

## Simultaneous Excitation of Two Noninteracting Atoms with Time-Frequency Correlated Photon Pairs in a Superconducting Circuit

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We report the first observation of simultaneous excitation of two noninteracting atoms by a pair of time-frequency correlated photons in a superconducting circuit. The strong coupling regime of this process enables the synthesis of a three-body interaction Hamiltonian, which allows the generation of the tripartite Greenberger-Horne-Zeilinger state in a single step with a fidelity as high as 0.95. We further demonstrate the inhibition of the simultaneous two-atom excitation by continuously measuring whether the first photon is emitted. This work provides a new route in synthesizing many-body interaction Hamiltonian and coherent control of entanglement.

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Two-photon absorption, where two photons successively induce a quantum transition, was predicted by Goepfert-Mayer in 1931 [1]. It was not experimentally observed [2] until the invention of the laser, which provides the necessary intense monochromatic radiation. This effect has been widely used in fluorescent imaging [3,4], micro-fabrication [5], and quantum light sources [6]. It is worth noting that time-frequency entangled two photons or squeezed light can efficiently enhance two-photon absorption [7–10]. Two photons can also simultaneously excite two atoms that are interacting with each other [11–13], which offers new methods in collective control of quantum systems [14] and in spectroscopy with nanoscale resolution [15]. However, the requisite of interaction between atoms hinders the application of this effect in quantum information processing, since it is difficult to make far apart qubits interact with each other [16,17]. To circumvent this obstacle, many proposals [18–23] on simultaneous two-atom excitation have emerged. In particular, a single photon can simultaneously excite two atoms [18] with the effect of the counterrotating terms in ultrastrong coupling [24]. However, it is still challenging to realize the ultrastrong coupling and to mitigate its side effect. Until now, the simultaneous excitation of two noninteracting atoms has never been observed experimentally.

While two uncorrelated photons with frequencies  $\nu_1$  and  $\nu_2$  cannot simultaneously excite two noninteracting atoms with frequencies  $\omega_1$  and  $\omega_2$  at the two-photon resonance,

$$\begin{aligned} \nu_1 + \nu_2 &= \omega_1 + \omega_2, \\ \nu_i &\neq \omega_j \quad \text{for all } i, j = 1, 2, \end{aligned} \quad (1)$$

it has been shown that the time-frequency entangled photon pairs generated in a cascade three-level system can excite two noninteracting atoms [10,25]. There are four quantum pathways for two atoms absorbing two photons, as shown in Fig. 1(a). With two independent photons, the probability amplitudes in these four pathways cancel out. However, if the two-photons are time-frequency entangled such that only two pathways are allowed, there is a finite probability that the two atoms are simultaneously excited. Unfortunately, the difficulty in collecting the photon pairs in free space greatly limits the efficiency of this process despite its promising applications in quantum spectroscopy [26,27]. In this Letter, we experimentally demonstrate that by confining the photon pairs in a cavity, two noninteracting artificial atoms can be simultaneously excited in a superconducting circuit. The three-body interaction between the light source and the two atoms is in the strong-coupling regime, such that the Greenberger-Horne-Zeilinger (GHZ) state [28] can be dynamically generated in a single step. We also show that this many-body dynamics can be coherently controlled by a continuous measurement [29–31]. This scheme requires neither counterrotating terms nor ultrastrong coupling and provides a new method

