Supplemental Material for "One photon simultaneously excites two atoms in ultrastrongly coupled light-matter"

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S1. Circuit Hamiltonian

Here we describe the circuit Hamiltonian calculation in detail. The branch fluxes across the circuit elements, which are junctions, the inductance L_r, and the capacitance C_r, follow Kirchhoff's voltage laws:

$$\varphi_{\beta 1} + \varphi_{\alpha 1} + \varphi_{a1} + \varphi_{b1} = \varphi_{\mathrm{e}1}\,, \tag{S1}$$

$$\varphi_{\beta 2} + \varphi_{\alpha 2} + \varphi_{a2} + \varphi_{b2} = \varphi_{\mathrm{e}2}\,, \tag{S2}$$

$$\varphi_{\mathrm{cr}} + \varphi_{\ell\mathrm{r}} + \varphi_{\beta 1} - \varphi_{\beta 2} = 0\,, \tag{S3}$$

where φ_{cr} and $\varphi_{\ell\mathrm{r}}$ represent fluxes between the resonator capacitor and the inductor. The total Lagrangian of the circuit is described as the summation of circuit components as

$$\mathcal{L}_{\mathrm{tot}} = \mathcal{K}_{\mathrm{J}1} + \mathcal{K}_{\mathrm{J}2} - \mathcal{U}_{\mathrm{J}1} - \mathcal{U}_{\mathrm{J}2} + \mathcal{L}_{\mathrm{r}}\,, \tag{S4}$$

where

$$\mathcal{K}_{\mathrm{J}k} = \frac{C_\mathrm{J}}{2}\left[\beta\dot{\phi}_{\beta k}^2 + \dot{\phi}_{ak}^2 + \dot{\phi}_{bk}^2 + \alpha_i(\dot{\phi}_{\beta k} + \dot{\phi}_{ak} + \dot{\phi}_{bk})^2\right], \tag{S5}$$

$$\begin{aligned}\mathcal{U}_{\mathrm{J}k} = -E_\mathrm{J}\,[&\beta_i\cos(\varphi_{\beta k}) + \cos(\varphi_{ak}) + \cos(\varphi_{bk}) \\ &+\alpha_i\cos(\varphi_{\mathrm{e}k} - \varphi_{\beta k} - \varphi_{ak} - \varphi_{bk})]\,,\end{aligned} \tag{S6}$$

$$\mathcal{L}_\mathrm{r} = \frac{C_\mathrm{r}}{2}\dot{\phi}_{\mathrm{cr}}^2 - \frac{1}{2L_\mathrm{r}}(\phi_{\mathrm{cr}} + \phi_{\beta 1} - \phi_{\beta 2})^2\,, \tag{S7}$$

and $k \in \{1, 2\}$ represents the index of a qubit. The sub-index cr refers to the capacitor of the resonator. The qubit kinetic energy part of the Lagrangian in Eq. (S4) becomes

$$\mathcal{K}_{\mathrm{J}i} = \frac{1}{2}\dot{\boldsymbol{\phi}}^\mathrm{T}\mathbf{M}\dot{\boldsymbol{\phi}}\,, \tag{S8}$$

where $\boldsymbol{\phi}_k \equiv \begin{pmatrix}\phi_{\beta k} & \phi_{ak} & \phi_{bk}\end{pmatrix}^\mathrm{T}$ and the mass matrix is given by

$$\mathbf{M}_k = C_\mathrm{J}\begin{pmatrix}\beta_k + \alpha_k & \alpha_k & \alpha_k \\ \alpha_k & 1+\alpha_k & \alpha_k \\ \alpha_k & \alpha_k & 1+\alpha_k\end{pmatrix}. \tag{S9}$$

Using the canonical conjugate $q_i = \partial L_{\mathrm{tot}}/\partial\dot{\phi}_i$ for $\dot{\phi}_i$, where $i \in \{\beta_k, a_k, b_k\}$, we can rewrite Eq. (S8) as

$$\mathcal{K}_{\mathrm{J}k} = \frac{1}{2}\mathbf{q}_k^\mathrm{T}\mathbf{M}^{-1}\mathbf{q}_k\,. \tag{S10}$$

