Mixing of coherent waves on a single three-level artificial atom

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We report coherent frequency conversion in the gigahertz range via three-wave mixing on a single artificial atom in open space. All frequencies involved are in vicinity of transition frequencies of the three-level atom. A cyclic configuration of levels is therefore essential, which we have realised with an artificial atom based on the flux qubit geometry. The atom is continuously driven at two transition frequencies and we directly measure the coherent emission at the sum or difference frequency. Our approach enables coherent conversion of the incoming fields into the coherent emission at a designed frequency in prospective devices of quantum electronics.

For a long time research in experimental quantum optics focused on studying ensembles of natural atoms [1, 2]. However, there have been huge advances in performing analogous quantum optics experiments using other systems [3–5]. In particular, superconducting artificial atoms are remarkably attractive to study quantum optics phenomena. The artificial atoms are nano-scale electronic circuits that can be fabricated using well established techniques and can therefore be easily scaled up to larger systems. Their energy levels can be engineered as desired, and strong coupling can be achieved with resonators and transmission lines [6–9]. This greater control of parameters allows one to reproduce quantum optics phenomena with improved clarity or even reach regimes, that are unattainable with natural atoms. For instance coherent population trapping [10], electromagnetically induced transparency [11, 12], Autlers-Townes splitting [13–17], and quantum wave mixing [18] have been experimentally observed in superconducting three-level systems [19–23]. Moreover, three-level atoms can be used to cool quantum systems [24, 25], amplify microwave signals [26] and generate single or entangled pairs of photons [27] – important applications for future quantum networks. Here we investigate three-wave mixing, a nonlinear optical effect that can occur in cyclic three-level atoms, which are lacking in nature [28], but can easily be realised with superconducting artificial atoms. The only suitable natural systems for the three-wave mixing are chiral molecular three-level systems without inversion symmetry [29]. However, these systems cannot be tuned in frequency. Different to Josephson junction based parametric three-wave mixing devices [30], that rely on mixing on a classical non-linearity, we implement here another method to generate three-wave mixing using a single cyclic or Δ-type artificial atom. This was considered theoretically in references [28, 31].

We directly measure the coherent emission of the cyclic three-level atom under two external drives corresponding to two atomic transitions. The emission occurs at a single mixed frequency (sum or difference). This emission is a corollary of coherent frequency conversion but inherently differs from classical frequency conversion [32, 33] which would result in sidebands at the sum and difference frequencies. Previously, coherent atomic excitations using two frequencies have been studied in a single dc-SQUID phase qubit circuit with two internal degrees of freedom [34]. However, in this work, we realise coherent frequency conversion with a cyclic artificial atom in open space, which offers some advantages over placing it in a cavity. In particular, it allows to directly detect the coherent (elastic) component of the emitted field at sum or difference frequencies of the artificial atom [35]. This work establishes innovative quantum electronics that enables three-wave mixing, and coherent frequency conversion.

Our device consists of a superconducting loop (∼10μm²) interrupted by four Josephson junctions. This geometry is based on the flux qubit [36] where one of the Josephson junctions, the α-junction, has a reduced geometrical overlap by a factor of α. It is capacitively coupled to a 1D transmission line via an interdigitated capacitance of C = 6 fF (see Fig 1(a)) resulting into a photon rate in the range from several MHz to a few tens of MHz depending on frequency. The device parameters (Josephson energy EJ/h = 65 GHz, charging energy (E_C = e^2/2C) E_C/h = 19 GHz, and α = 0.45) have been chosen such that the three lowest transition frequencies fall into the frequency measurement band of our experimental setup. The coupling to the transmission line is strong enough so that non-radiative atom relaxation is negligible and hence the majority of photons from the atom are emitted into the transmission line. The device was fabricated by means of electron-beam lithography and shadow evaporation technique with controllable oxidation.

The transition frequencies, ω_{12}, ω_{23}, and ω_{13} are controlled by the external magnetic flux threaded through the loop, Φ = Φ_0/2 + δΦ, where Φ_0 is the flux quan-
tum and $\delta \Phi$ is the detuning from the energy degeneracy point of the artificial atom. The atomic transition energies are found by performing transmission spectroscopy using a vector network analyser (VNA). We sweep the frequency of a probe microwave against the flux bias $\Phi$ is the detuning from the energy degeneracy point $\delta \omega_{31} = \omega_{31} - \omega_{32} = \delta \omega_{32}$ (Fig. 2(a)) and emission at $|2\rangle \rightarrow |1\rangle$ is measured. Here $\delta \omega_{ij}$ are small detunings from their corresponding atomic transition frequencies $\omega_{ij} = \omega_i - \omega_j$ with $i > j$. In the rotating wave approximation of the semi-classical picture, the three-level artificial atom under two drives $\omega_{31}^{d}$, $\omega_{32}^{d}$ coupling the atomic states through the dipole interaction $\hbar \Omega_{ij} = \phi_{ij} V_{ij}$, with $\phi_{ij}$ the atomic dipole moment, is described by the Hamiltonian

