Detecting the symmetry breaking of the quantum vacuum in a light–matter coupled system

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Hybrid quantum systems in the ultrastrong, and even more in the deep-strong, coupling regimes can exhibit exotic physical phenomena and promise new applications in quantum technologies. In these nonperturbative regimes, a qubit–resonator system has an entangled quantum vacuum with a nonzero average photon number in the resonator, where the photons are virtual and cannot be directly detected. The vacuum field, however, is able to induce the symmetry breaking of a dispersively coupled probe qubit. We experimentally observe this Higgs-like quantum-vacuum symmetry breaking induced by the field of a lumped-element superconducting resonator deep-strongly coupled with a flux qubit. This result opens a way to experimentally explore the novel quantum-vacuum effects emerging in the deep-strong coupling regime.

Superconducting quantum circuits based on Josephson junctions (JJs) [1–8] have developed rapidly in recent years and demonstrated quantum advantage, over classical counterparts, in information processing [9, 10]. Now they are considered to be one of the most promising experimentally-realizable systems for quantum computing [11–13]. Also, the experimental advancements in superconducting qubit–resonator systems have stimulated theoretical and experimental researches on quantum optics in the microwave regime [14, 15]. As a solid-state version of cavity quantum electrodynamics (QED) [16, 17], circuit QED [18–20] has greater flexibility and tunability, and it can achieve ultrastrong and even deep-strong light–matter couplings to individual qubits [21–27], owing to the large dipole moment of the superconducting qubit (i.e., artificial atom) and the small mode volume of the resonator. When the qubit–resonator coupling approaches the nonperturbative ultrastrong regime, novel quantum-optics phenomena occur [28–33], including puzzling modifications of the quantum vacuum of the system [34–39].

In the nonperturbative ultrastrong-coupling regime, the qubit–resonator system can be described by a quantum Rabi model. It is particularly interesting to harness controllable physical parameters to tune the quantum vacuum of the system, since it becomes a novel entangled ground state $|G\rangle$ rather than the trivial product ground state of the Jaynes-Cummings model. In such an exotic quantum vacuum, while the average photon number in the resonator is nonzero, i.e., $\langle G| a^\dagger a | G \rangle \neq 0$, where $a^\dagger (a)$ is the creation (annihilation) operator of the resonator mode, the ground-state photons are actually virtual (tightly bound to the artificial atom [40]) and cannot be directly detected. Theoretically, it was proposed to employ non-adiabatic modulations [35], sudden turn-off of qubit–resonator interaction [36] or a spontaneous decay mechanism of multi-level systems [37] to convert these virtual photons into real ones (similar to the dynamical Casimir effect [41, 42]), so as to generate radiation out of the resonator. However, these are still experimentally challenging.

In the standard model of particle physics, the $W^\pm$ and $Z$ weak gauge bosons obtain mass via the Higgs mechanism, in which the electroweak gauge symmetry $\text{SU}(2) \times \text{U}(1)$ is broken due to the interaction with a symmetry-broken vacuum field (the Higgs field) displaying a nonzero vacuum expectation value. In our experiment, we observe the parity symmetry breaking of a
probe superconducting circuit (Xmon) dispersively coupled to a qubit–resonator system in the deep-strong coupling regime. This effect is very similar to the Higgs mechanism, although in our case the broken symmetry is discrete. At the optimal point, both the flux qubit and the qubit–resonator system have a well-defined parity symmetry [43]. In this parity-symmetry case, the quantum-vacuum expectation value of the resonator field is zero, \( \langle G|(a + a^\dagger)|G \rangle = 0 \), where \( G \) is the qubit–resonator ground state. With the external flux tuned away from the optimal point, parity-symmetry breaking is induced for both the flux qubit and the qubit–resonator system, giving rise to \( \langle G|(a + a^\dagger)|G \rangle \neq 0 \) [39]. In our experiment, the achieved qubit–resonator system is in the deep-strong coupling regime, so the quantum-vacuum state is very different and, when away from the optimal point, this can produce a sizable nonzero value of \( \langle G|(a + a^\dagger)|G \rangle \) as well as observable symmetry breaking effects. Indeed, as demonstrated in our experiment, the qubit–resonator system is able to break the parity selection rule of the dispersively coupled Xmon, thus enabling forbidden transitions.

**Deep-strongly coupled qubit–resonator circuit.**—The system is composed of a five-junction flux qubit deep-strongly coupled to a superconducting lumped-element resonator via a common Josephson junction (JJ) (Fig. 1). In addition, we use an Xmon as a quantum detector, which capacitively coupled to the lumped-element resonator on the left and to a coplanar-waveguide resonator on the right. The whole device is placed in a dilution refrigerator cooled down to a temperature of \( \sim 30 \) mK.

Similar to the three-junction flux qubit [2], the five-junction flux qubit has both clockwise and counterclockwise persistent-current states. Away from the optimal point \( \Phi_{\text{ext}} = (n + \frac{1}{2})\Phi_0 \), where \( \Phi_{\text{ext}} \) is the external flux threading the loop of the flux qubit, \( \Phi_0 = h/2e \) is the superconducting flux quantum, and \( n \) is an integer, these two persistent-current states have an energy difference \( \varepsilon = 2I_p\delta\Phi_{\text{ext}} \), depending on the maximum persistent current \( I_p \) and the flux bias \( \delta\Phi_{\text{ext}} \equiv \Phi_{\text{ext}} - (n + \frac{1}{2})\Phi_0 \). Also, there is a barrier between these two persistent-current states, which removes their degeneracy at the optimal point by opening an energy gap \( \Delta \). In the basis of eigenstates, the Hamiltonian of the flux qubit can be written as (setting \( \hbar = 1 \)) \( H_q = \omega_q\sigma_z/2 \), where \( \omega_q = \sqrt{\Delta^2 + \varepsilon^2} \) is the transition frequency of the qubit and \( \sigma_z \) is a Pauli operator. The quantum two-level system is a good model for the flux qubit because of its relatively large anharmonicity.

