



One Photon Can Simultaneously Excite Two or More Atoms

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We consider two separate atoms interacting with a single-mode optical or microwave resonator. When the frequency of the resonator field is twice the atomic transition frequency, we show that there exists a resonant coupling between *one* photon and *two* atoms, via intermediate virtual states connected by counterrotating processes. If the resonator is prepared in its one-photon state, the photon can be jointly absorbed by the two atoms in their ground state which will both reach their excited state with a probability close to one. Like ordinary quantum Rabi oscillations, this process is coherent and reversible, so that two atoms in their excited state will undergo a downward transition jointly emitting a single cavity photon. This joint absorption and emission process can also occur with three atoms. The parameters used to investigate this process correspond to experimentally demonstrated values in circuit quantum electrodynamics systems.

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Multiphoton excitation and emission processes were predicted in 1931 by Göppert-Mayer in her doctoral dissertation on the theory of two-photon quantum transitions [1]. Two-photon absorption consists in the simultaneous absorption of two photons of identical or different frequencies by an atom or a molecule. Two-photon excitation is now a powerful spectroscopic and diagnostic tool [2,3]. One may wonder if the reverse phenomenon, i.e., joint multiatom emission of one photon or multiatom excitation with a single photon, is ever possible. We show that these processes not only can be enabled by the strong correlation between the states of the atoms and those of the field occurring in cavity quantum electrodynamics (QED) [4], but they can even take place with probability approaching one.

Cavity QED investigates the interaction of confined electromagnetic field modes with natural or artificial atoms under conditions where the quantum nature of light affects the system dynamics [5,6]. A high degree of manipulation and control of quantum systems can be reached in the strong-coupling regime, where the atom-field coupling rate is dominant with respect to the loss and decoherence rates. This paves the way for many interesting physical applications [6–9]. Cavity QED is also very promising for the realization of quantum gates [10–12] and quantum networks for quantum computational tasks [13–15]. Many of the proposed concepts, pioneered with flying atoms, have been adapted and further developed using superconducting artificial atoms in the electromagnetic field of microwave resonators, giving rise to the rapidly growing field of circuit QED, which is very promising for future quantum technologies [8,9,12,16–19]. In these systems, coupling rates

between an individual qubit and a single electromagnetic mode of the order of 10% of the unperturbed frequency of the bare subsystems have been experimentally reached [20–23]. Such a coupling rate is significantly higher than that obtained using natural atoms. Such an ultrastrong coupling (USC) opens the door to the study of the physics of virtual processes which do not conserve the number of excitations governed by the counterrotating terms in the interaction Hamiltonian [24–33]. Recently, it has been shown that these excitation-number-nonconserving processes enable higher-order atom-field resonant transitions, making possible coherent and reversible multiphoton exchanges between the qubit and the resonator [34–36].

Here we examine a quantum system constituted by two two-level atoms coupled to a single-mode resonator in the regime where the field-atom detuning $\Delta = \omega_c - \omega_q$ is large as compared to their coupling rate λ (ω_c and ω_q are the resonance frequency of the cavity mode and the qubit transition frequency). We investigate the situation where the two qubits are initially in their ground state and one photon is present in the resonator, corresponding to the initial state $|g, g, 1\rangle$. We find that, if $\omega_c \approx 2\omega_q$, a single cavity photon is able to excite simultaneously two independent atoms. During this process no parametric down-conversion, splitting the initial photon into observable pairs of photons at frequency $\omega_c/2$, occurs. The cavity photon is directly and jointly absorbed by the two atoms. As shown in Fig. 1, the initial state $|g, g, 1\rangle$ goes to virtual intermediate states that do not conserve the energy, but comes back to the real final state $|e, e, 0\rangle$ that does conserve energy (the additional virtual transitions contributing to the process are shown in Fig. S1 of the Supplemental Material

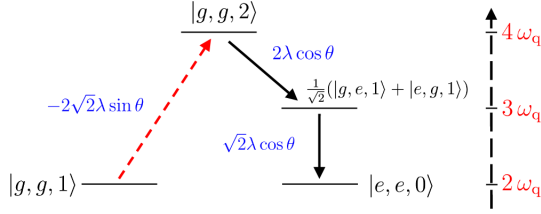


FIG. 1. Sketch of the process giving the main contribution to the effective coupling between the bare states $|g, g, 1\rangle$ and $|e, e, 0\rangle$, via intermediate virtual transitions. Here, the excitation-number-nonconserving processes are represented by arrowed dashed line. The transition matrix elements are also shown.