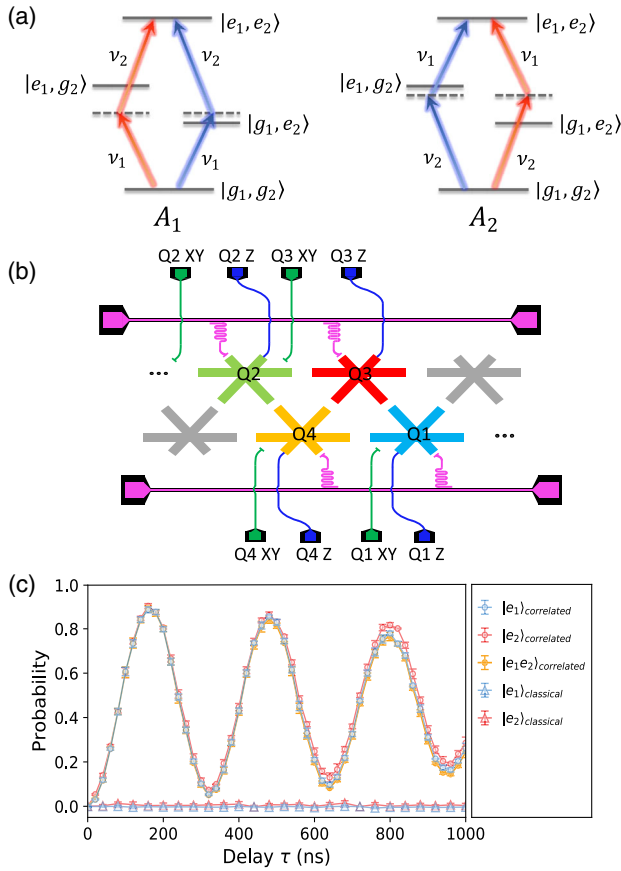


FIG. 1. Simultaneous excitation of two noninteracting qubits. (a) The quantum paths for  $A_1$  and  $A_2$  in Eq. (1). The pathways with the same color (blue or red) in  $A_1$  and  $A_2$  cancel each other, evident from their opposite values of the detunings from the atomic levels. (b) Sketch of part of the multiqubit superconducting circuit. Four qubits, labeled  $Q_1$ ,  $Q_2$ ,  $Q_3$ , and  $Q_4$ , are used in this experiment. Each qubit has its own flux bias line (blue) for fast frequency tuning and microwave line (green) for SU(2) spin rotations. For qubit readout, each qubit is capacitively coupled to its own readout resonator which is coupled to one of the two common transmission lines (pink). (c) The single-qubit probabilities of finding  $Q_1$  (blue) and  $Q_2$  (red) in their excited states when they are simultaneously driven by correlated photon pairs (circles) and uncorrelated classical microwaves (triangles) as functions of the interaction time  $\tau$ . The joint probability of finding  $Q_1$  and  $Q_2$  in  $|e_1, e_2\rangle$  by absorbing the correlated photon pairs is denoted by the filled yellow circles, which almost overlap with the empty blue and red circles. The simultaneous excitation of the two qubits is in conformity with the prediction of Ref. [10].

of synthesizing three-body interactions and entanglement generation.

Intuitively, it is easy to understand that two uncorrelated photons cannot simultaneously excite two noninteracting atoms under the condition in Eq. (1). Since each atom independently interacts with the two photons and can be considered separately, they cannot be excited because neither of them is in resonance with the photons. For the

convenience of the discussion on the correlated photon pairs, we alternatively regard the two atoms as an entity and the transition probability can be calculated by considering which photon is absorbed first. The two-photon transition matrix element consists of two parts,  $A_1$  and  $A_2$ , corresponding to the quantum pathways where  $\nu_1$  and  $\nu_2$  are first absorbed, as shown in Fig. 1(a). From the second order perturbation theory, we obtain [10],

$$A_1 = \frac{\kappa_{22}\kappa_{11}}{\nu_1 - \omega_1} + \frac{\kappa_{12}\kappa_{21}}{\nu_1 - \omega_2},$$

$$A_2 = \frac{\kappa_{21}\kappa_{12}}{\nu_2 - \omega_1} + \frac{\kappa_{11}\kappa_{22}}{\nu_2 - \omega_2}, \quad (2)$$