$$H = -\hbar \omega_{31} \sigma_1 + \delta \omega_{32} \sigma_2 - \hbar \left[ \frac{\Omega_{12}^2}{2} (\sigma_{13} + \sigma_{31}) + \frac{\Omega_{23}^2}{2} (\sigma_{32} + \sigma_{23}) \right].$$

where $\sigma_{ij} = |i\rangle \langle j|$ is the transition operator. The dynamics of the system are governed by the Markovian master equation.

The atom interacting with 1D open space emits a coherent wave [8, 35]

$$V_{ji}^{\text{em}}(x,t) = \frac{i}{\hbar} \frac{\Gamma_{ji}}{\phi_{ji}} \langle \sigma_{ij} \rangle e^{i(k_{ji}x) - \omega_{ji}t}$$

where $\langle \sigma_{ij} \rangle = \rho_{ji}$ is found from the stationary solution ($\dot{\rho} = 0$) of the master equation. The spectral density
shown above Fig. 2(d). The narrow peak position frequency $\omega_P$ is expected to be due to level splitting induced by driving fields. This is restrained by the transition frequencies of the cyclic Hamiltonian $\mathcal{H}$, for varying values of $\Omega_{ij}$, while keeping $\Omega_{23}$ constant. Splitting of the coherent emission under large driving amplitude $\Omega_{ij}$ is observed which appears due to level splitting induced by driving fields. This splitting is investigated further by recording the coherent emission versus detuning of the two drives for various combinations of powers. As seen in Fig. 3(a), the direction of the splitting is determined by the stronger drive: $\Omega_{13} >> \Omega_{23}$ leads to $\Omega_{13}$ splitting level [1]; $\Omega_{23} >> \Omega_{13}$ leads to $\Omega_{23}$ splitting level [2] and the splitting pattern in the coherent emission is turned by 90 degrees.

In an analogous way, we pump transitions between states [3] and [1] with driving frequency $\omega_{23}^d = \omega_{31} + \delta \omega_{31}$ and transitions between states [2] and [1] with driving frequency $\omega_{21}^d = \omega_{21} + \delta \omega_{21}$ (Fig. 2(b)) resulting in the Hamiltonian

$$H = -\hbar(\delta \omega_{21}\sigma_{22} + \delta \omega_{31}\sigma_{33}) - \hbar\left[\frac{\Omega_{12}}{2}(\sigma_{12} + \sigma_{21}) + \frac{\Omega_{13}}{2}(\sigma_{32} + \sigma_{23})\right].$$

In this pumping scheme, the emission power of the coherent emission of transitions between states [3] and [2], $\nu_{32}^c$, is extracted and a narrow peak in the power spectrum at $\omega_{32}/2\pi = 8.35$ GHz is recorded. The coherent emission between states [3] and [2] is monitored as a function of detuning of the drives, $\delta \omega_{13}^d$ and $\delta \omega_{21}^d$, for several combinations of driving amplitudes, $\Omega_{13}$ and $\Omega_{12}$, Fig. 3(b), the result being more complex than in the previous driving configuration. It becomes

$$P(\omega) = \frac{\hbar \omega T_{ji}}{2} |\langle \sigma_{ij} \rangle|^2,$$

where $P = |V_{jm}^{em}|^2/2\omega_0$. Here $\omega$ is in the vicinity of the transition frequency $\omega_{21}/2\pi = 6.48$ GHz as schematically shown above Fig. 2(d). The narrow peak power $P(\omega)$ can be measured by a spectrum analyser or any equivalent methods (see Supplementary Methods). Further we refer to the Voltage amplitude $V_{jm}^{em}$, which can be extracted from $P(\omega)$, as the coherent emission. The linewidth of the emission peak is as narrow as the linewidths of the generator emission that is driving the artificial atom, in-
apparent that the coherent emission from the atom depends on all relaxation and dephasing rates. The bright coherent emission line stretching diagonally from the bottom left to the top right corner in Fig. 3(b) is primarily determined by dephasing on the \( |3 \rangle \rightarrow |1 \rangle \), \( \nu_{31}^{\text{em}} \), with Rabi frequencies corresponding to the respective field strengths \( \Omega_{13}, \Omega_{23} \). The vertical coherent emission line that appears for some transitions between states \( |1 \rangle \) and \( |2 \rangle \) is primarily determined by dephasing on the \( |1 \rangle \rightarrow |2 \rangle \) transition, \( \gamma_{23} \). Emission lines broaden when the two driving frequencies are comparable to each other and larger than their dephasing rates. The dependency of the parameters on the emission line was developed from experimental data and numerical simulations. In contrast to the experiment, all input parameters of the numerical simulations can be varied independently.