[37]). If $\omega_c \approx 3\omega_q$, the simultaneous excitation of three atoms, $|g, g, g, 1\rangle \rightarrow |e, e, e, 0\rangle$, is also possible (see Fig. S3 of the Supplemental Material [37]). If the coupling is sufficiently strong, even a higher number of atoms can be excited with a single photon. Owing to optical selection rules, the two-atom process requires parity-symmetry breaking of the atomic potentials, which can be easily achieved in superconducting artificial atoms [34,38,39]. On the contrary, the three-atom process does not need broken symmetry.

The Hamiltonian describing the system consisting of a single cavity mode interacting with two or more identical qubits with possible symmetry-broken potentials is given by [20,31]

$$\hat{H}_0 = \hat{H}_q + \hat{H}_c + \lambda \hat{X} \sum_i (\cos \theta \hat{\sigma}_x^{(i)} + \sin \theta \hat{\sigma}_z^{(i)}), \quad (1)$$

where $\hat{H}_q = (\omega_q/2) \sum_i \hat{\sigma}_z^{(i)}$ and $\hat{H}_c = \omega_c \hat{a}^\dagger \hat{a}$ describe the qubit and cavity Hamiltonians in the absence of interaction, $\hat{X} = \hat{a} + \hat{a}^\dagger$, $\hat{\sigma}_x^{(i)}$ and $\hat{\sigma}_z^{(i)}$ are Pauli operators for the i th qubit, and λ is the coupling rate of each qubit to the cavity mode. For $\theta = 0$, parity is conserved. For flux qubits, this angle, as well as the transition frequency ω_q , can be continuously tuned by changing the external flux bias [20,38]. For the sake of simplicity, Eq. (1) describes identical qubits, but this is not an essential point. In contrast to the Jaynes-Cummings model, the Hamiltonian in Eq. (1) explicitly contains counterrotating terms of the form $\hat{\sigma}_+^{(i)} \hat{a}^\dagger$, $\hat{\sigma}_-^{(i)} \hat{a}$, $\hat{\sigma}_z^{(i)} \hat{a}^\dagger$, and $\hat{\sigma}_z^{(i)} \hat{a}$. The first (second) term creates (destroys) two excitations while the third (fourth) term creates (destroys) one excitation. The presence of counterrotating terms in the interaction Hamiltonian enables four different paths which, starting from the initial state $|g, g, 1\rangle$, reach the final state $|e, e, 0\rangle$ (see Supplemental Material [37]). Each path includes three virtual transitions involving out-of-resonance intermediate states. Figure 1 displays only the process that gives the main contribution to the effective coupling between the bare states $|g, g, 1\rangle$ and $|e, e, 0\rangle$. Higher-order processes, depending on the atom-field interaction strength, can also

contribute. By applying standard third-order perturbation theory, we obtain the following effective coupling rate: $\Omega_{\text{eff}}/\omega_q \equiv (8/3)(\lambda/\omega_q)^3 \sin \theta \cos^2 \theta$. The analytical derivation of the effective coupling rate as a function of λ/ω_q is presented in Sec. I of Supplemental Material [37]. Already at a coupling rate $\lambda/\omega_q = 0.1$, an effective (two qubits)-(one photon) coupling rate $\Omega_{\text{eff}}/\omega_q \sim 10^{-3}$ can be obtained, corresponding, e.g., to an effective Rabi splitting $2\Omega_{\text{eff}} \approx 12$ MHz, for a flux qubit with transition frequency $\omega_q = 6$ GHz. We observe that Ω_{eff} strongly depends on the mixing angle θ , and it is maximum for $\theta = \arccos \sqrt{2/3}$.

