Ultrastrong-coupling effects induced by a single classical drive in Jaynes-Cummings-type systems

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We propose the effective simulation of light-matter ultrastrong-coupling phenomena with strong-coupling systems. Recent theory and experiments have shown that the single-atom quantum Rabi model can be simulated by the Jaynes–Cummings model with two additional classical drives. Here, we show that quantum nonlinear optical phenomena, relying on the counter-rotating terms of the Rabi model, can be implemented by the Jaynes-Cummings and Tavis–Cummings models with only a single classical drive. We analyze three examples: a single atom exciting two photons, frequency conversion, and a single photon exciting two atoms.

Introduction.—The ultrastrong coupling (USC) of light and matter is attracting increasing interest beyond the fields of cavity [1] and circuit [2, 3] quantum electrodynamics (QED) [4, 5]. This interest has been stimulated in the last decade by several experiments finally reaching USC in a variety of physical systems, e.g., superconducting quantum circuits, intersubband polaritons, Landau polaritons, organic molecules, and quantum optomechanical systems [4, 5]. The USC of light and matter (e.g., a cavity mode and a natural or artificial atom) occurs when their coupling strength g becomes comparable to the atomic (ωa) or cavity (ωc) frequencies. More precisely, according to the usual convention, the USC regime occurs when η = max(g/ωa, g/ωc) is in the range [0, 1]. The regime η ≥ 1 is often referred to as deep strong coupling (DSC) [6].

Compared to strong coupling (SC; η < 0.1, but g larger than the loss rates in the system), USC opens new perspectives for efficiently simulating known effects and observing fundamentally new phenomena in quantum nonlinear optics [7–17], quantum field theory, supersymmetric (SUSY) field theories [18], cavity optomechanics [19–26], quantum plasmonics [21, 27–29], light-induced superconductivity [30, 31], quantum thermodynamics [32], photochemistry (chemistry QED) [33–36], as well as metamaterial and material sciences. For a more detailed review, see Refs. [4, 5]. Ultrastrong coupling also has applications in quantum metrology and spectroscopy [37] and quantum information processing (QIP), including novel and efficient realizations of protected QIP [38], holonomic QIP [39], quantum gates [40, 41], quantum memories [42, 43], and quantum error correction codes [13].

The basic model for USC of a single two-level atom to a single-mode cavity is the quantum Rabi model [44, 45] (QRM). Its multi-atom or multi-mode generalizations include the Dicke [46] and Hopfield [47] models. When η < 0.1, these models for USC can be reduced to the simpler Jaynes–Cummings model [48] (JCM) and its multi-mode or multi-atom generalizations (e.g., the Tavis–Cummings model [49]). Since SC is easier to realize in experiment than USC, the question arises whether the predicted USC phenomena can be observed or at least simulated also in the SC regime, e.g., by adding classical drives applied to atom(s) or cavity mode(s) in the SC models. We note that simulating the QRM could also enable simulating other closely related fundamental quantum models, which include the spin-boson [50, 51] and Kondo [50, 52, 53] renormalization-group models, the Rashba-Dresselhaus model [18], and a Jahn-Teller model [54–58] among others. Even vacuum-induced symmetry breaking [59], which is analogous to the Higgs mechanism, has been predicted in the USC regime.

Quantum simulations are among the most important applications of quantum technologies [60, 61]. Quantum simulations of the atom-cavity dynamics in the USC and DSC regime in the Rabi and Dicke models have recently attracted much theoretical [62–74] and experimental [75–78] interest. The methods described in Refs. [63, 65] and implemented in circuit-QED [76] and trapped-ion experiments [78] simulate the QRM in the USC regime with a light-matter system described by the JCM in the SC regime. These quantum simula-

FIG. 1: Sketches of the two setups that we consider for observing ultrastrong-coupling phenomena. (a) A single two-level atom of frequency ωa coupled to two cavities of frequencies ω1 and ω2. (b) Two two-level atoms of frequency ωa coupled to a cavity of frequency ωc. In both setups, a single coherent drive of frequency ω1 and amplitude Ω is applied to each atom.
tions require two drives to be applied to a system with a single atom and a single-mode resonator.

In this Letter, we propose a method for quantum simulations of USC light-matter phenomena using only a single drive applied to a multi-atom or multi-mode system, as illustrated in Fig. 1. We show how this method can be used to simulate hallmark USC processes that do not conserve the number of excitations in the light-matter system [11]: a single two-level atom emitting two photons [8], frequency conversion of two photonic modes coupled to a two-level atom [12], and a single photon exciting two atoms [10]. We also give a protocol for an experimental implementation and show that different well-developed experimental systems can be used for such an implementation. Given the breadth of USC research areas outlined above, we expect that this new simulation method will find many more applications.