Then, we obtain the total Hamiltonian of the circuit as

$$\begin{aligned}\mathcal{H}_{\mathrm{tot}} &= \sum_k \mathbf{q}_k^{\mathrm{T}}\dot{\boldsymbol{\Phi}}_k - \mathcal{L}_{\mathrm{tot}} \\ &= \sum_k (4E_{\mathrm{c}}\tilde{\mathbf{q}}_k^{\mathrm{T}}\tilde{\mathbf{M}}_k^{-1}\tilde{\mathbf{q}}_k + E_{\mathrm{Lr}}\varphi_{\beta k}^2 + \mathcal{U}_{\mathrm{J}k}) + \mathcal{H}_{\mathrm{r}} + \mathcal{H}_{\mathrm{int}}\,,\end{aligned} \tag{S11}$$

where $2e\tilde{q} = q$ and $C_{\mathrm{J}}\tilde{\mathbf{M}} = \mathbf{M}$, and the term $E_{\mathrm{Lr}}\varphi_{\beta k}$ originates from Eq. (S7) because we define

$$\mathcal{H}_{\mathrm{r}} \equiv \frac{C_{\mathrm{r}}}{2}\dot{\phi}_{\mathrm{cr}}^2 + \frac{1}{2L_{\mathrm{r}}}\phi_{\mathrm{cr}}^2\,, \tag{S12}$$

as a bare LC resonator. We expand the total Hamiltonian Eq. (S11) using the eigenvectors $|i\rangle_k$ ($i \in \mathbb{N}$) of the atom Hamiltonians ($\mathcal{H}_{\mathrm{tot}} - \mathcal{H}_{\mathrm{r}} - \mathcal{H}_{\mathrm{int}}$),

$$\begin{aligned}\hat{\mathcal{H}}_{\mathrm{tot}} =& \hbar\sum_i (\Omega_i^{(1)}\,|i\rangle_1\,\langle i|_1 + \Omega_i^{(2)}\,|i\rangle_2\,\langle i|_2) + \hat{\mathcal{H}}_{\mathrm{r}} \\ &- \hbar\sum_{i,j}(g_{ij}^{(1)}\,|i\rangle_1\,\langle j|_1 - g_{ij}^{(2)}\,|i\rangle_2\,\langle j|_2)(\hat{a}^\dagger + \hat{a}) \\ &- E_{\mathrm{L}}\sum_{i,j} g_{ij}^{(1)}g_{ij}^{(2)}\,|i\rangle_1\,|i\rangle_2\,\langle j|_1\,\langle j|_2\,,\end{aligned} \tag{S13}$$

where $\hbar\Omega_i^{(k)}$ is the i-th eigenenergy of atom k and $\hbar g_{ij}^{(k)} = I_{\mathrm{zpf}}\Phi_0\,\langle i|\hat{\varphi}_{\beta k}|j\rangle$ is the coupling matrix element ($I_{\mathrm{zpf}} = \sqrt{\hbar\omega_{\mathrm{r}}/2L_{\mathrm{r}}}$). When we use the two lowest eigenstates for each qubit, we can obtain the Hamiltonian with the two-level system qubits and the N-level system resonator as shown in Eqs. (5) and (6) in the main text. In Eqs. (5) and (6) in the main text, the spin-spin interaction reduces the current flowing in the resonator loop, which in this system is the ferromagnetic coupling.

For the fitting, we use Eqs. (6) in the main text, and 11 fitting parameters, this includes the offset value when $\varepsilon_1 = 0$ and the persistent current I_{p1} in qubit 1 to derive $\varepsilon_1 = I_{\mathrm{p1}}\Phi_0(\varphi_{\mathrm{e1}} - 0.5)$, where Φ_0 is the flux quantum.

S2. Asymmetric spectrum in the generalized Dicke Hamiltonian

Consider the system Hamiltonian in the main text [Eq. (6)] with $\Delta_k = 0$, $g_1 = g_2 = g$, and substituting σ_{zk} with its eigenvalue $m_k = \pm 1$, we can write:

$$\mathcal{H}_s/\hbar = \frac{|\varepsilon_1|}{2} m_1 + \frac{|\varepsilon_2|}{2} m_2 + \omega_{\mathrm{r}} a^\dagger a + gM(a^\dagger + a)\,, \tag{S14}$$

with $M = \mathrm{sgn}(\varepsilon_2) m_2 - \mathrm{sgn}(\varepsilon_1) m_1$. Performing the substitution $\hat{a} = \hat{b} - Mg/\omega_{\mathrm{r}}$ in Eq. S19,