We diagonalize numerically the Hamiltonian in Eq. (1) for the case of two qubits and indicate the resulting energy eigenvalues and eigenstates as $\hbar\omega_i$ and $|i\rangle$, with $i = 0, 1, \dots$, choosing the labeling of the states such that $\omega_k > \omega_j$ for $k > j$. We use a normalized coupling rate $\lambda/\omega_q = 0.1$ and an angle $\theta = \pi/6$. Figure 2(a) shows the frequency differences $\omega_{i,0} = \omega_i - \omega_0$ for the lowest energy states as a function of the resonator frequency. Starting from the lowest excited states of the spectrum, a large splitting anticrossing around $\omega_c/\omega_q = 1$ can be observed [see arrows in Fig. 2(a)]. It corresponds to the standard

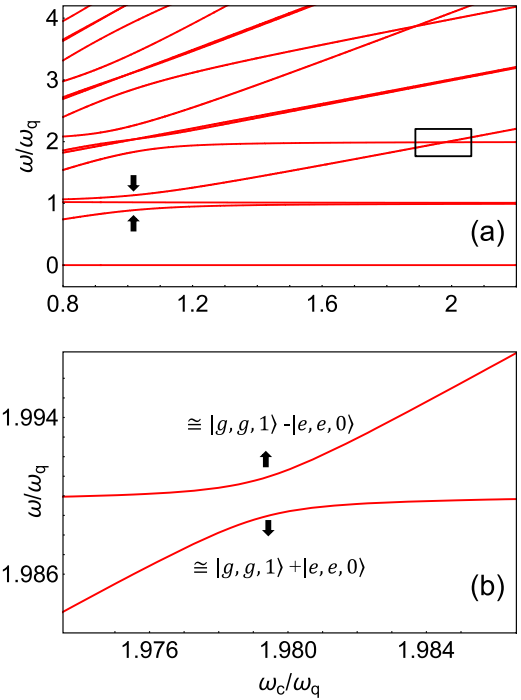


FIG. 2. (a) Frequency differences $\omega_{i,0} = \omega_i - \omega_0$ for the lowest energy eigenstates of Hamiltonian Eq. (1) as a function of ω_c/ω_q . Here, we consider a normalized coupling rate $\lambda/\omega_q = 0.1$ between the resonator and each of the qubits. We used $\theta = \pi/6$. The black arrows indicate the ordinary vacuum splitting arising from the coupling between the states $|g, g, 1\rangle$ and $(1/\sqrt{2})(|g, e, 0\rangle + |e, g, 0\rangle)$. (b) Enlarged view of the spectral region delimited by a square in (a). This shows an avoided-level crossing, demonstrating the coupling between the states $|g, g, 1\rangle$ and $|e, e, 0\rangle$ due to the presence of counterrotating terms in the system Hamiltonian.

vacuum Rabi splitting, which appears also when neglecting the counterrotating terms. The straight line at $E/\omega_q = 1$ corresponds to the dark antisymmetric state $(|g, e, 0\rangle - |e, g, 0\rangle)/\sqrt{2}$. Even larger splitting anticrossings around $\omega_c/\omega_q = 1$ can be observed at higher E values. These correspond to the second and third rung of the Jaynes-Cummings ladder. We are interested in the region around $\omega_c/\omega_q = 2$, where the levels three and four display an apparent crossing at $E/\omega_q \approx 2$. Actually, what appears as a crossing on this scale turns out to be a splitting anticrossing on an enlarged view as in Fig. 2(b). Observing that just outside this avoided-crossing region one level remains flat as a function of ω_c with energy $\omega \approx 2\omega_q$, while the other grows as ω_c , this splitting clearly originates from the hybridization of the states $|e, e, 0\rangle$ and $|g, g, 1\rangle$. The resulting states are well approximated by the states $(|e, e, 0\rangle \pm |g, g, 1\rangle)/\sqrt{2}$. This splitting is not present in the rotating-wave approximation (RWA), where the coherent coupling between states with a different number of excitations is not allowed, nor does it occur in the absence of symmetry breaking ($\theta = 0$). The normalized splitting has a value $2\Omega_{\text{eff}}/\omega_q = 1.97 \times 10^{-3}$, which is in good agreement with 2×10^{-3} , obtained within perturbation theory. This observed hybridization opens the way to the observation of weird effects such as the simultaneous excitations of *two* qubits with only *one* cavity photon. Such a coupling between the states $|e, e, 0\rangle$ and $|g, g, 1\rangle$ can be analytically described by the effective interaction Hamiltonian $H_{\text{eff}} = -\Omega_{\text{eff}}(|e, e, 0\rangle\langle g, g, 1| + \text{H.c.})$.