Hamiltonians for light-matter coupling.—The QRM describes the interaction between a two-level atom (qubit) of frequency $\omega_c$ and a cavity mode of frequency $\omega_c$ by the Hamiltonian ($\hbar = 1$)

$$H_R = H_0 + \sigma_z X = H_0 + g(\sigma + \sigma^\dagger)(a + a^\dagger),$$

where $H_0 = (\omega_a/2)^\sigma_z + \omega_c a^\dagger a$ is the free Hamiltonian, $a$ ($a^\dagger$) is the annihilation (creation) operator of the cavity mode, $X = a + a^\dagger$ is the canonical position operator, $x = \sigma + \sigma^\dagger$ and $\sigma^\circleslash = \sigma^\dagger$ are Pauli operators, $a\sigma x$ is the atomic lowering (raising) operator, and $g$ is the atom–cavity coupling constant. Under the rotating-wave approximation (RWA), which is valid if $\{\omega_c, \omega_a\} \gg \{g, |\omega_c - \omega_a|\}$, the counter-rotating terms, $\sigma^\dagger a^\dagger$ and $\sigma a$, in Eq. (1) can be ignored. This leads to the standard JCM described by the Hamiltonian $H_{JC} = H_0 + g(\sigma a^\dagger + \sigma^\dagger a)$. The counter-rotating terms can be effectively restored in the JCM in various ways, e.g., using cavity-light squeezing [71, 72] to enhance the coupling strength $g$. A simpler method is to apply classical drives, as suggested in Ref. [63]. Indeed, by applying two time-dependent classical drives to the atom, $H_{d\text{riv}} = \sum_{n=1,2} \Omega_n \sigma e^{i\omega_n t} + \sigma^\circleslash e^{-i\omega_n t}$, with driving strengths $\Omega_n$ and frequencies $\omega_n$, in addition to $H_{JC}$, the effective interaction Hamiltonian $H_{R}^\circ$ can simulate the QRM [63]. Note that $H_{R}^\circ$ is given as an approximate interaction in a rotated frame (where the first drive is time independent and spins are in the basis rotated by the Hadamard gate), assuming that $\Omega_1$ is relatively strong, and choosing the resonance condition for the second drive strength as $\Omega_2 = (\omega_1 - \omega_2)/2$. Under these assumptions and approximations, $H_{R}^\circ$ can be given by Eq. (1), but with a rescaled effective cavity frequency $\omega_c' = \omega_c - \omega_1$ and the effective coupling constant $g' = g/2$. Thus, the JCM with two classical drives can effectively simulate the QRM, where the ratio $\eta \equiv g/\omega_c$ can be effectively increased as $\eta' \equiv g'/\omega_c' = g/[2(\omega_c - \omega_1)]$ from the SC regime up to the USC regime, or even the DSC regime.

Here we show that, instead of simulating the QRM, we can enable particular nonlinear processes [8, 10–13] that have been reported in the USC, and that rely on the effect of counter-rotating terms, by using a simpler approach based on a single driving field. Our approach is inspired by earlier work on creating multi-photon states in cavity QED [79–81].

**USC effect I: Two photons excited by a single atom.**—We first consider the setup in Fig. 1(a), i.e., two cavities coupled to a single qubit that is coherently driven by a classical field. In a frame rotating with the frequency $\omega_L$ of the driving field, the Hamiltonian is given by

$$H = \Delta_1 a_1^\dagger a_1 + \Delta_2 a_2^\dagger a_2 + \Delta_\sigma \sigma^\dagger \sigma + \Omega(\sigma + \sigma^\dagger) + g[\sigma(a_1^\dagger + a_2^\dagger) + \text{h.c.}],$$

with $a_{1,2}$ the bosonic annihilation operators of the cavity modes; $\Delta_1$, $\Delta_2$ and $\Delta_\sigma$ are the frequency detunings between the cavities or qubit and the drive ($\Delta_x \equiv \omega_x - \omega_L$), $\Omega$ is the amplitude of the driving field, and $g$ is the coupling rate between the cavities and the qubit (considered to be equal for simplicity).

The part of Eq. (2) that only depends on $\sigma$ can be easily diagonalized. Denoting the ground and excited eigenstates of a non-driven qubit $|g\rangle$ and $|e\rangle$, respectively, the new eigenstates with the driving applied correspond to a rotated spin basis, i.e.,

$$|\pm\rangle = \cos \theta |g\rangle + \sin \theta |e\rangle = e^{i\eta g^2} |g\rangle, \hspace{1cm} (3)$$

$$|\mp\rangle = \sin \theta |g\rangle - \cos \theta |e\rangle = e^{i\eta g^2(n+\pi)} |g\rangle, \hspace{1cm} (4)$$

with $\cos \theta \equiv 1/\sqrt{1+\xi^2}$, $\sin \theta \equiv 1/\sqrt{1+\xi^2}$, $\theta \in [0, \pi/2]$, and $\xi \equiv \Omega/(|\Delta_2/2 + R|)$, where $R$ is the Rabi frequency given by $R \equiv \sqrt{\Omega^2 + (\Delta_\sigma/2)^2}$.