$$\mathcal{H}_s/\hbar = \frac{|\varepsilon_1|}{2} m_1 + \frac{|\varepsilon_2|}{2} m_2 + \omega_{\mathrm{r}}(\hat{b}^\dagger - M\frac{g}{\omega_{\mathrm{r}}})(\hat{b} - M\frac{g}{\omega_{\mathrm{r}}}) + gM(\hat{b}^\dagger - M\frac{g}{\omega_{\mathrm{r}}} + \hat{b} - M\frac{g}{\omega_{\mathrm{r}}})\,, \tag{S15}$$

$$= \omega_{\mathrm{r}} b^\dagger b + \frac{|\varepsilon_1|}{2} m_1 + \frac{|\varepsilon_2|}{2} m_2 - M^2 \frac{g^2}{\omega_{\mathrm{r}}}\,, \tag{S16}$$

we obtain the Hamiltonian of a harmonic oscillator. By applying the annihilation operator $\hat{b}$ to its ground state $|0\rangle_M$ (i.e. $\hat{b}\,|0\rangle_M = 0$), we thus obtain

$$\hat{a}\,|0\rangle_M = -M\frac{g}{\omega_{\mathrm{r}}}\,|0\rangle_M\ . \tag{S17}$$

From Eq. S17, we see that atomic states with $M = 0$ are associated with the zero-photon state; while atomic states with $M = \pm 2$ are associated with the photonic coherent states $|\pm\alpha\rangle$. In turn, M depends on the sign of ε_1 and ε_2. If $\varepsilon_1 < 0$ and $\varepsilon_2 < 0$, then $M_- \equiv M = m_1 - m_2$. If $\varepsilon_1 > 0$ and $\varepsilon_2 < 0$, then $M_+ \equiv M = -(m_1 + m_2)$. Table SI shows the eight possible states as a function of the sign of ε_1 when the interaction is longitudinal. This explains the asymmetry of the spectra respect the sign of ε_1.

TABLE SI. Possible values of M and relative states. We consider $\varepsilon_2 < 0$.

m_1	-1	+1	-1	+1
m_2	-1	-1	+1	+1
M_-	0	+2	-2	0
States if $\varepsilon_1 < 0$	$\lvert gg\rangle\,\lvert 0\rangle$	$\lvert eg\rangle\,\lvert -\alpha\rangle$	$\lvert ge\rangle\,\lvert +\alpha\rangle$	$\lvert ee\rangle\,\lvert 0\rangle$
M_+	+2	0	0	-2
States if $\varepsilon_1 > 0$	$\lvert gg\rangle\,\lvert +\alpha\rangle$	$\lvert eg\rangle\,\lvert 0\rangle$	$\lvert ge\rangle\,\lvert 0\rangle$	$\lvert ee\rangle\,\lvert -\alpha\rangle$

S3. Coupling ratio dependence of one-photon two-atom excitation effect

Figure S1 shows the numerically calculated transition frequency ω_{i0} (spectrum) for the coupling ratios $g_{1,2}/\omega_{\mathrm{r}} = 0$, 0.65 (fitted value), and 1. We use the same Hamiltonian in the main text,

$$\begin{aligned}\hat{\mathcal{H}}_{\mathrm{tot}}/\hbar \simeq\ & \omega_{\mathrm{r}}\hat{a}^{\dagger}\hat{a} + \frac{\varepsilon_1}{2}\hat{\sigma}_{z1} + \frac{\Delta_1}{2}\hat{\sigma}_{x1} + \frac{\varepsilon_2}{2}\hat{\sigma}_{z2} + \frac{\Delta_2}{2}\hat{\sigma}_{x2} \\ & - (g_1\hat{\sigma}_{z1} - g_2\hat{\sigma}_{z2})(\hat{a}^{\dagger} + \hat{a}) - \frac{2g_1g_2}{\omega_{\mathrm{r}}}\hat{\sigma}_{z1}\hat{\sigma}_{z2}\,, \end{aligned} \tag{S18}$$

and fitting parameters. We can see that the asymmetry of the spectrum increases with $g_{1,2}/\omega_{\mathrm{r}}$, and the antisplitting gap between ω_{30} and ω_{40} in $g_{1,2}/\omega_{\mathrm{r}} \simeq 0.65$ $(-1 < \varepsilon_1/\omega_{\mathrm{r}} < 0)$ is larger than that of the other two coupling ratios.