A key theoretical issue of the USC regime is the distinction between bare (unobservable) excitations and physical particles that can be detected [28,40]. For example, when the counterrotating terms are taken into account, the mean photon number in the system ground state becomes different from zero: $\langle 0|\hat{a}^\dagger\hat{a}|0\rangle \neq 0$. However, these photons are actually virtual [40] because they do not correspond to real particles that can be detected in a photon-counting experiment. The same problem holds for the excited states. According to these analyses, the presence of an n -photon contribution in a specific eigenstate of the system does not imply that the system can emit n photons when prepared in this state.

In order to fully understand and characterize this anomalous avoided crossing not present in the RWA, a more quantitative analysis is required. In the following, we therefore calculate the output signals and correlations which can be measured in a photodetection experiment. We fix the cavity frequency at the value where the splitting between level 3 and 4 is minimum. Instead of starting from the ideal initial state $(|3\rangle - |4\rangle)/\sqrt{2} \approx |g, g, 1\rangle$, more realistically, we consider the system initially in its ground state $|0\rangle \approx |g, g, 0\rangle$ and study the direct excitation of the cavity by an electromagnetic Gaussian pulse with central frequency $\omega_d = (\omega_{4,0} + \omega_{3,0})/2$. In this strongly dispersive regime, the resonator displays very low anharmonicity, so

that for a strong system excitation such as that induced by a π pulse, higher-energy states of the resonator (as the state $|8\rangle \approx |g, g, 2\rangle$) can be resonantly populated. This problem can be avoided by feeding the system with a single-photon input or by probing the system in the weak-excitation regime. However, in order to achieve a deterministic transition $|g, g, 1\rangle \rightarrow |e, e, 0\rangle$, a useful route involves introducing a Kerr nonlinearity into the resonator, able to activate a photon blockade. In circuit QED this can be realized by introducing some additional Josephson junction, or coupling the resonator with weakly detuned artificial atoms [41]. This additional nonlinearity can be described by the Hamiltonian term $\hat{H}_K = \mu\hat{a}^{\dagger 2}\hat{a}^2$. The driving Hamiltonian, describing the system excitation by a coherent electromagnetic pulse, is $\hat{H}_d(t) = \mathcal{E}(t)\cos(\omega t)\hat{X}$, where $\mathcal{E}(t) = A \exp[-(t-t_0)^2/(2\tau^2)]/(\tau\sqrt{2\pi})$. Here, A and τ are the amplitude and the standard deviation of the Gaussian pulse, respectively. A includes the factor $\sqrt{\kappa}$, where κ is the loss rate through the cavity port. The system is thus under the influence of the total Hamiltonian $\hat{H} = \hat{H}_0 + \hat{H}_K + \hat{H}_d(t)$.

The output photon flux emitted by a resonator can be expressed as $\Phi_{\text{out}} = \kappa\langle\hat{X}^-\hat{X}^+\rangle$, where $\hat{X}^+ = \sum_{j,k>j} X_{jk}|j\rangle\langle k|$ and $\hat{X}^- = (\hat{X}^+)^\dagger$, with $X_{jk} \equiv \langle j|(\hat{a}^\dagger + \hat{a})|k\rangle$, are the positive and negative frequency cavity-photon operators [30,36]. Neglecting the counterrotating terms, or in the limit of negligible coupling rates, they coincide with \hat{a} and \hat{a}^\dagger , respectively. The signal directly emitted from the qubit is proportional to the qubit mean excitation number $\langle\hat{C}^-\hat{C}^+\rangle$, where \hat{C}^\pm are the qubit positive and negative frequency operators, defined as $\hat{C}^+ = \sum_{j,k>j} C_{jk}|j\rangle\langle k|$ and $\hat{C}^- = (\hat{C}^+)^\dagger$, with $C_{jk} \equiv \langle j|(\hat{\sigma}_- + \hat{\sigma}_+)|k\rangle$ [30,36]. Neglecting the counterrotating terms, or in the limit of negligible coupling rates, they coincide with $\hat{\sigma}_-$ and $\hat{\sigma}_+$, respectively. In circuit QED systems, this emission can be detected by coupling the qubit to an additional microwave antenna [8].