Compared to the coplanar-waveguide resonator, the lumped-element resonator has the advantage of only a single resonator mode [24]: \( H_r = \omega_r a^\dagger a \), where \( \omega_r \) is the resonance frequency of the resonator mode. This \( \omega_r \) is V-shaped versus \( \delta\Phi_{\text{ext}} \) around the optimal point [24] because the inductance across the qubit loop, as part of the total inductance of the lumped-element resonator, depends approximately linearly on \( |\delta\Phi_{\text{ext}}| \). The large JJ shared by the flux qubit and the lumped-element resonator acts as an effective inductance to produce an interaction between them, \( H_{\text{int}} = g[\cos\theta\sigma_z - \sin\theta\sigma_x](a^\dagger + a) \), where \( \tan\theta = \Delta/\varepsilon \), and \( g = MI_pI_r \) is the coupling strength, with \( M \) being the mutual inductance and \( I_r \) the vacuum fluctuation current along the center conductor of the lumped-element resonator. When the qubit–resonator coupling is in the ultrastrong or deep-strong regime, one cannot apply the rotating-wave approximation (RWA) to \( H_{\text{int}} \), and the Hamiltonian of the qubit–resonator system is written as

\[
H_s = \frac{1}{2}\omega_p\sigma_z + \omega_r a^\dagger a + g[\cos\theta\sigma_z - \sin\theta\sigma_x](a^\dagger + a),
\]

i.e., the generalized quantum Rabi model [15].

We can extract the parameters in \( H_s \) by fitting the reflection spectra of the qubit–resonator system, as measured by applying a probe tone to the system. Around \( \omega_p/2\pi = 4.8 \) GHz (near the bare frequency of the lumped-element resonator) and 5.6 GHz, clear transitions are observed, of which the corresponding frequencies are found to be consistent with the transition frequencies from the ground state \( |G \rangle \equiv |0\rangle_s \) to the first- and second-excited states, \( |1\rangle_s \) and \( |2\rangle_s \) of the qubit–resonator system, respectively, i.e., \( \omega_{01} \) and \( \omega_{02} \) (see the solid fitting curves in Figs. 2c and 2b). Around \( \omega_p/2\pi = 11.9 \) GHz, we observe the transition from the ground state \( |G \rangle \) to the
FIG. 2. | Reflection spectra. Reflection spectra of the deep-strongly coupled qubit–resonator system versus the external flux bias \( \Phi_{\text{ext}} \) and the probe frequency \( \omega_p \) around \( \Phi_{\text{ext}} = (3 + \frac{1}{2})\Phi_0 \) (which is a more stable flux bias point than \( \Phi_{\text{ext}} = \frac{1}{2}\Phi_0 \) in our system). The solid blue curves in a–c are the fitted transition frequencies between the ground state to the third-, second- and first-excited states of the qubit–resonator system, respectively (i.e., \( \omega_{01}, \omega_{02}, \) and \( \omega_{03} \)). In a, the additional transitions indicated by the dashed red curves correspond to sideband transitions (assisted by the Xmon levels) in the system.

By fitting the transition frequencies \( \omega_{01}, \omega_{02} \) and \( \omega_{03} \) with experimental results in Fig. 2, we can derive the parameters of the generalized Rabi model in Eq. (1), which are \( I_p = 245 \) nA and \( \Delta/2\pi = 15.0 \) GHz for the flux qubit, \( \omega_f/2\pi = 4.82 \) GHz for the lumped-element resonator, and \( g/2\pi = 4.55 \) GHz for the qubit–resonator coupling. Here the obtained resonance frequency \( \omega_f/2\pi = 4.82 \) GHz is the value when the external flux bias is at the optimal point \( \delta \Phi_{\text{ext}} = 0 \). In our qubit–resonator system, we achieve \( g/\omega_f \approx 0.944 \), indicating that it indeed reaches the deep-strong coupling regime \( g/\omega_f \approx 1 \).

When \( \delta \Phi_{\text{ext}} = 0 \) (\( \theta = \pi/2 \)), the deep-strongly coupled system in Eq. (1) reduces to the standard quantum Rabi model. Instead of a trivial (product) ground state \( |g, 0\rangle \) in the Jaynes-Cummings model, it has a quantum vacuum (i.e., entangled ground state) \( |G\rangle \), with \( \langle G|a^\dagger a|G\rangle \neq 0 \). This standard Rabi model has a well-defined parity symmetry, characterized by \( \sigma_z e^{i\pi a^\dagger a} \), which ensures that the ground state is a superposition of all states with an even number of excitations [28]. For this quantum vacuum, \( \langle G|a^\dagger a|G\rangle = 0 \). When \( \delta \Phi_{\text{ext}} \neq 0 \), \( H_s \) in Eq. (1) has an extra longitudinal coupling term proportional to \( \sigma_z \). It breaks the parity symmetry of the model, and hence both even and odd number of excitations are all allowed in the new ground state [39]. Now, both \( \langle G|a^\dagger a|G\rangle \neq 0 \) and \( \langle G|(a + a^\dagger)|G\rangle \neq 0 \).

Detection of the induced symmetry breaking.—Below we harness an Xmon [7] to detect this quantum vacuum, which is both largely detuned with the lumped-element resonator and weakly coupled to it via a small capacitor (cf. Fig. 1a). In such a dispersive regime, the effect of the Xmon on the qubit–resonator system is greatly reduced. The Xmon can be modeled by the Hamiltonian \( H_X = 4E_c n^2 - E_J \cos \varphi \), where \( E_c \) is the single-electron charging energy of the JJ, \( E_J \) is the Josephson coupling energy, \( n = -i\partial/\partial \varphi \), and \( \varphi \) is the phase drop across the JJ. In the Xmon, the metallic cross and the ground metal provides the JJ with a large shunt capacitor to reduce its sensitivity to the charge noise [5, 6].

The Xmon’s parameters can be determined with the dispersive readout technique by coupling the Xmon to a coplanar-waveguide resonator (see Fig. 1c and Fig. S1 in Supplementary Information). The resonance frequency of this waveguide resonator is measured to be \( \omega_{\text{CPW}}/2\pi = 3.554 \) GHz and the coupling strength between the waveguide resonator and the Xmon qubit is \( g_X/2\pi = 28 \) MHz. Then, we obtain the transition frequency \( \omega_X/2\pi = 5.181 \) GHz of the Xmon qubit and its anharmonicity \( A/2\pi = -0.16 \) GHz. With these parameters as well as the relations \( \omega_X = \sqrt{8E_cE_J/E_c} \), \( A = -E_c \), we have \( E_c/2\pi = 0.16 \) GHz and \( E_J/2\pi = 20.97 \) GHz. Because the lumped-element resonator couples to the Xmon, it induces an offset charge to the Josephson junction, leading \( H_X \) to \( H_X = 4E_c(n-n_R)^2- \).
$E_1 \cos \varphi$, where $n_R = i g' (a - a^\dagger)$, and $g' \approx g_X$ (by a symmetric design) is the coupling strength between the lumped-element resonator and the Xmon qubit.