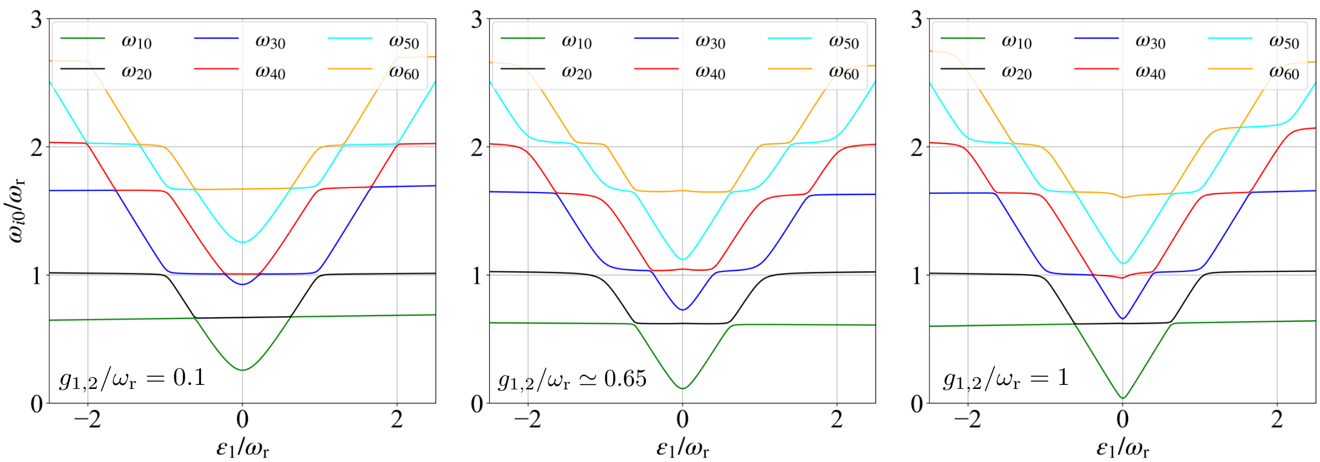

FIG. S1. Frequency difference ω_{i0} for the lowest-energy eigenstates of Hamiltonian Eq. (S18) using different coupling ratio $g_{1,2}/\omega_{\mathrm{r}} \simeq 0$, 0.65 (fitted value), and 1. Other parameters for Eq. (S18) are obtained from fitting the spectrum in the main text. The tilt in the horizontal axis originates from the fitting parameter A (shown in the main text) caused mainly by crosstalk.

We numerically calculate the projection $P_j \equiv |\langle\Psi_j|j\rangle|^2$ of the superposition states $|\Psi_{gg1}\rangle \simeq (|\psi_3\rangle - |\psi_4\rangle)/\sqrt{2}\,(\simeq |gg1\rangle)$ and $|\Psi_{ee0}\rangle \simeq (|\psi_3\rangle + |\psi_4\rangle)/\sqrt{2}\,(\simeq |ee0\rangle)$ at the anticrossing point on the bare states $|j\rangle = \{|gg1\rangle\,, |ee0\rangle\}$ as a function of the coupling ratio, and the result is shown in Fig. S2(a). As mentioned in the theoretical prediction in Ref. 1, a lower g/ω_{r} maximizes the projection. However, the effective coupling strength below $g/\omega_{\mathrm{r}} = 0.1$ is much smaller than that at larger coupling ratios, see Fig. S2(b). Thus, when g/ω_{r} is below 0.1, we cannot clearly see the antisplitting between $|gg1\rangle$ and $|ee0\rangle$ and the OPETA effect. As shown in the right panel of Fig. S2, the effective coupling is maximum at around $g_{1,2}/\omega_{\mathrm{r}} \simeq 0.66$, which is close to our system.

According to the theoretical prediction in Ref. 1, when the Hamiltonian has no direct spin-spin interaction, which