Working in the eigenbasis $|\pm\rangle$, the original lowering operator $\sigma$ can be written in terms of the new operators $\tilde{\sigma} \equiv |\mp\rangle\langle +|$ as $\sigma = s\tilde{\sigma} - c\tilde{\sigma}^\circleslash + cs\tilde{\sigma}_z$, with $s = \sin \theta, c = \cos \theta, \tilde{\sigma}_z = 2\tilde{\sigma}^\circleslash - 1$. Therefore, the resulting Hamiltonian in the rotated spin basis reads

$$H = \Delta_1 a_1^\dagger a_1 + \Delta_2 a_2^\dagger a_2 + \Omega\tilde{\sigma}_z + g\left(s^2\tilde{\sigma} - c^2\tilde{\sigma}^\circleslash + cs\tilde{\sigma}_z\right)(a_1^\dagger + a_2^\dagger) + \text{h.c.}. \hspace{1cm} (5)$$

The transition energy of the effective qubit is now given by $R$, which can be made small enough that counter-rotating terms of the kind $\tilde{\sigma}^\circleslash a_1^\dagger$ and $\tilde{\sigma}_z a^\dagger$ play a relevant role in the dynamics. The presence of the latter type of coupling terms, involving $\tilde{\sigma}_z$, makes $H$ reminiscent of the generalized QRM, where a coupling term proportional to $\tilde{\sigma}_z(a + a^\dagger)$ is added to the QRM in Eq. (1) [7, 8, 10, 11, 82, 83]. The presence of the $\tilde{\sigma}_z$ coupling term breaks parity symmetry and enables processes that changes the number of excitations in the system by an odd number [4, 5, 11].

We will now see how, in the limit of $\alpha \equiv g/R \ll 1$, the counter-rotating terms in Eq. (5) lead to Rabi oscillations between pairs of eigenstates of the bare Hamiltonian that are not directly coupled by the interactions [11], with Rabi frequencies $\propto g\eta$. In the effective USC regime when $\alpha \sim 0.1$, we find the optimal condition in which $g/R \ll 1$ remains valid, while the effective Rabi frequencies $\sim 0.1g$ can be significant compared to decoherence rates. The normal USC condition $\eta \gtrsim 0.1$ for observing these phenomena is thus lifted.
FIG. 2: USC effect I: Energy-level diagrams and transitions for the process where a single atom emits two photons. (a) Energy levels $E_n$, for the Hamiltonian in Eq. (5) as a function of $\Delta_1$. Parameters: $\Omega = 80\gamma$, $\Delta_2 = 3\Omega/2$, with $R = \sqrt{18}$. The Hilbert space is truncated at three photons for simplicity. (b) Zoom-in on the anti-crossing between the energy levels corresponding to $|+\rangle$ and $|-\rangle$, which couples the states $|\pm\rangle$ and $|\mp\rangle$ of the effective interaction between these two states. (c) The transitions in the second-order process that creates the effective coupling between $|+, m\rangle$ and $|-, n + 1, m + 1\rangle$. Red dashed (blue solid) arrows indicate transitions that change the total number of excitations in the system by one (zero), i.e. transitions mediated by counter-rotating (non-rotating) terms. The transition cannot take place without involving counter-rotating terms.

One example of such a nonlinear process is the simultaneous excitation of one photon in each cavity by the single qubit. By plotting the energy levels of Eq. (5) [Fig. 2(a)], and zooming in around $\Delta_1 + \Delta_2 \approx 2R$, we find an avoided-level crossing [Fig. 2(b)]. The interaction around this point is described by the effective Hamiltonian [84]:

$$H_{\text{eff}}^I = \Delta_1 a_1^+ a_1 + \Delta_2 a_2^+ a_2 + (R + \lambda)\sigma_z + \left(\chi_1 a_1^+ a_1 + \chi_2 a_2^+ a_2\right)\sigma_z + g_{\text{eff}}^I\left(a_1^+ a_1 \sigma^- + \text{h.c.}\right),$$

which couples the states $|+, n, m\rangle \leftrightarrow |-, n + 1, m + 1\rangle$, confining the dynamics inside that manifold. This effective interaction requires both states to be quasi-resonant, which implies, ignoring for now small dispersive energy shifts, the two conditions:

$$\Delta_1 + \Delta_2 \approx 2R,$$  
$$\Delta_1 \neq \Delta_2 \neq (\pm R, \pm 2R).$$

The second condition is imposed in order to be detuned from first-order processes (e.g., $\sigma a_1^+ + \text{h.c.}$ if $\Delta_1 = 2R$) and competing second-order processes (e.g., $\sigma a_1^2 + \text{h.c.}$ for $\Delta_1 = R$) exciting degenerate photon pairs within a single cavity [79–81]. The effective two-photon coupling rate in Eq. (6) is given by

$$g_{\text{eff}}^I = \frac{g^2}{Rf(1-f)} \cos^2\theta,$$

where we defined $\Delta_1 = 2Rf$ and $\Delta_2 = (1 - f)2R$, $f \in (0, 1)$, so that Eq. (7a) is automatically fulfilled. This effective interaction is mediated by the second-order processes shown in Fig. 2(c). The Lamb shift of the qubit is

$$\lambda = g^2\left[\frac{c^4}{2} \left(\frac{1}{\Delta_1} + \frac{1}{\Delta_2}\right) - \frac{s^4}{2} \left(\frac{1}{\Delta_1} + \frac{1}{\Delta_2}\right)\right],$$

and the dispersive coupling rates are

$$\chi_i = g^2\left(\frac{c^4}{\Delta_i^4} - \frac{s^4}{\Delta_i^4}\right),$$

with $\Delta_i^\pm \equiv \Delta_i \pm 2R$.