The total Hamiltonian of the deep-strongly coupled qubit–resonator system plus the Xmon can be expressed as $H_{\text{tot}} = H_s + H_{X}$. Owing to the large transition frequency between the ground and first-excited states, the deep-strongly coupled qubit–resonator system nearly stays in the ground state $|G\rangle$ at the temperature $\sim 30$ mK.

Note that in the dispersive regime, where the Xmon–resonator coupling rate is much lower than the corresponding detuning, the energy transitions of the Xmon are almost unaffected by the interaction and can be easily identified with standard spectroscopic techniques. We also observe that, neglecting the interaction between the resonator and the flux qubit, or considering a flux qubit at the optimal point, the Xmon–resonator system displays parity symmetry. On the contrary, when the qubit is brought out of the optimal point, the very strong qubit–resonator coupling strength can induce a symmetry breaking of the Xmon, even for moderate Xmon–resonator coupling strengths (see Supplementary Information).

In Figs. 3a and 3b, we show the single- and two-photon transitions between the lowest two levels of the Xmon using two-tone spectroscopy. Here the resonance frequency of the single-photon transition corresponds to the transition frequency $\omega_X$ of the Xmon qubit, and the resonance frequency of the two-photon transition is $\frac{1}{2} \omega_X$. In our chip, $\frac{1}{2} \omega_X$ is designed to be well separated from both $\omega_{\Phi}$ and $\frac{1}{2} \omega_{\Phi2}$ of the deep-strongly coupled qubit–resonator system to avoid any unwanted transitions. The drive power ($-65$ dBm) applied at the local drive port for exciting the two-photon transition is much stronger than that for the single-photon transition ($-120$ dBm). In Fig. 3b, the signal of the two-photon transition is found to disappear at the optimal point $\delta \Phi_{\text{ext}} = 0$, evidencing that the well-defined parity symmetry of the standard Rabi model preserves the parity selection rule of the Xmon. When deviating from the optimal point, the parity-symmetry breaking in $H_s$, in addition to producing a nonzero vacuum expectation value $v = \langle G|(a + a^\dagger)|G\rangle \neq 0$, is able to break the parity symmetry of the Xmon artificial atom.

These results can be described by adopting a simplified model for the Xmon using only its four lowest energy levels. Considering the large detunings ($\gtrsim 20$ GHz), the effects from higher levels are negligible. With the Xmon now approximated as a four-level system (qudit), the total Hamiltonian of the qubit–resonator system plus the Xmon can be written as

$$H_{\text{tot}} = H_s + H_X^{(4)} - g' (a - a^\dagger) (b - b^\dagger),$$

where $b = \sum_{n=0}^{3} \sqrt{n + 1} |n\rangle \langle n + 1|$ is the annihilation operator for the Xmon, and $H_X^{(4)} = \sum_{n=0}^{3} \epsilon_n |n\rangle \langle n|$ is the bare Xmon energy ($H_X$), projected into the reduced 4-dimensional Hilbert space. We can evaluate the single- and two-photon absorptions under coherent drive of the Xmon by studying its effective polarization $\langle P(\omega_d) \rangle = \text{Tr}[\rho (\omega_d)]$, with $\rho$ being the density operator of the system. The latter can be calculated using the master equation approach in the dressed picture [46] (see Supplementary Information). The simulated results are shown in Figs. 3c and 3d, which are in a good agreement with the experimental observations.

![FIG. 3. Excitation spectra.](image) Excitation spectra of the Xmon qubit versus the external flux bias $\delta \Phi_{\text{ext}}$ and the drive frequency $\omega_d$ (a) $\Phi_{\text{ext}} = (3 + \frac{1}{2})\Phi_0$. The frequency of the probe tone is fixed at 3.554 GHz, in resonance with the $\lambda/2$ mode of the coplanar-waveguide resonator. (b) $\Phi_{\text{ext}} = 3\Phi_0$. The frequency of the probe tone is fixed at 3.554 GHz, in resonance with the $\lambda/2$ mode of the coplanar-waveguide resonator. (c) $\Phi_{\text{ext}} = (3 + \frac{1}{2})\Phi_0$. The frequency of the probe tone is fixed at 3.554 GHz, in resonance with the $\lambda/2$ mode of the coplanar-waveguide resonator. (d) $\Phi_{\text{ext}} = 3\Phi_0$. The frequency of the probe tone is fixed at 3.554 GHz, in resonance with the $\lambda/2$ mode of the coplanar-waveguide resonator.

We observe that the interaction-induced symmetry breaking mechanism detected here is more complex with
respect to the Higgs mechanism and to that described in Ref. 39. In these two cases, the effect is directly induced by the vacuum expectation value of the field. For example, for \( w = \langle G \rangle (a - a^\dagger) \langle G \rangle \neq 0 \), the Xmon–resonator interaction in Eq. (2) could be approximated as \(~= -g_w (b - b^\dagger)\). It can be shown that this term directly determines the symmetry breaking of the Xmon in Ref. 39. However, in the present case, it turns out that \( w = 0 \), since the inductive coupling between the resonator and the flux qubit determines \( w = 0 \) and \( v \neq 0 \). Nonetheless, a full quantum analysis (see Supplementary Information) shows that in such a case \( (w = 0) \) as well, the Xmon can undergo symmetry breaking, when interacting with a vacuum field displaying symmetry breaking. Specifically, the symmetry-breaking of the qubit–resonator system determines a nonzero matrix element entering the two-photon transition rate, thus enabling the two-photon transitions in the Xmon.