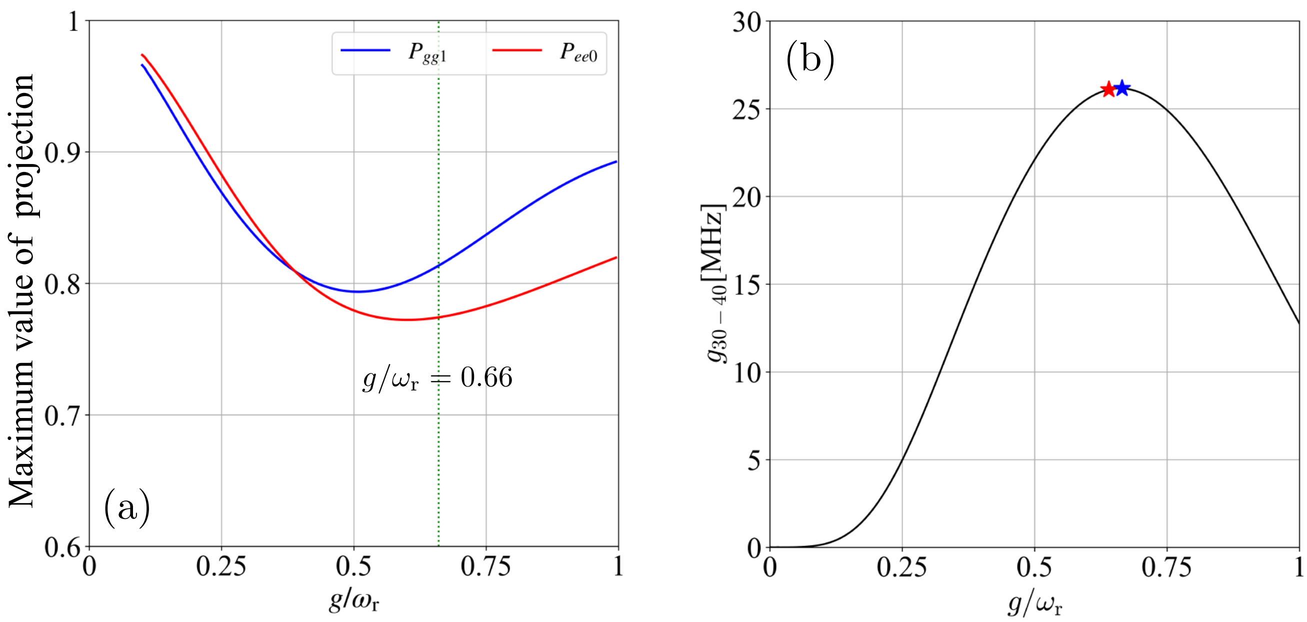

FIG. S2. The left panel shows the maximum value of the projections $P_{gg1}(g/\omega_{\rm r})$ and $P_{ee0}(g/\omega_{\rm r})$ at the antisplitting point. The green dotted vertical line corresponds to the value that maximizes g_{30-40}. the right panel shows the effective coupling constant g_{30-40} plotted against the coupling ratio $g/\omega_{\rm r}$. The red and blue star represent $g_1/\omega_{\rm r} = 0.64$ and $g_2/\omega_{\rm r} = 0.67$, respectively.

is written as

$$\mathcal{H}_{\rm ideal}/\hbar = \frac{\omega_{\rm q}}{2} \sum_{i=1,2} \hat{\sigma}_{zi} + \omega_{\rm r} a^\dagger a + g \sum_{i=1,2} (\hat{\sigma}_{zi} \cos\theta + \hat{\sigma}_{xi} \sin\theta)(\hat{a}^\dagger + \hat{a})\,, \tag{S19}$$

the effective coupling strength Ω between $|gg1\rangle$ and $|ee0\rangle$ can be approximate to

$$\Omega \simeq \frac{8}{3} \frac{g^3}{\omega_{\rm q}^2} \sin\theta \cos\theta^2\,, \tag{S20}$$

where $\omega_{\rm q}$ is the qubit frequency, and $\theta = \arctan(-\Delta/\varepsilon)$. The parameters in our system are $\theta_1 \simeq 0.09 \times 2\pi$, $\theta_2 \simeq 0.06 \times 2\pi$, $\omega_{\rm q1} \simeq 3.42$ GHz, and $\omega_{\rm q2} \simeq 2.48$ GHz. From Eq. (S20), the effective coupling constant Ω obtained using the parameters in our system is expected to be more than 200 MHz. The measured effective coupling constant g_{30-40} is much suppressed. Besides, if the Hamiltonian has no direct spin-spin interaction, the projections on $|gg1\rangle$ and $|ee0\rangle$ at the anticrossing point is less than 0.8 when $g/\omega_{\rm r} = 0.25$, and when $g/\omega_{\rm r} \gg 0.25$, the OPETA effect is no longer observed.

[1] L. Garziano, V. Macrì, R. Stassi, O. Di Stefano, F. Nori, and S. Savasta, Physical Review Letters **117**, 043601 (2016).