Equation (8) shows that the resonant-driving condition $\Delta_\sigma = 0$ ($\theta = \pi/4$) does not provide the maximum possible two-photon coupling rate. In particular, for a fixed $R$, we see that the optimal angle maximizing $g_{\text{eff}}^I$ is $\theta^* = \pi/3$. This angle yields the optimum value $g_{\text{eff}}^I(\theta^*) \approx 1.39g_{\text{eff}}^I(\theta = \pi/4)$. Alternatively, we can compute the optimal detuning $\Delta_\sigma$ for a fixed $\Omega$, which is experimentally more meaningful since varying $\Delta_\sigma$ for a fixed $\Omega$ is more straightforward than varying $\theta$ for a fixed $R$. By writing Eq. (8) explicitly in terms of $\Delta_\sigma$ and $\Omega$, we obtain the optimal detuning $\Delta_\sigma^* = \Omega/\sqrt{2}$. The corresponding value of $g_{\text{eff}}^I$ is then given by $g_{\text{eff}}^I(\Delta_\sigma^*) \approx 1.18g_{\text{eff}}^I(\Delta_\sigma = 0)$.

In order to obtain full two-photon Rabi oscillations between the two states $|1\rangle = |+, n, m\rangle, |2\rangle = |-, n + 1, m + 1\rangle$, the quasi-resonance condition Eq. (7a) needs to be fine-tuned to account for the Lamb shift of the qubit and the dispersive qubit-cavity couplings in Eq. (6), given by $\lambda$ and $\chi_1, \chi_2$. In other words, $\Delta_1$ and $\Delta_2$ must be chosen such that $\langle 1|H_{\text{eff}}|1\rangle = \langle 2|H_{\text{eff}}|2\rangle$. Introducing a correction $\delta$ such that $\Delta_1 = 2Rf + \delta$, we solve this equation for the Hamiltonian in Eq. (6) and obtain

$$\delta = 2\lambda + \chi_1(2n + 1) + \chi_2(2m + 1).$$

Experimental protocol.—We now discuss an experimental protocol for implementing and measuring the non-linear process. This protocol is shown in Fig. 3. Starting with no photons in the cavities and the qubit in its ground state $|g\rangle$, the first step is to apply a rotation of $2\theta$ around the $y$-axis to bring the qubit into the eigenstate $|+\rangle$. At this stage, the cavities and the qubit are detuned and no interaction takes place. Then, the
we obtain the effective Hamiltonian off-resonance. Following the same procedure outlined in [84], order processes introducing photon pairs into the cavities are where, again, the second condition guarantees that second-

Fig. 1(a) can also be exploited to engineer other processes, 

The setup in USC effect II: Frequency conversion.—The setup in Fig. 1(a) can also be exploited to engineer other processes, e.g., frequency conversion. In that case, we want to couple the states \( |n+1, m, -\rangle \) and \( |n, m+1, +\rangle \). The resonance condition then becomes

\[
\Delta_1 \approx 2R + \Delta_2, \\
\Delta_1 \neq \Delta_2 \neq \pm R, 
\]

where, again, the second condition guarantees that second-order processes introducing photon pairs into the cavities are off-resonance. Following the same procedure outlined in [84], we obtain the effective Hamiltonian

\[
H_{\text{eff} \text{III}}^{\text{II}} = \Delta_1 a_1^\dagger a_1 + \Delta_2 a_2^\dagger a_2 + (R + \lambda)\hat{\sigma}_z \\
+ \left( \chi_1 a_1^\dagger a_1 + \chi_2 a_2^\dagger a_2 \right)\hat{\sigma}_z + g_{\text{eff} \text{III}}^{\text{II}} (a_1^\dagger a_2 \hat{\sigma} + \text{h.c.}),
\]

having now defined \( \Delta_1 = 2fR \) and \( \Delta_2 = (f - 1)2R \), \( f \in (0, 1) \). Once again, driving the qubit on resonance does not maximize \( g_{\text{eff} \text{III}}^{\text{II}} \). Frequency-conversion-rate increases by 50-70% compared to resonant driving can be achieved by using the optimal angle \( \theta^* \) or the optimal detuning \( \Delta^* \), whose analytical expressions can be found in the Supplemental Material [84].

USC effect III: Two atoms excited by a single photon.—The last process that we demonstrate is the excitation of two atoms by a single photon, i.e., the direct coupling between the states \( |+, +, n\rangle \) and \( |--, n+1\rangle \). We now consider the setup in Fig. 1(b), i.e., a cavity coupled to two coherently driven qubits, with lowering operators \( \sigma_{1,2} \). For simplicity and without loss of generality, we consider both qubits to have the same transition frequencies. In the rotating frame of the driving, the Hamiltonian is

\[
H = \Delta_0 a_1^\dagger a_1 + \Delta_\sigma \left( \sigma_{1,2}^\dagger \sigma_{1,2} + \Omega (\sigma_1 + \sigma_2 + \text{h.c.}) \right) \\
+ g \left[ a \left( \sigma_{1,2}^\dagger + \sigma_{1,2}^\dagger \right) + \text{h.c.} \right],
\]  

As in the previous cases, the effective coupling can be maximized by driving the qubit slightly off resonance, using either the optimal angle \( \theta^* \) or the optimal detuning \( \Delta^* \), whose expressions we provide in [84].