Considering the eigenstates of the total Hamiltonian \( H_{\text{tot}} \), the two-photon transition rate is proportional to the product \( |Y_{0,1,2} \rangle \), where \( Y_{i,j} = \langle E_i | - i (b - b^\dagger)|E_j \rangle \), with \( |E_j \rangle \) eigenvectors of \( H_{\text{tot}} \) sorted from the lower to the higher corresponding energy levels. Thus, with \( |E_0 \rangle \) being the ground state of the whole interacting system, we identify \( |E_1 \rangle \) as the first excited level of \( H_{\text{tot}} \) (slightly dressed by the interaction with the Xmon) and \( |E_2 \rangle \) the corresponding first excited dressed level of \( H^{(4)}_{\text{tot}} \). It turns out that \( Y_{0,1} \) as well as \( Y_{0,2} \) are nonzero and almost constant in the interval of flux offset here reported, while \( Y_{1,2} \) is very well approximated by a linear function of \( \delta \Phi_{\text{ext}} \) (see Fig. S4 in Supplementary Information) and it is zero for \( \delta \Phi_{\text{ext}} = 0 \), due to the parity symmetry. This explains the onset of the parity-symmetry breaking felt by the Xmon.

In conclusion, we have experimentally demonstrated a Higgs-like quantum-vacuum-induced symmetry breaking of a probe artificial atom (Xmon) coupled to a resonator field with no real excitation, but with a nonzero vacuum expectation value. The violation of the Xmon parity selection rule comes from virtual paths enabled by its interaction with an electromagnetic resonator whose parity symmetry is significantly broken by the deep-strong light–matter interaction with a flux qubit. The experimental results are in very good agreement with our theoretical analysis. The proposed setting offers a novel way to explore quantum-vacuum effects emerging in the light–matter ultra-strong and deep-strong coupling regimes and can be used as a tool to explore the coherence properties of quantum vacua in these exotic hybrid quantum systems [47–49], and the occurrence of super-radiant phase transitions in Dicke-like systems [34].

