delta - 2A_1, \\
 E_{[2,0]g} &= E_{[3,0]g} - A_1, \\
 E_{[1,0]g} &= E_{[3,0]g} - 2A_1.
\end{align*}
\] (S.28)

By doing some algebra and keeping only the leading terms in $E_{[3,0]g}$, the $(2 \times 2)$ effective Hamiltonian can be approximated as,

\[
\begin{align*}
 \hat{H}_F^{(3,0)} &= \begin{bmatrix} E_{[3,0]g} g_{\text{eff}(3,0)}^{(3,0)} \\ g_{\text{eff}(3,0)}^{(3,0)} E_{[0,0]e} \end{bmatrix}, \\
 g_{\text{eff}(3,0)}^{(3,0)} &= -\frac{4A_1^3 \Delta}{E_{[3,0]g}^3}.
\end{align*}
\] (S.29)

where $E_{[3,0]g} = -\frac{\omega q}{2} + 3\omega_1$, and $E_{[0,0]e} = \frac{\omega q}{2}$. It is worth noting that in Eqs. (S.25)-(S.27) all the four paths share the same nominator, which remains the same in Eq. (S.29).
The same approach can be used to obtain the $(2\times2)$ effective Hamiltonian for the $(2,1)$ transition manifold. As can be seen in Fig. (S2), there are ten possible paths that connect the $|2,1,g\rangle$ vertex to $|0,0,e\rangle$ vertex. The only task here would be to count for the weighted edges for one of these paths. It should be mentioned again that the nominator of the effective coupling in all these ten paths are the same as it appears in Eq. (S.30). Within the same approximation discussed above, one could simply write,

\[
\hat{H}_F^{(2,1)} = \begin{bmatrix} E_{[2,1,g]}^{(2,1)} & g_{e_{ff}}^{(2,1)} \\ g_{e_{ff}}^{(2,1)} & E_{[0,0,e]}^{(2,1)} \end{bmatrix}, \quad g_{e_{ff}}^{(2,1)} = -\frac{10 A_1^2 A_2 \Delta}{E_{[2,1,g]}^{(2,1)}}
\]  
(S.30)

where $E_{[2,1,g]} = -\frac{\omega q^2}{2} + 2\omega_1 + \omega_2$.

The $(2\times2)$ effective Hamiltonian for $(4,-1)$ is obtained similarly, considering the fact that there will be 20 possible pathways connecting $E_{[4,-1,g]}$ and $E_{[0,0,e]}$:

\[
\hat{H}_F^{(4,-1)} = \begin{bmatrix} E_{[4,-1,g]}^{(4,-1)} & g_{e_{ff}}^{(4,-1)} \\ g_{e_{ff}}^{(4,-1)} & E_{[0,0,e]}^{(4,-1)} \end{bmatrix}, \quad g_{e_{ff}}^{(4,-1)} = -\frac{20 A_1^4 A_2 \Delta}{E_{[4,-1,g]}^{(4,-1)}}
\]  
(S.31)

where $E_{[4,-1,g]} = -\frac{\omega q^2}{2} + 4\omega_1 - \omega_2$. 