where  $\kappa_{ij}$  (much smaller than the one-photon detunings  $|\nu_j - \omega_i|$  to avoid single-photon excitation and make second order process dominate) is the coupling strength between the atom and photon with frequencies  $\omega_i$  and  $\nu_j$ . The two-photon resonance in Eq. (1) renders that  $A_1 = -A_2$  and the total two-photon transition matrix element is zero. However, if the two photons are correlated in such a way that some of the pathways are absent, we can remove the cancellation between the four pathways [32] (see [25] for more general discussion). A famous example is the time-frequency entangled photon pair generated from a cascade transition of a three-level atom [10], where the photon with frequency  $\nu_1$  is guaranteed to arrive at the atoms first. In this case only  $A_1$  is present and it is generally nonzero. Consequently, the two atoms can be simultaneously excited. However, although time-frequency correlated photon pairs can be generated in nonlinear crystals [33] and atomic media [34], the simultaneous excitation of two noninteracting atoms has never been experimentally observed due to the difficulty both in finding the proper atoms and in collecting the entangled photon pairs.

Superconducting circuits offer a versatile platform to verify this interesting process, thanks to the flexibility of the artificial atoms (superconducting qubits) and the strong atom-photon coupling. The experiment is performed in a superconducting circuit consisting of multiple qubits interconnected in a zigzag triangular ladder [35] [see Fig. 1(b)], where the couplings between neighboring qubits are measured to be 10–13 MHz ( $\times 2\pi$ ) and otherwise the couplings are weaker by about an order of magnitude. Each qubit is a frequency tunable transmon [36] circuit, whose sinusoidal potential well hosts multiple energy levels. The lowest three energy levels of the  $j$ th qubit  $Q_j$  are referred to as the ground state  $|g_j\rangle$ , the first excited state  $|e_j\rangle$ , and the second excited state  $|f_j\rangle$ , the latter of which is only involved in the experiment for  $Q_3$ . In our experimental setup, nonadjacent  $Q_1$  and  $Q_2$  are the two noninteracting atoms, and  $Q_3$  is a qutrit that generates the cascade photon pairs.  $Q_4$  is used as a measuring qubit in demonstrating the quantum Zeno effect. We can tune the resonance frequencies, perform SU(2) rotations and measure the quantum

states of these qubits through their own flux bias lines,  $XY$  lines and readout resonators, respectively. At the experimental frequencies, the coherence performance of these qubits is characterized by the lifetime and Ramsey interference measurements, which yield the typical relaxation time  $T_1 > 20 \mu\text{s}$  and the Gaussian dephasing time  $T_2^* \approx 1 \mu\text{s}$  (see the Supplemental Material [37]).

The key experimental result is shown in Fig. 1(c). We first prepare  $Q_3$  in the state  $|f_3\rangle$ , which subsequently emits two cascade photons with frequencies  $\nu_1 \approx 2\pi \times 4.865 \text{ GHz}$  (for  $|f_3\rangle \rightarrow |e_3\rangle$  transition) and  $\nu_2 \approx 2\pi \times 5.100 \text{ GHz}$  (for  $|e_3\rangle \rightarrow |g_3\rangle$  transition). These two photons simultaneously excite  $Q_1$  and  $Q_2$  with transition frequencies  $\omega_1 \approx 2\pi \times 5.183 \text{ GHz}$  and  $\omega_2 \approx 2\pi \times 4.781 \text{ GHz}$ , which satisfy Eq. (1). The strong coupling regime of this process is demonstrated by the well observed three-body Rabi oscillation in Fig. 1(c). We notice that the probability of finding  $Q_1$  and  $Q_2$  jointly in  $|e_1, e_2\rangle$  (filled yellow circles) are almost the same as those of finding them individually excited (empty blue and red circles), which is evidence of the simultaneity of the two-atom excitation. In contrast, by simultaneously driving  $Q_1$  and  $Q_2$  with uncorrelated classical microwave pulses with the same frequencies  $\nu_1$  and  $\nu_2$ , we observe no excitation of the qubits, as shown by the triangles in Fig. 1(c).