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[1] Y. Nakamura, Y. A. Pashkin, and J. S. Tsai, Coherent control of macroscopic quantum states in a single-Cooper-pair box, Nature 398, 786 (1999).
[2] J. E. Mooij, T. P. Orlando, L. Levitov, L. Tian, C. H. van der Wal, and S. Lloyd, Josephson persistent-current qubit, Science 285, 1036 (1999).
[3] Y. Yu, S. Han, X. Chu, S. I. Chu, and Z. Wang, Coherent temporal oscillations of macroscopic quantum states in a Josephson junction, Science 296, 889 (2002).
[4] J. M. Martinis, S. Nam, J. Aumentado, and C. Urbina, Rabi Oscillations in a Large Josephson-Junction Qubit, Phys. Rev. Lett. 89, 117901 (2002).
[5] J. Q. You, X. Hu, S. Ashhab, and F. Nori, Low-coherence flux qubit, Phys. Rev. B 75, 140515(R) (2007).
[6] J. Koch, T. M. Yu, J. Gambetta, A. A. Houck, D. I. Schuster, J. Majer, A. Blais, M. H. Devoret, S. M. Girvin, and R. J. Schoelkopf, Charge-insensitive qubit design derived from the Cooper pair box, Phys. Rev. A 76, 042319 (2007).
[7] R. Barends, J. Kelly, A. Megrant, D. Sank, E. Jeffrey, Y. Chen, Y. Yin, B. Chiaro, J. Mutus, C. Neill, P. O’Malley, P. Roushan, J. Wenner, T. C. White, A. N. Cleland, and J. M. Martinis, Coherent Josephson Qubit Suitable for Scalable Quantum Integrated Circuits, Phys. Rev. Lett. 111, 080502 (2013).
[8] F. Yan, S. Gustavsson, A. Kannal, J. Birenbaum, A. P. Sears, D. Hover, T. J. Gudmundsen, D. Rosenberg, G. Samach, S. Weber, J. L. Yoder, T. P. Orlando, J. Clarke, A. J. Kerman, and W. D. Oliver, The flux qubit revisited to enhance coherence and reproducibility, Nat. Commun. 7, 12964 (2016).
[9] F. Arute et al., Quantum supremacy using a programmable superconducting processor, Nature 574, 505
(2019).
[10] Yulin Wu et al., Strong Quantum Computational Advantage Using a Superconducting Quantum Processor, Phys. Rev. Lett. 127, 180501 (2021).
[11] G. Wendin, Quantum information processing with superconducting circuits: A review, Rep. Prog. Phys. 80, 106001 (2017).
[12] M. Kjaergaard, Mollie E. Schwartz, J. Braumüller, P. Krantz, J. I.-J. Wang, S. Gustavsson, and W. D. Oliver, Superconducting Qubits: Current State of Play, Annu. Rev. Condens. Matter Phys. 11, 369 (2020).
[13] S. Kwon, A. Tomonaga, G. L. Bhai, S. J. Devitt, and J. S. Tsai, Gate-based superconducting quantum computing, J. Appl. Phys. 129, 041102 (2021).
[14] J. Q. You and F. Nori, Atomic physics and quantum optics using superconducting circuits, Nature 474, 589 (2011).
[15] X. Gu, A. F. Kockum, A. Miranowicz, Y.-X. Liu, and F. Nori, Microwave photonics with superconducting quantum circuits, Phys. Rep. 718, 1 (2017).
[16] H. Mabuchi, and A.C. Doherty, Cavity quantum electrodynamics: coherence in context, Science 298, 1372 (2002).
[17] J.-M. Raimond, M. Brune, and S. Haroche, Manipulating quantum entanglement with atoms and photons in a cavity, Rev. Mod. Phys. 73, 565 (2001).
[18] I. Chiocrescu, P. Bertet, K. Semba, Y. Nakamura, C. J. P. M. Harmans, and J. E. Mooij, Coherent dynamics of a flux qubit coupled to a harmonic oscillator, Nature 431, 159 (2004).
[19] A. Wallraff et al., Strong coupling of a single photon to a superconducting qubit using circuit quantum electrodynamics, Nature 431, 162 (2004).
[20] M. H. Devoret, S. Girvin, and R. Schoelkopf, Circuit-QED: how strong can the coupling between a Josephson junction atom and a transmission line resonator be? Ann. Phys. (Leipzig.) 16, 767 (2007).
[21] T. Niemczyk, F. Deppe, H. Huebl, E. P. Menzel, F. Hocke, M. J. Schwarz, J. J. Garcia-Ripoll, D. Zueco, T. Hämmer, E. Solano, A. Marx, and R. Gross, Circuit quantum electrodynamics in the ultrastrong-coupling regime, Nat. Phys. 6, 772 (2010).
[22] P. Forst-Díaz, J. Lisenfeld, D. Marcos, J. J. García-Ripoll, E. Solano, C. J. P. M. Harmans, and J. E. Mooij, Observation of the Bloch-Siegert shift in a qubit-oscillator system in the ultrastrong coupling regime, Phys. Rev. Lett. 105, 237001 (2010).
[23] P. Forst-Díaz, J. J. García-Ripoll, B. Peropadre, J. L. Or-giazzi, M. A. Yurtalan, R. Belyansky, C. M. Wilson, and A. Lupascu, Ultrastrong coupling of a single artificial atom to an electromagnetic continuum in the nonperturbative regime, Nat. Phys. 13, 39 (2017).
[24] F. Yoshihara, T. Fuse, S. Ashhab, K. Kakuyanagi, S. Saito, and K. Semba, Superconducting qubit-oscillator circuit beyond the ultrastrong-coupling regime, Nat. Phys. 13, 44 (2017).
[25] F. Yoshihara, T. Fuse, Z. Ao, S. Ashhab, K. Kakuyanagi, S. Saito, T. Aoki, K. Koshino, and K. Semba, Inversion of qubit energy levels in qubit-oscillator circuits in the deep-strong-coupling regime, Phys. Rev. Lett. 120, 183601 (2018).
[26] Z. Chen, Y. Wang, T. Li, L. Tian, Y. Qiu, K. Inomata, F. Yoshihara, S. Han, F. Nori, J. S. Tsai, and J. Q. You, Single-photon-driven high-order sideband transitions in an ultrastrongly coupled circuit-quantum-electrodynamics system, Phys. Rev. A 96, 012325 (2017).
[27] Shuai-Peng Wang, Guo-Qiang Zhang, Yimin Wang, Zhen Chen, Tiefu Li, J. S. Tsai, Shi-Yao Zhu, and J. Q. You, Photon-Dressed Bloch-Siegert Shift in an Ultrastrongly Coupled Circuit Quantum Electrodynamical System Phys. Rev. Applied 13, 054063 (2020).
[28] A. F. Kockum, A. Miranowicz, S. D. Liberato, S. Savasta and F. Nori, Ultrastrong coupling between light and matter, Nat. Rev. Phys. 1, 19 (2019).
[29] P. Forst-Díaz, L. Lamata, E. Rico, J. Kono, and E. Solano, Ultrastrong coupling regimes of light-matter interaction, Rev. Mod. Phys. 91, 025005 (2019).
[30] D. Zueco, G. M. Reuther, S. Kohler, and P. Hänggi, Qubit-oscillator dynamics in the dispersive regime: Analytical theory beyond the rotating-wave approximation, Phys. Rev. A 80, 033846 (2009).
[31] S. Ashhab and F. Nori, Qubit-oscillator systems in the ultrastrong-coupling regime and their potential for preparing nonclassical states, Phys. Rev. A 81, 042311 (2010).
[32] J. Casanova, G. Romero, I. Lizuain, J. J. García-Ripoll, and E. Solano, Deep Strong Coupling Regime of the Jaynes-Cummings Model, Phys. Rev. Lett. 105, 263603 (2010).
[33] D. Z. Rosatto, C. J. Villas-Bôas, M. Sanz, and E. Solano, Spectral classification of coupling regimes in the quantum Rabi model, Phys. Rev. A, 96, 013849 (2017).
[34] P. Nataf and C. Ciuti, Vacuum degeneracy of a circuit QED system in the ultrastrong coupling regime, Phys. Rev. Lett. 104, 023601 (2010).
[35] G. Vacanti, S. Pugnetti, N. Didier, M. Paternostro, G. M. Palma, R. Fazio, and V. Vedral, Photon Production from the Vacuum Close to the Superradiant Transition: Linking the Dynamical Casimir Effect to the Kibble-Zurek Mechanism, Phys. Rev. Lett. 108, 093603 (2012).
[36] L. Garziano et al., Switching on and off of ultrastrong light-matter interaction: Photon statistics of quantum vacuum radiation, Phys. Rev. A 88, 063829 (2013).
[37] R. Stassi et al., Spontaneous conversion from virtual to real photons in the ultrastrong-coupling regime, Phys. Rev. Lett. 110, 243601 (2013).
[38] M. Cirio et al., Amplified optomechanical transduction of virtual radiation pressure, Phys. Rev. Lett. 119, 053601 (2017).
[39] L. Garziano et al., Vacuum-induced symmetry breaking in a superconducting quantum circuit, Phys. Rev. A 90, 043817 (2014).
[40] C. S. Munoz et al., Resolution of superluminal signalling in non-perturbative cavity quantum electrodynamics, Nat. Commun. 9, 1924 (2018).
[41] J. R. Johansson et al., Dynamical Casimir effect in a superconducting coplanar waveguide, Phys. Rev. Lett. 103, 147003 (2009).
[42] C. M. Wilson et al., Observation of the dynamical Casimir effect in a superconducting circuit, Nature 479, 376-379 (2011)
[43] Y. X. Liu et al., Optical Selection Rules and Phase-Dependent Adiabatic State Control in a Superconducting Quantum Circuit, Phys. Rev. Lett. 95, 087001 (2005).
[44] A. Goban et al., Atom-light interactions in photonic crys-
[46] A. Ridolfo et al., Photon Blockade in the Ultrastrong Coupling Regime, Phys. Rev. Lett. 109, 193602 (2012).

[47] M.-J. Hwang, R. Puebla, and M. B. Plenio, Quantum phase transition and universal dynamics in the Rabi model, Phys. Rev. Lett. 115, 180404 (2015).

[48] M. Liu, S. Chesi, Z.-J. Ying, X. Chen, H.-G. Luo, and H.-Q. Lin, Universal scaling and critical exponents of the anisotropic quantum Rabi model, Phys. Rev. Lett. 119, 220601 (2017).

[49] Q.-T. Xie, S. Cui, J.-P. Cao, L. Amico, and H. Fan, Anisotropic Rabi model, Phys. Rev. X 4, 021046 (2014).
Supplementary Information for “Detecting the symmetry breaking of the quantum vacuum in a light–matter coupled system”

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ADDITIONAL EXPERIMENTAL DETAILS

The experimental setup is shown in Fig. S1. The superconducting lumped-element and coplanar-waveguide resonators are made by patterning a niobium thin film of thickness 50 nm deposited on a 10×3 mm2 silicon chip via electron beam lithography. The flux qubit is also fabricated on the silicon substrate in the middle of the center conductor of the lumped-element resonator by using both electron beam lithography and double-angle evaporation of aluminium. An external magnetic field generated by a magnetic coil surrounding the device is applied to tune the magnetic flux threading through the qubit loop. The Josephson junction in the Xmon is connected to the cross-shaped capacitor at one end and fabricated using separate steps of electron beam lithography and double-angle evaporation. Reflection spectra of the deep-strongly coupled qubit–resonator system at the frequency ωp of the probe tone are measured with a vector network analyser (VNA). Another microwave signal at δtuned to the driving frequency of the Xmon for two-tone spectroscopy measurements. The input signals are attenuated and filtered at various temperature stages before finally reaching the sample. Also, two isolators and a low-pass filter (LPF) are used to protect the sample from the amplifier’s noise.