Experimental implementations.—The results presented here are based on very fundamental models that describe the exchange of single excitations between a qubit and a harmonic oscillator, and can therefore be applied in many different systems. In Table I, we compare, under experimentally feasible assumptions, the effective coupling strengths \( g_{\text{eff}} \) and decoherence rates \( \gamma \) that can be obtained in five experimental platforms. An experimental implementation is feasible when \( g_{\text{eff}}/\gamma > 1 \), i.e., when the effective coupling is strong. Table I shows that the second-order processes we have proposed...
The table below lists the experimentally feasible effective rates for the three processes discussed in the text: (I) a single photon exciting two atoms, (II) frequency conversion, and (III) a single atom exciting two photons. We have set $\Omega/g = 20$: $\gamma$ refers to the largest decoherence rate in the system.

| System                      | $g/(2\pi)$ | $\gamma/(2\pi)$ | $(g_{\text{eff}}, g_{\text{H}}, g_{\text{III}})/\gamma$ |
|----------------------------|------------|------------------|-------------------------------------------------|
| Natural atoms [85]         | 34 MHz     | 4.1 MHz          | $(0.6, 0.4, 0.004)$                              |
| Trapped ions [86]          | 10 kHz     | 100 MHz          | $(7.9, 5.1, 0.05)$                               |
| Quantum acoustics [87]     | 16 MHz     | 0.6 MHz          | $(2.1, 1.3, 0.01)$                               |
| Circuit QED [88]           | 335 MHz    | 0.5 MHz          | $(52.9, 33.8, 0.36)$                             |
| Quantum dots [89]          | 19.3 GHz   | 6.0 GHz          | $(0.3, 0.2, 0.002)$                              |

TABLE I: Experimentally feasible effective rates for the three processes discussed in the text: (I) a single photon exciting two atoms, (II) frequency conversion, and (III) a single atom exciting two photons. We set $\Omega/g = 20$: $\gamma$ refers to the largest decoherence rate in the system.

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Conclusions.---We have presented a experimentally simple method for quantum simulations of phenomena in the USC regime, requiring only a single qubit drive to be applied on a system in the SC regime. Our method is ready for its implementation on several existing experimental platforms and opens up new possibilities for exploring USB physics.

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Supplementary Material

Contents

Effective Hamiltonians 8
USC effect I: Details for two photons excited by a single atom 9
USC effect II: Details for frequency conversion 9
USC effect III: Details for two atoms excited by a single photon 10
Validity of the perturbation theory 11
Effect of decoherence 12

Effective Hamiltonians

In this work, we use a matrix form of perturbation theory that allows one to obtain energy corrections to arbitrary orders with a single matrix inversion. Let us consider a Hilbert subspace \( \mathcal{A} \) consisting of \( N_A \) states \( \{|a_1\rangle, |a_2\rangle, \ldots \} \) whose effective dynamics we wish to describe. This subspace is coupled to another subspace \( \mathcal{B} \) consisting of \( N_B \) states \( \{|b_1\rangle, |b_2\rangle, \ldots \} \) that we want to adiabatically eliminate. We define the projectors onto the respective subspaces as \( P_A \) and \( P_B \). The total Hamiltonian of the combined system is given by

\[
H = \begin{pmatrix} h & V \\ V^\dagger & \tilde{H} \end{pmatrix},
\]

(S1)

where \( h = P_A \hat{H} P_A \) is an \((N_A \times N_A)\) matrix acting only on \( \mathcal{A} \), \( \hat{H} = P_B \hat{H} P_B \) is an \((N_B \times N_B)\) matrix acting only on \( \mathcal{B} \), and \( V = P_B \hat{H} P_A \) is an \((N_A \times N_B)\) matrix coupling both subspaces. Our objective is to obtain an effective Hamiltonian \( h^{\text{eff}} \) describing the dynamics within \( \mathcal{A} \). The underlying assumption is that the eigenvalues of \( h \) are close to the energy \( E \), while the eigenvalues of \( \tilde{H} \) are detuned from \( E \) by values much larger than the elements of \( V \), and therefore can be adiabatically eliminated. This is done by writing the eigenvalue problem:

\[
\begin{pmatrix} h & V \\ V^\dagger & \tilde{H} \end{pmatrix} \begin{pmatrix} \phi \\ \chi \end{pmatrix} = E \begin{pmatrix} \phi \\ \chi \end{pmatrix},
\]

(S2)

where \( \phi \) and \( \chi \) are column vectors of length \( N_A \) and \( N_B \), respectively. After matrix multiplication, we obtain the following system of two equations for \( \phi \) and \( \chi \):

\[
(E - h)\phi = V\chi, \quad (S3a)
\]

\[
(E - \tilde{H})\chi = V^\dagger\phi. \quad (S3b)
\]

By solving Eq. \( (S3b) \) and substituting into Eq. \( (S3a) \), we obtain:

\[
(E - H^{\text{eff}})\phi = 0, \quad (S4)
\]

where \( H^{\text{eff}}(E) = h + \delta h \), and

\[
\delta h = V \frac{1}{E - \tilde{H}} V^\dagger. \quad (S5)
\]

\( H^{\text{eff}} \) corresponds to effective Hamiltonians that we have presented in the main text. Notably, this simple expression includes contributions from processes beyond second-order perturbation theory; the order of such processes is encoded in the size of the matrix. In the following sections we provide further details on how the effective Hamiltonian was obtained in the three cases studied in the main text.
For simplicity, those processes are neither depicted in Fig. 2 of the main text nor in Fig. S1 and S2. Using shifts and dispersive cavity-qubit couplings, through processes such as

\[ |n+1,0,+\rangle \rightarrow |n,1,−\rangle \]