During the process of the simultaneous excitation of the two atoms, three-body entanglement between the light source and the two atoms is generated. This can be seen from the effective Hamiltonian between  $Q_1$ ,  $Q_2$ , and  $Q_3$  ( $\hbar = 1$ ),

$$H_{\text{eff}} = A_1 \sigma_1^+ \sigma_2^+ \Xi_3^- + \text{H.c.}, \quad (3)$$

where  $A_1$  is shown in Eq. (2) and its value is approximately  $2\pi \times 2 \text{ MHz}$ . The raising operators for the three qubits are defined as  $\sigma_j^+ = |e_j\rangle\langle g_j|$  for  $j = 1, 2$  and  $\Xi_3^+ = |f_3\rangle\langle g_3|$ , and the lowering operators are their Hermitian conjugate. The Hamiltonian in Eq. (3) is a three-body interaction Hamiltonian that can entangle the three qubits in a single step [18,19,21], which has never been realized experimentally. In the present scheme, the large value of  $A_1$  allows a high-fidelity GHZ state to be generated within a timescale much shorter than the typical coherence times of these qubits [37]. The experimental result is shown in Fig. 2.

The pulse sequence for generating the GHZ state is shown in Fig. 2(a). We first prepare  $Q_1$ - $Q_2$ - $Q_3$  in the initial state  $|\psi(0)\rangle = |g_1, g_2, f_3\rangle$  at their respective idle frequencies. Then the three qubits are quickly biased to their operating frequencies, so that Eq. (1) is satisfied [37]. After the dynamic evolution with a variable time  $\tau$ , we bring the qubits back to their idle frequencies for measurement. The results of the joint measurement of the wave function  $|\psi(\tau)\rangle = c_1(\tau)|g_1, g_2, f_3\rangle + c_2(\tau)|e_1, e_2, g_3\rangle$ , ignoring the insignificant terms, are shown in Fig. 2(b), in which the experimentally obtained probabilities of  $|c_1(\tau)|^2$  and

$|c_2(\tau)|^2$  are plotted as functions of the interaction time  $\tau$ . At  $\tau \approx 88 \text{ ns}$ , ideally a three-qubit GHZ state  $|\psi_{\text{GHZ}}\rangle = (|g_1, g_2, f_3\rangle + e^{i(\pi/2+\phi)}|e_1, e_2, g_3\rangle)/\sqrt{2}$  is generated, where  $\phi$  is the dynamical phase picked up as the three qubits are tuned back to their idle frequencies right after the three-body dynamics. Quantum state tomography of a