The dependency of the resonance frequency of the lumped-element resonator ωr on the external flux bias δΦext around the optimal point can be approximately described as

\[
\omega_r(\delta\Phi_{\text{ext}}) = \frac{\omega_r(0)}{\sqrt{1 + D \cos \theta |\delta\Phi_{\text{ext}}|}}.
\]

where the constant \(D = -0.18\) and is determined by tuning the fitting curve of \(\omega_0\) in Fig. 2c of the main text when \(\omega_r(0)\) is fixed.

Figure S2 displays the bare and normalized reflection spectra without the fitting curves of Figs. 2a and 2b in the main text. The abrupt changes in the transmission background around 5.6 GHz and 11.9 GHz can be seen in the bare spectra in Figs. S2a and S2b.

Figure S3 displays the calculated excitation spectra \(\omega_{0,n}\) of the total system (the deep-strongly coupled qubit–resonator system plus the Xmon). The observed transitions \(\omega_{01}, \omega_{02}\) and \(\omega_{03}\) in Fig. 2 of the main text correspond to \(\omega_{0,1}, \omega_{0,3}\) and \(\omega_{0,6}\). The sideband transitions in Fig. 2a of the main text correspond to (from inside to outside) \(\omega_{18} \approx \omega_{04,X} - (\omega_{01} + \omega_{10,Y}), \omega_{5,20} \approx (\omega_{02} + \omega_{03,Y}) - \omega_{12,X}, \omega_{5,16} \approx (\omega_{04} + \omega_{01,Y}) - \omega_{02,X}\) and \(\omega_{5,15} \approx \omega_{05} - \omega_{02,X}\), where \(\omega_{n,m} = \omega_{0,m} - \omega_{0,n}\) and \(\omega_{0,n,X}\) denotes the dressed excitation energy levels of the Xmon. The observation of these high-order sideband transitions near the band edge is a surprise, which may demand a further theoretical study.

THEORY

Theoretical description

We consider a deep-strongly coupled (DSC) system constituted by a flux qubit coupled inductively to a lumped-element resonator through a shared Josephson junction. This system can display a photonic vacuum symmetry breaking [1]. Here we demonstrate that the presence of such symmetry breaking can induce the
FIG. S1. Schematic of the experimental setup. Figure 1a in the main text is the area denoted by the blue rectangular box.

FIG. S2. a(c) and b(d) are bare (normalized) reflection spectra without the fitting curves of Figs. 2a and 2b in the main text.

FIG. S3. Calculated excitation spectra of the total system ($\omega_{0,n}$). The parameters used in the calculation are the same as in the main text. The dressed excitation energy levels of the deep-strongly coupled qubit–resonator system $\omega_{0n}$ (Xmon $\omega_{0m,X}$) are in blue (magenta). The black levels correspond to multiple excitation levels $\omega_{0n} + \omega_{0m,X}$. The red dashed double-arrowed lines indicate the observed sideband transitions in Fig. 2a of the main text.

breaking of parity selection rules of an Xmon artificial atom interacting dispersively with the ultrastrongly coupled system via the lumped-element resonator. This selection-rule breaking can then be probed by applying a driving field on the coupled Xmon.

The total Hamiltonian for our three-component system
can be written as
\[ H_{\text{tot}} = H_s + H_X^{(4)} - g'(a - a^\dagger)(b - b^\dagger), \quad (S2) \]
where \( H_s \) is the Hamiltonian of the DSC qubit–resonator system in the main text, and
\[ b = \sum_{n=0}^{3} \sqrt{n+1} |n\rangle\langle n+1| \quad (S3) \]
is the annihilation operator for the Xmon modeled here as a four-level artificial atom (qudit). The bare Xmon as a qudit is
\[ H_X^{(4)} = \sum_{n=0}^{3} \varepsilon_n |n\rangle\langle n|, \quad (S4) \]
where we choose \( \varepsilon_0 = 0 \) for convenience.

Owing to the parity symmetry, two-photon transitions in the bare Xmon artificial atom are forbidden. Thus, an observation of the two-photon transition represents a signature of induced breaking of the selection rules in the Xmon coupled to the qubit–resonator system. Below we show that the two-photon transition rate can be directly calculated using second-order perturbation theory.

Let us consider a single-tone external drive exciting the Xmon,
\[ H_1(t) = (\Omega/2)(Y^- e^{-i\omega_d t} + Y^+ e^{i\omega_d t}), \quad (S5) \]
where
\[ Y^+ = -i \sum_{j<k} \langle E_j|(b - b^\dagger)|E_k\rangle |E_j\rangle\langle E_k|, \quad (S6) \]
with \( |E_j\rangle \) being the eigenstates of the Hamiltonian \( (S2) \), which are ordered so that \( j < k \) if \( \omega_j < \omega_k \).

We choose system’s ground state \( |E_0\rangle \) as the initial state. The first-excited state of the system corresponds to the dressed first-excited state of the DSC qubit–resonator system. The second-excited state of the system corresponds to the dressed first-excited state of the Xmon. The transition rate from \( |E_0\rangle \) to \( |E_2\rangle \) can be written as
\[ W_2(\omega_d) = 2\pi |\mathcal{T}_{02}|^2 \delta(\omega_{02} - 2\omega_d), \quad (S7) \]
where \( \omega_{j,k} = \omega_k - \omega_j \), and
\[ \mathcal{T}_{02} = \sum_k \frac{V_{0,k}V_{k,2}}{\omega_{0,k} - \omega_d}. \quad (S8) \]
Here, the matrix elements of the perturbation potential are
\[ V_{j,k} = \frac{\Omega}{2} \langle E_j|Y^+|E_k\rangle. \quad (S9) \]
The losses of the system components can be included by converting the Dirac delta function in Eq. \((S7)\) into a Lorentzian:
\[ \delta(\omega_{02} - 2\omega_d) \rightarrow \frac{1}{2\pi} \frac{\Gamma_{02}}{\omega_{02} - 2\omega_d} + \frac{\Gamma_{02}^2}{4}. \quad (S10) \]

The definition of the loss rates as \( \Gamma_{2,0} \) can be found in the next section.