Note that states such as

\[ |n,1,−\rangle \]

The transitions in the second-order process that creates the effective coupling between

\[ \chi \]

in the absence of coupling

\[ E_0 = \Delta_2 \]

The Hilbert space is truncated at 1 photon for simplicity. (b) Zoom-in on the anti-crossing between the energy levels corresponding to \(|1,0,\pm\rangle\) and \(|0,1,−\rangle\). The size of the level splitting at the resonance \(\Delta_1 = \Delta_1^f = 2R + \Delta_2 + \delta = 2RF + \delta\) — where both states have the same energy in the absence of coupling

\[ E_0 = \Delta_2 + \lambda = R(2F + \delta) \]

— indicates the strength \(g_{\text{eff}}\) of the effective interaction between these two states. (c) The transitions in the second-order process that creates the effective coupling between \(|n,m,−\rangle\) and \(|n−1,m,+,+\rangle\). Blue solid arrows: transitions that conserve the number of excitations. Red dashed arrows: transitions that change the number of excitations by one. Blue dashed arrows: transitions that change the number of excitations by two.

### USC effect I: Details for two photons excited by a single atom

We consider the following two subspaces, with \(N_A = 2\) and \(N_B = 12\):

- \(A = \{|n, m, +\rangle, |n+1, m+1, −\rangle\}\),
- \(B = \{|n+1, m, ±\rangle, |n, m, ±\rangle, |n+2, m+1, ±\rangle, |n+1, m+2, ±\rangle, |n, m−1, ±\rangle, |n−1, m, ±\rangle\}\).

Note that states such as \(|n+2, m+1, ±\rangle\) do not contribute to the effective coupling between the two states in \(A\), but to the Lamb shifts and dispersive cavity-qubit couplings, through processes such as \(|n+1, m+1, −\rangle \rightarrow |n+2, m+1, ±\rangle \rightarrow |n+1, m+1, −\rangle\).

For simlicity, those processes are neither depicted in Fig. 2 of the main text nor in Fig. S1 and S2. Using \(E = \Delta_1 n + \Delta_2 m + R\), the correction to the effective Hamiltonian in the subspace \(A\), given by Eq. (S5), has the form:

\[
\delta h^I = \left( \begin{array}{c}
\chi_1 n + \chi_2 m + \lambda + C \\
g_{\text{eff}}^I \sqrt{(n+1)(m+1)} - \chi_1 (n+1) - \chi_2 (m+1) - \lambda + C
\end{array} \right),
\]

which allows us to extract \(\chi_1\), \(\chi_2\), \(\lambda\) and \(g_{\text{eff}}^I\), and, omitting any overall shift \(C\), to write the effective Hamiltonian \(H_{\text{eff}}^I\) in the general form

\[
H_{\text{eff}}^I = \Delta_1 a_1^\dagger a_1 + \Delta_2 a_2^\dagger a_2 + (R + \lambda) \sigma_z + \left(\chi_1 a_1^\dagger a_1 + \chi_2 a_2^\dagger a_2\right) \sigma_z + g_{\text{eff}}^I \left(a_1^\dagger a_2^\dagger \sigma + \text{h.c.}\right).
\]

The expressions for \(g_{\text{eff}}^I\), \(\chi_1\), \(\chi_2\) and \(\lambda\) are provided in the main text.

### USC effect II: Details for frequency conversion

We consider the following two subspaces, with \(N_A = 2\) and \(N_B = 12\):

- \(A = \{|n+1, m, −\rangle, |n, m+1, +\rangle\}\),
- \(B = \{|n+1, m+1, ±\rangle, |n, m, ±\rangle, |n+2, m, ±\rangle, |n+1, m−1, ±\rangle, |n, m+2, ±\rangle, |±, n−1, m\rangle\}\).

Here, the correction that we obtain is

\[
\delta h^{II} = \left( \begin{array}{c}
-(n+1)\chi_1 - m\chi_2 - \lambda + C \\
g_{\text{eff}}^{II} \sqrt{(n+1)(m+1)} - \chi_1 n - \chi_2 m - \lambda + C
\end{array} \right),
\]

which allows us to write the general form of the effective Hamiltonian,

\[
H_{\text{eff}}^{II} = \Delta_1 a_1^\dagger a_1 + \Delta_2 a_2^\dagger a_2 + (R + \lambda) \sigma_z + \left(\chi_1 a_1^\dagger a_1 + \chi_2 a_2^\dagger a_2\right) \sigma_z + g_{\text{eff}}^{II} \left(a_1^\dagger a_2^\dagger \sigma + \text{h.c.}\right).
\]
The expression for $g_{\text{eff}}^{\text{II}}$ is given in the main text. The dispersive coupling rates are in this case given by

$$\chi_i = g^2 \left( \frac{c^4}{2R + \Delta_i} + \frac{s^4}{2R - \Delta_i} \right),$$

(S8)

and the Lamb shift is

$$\lambda = \frac{g^2}{2} \left[ \frac{c^4(4R + \Delta_1 + \Delta_2)}{2(R + \Delta - 1)(2R + \Delta_2)} + \frac{s^4(4R - \Delta_1 - \Delta_2)}{(2R - \Delta_1)(2R - \Delta_2)} \right].$$

(S9)

The dispersive couplings and the Lamb shift make the diagonal elements of $\delta h^{\text{II}}$ unequal. Since both elements need to be equal in order to achieve complete Rabi oscillations, one needs to introduce a small correction the resonance condition $\Delta_1 + \Delta_2 = 2R$.