To achieve coherent frequency upconversion we pump transitions between states \( |2 \rangle \) and \( |1 \rangle \) with driving frequency \( \omega_{21}^{d} = \omega_{21} + \delta \omega_{21} \) and transitions between states \( |3 \rangle \) and \( |2 \rangle \) with driving frequency \( \omega_{32}^{d} = \omega_{32} + \delta \omega_{32} \), see Fig. 2(c). The Hamiltonian for this configuration is

\[
H = -\hbar (\delta \omega_{21} \sigma_{11} + \delta \omega_{32} \sigma_{33}) \\
- \hbar \left[ \Omega_{12}^{2} (\sigma_{12} + \sigma_{21}) + \frac{\Omega_{23}}{2} (\sigma_{32} + \sigma_{23}) \right].
\]

As expected, we observe a single narrow coherent emission peak in the emission power spectrum only at the sum frequency \( \omega_{13}/2\pi + \omega_{23}/2\pi = 14.83 \) GHz, but not at the difference frequency \( \omega_{13} - \omega_{23} \), confirming that our results cannot be explained by mixing with a classical nonlinearity. Similar to the previous pumping configurations, the coherent emission peak is split under a strong driving amplitude. Fig. 3 (c) shows the behaviour of the coherent emission \( V_{13}^{\text{em}} \) expressed as photon rate \( \nu_{31} \)

Finally, we numerically simulate our experimental results using the master-equation formalism with the Lindblad term

\[
L[\rho] = (\Gamma_{31} \rho_{33} + \Gamma_{21} \rho_{22}) \sigma_{11} + (\Gamma_{32} \rho_{33} - \Gamma_{21} \rho_{22}) \sigma_{22} \\
- (\Gamma_{31} \rho_{33} + \Gamma_{23} \rho_{22}) \sigma_{33} - \sum_{i \neq j} \gamma_{ij} \rho_{ij} \sigma_{ij}.
\]

Here \( \gamma_{ij} = \gamma_{ji} \) is the damping rate of the off-diagonal terms (dephasing) and \( \Gamma_{ij} \) is the relaxation rate between the levels \( |i \rangle \) and \( |j \rangle \). In the numerical simulations dephasing and relaxation rates are arbitrary numbers. The constraints are that dephasing and relaxation rates are fixed by our sample (the three-level atom) throughout the experiment, and the input driving powers are varied but known (we set them at the generators). By finding the correspondence between the simulations, Fig. 4, and our measurement results, Fig. 3, we extract \( \Gamma_{21}/2\pi = 8 \) MHz, \( \gamma_{21}/2\pi = 8 \) MHz, \( \Gamma_{32}/2\pi = 38 \) MHz, \( \gamma_{32}/2\pi = 42 \) MHz, \( \Gamma_{31}/2\pi = 41 \) MHz, and \( \gamma_{31}/2\pi = 39.5 \) MHz agreeing with our expectations.

Since our measurement set-up has not been pre-calibrated, experimental results include gain and attenuation coefficients and are therefore presented in arbitrary units. Comparing the experimental results with the numerical simulations would yield a calibration of our output line. This calibration depends on frequency and was not the focus of this work. Nevertheless, we obtain a calibration factor of our output line of \( G_{21} = 2 \times 10^{5} \), \( G_{31} = 1.3 \times 10^{5} \), \( G_{31} = 10^{5} \) for each of the three transition frequencies, 6.48 GHz, 8.35 GHz, 14.83 GHz respectively. The visible difference between our experimental measurement results, Fig. 3, and the numerical simulations, Fig. 4, are noise in the experiment.

In conclusion, we have demonstrated three-wave mixing and coherent frequency conversion using a single cyclic three-level artificial atom. The fundamental difference from classical Josephson junction based parametric three-wave mixing devices [30] is that here transition frequencies of the artificial atom are mixed to generate a single coherent emission peak at the sum or difference frequency. A requirement for this phenomena to occur is a cyclic-type atom, which is absent in nature due to electric-dipole selection rules, but can easily be realised with superconducting artificial atoms. Thus we suggest a
unique method of generating coherent fields at a designed frequency by mixing on the single artificial atom.

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