The two-photon Xmon power spectrum can be obtained as
\[ \langle Y^-Y^+ \rangle = \text{Tr}[\rho Y^-Y^+], \quad (S11) \]
where \( \rho \) is system’s density operator. It can be written as
\[ \langle Y^-Y^+ \rangle = \frac{W_2(\omega_d)}{\Gamma_{02}} (|Y_{0,2}\rangle^2 + |Y_{1,2}\rangle^2), \quad (S12) \]
where \( Y_{j,k} = \langle E_j|Y^+|E_k\rangle \). In comparison with \( Y_{0,2} \), the matrix element \( Y_{1,2} \) is almost negligible.

### Density matrix approach

Here we derive the two-photon emission rate of the three-component system described in the previous section, by using a perturbative master equation approach. We start from the master equation for the coupled system in the basis of its eigenstates (dressed-states approach \([2]\)), including the external drive (that is the perturbation):
\[ \dot{\rho} = i[\rho, H_0 + H_1(t)] + \mathcal{L}[\rho], \quad (S13) \]
where \( H_0 = \sum_n \omega_n |E_n\rangle\langle E_n| \) is the diagonalized Hamiltonian of Eq. \((S2)\). The external driving field is described by the time-dependent interaction
\[ H_1(t) = \Omega \cos(\omega_d t)(Y^- + Y^+), \quad (S14) \]
and \( \mathcal{L}[\rho] \) is the dissipator expressed in the eigenstates of the DSC system (see, e.g., Ref. \([2]\)). Applying the rotating-wave approximation, we can write
\[ H_1(t) = (\Omega/2)(Y^- e^{-i\omega_d t} + Y^+ e^{i\omega_d t}). \quad (S15) \]
We will also consider an expansion of the density matrix in terms of powers of the drive amplitude, so that we can separate each contribution, i.e.
\[ \rho = \rho^{(0)} + \rho^{(1)} + \rho^{(2)} + ... = \sum_{n=0}^{\infty} \rho^{(n)}. \quad (S16) \]
Moreover, since the drive can be expanded in Fourier components as
\[ H_1(t) = \sum_k H^{(1)}_{\omega_k} \exp(i\omega_k t), \quad (S17) \]
(actually, we will use here only a single-tone drive) the same can be done with \( \rho^{(n)} \), leading to the expression
\[ \rho = \sum_{n=0}^{\infty} \sum_k \rho^{(n)}_{\omega_k} \exp(i\omega_k t), \quad (S18) \]
where $\omega_k = \pm k \omega_d$, with $k$ being an integer number. Considering the steady state, plugging Eq. (S18) into the master equation, we obtain a set of closed equations separated in driving orders and Fourier components, that at the lowest order looks like:

$$ \rho^{(0)}(0) = 0 = i[\rho^{(0)}, H_0] + \mathcal{L}[\rho^{(0)}], \quad (S19) $$

which leads to the thermal density operator

$$ \rho^{(0)} = \frac{1}{Z} \sum_j \exp \left( -\frac{\hbar \omega_j}{k_B T} \right) |E_j\rangle\langle E_j|, \quad (S20) $$

with the partition function $Z = \sum_j \exp \left( -\frac{\hbar \omega_j}{k_B T} \right)$. The next two sets of equations for the first- and second-order density matrices can be formally derived from the master equation, and they have the following forms:

$$ \dot{\rho}^{(1)}(\omega_k) = i\omega_k \rho^{(1)}(\omega_k) = i[\rho^{(0)}, H^{(1)}_\omega] + i[\rho^{(1)}_\omega, H_0] + \mathcal{L}[\rho^{(1)}_\omega], \quad (S21a) $$

$$ \dot{\rho}^{(2)}(\omega_j + \omega_k) = i(\omega_j + \omega_k) \rho^{(2)}(\omega_j + \omega_k) = i[\rho^{(1)}_j, H^{(1)}_\omega] + i[\rho^{(2)}_j, H_0] + \mathcal{L}[\rho^{(2)}_j + \omega_k]. \quad (S21b) $$

The solutions of the above expressions can be derived sequentially, by plugging the already found solution for the previous order. In this way we find for the first- and second-order steady-state density matrices:

$$ (\rho^{(1)}_{\omega_k})_{n,m} = \frac{(H^{(1)}_{\omega_k})_{n,m}}{\omega_k - \omega_n,m + i\Gamma_{n,m}/2} (\rho^{(0)}_{n,m} - (\rho^{(0)}_{m,n}), \quad (S22a) $$

$$ (\rho^{(2)}_{\omega_j + \omega_k})_{n,m} = \frac{([H^{(1)}_{\omega_k}, \rho^{(1)}_{\omega_k}])_{n,m} + ([H^{(1)}_{\omega_j}, \rho^{(1)}_{\omega_j}])_{m,n}}{\omega_j + \omega_k - \omega_n,m + i\Gamma_{n,m}/2}. \quad (S22b) $$

The decay rates $\Gamma_{n,m}$ are the sum of all the lossy channels involved in the $|E_m\rangle \rightarrow |E_n\rangle$ transition and its expression is shown below. For a nonzero temperature reservoir, one has to take care of the modified decay rates for the generic transition $|E_j\rangle \rightarrow |E_i\rangle$ (with $j > i$). For the sake of simplicity, we assume that both the artificial atoms and the lumped-element resonator are thermalized at the same temperature. Thus, the rates can be calculated via the Fermi’s golden-rule. At zero-temperature, we obtain $\Gamma_{i,j} = \gamma_{i,j}^{(a)} + \gamma_{i,j}^{(b)}$, where $\gamma_{i,j}^{(a)} = \gamma_a |\langle E_i|\sigma_+|E_j|\rangle|^2$, $\gamma_{i,j}^{(b)} = \gamma_a |\langle E_i|(a + a^\dagger)|E_j|\rangle|^2,$ and $\gamma_{i,j}^{(b)} = \gamma_b |\langle E_i|(b + b^\dagger)|E_j|\rangle|^2$. At $T > 0$, the loss rates become

$$ \Gamma_{i,j} = \sum_{s=q,a,b} \left( \sum_{k<i} \gamma_{i,k}^{(s)}(1+2\bar{n}_{i,k}) + \sum_{k>j} \gamma_{j,k}^{(s)}(1+2\bar{n}_{j,k}) \right), \quad (S23) $$

where $\bar{n}_{i,j}$ is the thermal noise that affects the $|E_j\rangle \rightarrow |E_i\rangle$ transition, i.e., $\bar{n}_{i,j} = 1/[\exp(\hbar \omega_{i,j}/k_B T) - 1]$. In the following theoretical analysis, corroborated by the standard working temperature of the DSC system ($T \sim 30$ mK), we can safely ignore the enhancement induced rates, by choosing the relevant $\bar{n}_{i,j} = 0$.