Introducing the correction $\delta$ such that $\Delta_1 = 2Rf + \delta$ into the final expression of $H_{\text{eff}}$ (which implies the approximation of ignoring $\delta$ during the derivation of $H_{\text{eff}}$), and imposing that the diagonal elements are equal, we are left with the expression for the correction to the general resonance condition:

$$\delta = (2n + 1)\chi_1 + (2m + 1)\chi_2 + 2\lambda.$$

(S10)

This expression in terms of $\chi_i$ and $\lambda$ coincides with the one obtained for the case of two photons excited by a single atom. Similarly, driving the qubit on resonance does not maximize $g_{\text{eff}}^{\text{II}}$ either. Optimizing the angle gives

$$\theta^*(f \leq \frac{1}{2}) = \arccos \left[ \frac{\sqrt{3 + 2f \pm \sqrt{9 - 4f + 4f^2}}}{2\sqrt{2}} \right].$$

(S11)

The factor gained with respect to $\theta = \pi/4$ is also $f$-dependent and has the following expression:

$$\frac{g_{\text{eff}}^{\text{II}}(\theta^*)}{g_{\text{eff}}^{\text{II}}(\theta = \pi/4)} = -\frac{(-6f + f' + 3) \sqrt{-2f^2(2f + f' - 2) + f' + 3}}{8\sqrt{2}(2f - 1)}$$

(S12)

where $f' = \sqrt{4(1-f)f + 9}$. For the particular case $f = 1/4$, we find

$$g_{\text{eff}}^{\text{II}}(\theta^*) \approx 1.76 g_{\text{eff}}^{\text{II}}(\theta = \pi/4).$$

(S13)

Alternatively, we can compute the optimal detuning $\Delta_\sigma$ for a fixed $\Omega$ (instead of fixed $R$), which is experimentally more meaningful given that varying $\Delta_\sigma$ for a fixed $\Omega$ is more straightforward than varying $\theta$ for a fixed $R$:

$$\Delta_\sigma^* = -\Omega \frac{(1 - 2f)}{|1 - 2f|} \sqrt{-2 + \frac{1 - \sqrt{1 - f(1 - f)|1 - 2f|}}{f(1 - f)}}.$$

(S14)

For the particular case $f = 1/4$, $\Delta_\sigma^* \approx -0.96 \Omega$, which leads to

$$g_{\text{eff}}^{\text{II}}(\Delta_\sigma^*) \approx 1.52 g_{\text{eff}}^{\text{II}}(\Delta_\sigma = 0).$$

(S15)

**USC effect III: Details for two atoms excited by a single photon**

We consider the following two subspaces, with $N_A = 2$ and $N_B = 12$:

- $\mathcal{A} = \{|+,+,n\},\{-,+,n+1\}$,

- $\mathcal{B} = \{|+,+,n+1\},\{-,+,n+1\},\{|+,+,n+1\},\{-,+,-n\},\{-,+,-n\},\{|+,+,n+2\},\{-,+,-n+2\},\{|+,+,n+2\},\{-,+,-n+2\}.$

Here, the correction that we obtain is

$$\delta h^{\text{III}} = \begin{pmatrix} 2n\chi + 2\lambda + C \\ g_{\text{eff}}^{\text{III}} \sqrt{n + 1} \\ -2(n + 1)\chi - 2\lambda + C \end{pmatrix}.$$
FIG. S2: USC effect III: Energy-level diagrams and transitions for the process of exciting two atoms with a single photon. (a) Energy levels $E_n$ for the Hamiltonian in Eq. (15) as a function of $\Delta_a$. Parameters: $\Omega = 20g$, $\Delta_\sigma = \Delta^*_{\sigma}$ [see Eq. (S21)], with $R = \sqrt{\Omega^2 + \Delta^2_{\sigma}/4}$. The Hilbert space is truncated at 4 photons for simplicity. (b) Zoom-in on the anti-crossing between the energy levels corresponding to $|+\,+,\,0\rangle$ and $|-\,-,\,1\rangle$. The size of the level splitting at the resonance $\Delta_a = \Delta^*_{\sigma} = 4R + \delta$—where both states have the same energy in the absence of coupling $E_0 = 2R$—indicates the strength $g_{\text{III}}$ of the effective interaction between these two states. (c) The transitions in the third-order process that creates the effective coupling between $|-,\,-,n+1\rangle$ and $|+,\,+,n-1\rangle$. Blue solid arrows: transitions that conserve the number of excitations. Red dashed arrows: transitions that change the number of excitations by one. Blue dashed arrows: transitions that change the number of excitations by two.

