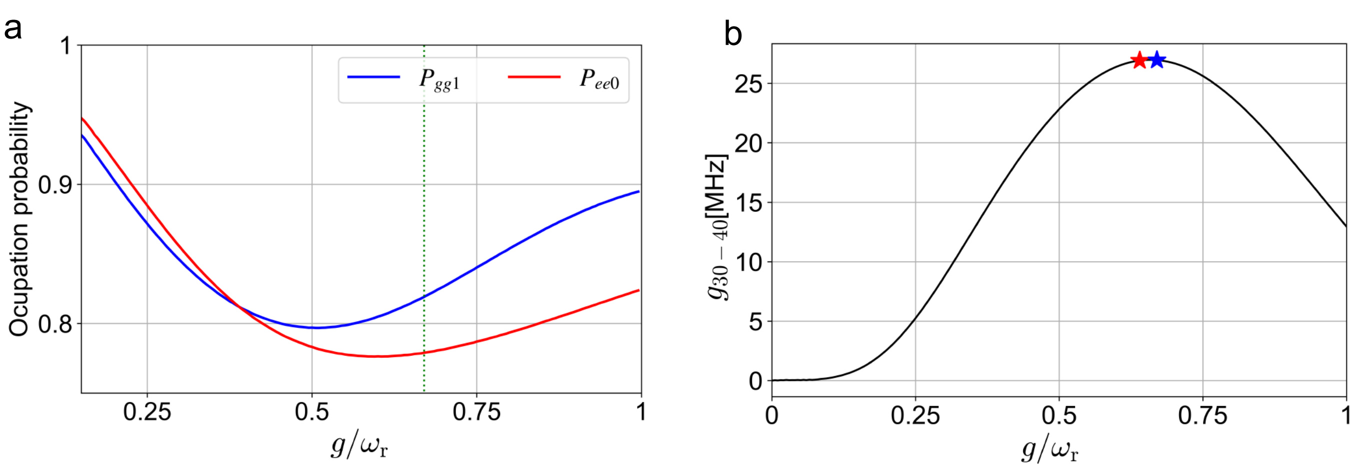
Supplemental Material for "One photon simultaneously excites two atoms in ultrastrongly coupled light-matter"

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

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 Supplementary Fig. 1(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 Supplementary Fig. 1(b). Thus, when g/ω_r is below 0.1, we cannot clearly see the antisplitting between $|gg1\rangle$ and $|ee0\rangle$ and the "one–photon–exciting–two–atoms" effect. As shown in the right panel of Supplementary Fig. 1, the effective coupling is maximum at around $g_{1,2}/\omega_\mathrm{r} \simeq 0.7$, which is close to our system.

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



Supplementary Fig. 1. The left panel shows the maximum value of the projections $P_{gg1}(g/\omega_\mathrm{r})$ and $P_{ee0}(g/\omega_\mathrm{r})$ 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/ω_r. The red and blue star represent $g_1/\omega_\mathrm{r} = 0.64$ and $g_2/\omega_\mathrm{r} = 0.67$, respectively.

is written as

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

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

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

where ω_{q} is the qubit frequency, and $\theta = \arctan(-\Delta/\varepsilon)$. The parameters in our system are $\theta_1 \simeq 0.09 \times 2\pi$, $\theta_2 \simeq 0.06 \times 2\pi$, $\omega_{\text{q1}} \simeq 3.42$ GHz, and $\omega_{\text{q2}} \simeq 2.48$ GHz. From Eq. (2), 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 at $g/\omega_{\text{r}} = 0.25$, and when $g/\omega_{\text{r}} \gg 0.25$, the "one–photon–exciting–two–atoms" 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).