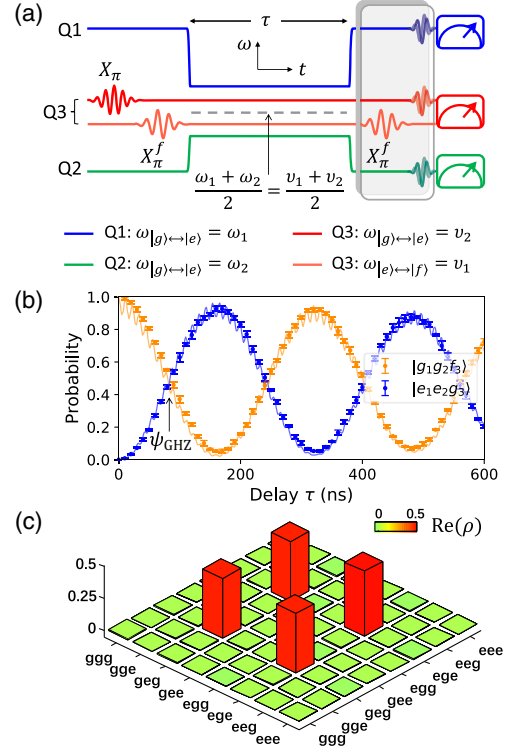


FIG. 2. Generating the GHZ state with the three-body interaction Hamiltonian. (a) Illustrative pulse sequence drawn in the frequency versus time frame. After initializing  $Q_1$ - $Q_2$ - $Q_3$  in  $|g_1, g_2, g_3\rangle$  at their respective idle frequencies, we prepare the state  $|g_1, g_2, f_3\rangle$  by successively applying to  $Q_3$  an  $X_\pi$  rotation (a  $\pi$  rotation around  $X$  axis transferring  $|g_3\rangle$  to  $|e_3\rangle$ , red sinusoid) and an  $X_\pi^f$  rotation (transferring  $|e_3\rangle$  to  $|f_3\rangle$ ), light red sinusoid), following which we apply square pulses to tune the qubit frequencies, so that Eq. (1) is satisfied, for a dynamical evolution with a variable time  $\tau$ . Finally we bring the qubits back to their idle frequencies for simultaneous readout: The same pulse sequence is repeated 1500 times to count the 12 probabilities  $\{P_{ggg}, P_{gge}, P_{ggf}, \dots, P_{gef}, P_{eef}\}$ . The tomographic pulses in the shaded box is only inserted for QST of the GHZ state,  $|\psi_{\text{GHZ}}\rangle$ , where the  $X_\pi^f$  rotation transforms  $|\psi_{\text{GHZ}}\rangle$  to its equivalent  $|\psi'_{\text{GHZ}}\rangle = (|g_1, g_2, e_3\rangle + e^{i\phi}|e_1, e_2, g_3\rangle)/\sqrt{2}$ , followed by null or  $\pi/2$  rotations running through the Pauli set  $\{I, X_{\pi/2}, Y_{\pi/2}\}$  for all three qubits. (b) The dominant occupational probabilities  $|c_1(\tau)|^2$  for  $|g_1, g_2, f_3\rangle$  ( $P_{ggf}$ , orange circles) and  $|c_2(\tau)|^2$  for  $|e_1, e_2, g_3\rangle$  ( $P_{eeg}$ , blue circles) measured as functions of the interaction time  $\tau$ , in comparison with the numerical simulation (lines). (c) QST of the experimental  $|\psi'_{\text{GHZ}}\rangle$  at  $\tau \approx 88 \text{ ns}$ . Shown is the real part of the three-qubit density matrix,  $\rho$ , after a numerical rotation to remove the phase  $\phi$  in  $|\psi'_{\text{GHZ}}\rangle$ .