Thanks to the photon-blockade effect, induced by the Kerr interaction \hat{H}_K , it is possible to resonantly excite the split states $|3\rangle$ and $|4\rangle$ with a π pulse, so that after the pulse arrival the population is completely transferred from the ground state to only these two energy levels. We use a pulse width $\tau = 1/(4\omega_{43})$. Figure 3(a) displays the numerically calculated dynamics of the photon number $\langle\hat{X}^-\hat{X}^+\rangle$, of the mean excitation number $\langle\hat{C}_1^-\hat{C}_1^+\rangle$ for qubit 1 (which, of course, coincides with that of qubit 2), and of the two-qubit correlation $G_q^{(2)} \equiv \langle\hat{C}_1^-\hat{C}_2^-\hat{C}_2^+\hat{C}_1^+\rangle$. Vacuum Rabi oscillations showing the reversible excitation exchange between the qubits and the resonator are clearly visible. We observe that, after a half Rabi period, $\Omega_{\text{eff}}t = \pi/2$, the excitation is fully transferred to the two qubits which reach an excitation probability approaching one. Hence, not only the multiatom absorption of a single photon is possible, but it can essentially be deterministic. We observe that the

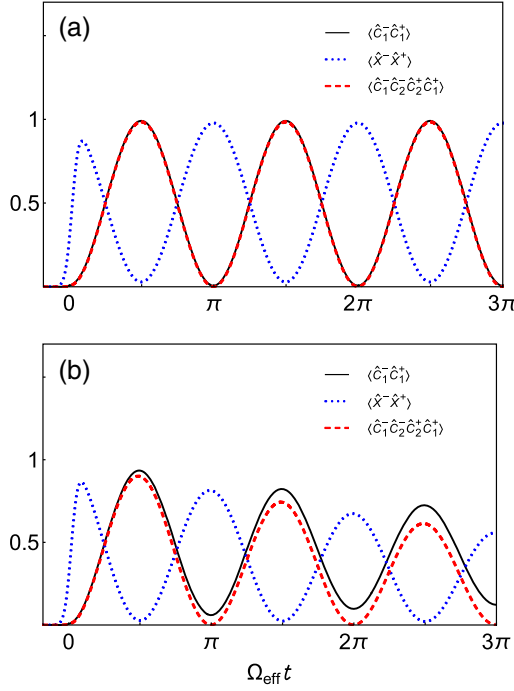


FIG. 3. (a) Time evolution of the cavity mean photon number $\langle \hat{X}^- \hat{X}^+ \rangle$ (dotted blue curve), qubit 1 mean excitation number $\langle \hat{C}_1^- \hat{C}_1^+ \rangle$ (continuous black curve), and the zero-delay two-qubit correlation function $G_q^{(2)} = \langle \hat{C}_1^- \hat{C}_2^- \hat{C}_2^+ \hat{C}_1^+ \rangle$ (dashed red curve) after the arrival of a π -like Gaussian pulse initially exciting the resonator. After the arrival of the pulse, the system undergoes vacuum Rabi oscillations showing the reversible joint absorption and reemission of one photon by two qubits. $\langle \hat{C}_1^- \hat{C}_1^+ \rangle$ and $G_q^{(2)}(t)$ are almost coincident. This perfect two-qubit correlation is a signature that the two qubits are jointly excited. (b) Time evolution of the cavity mean photon number (dotted blue curve), the qubit mean excitation number, and the two-qubit correlation as in (a), but including the effect of cavity damping and atomic decay. The corresponding rates are $\kappa = \gamma = 4 \times 10^{-5} \omega_q$.

single-qubit excitation $\langle \hat{C}_i^- \hat{C}_i^+ \rangle$ and $G_q^{(2)}$ almost coincide at any time. This almost-perfect two-qubit correlation is a clear signature of the joint excitation: if one qubit gets excited, the probability that the other one is also excited is very close to one. In summary, an electromagnetic pulse is able, thanks to the photon-blockade effect, to generate a *single* cavity photon, which then gets jointly absorbed by a *couple* of qubits. The resonant coupling can be stopped at this time, e.g., by changing the resonance frequency of the qubits. If not, the reverse process starts, where two qubits jointly emit a *single* photon: $|e, e, 0\rangle \rightarrow |g, g, 1\rangle$. We observe that $\langle \hat{X}^- \hat{X}^+ \rangle$ is not exactly zero at the photon minima. This occurs because the two-qubit excited state, owing to the same processes inducing its coupling with the one-photon state, acquires a dipole transition matrix element, so that this state is able to emit photons. We find that this effects increases when increasing the atom-field coupling strength λ (see Fig. S6 of Supplemental Material