In our calculations, we stop the perturbative development up to the second order, as the essential physics is fully caught. In particular, we can obtain the single- and two-photon absorption by looking at the polarization of the Xmon as a function of the drive frequency, which can be defined as $\langle P(\omega_d) \rangle = \text{Tr}[-i(b-b^\dagger)\rho(\omega_d)]$. In this formula, we can apply further simplifications by (i) considering the case of a ground state environment ($T = 0$) and (ii) dropping the negligible terms of $\langle P(\omega_d) \rangle$, finding that the leading terms that drive the onset of the sought effect yield

$$ |\langle P(\omega_d) \rangle| \approx |\langle Y_{2,0}\rho^{(2)}_{0,2} + Y_{0,2}\rho^{(2)}_{2,0} \rangle| = 2\Re[Y_{2,0}\rho^{(2)}_{0,2}]. \quad (S24) $$

Terms like $Y_{2,0}\rho^{(1)}_{0,2} + Y_{0,2}\rho^{(1)}_{2,0}$ represent a constant background as they are far-detuned resonances, and thus they can be ignored. Furthermore, we also find that $|Y_{2,0}| \approx 1$, as it represents the transition matrix element of the weakly coupled Xmon, from its ground to the first-excited level. Following our perturbative approach, we find that

$$ \rho^{(2)}_{0,2} = -\frac{\Omega^2}{\omega_d - \omega_{0,1} + i\Gamma_{0,2}/2} \frac{Y_{0,1}Y_{1,2}}{(2\omega_d - \omega_{0,2} + i\Gamma_{0,2}/2)}. \quad (S25) $$

Finally, as the detected field has frequency $\omega_d \sim \frac{1}{2}\omega_{2,0}$, the non-resonant part of the denominator can be expanded in series, leading to the final expression:

$$ |\langle P(\omega_d) \rangle| = \frac{\Omega^2|Y_{0,1}Y_{1,2}|}{\omega_{1,2} - \omega_{0,1}} \frac{\Gamma_{0,2}|Y_{2,0}|}{(2\omega_d - \omega_{0,2})^2 + \Gamma_{0,2}^2/4}. \quad (S26) $$

**Results**

Below we show the numerical results achieved with parameters extrapolated by the experimental data.
FIG. S4. Calculated most relevant terms in the two-photon absorption rate as a function of the external flux bias $\delta \Phi_{\text{ext}}$. The detected signal is approximately proportional to the product of these quantities. The calculated $Y_{1,2}$ term goes to zero for $\delta \Phi_{\text{ext}} \to 0$, when the parity selection rule for the dressed Xmon is restored. The $Y_{3,2}$ is not shown in the figure as it is almost unity in the whole range investigated here.

Namely, we use $\omega_r/2\pi = 4.82$ GHz, $\Delta/2\pi = 15$ GHz, $g/2\pi = 4.55$ GHz, $g'/2\pi \approx g_X/2\pi = 28$ MHz, and loss rates for the flux qubit, lumped-element resonator and Xmon are chosen to be $\gamma^{(\text{q})}/2\pi = \gamma^{(\text{a})}/2\pi = \gamma^{(b)}/2\pi = 2$ MHz. Also, we consider a thermalized DSC system at a temperature of 30 mK. Our model is able to reproduce the experimental results with a very good agreement. In the simulations, we use a quite strong external field exciting the Xmon, in order to produce a two-photon absorption, and its value in terms of linewidth is $\Omega = 200 \times \gamma^{(b)}$. Such a value is large enough to ensure the onset of the second-order processes. While for the direct excitation of the single-photon absorption, we reduce $\Omega$ by a factor $2 \times 10^6$, in doing so we can get the same order of the amplitudes of the single- and two-photon signals, which is consistent with the experimental observation.

Figure 3 in the main text shows the comparison between experimental (top) and theoretical (middle) results calculated for the reported parameters. The left panel shows the weak-excitation, two-tone spectroscopy of the dressed Xmon ($|E_0\rangle \to |E_2\rangle$) transition, while in the right panel, a strong-excitation spectroscopy shows the two-photon resonance at half of the Xmon ($|E_0\rangle \to |E_2\rangle$) transition. Both the experimental and theoretical signals are displayed as a function of the drive frequency $\omega_d$ and of the external flux bias, $\delta \Phi_{\text{ext}}$. For zero flux bias $\delta \Phi_{\text{ext}} = 0$, the two-photon resonance disappears as the parity symmetry is completely restored. The theoretical calculations display the changes in the amplitude of the Xmon polarization $|\langle P(\omega_d)\rangle|$. Figure S4 displays the most relevant term in the two-photon absorption rate as a function of the flux bias $\delta \Phi_{\text{ext}}$. It shows the origin of the dependence on the flux bias of the two-photon transition rate for the dressed Xmon. In particular, we observe that the matrix element $Y_{1,2}$ goes to zero for $\delta \Phi_{\text{ext}} \to 0$, owing to the restoration of parity symmetry. This disables two-photon transitions from the ground state to the dressed first excited state of the Xmon ($|E_0\rangle \to |E_2\rangle$).

[1] L. Garziano, R. Stassi, A. Ridolfo, O. Di Stefano, and S. Savasta, Vacuum-induced symmetry breaking in a superconducting quantum circuit, Phys. Rev. A 90, 043817 (2014).

[2] A. Ridolfo, M. Leib, S. Savasta, and M. J. Hartmann, Photon Blockade in the Ultrastrong Coupling Regime, Phys. Rev. Lett. 109, 193602 (2012).