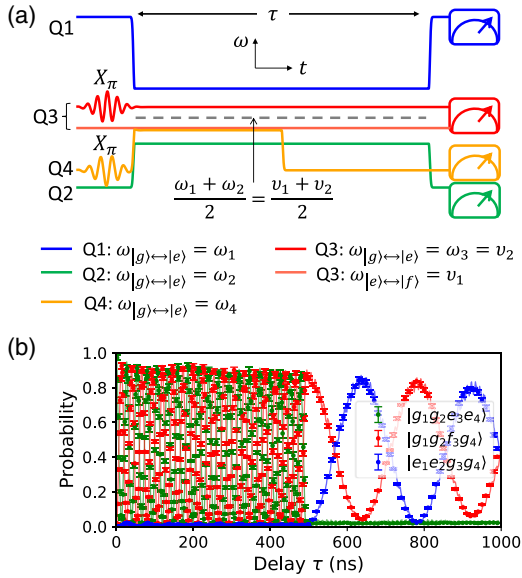


FIG. 3. The quantum Zeno effect of inhibiting the simultaneous two-atom excitation by continuous observation. (a) Illustrative pulse sequence drawn in the frequency versus time frame. Starting from all four qubits in their ground state, we first apply two  $X_\pi$  rotations to  $Q_3$  and  $Q_4$  (red and orange sinusoids). Then, we tune the frequencies of the four qubits into two-photon resonance, i.e.,  $\omega_1 + \omega_2 = \omega_3 + \omega_4$  where  $\omega_3 = \nu_2$  and  $\omega_4 = \nu_1$ , by applying square pulses to the qubit flux bias lines. At  $\tau < 500$  ns,  $Q_4$  is continuously measuring the state of  $Q_3$ . At  $\tau \approx 500$  ns when  $Q_3$  is in state  $|f_3\rangle$ , we tune  $Q_4$  to its idle frequency by applying a rectangle pulse. (b) The probabilities of finding the qubits in the state  $|g_1, g_2, e_3, e_4\rangle$  (green circles),  $|g_1, g_2, f_3, g_4\rangle$  (red circles), and  $|e_1, e_2, g_3, g_4\rangle$  (blue circles) as functions of the interaction time  $\tau$ , in comparison with the numerical simulation (lines). At  $\tau < 500$  ns, we observe the Rabi oscillation between  $|g_1, g_2, e_3, e_4\rangle$  and  $|g_1, g_2, f_3, g_4\rangle$ , without the state  $|e_1, e_2, g_3, g_4\rangle$  being noticeably excited due to the constant observation of  $Q_3$  by  $Q_4$ . At  $\tau > 500$  ns when  $Q_4$  is tuned off-resonant with  $Q_3$ , we observe the simultaneous excitation of  $Q_1$  and  $Q_2$  by the two cascade photons from  $Q_3$ .

$\psi_{\text{GHZ}}$ -equivalent state is shown in Fig. 2(c), which has a state fidelity of  $0.9491 \pm 0.0073$  [37]. The fidelity and preparation time of the GHZ state are comparable with those obtained via two-qubit gates [43].

Our superconducting circuit allows us to coherently control the simultaneous excitation of the two qubits by continuously measuring whether the first photon  $\nu_1$  is emitted. This is similar to the quantum Zeno effect [29–31,44–50], which was originally proposed as a fundamental question on whether frequent measurements can stop the spontaneous decay of a quantum system. The measurement is conducted by resonantly coupling qubit  $Q_4$  to the transition  $|f_3\rangle \leftrightarrow |e_3\rangle$  of  $Q_3$ , i.e., by setting  $\omega_4 \approx \nu_1$ , so that  $Q_3$  in  $|f_3\rangle$  can excite  $Q_4$  in about 14 ns, which is an order of magnitude shorter than the period of the three-body Rabi oscillation. By continuously observing  $Q_3$ , the three-body Rabi oscillation is totally inhibited, as shown in Fig. 3.

In contrast, once the observation is stopped by tuning  $\omega_4$  far away from  $\nu_1$ , the three-body Rabi oscillation appears.

The negligible simultaneous excitation of the two qubits  $Q_1$  and  $Q_2$  when being observed is reminiscent of the quantum Zeno effect, though as argued in literature especially in connection with the original experiment [30,51], the results of the experiment can be explained without invoking the wave function collapse [52,53]. We also notice that the state  $|g_1, g_2, e_3, e_4\rangle$  has negligible coupling with  $|e_1, e_2, g_3, g_4\rangle$ , so that  $Q_4$  does not have any direct effect on  $Q_1$  and  $Q_2$ , which is ideal for measurement with a minimal impact on the observed system.

The simultaneous excitation of two noninteracting atoms is of fundamental importance to the basic law of physics. Since the two atoms are not required to have interactions, in principle they can be placed arbitrarily far apart [10]. This indicates that the law of energy conservation can be locally violated (the absence of the one-photon resonance), although the energy is still conserved globally (i.e., the two-photon resonance). This is achieved by breaking the symmetry in the arrival time of the two photons. Since local energy conservation originates from the time-translational symmetry according to the Noether's theorem, the asymmetric arrival time of the two photons relieves the constraint of the local energy conservation. This effect also offers new freedom in engineering many-body interaction Hamiltonians, complementing the two-body interaction Hamiltonians that have been intensively investigated and utilized in quantum simulation [54–57]. Following the same line, four-body and five-body interaction Hamiltonians can also be synthesized and the current multiqubit superconducting circuit can be used to demonstrate novel topological orders [58,59].

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