The dispersive coupling and the Lamb shift are given by second-order processes:

$$ \chi = g^2 \frac{(2R - \Delta_a)c^4 + (2R + \Delta_a)s^4}{4R^2 - \Delta^2_a}, \quad \lambda = \chi/2. $$

(S17) (S18)

Setting $\Delta_a = 4R + \delta$, the optimum resonance condition is given by

$$ \delta = (4n + 2)\chi + 4\lambda = 4(n + 1)^2. $$

(S19)

For a fixed $R$, we can see that the optimal angle $\theta$ maximizing $g_{\text{eff}}$ is $\theta^* = \arctan\left(\sqrt{\frac{1+\sqrt{5}}{3-\sqrt{5}}}\right) \approx 0.356\pi$, giving the following maximum value of $g_{\text{eff}}^{\text{III}}$:

$$ g_{\text{eff}}^{\text{III}}(\theta^*) = \sqrt{\frac{11 + 5\sqrt{5}}{8}} \frac{g^3}{6R^2} \approx 1.67 g_{\text{eff}}^{\text{III}}(\theta = \pi/4). $$

(S20)

That is, when choosing the optimal angle $\theta^*$ we obtain 1.67× enhancement with respect to the resonant case $\Delta_{\sigma} = 0$, which corresponds to $\theta = \pi/4$. In a similar way, we can express this in terms of the optimal detuning:

$$ \Delta_{\sigma}^* = \Omega \sqrt{\frac{14\sqrt{109} - 122}{45}} \approx 0.73\Omega. $$

(S21)

The corresponding value of $g_{\text{eff}}$ is then given by

$$ g_{\text{eff}}^{\text{III}}(\Delta_{\sigma}^*) \approx -1.3 \frac{g^4}{6\Omega^2} = 1.3 g_{\text{eff}}^{\text{III}}(\Delta_{\sigma} = 0) $$

(S22)

giving 1.3× enhancement with respect to the resonant case, for the same driving amplitude $\Omega$.

Validity of the perturbation theory

In this section, we address the question of the validity of the perturbation theory for the three studied USC effects for large values of the perturbation parameter $g/\Omega$. To do so, we study the energy-level splitting $\Delta E_k$ between the two eigenstates $|\phi_k\rangle$. 
FIG. S3: Validity of the applied perturbation theory for the three examples. (a) Examples of the avoided crossings at two different values of the perturbation parameter $g/\Omega$. Smaller values of $\Omega$ imply larger splitting. (b-c) Splitting at the avoided crossing (solid) versus $\Omega/g$, compared to the effective rates computed from perturbation theory (dashed). The perturbation theory works when the two curves overlap. (e-f) The overlap between the two eigenstates at the avoided crossing and the two states involved in the nonlinear process; when the perturbation theory starts failing, the overlap is reduced, meaning that eigenstates contain contributions from other states.

and $|\varphi_{k-1}\rangle$ at the avoided-level crossing that we associate to each nonlinear process, see Fig. S3 (a). The resulting splitting is compared to the effective coupling rates $g_{\text{eff}}^{I}, g_{\text{eff}}^{II}$, and $g_{\text{eff}}^{III}$ that we have computed from perturbation theory, as we show in Fig. S3 (b-c). In addition, we compute the overlap between the two eigenstates $|\varphi_{k/k-1}\rangle$ and the two states between which we expect the Rabi oscillations to occur in each of the three cases considered in the text, see Fig. S3 (e-g). As $\Omega/g$ is reduced, the effective coupling rates increase. The perturbation theory starts failing at $\Omega/g \lesssim 10$ for cases I and II, and $\Omega/g \lesssim 2$ for case III (which is a third-order process). Below these values of $\Omega/g$, $|\varphi_{k,k-1}\rangle$ stop being composed exclusively of the two isolated states that constitute the desired nonlinear process, and the effective coupling rate predicted from perturbation theory departs from the real half-splittings between these eigenstates.

**Effect of decoherence**

Here we provide further study of the effect of decoherence (beyond Table I in the main text) computing the dynamics of the nonlinear processes in the presence of cavity losses. This is done by using the standard Lindblad master equation for the...
FIG. S4: Effect of decoherence induced by radiative decay in the cavity modes for the three nonlinear processes considered in this work. In each case, we monitor the population $n_a = \langle a^\dagger a \rangle$ of a cavity undergoing Rabi oscillations under the nonlinear processes with and without decay. Lossless dynamics are represented by straight, blue curves. Dissipative dynamics are shown in dashed curves, with lighter color representing higher decay rates. Decay rates used are $g_{\text{eff}} \times (0.25, 0.5, 1), \Omega/g = 20$ (a), 20 (b) and 10 (c).

Dynamics of the density matrix:

$$\dot{\rho} = -i[H, \rho] + \frac{\gamma_a}{2} \sum_{i=1}^{N_{\text{cav}}} \left(2a_i \rho a_i - a_i^\dagger a_i \rho - \rho a_i^\dagger a_i \right).$$

(S23)

where $\gamma_a$ is a cavity decay rate, and the sum runs over the total number of cavities (two in cases I and II, one in case III). In Fig. S4 we show that the effect of decoherence is the expected damping of the Rabi oscillations. The apparent higher robustness to losses of case III (two atoms excited by a single photon) can be explained by the fact that there is only one cavity, which is the only lossy subsystem, as compared to two cavities in cases I and II. We have assumed the cavity decay to be the main decoherence mechanism, therefore ignoring the decay or dephasing of the atoms to simplify the discussion.