[37]). In order to exclude that this joint qubit excitation does not occur via more conventional paths, involving the creation of photon pairs and/or a one-qubit-one-photon excitation, we have also calculated the photonic second-order correlation function $G_c^{(2)} \equiv \langle (\hat{X}^-)^2 (\hat{X}^+)^2 \rangle$ and the qubit-cavity correlation $G_{qc}^{(2)} \equiv \langle \hat{C}_i^- \hat{X}^- \hat{X}^+ \hat{C}_i^+ \rangle$. We find that their value is more than 2 orders of magnitude lower than that of the two-qubit correlation $G_q^{(2)}$. Calculations have been performed considering two-level atoms [see Eq. (1)]. Although flux qubits (generally employed to realize the USC regime with individual atoms) display very high anharmonicity (see, e.g., Ref. [19]), it is interesting to investigate if significant competing effects, lowering the correlation, can arise from the presence of additional atomic transitions. This analysis is carried out in Sec. IV of Supplemental Material [37].

Figure 3(a) has been obtained without including loss effects. The influence of cavity field damping and atomic decay on the process can be studied by the master equation approach. We consider the system interacting with zero-temperature baths. By using the Born-Markov approximation without the post-trace RWA [35], the resulting master equation for the reduced density matrix of the system is

$$\dot{\hat{\rho}} = i[\hat{\rho}(t), \hat{H}] + \kappa \mathcal{D}[\hat{X}^+] \hat{\rho} + \gamma \sum_i \mathcal{D}[\hat{C}_i^+] \hat{\rho}, \quad (2)$$

where the superoperator \mathcal{D} is defined as $\mathcal{D}[\hat{O}] \hat{\rho} = \frac{1}{2} (2\hat{O} \hat{\rho} \hat{O}^\dagger - \hat{\rho} \hat{O}^\dagger \hat{O} - \hat{O}^\dagger \hat{O} \hat{\rho})$. We use $\kappa = \gamma = 3 \times 10^{-5} \omega_q$. Figure 3(b) shows how the cavity losses and the atomic decay affect the system dynamics. As expected, the vacuum Rabi oscillations undergo damping and, as expected, the two-qubit correlation is more fragile to losses. Finally, we have also considered the case of nonidentical qubits. We find that also in this case, for qubit transition frequencies such that $\omega_{q1} + \omega_{q2} \simeq \omega_c$, it results $\langle \hat{C}_1^- \hat{C}_1^+ \rangle = \langle \hat{C}_2^- \hat{C}_2^+ \rangle \simeq G_q^{(2)}$ (see Fig. S9 in Supplemental Material [37]). This result further confirms the simultaneous and joint nature of this multiatom process.

The processes described here can be observed by placing two superconducting artificial atoms at opposite ends of a superconducting transmission line resonator [42]. These multiatom excitation and emission processes can find useful applications for the development of novel quantum technologies. Conditional quantum-state transfer is a first possible application: the quantum information stored in one of the two qubits can be transferred to the resonator conditioned by the state of the second qubit. We also observe that the quantum Rabi oscillations displayed in Fig. 3 imply that a hybrid entangled Greenberger-Horne-Zeilinger (GHZ) state, $(|g, g, 1\rangle + |e, e, 0\rangle)/\sqrt{2}$, can be obtained by an elementary quantum Rabi process after a time $t = \pi/(4\Omega_{\text{eff}})$. This state can be stored by just changing the transition frequency of one of the two qubits. Besides possible applications, the puzzling results

presented here, showing that *one* photon can divide its energy into *two* spatially separated atoms, and that vacuum fluctuations [43] can induce separate atoms to behave as a single quantum entity (as testified by the one-photon transition matrix element acquired by the transition $|g, g\rangle \rightarrow |e, e\rangle$), provide new insights into the quantum aspects of the interaction between light and matter.

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