Regular Article

Photon pair production by STIRAP in ultrastrongly coupled matter-radiation systems

A. Ridolfo^{1,2,3}, G. Falci^{1,3,4,a}, F.M.D. Pellegrino^{1,3}, G.D. Maccarrone¹, and E. Paladino^{1,3,4}

¹ Dipartimento di Fisica e Astronomia "Ettore Majorana", Università di Catania, Via Santa Sofia 64, Catania (I), Italy

² RIKEN, Saitama 351-0198, Japan

³ Istituto Nazionale di Fisica Nucleare, Via Santa Sofia 64, Catania (I), Italy

⁴ CNR-IMM Catania Universitá (MATIS), Via Santa Sofia 64, Catania (I), Italy

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Abstract. Artificial atoms (AAs) offer the possibility to design physical systems implementing new regimes of ultrastrong coupling (USC) between radiation and matter [C. Ciuti et al., Phys. Rev. B **72**, 115303 (2005)], where previously unexplored non-perturbative physics emerges. While experiments so far provided only spectroscopic evidence of USC, we propose the dynamical detection of virtual photon pairs in the dressed eigenstates, which is a "smoking gun" of the very existence of USC in nature. We show how to coherently amplify this channel to reach 100% efficiency by operating advanced control similar to stimulated Raman adiabatic passage (STIRAP) [N.V. Vitanov et al., Rev. Mod. Phys. **89**, 015006 (2017)].

1 Introduction

Since 1983 new fundamental phenomena have been demonstrated in systems of twolevel atoms coupled to a high-Q cavity [3]. In the simplest cases, the strong coupling (SC) regime is described by the quantum Rabi model [4,5]

$$H_R = \varepsilon |e\rangle \langle e| + \omega_c a^{\dagger} a + g \left[a|e\rangle \langle g| + a^{\dagger}|g\rangle \langle e| \right] + g \left[a^{\dagger}|e\rangle \langle g| + a|g\rangle \langle e| \right].$$
(1)

Here, $\{|g\rangle, |e\rangle\}$ are the eigenstates of the two-level atom, ϵ being their energy splitting, and the energy of a quantum in the cavity is $\omega_c \sim \epsilon$. In the SC regime the coupling constant g between the atomic dipole and the quantized electric field, $\propto a^{\dagger} + a$ is larger than both the decay rate of the atom, γ , and that of the cavity, κ . In the last decade, this model has been implemented in architectures of artificial atoms (AAs), such as quantum dots and superconducting Josephson "circuit-QED" systems [6–8]. In the SC regime g/ω_c is small, typically $\sim 10^{-6} - 10^{-2}$ depending on the implementations, and the "counterrotating" term in the Hamiltonian equation (1) can be neglected,

^a e-mail: gfalci@dmfci.unict.it

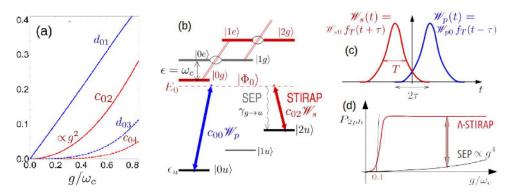


Fig. 1. (a) Amplitudes c_{02} and $d_{1\pm,2}$ which are nonvanishing for eigenstates $|\Phi_0\rangle$ and $|\Phi_{1\pm}\rangle$ in the USC regime. (b) Scheme of a three-level system coupled to an oscillator in the USC regime: population is pumped from $|0 u\rangle$ to the dressed ground state $|\Phi_0\rangle$, virtual photons pairs being converted to real photons by spontaneous emission or by STIRAP-induced coherent population transfer to $|2 u\rangle$. The pump pulse is resonant with the $|0u\rangle \leftrightarrow |\Phi_0\rangle$ transition, $\omega_p = E_0 - \epsilon_u$, and the Stokes pulse with the $|\Phi_0\rangle \leftrightarrow |2u\rangle$ transition, $\omega_s = E_0 - (\epsilon_u + 2\omega_c)$. (c) STIRAP is operated by shining the Stokes pulse (red) before the pump pulse (blue); here $\mathscr{W}_k(t)$ for k = p, s are the pulse envelopes and we used a Gaussian $f_T(t)$. (d) STIRAP amplifies coherently the photon pair production channel, up to 100% efficiency.

yielding the Jaynes–Cummings (JC) model [8]. In this limit, the dynamics is described in terms of single excitations coherently exchanged between atom and cavity, thus making circuit-QED systems fundamental building blocks for the design of quantum architectures [9].

AAs offer the possibility to enter the new "ultrastrong coupling" (USC) regime, $g \sim \omega_c, \epsilon$ [1,8,10–13,16,17] where several new features appear. For instance while the JC ground state is factorized, $|n = 0\rangle \otimes |g\rangle$, in the USC regime it contains virtual photons [19,20], $|\Phi_0\rangle = \sum_{n=0}^{\infty} c_{0n} |ng\rangle + d_{0n} |ne\rangle$ with vanishing c_{0n} for odd number of photons n and vanishing d_{0n} for even n (see Fig. 1a). Dynamics in the USC regime is expected to display several new phenomena [1,8,18,19,21], which however have not been observed so far, experiments being limited to spectroscopy. Therefore, demonstrating dynamics in the USC regime remains an outstanding challenge. In this work, we will analyze an advanced control protocol allowing to detect dynamically virtual photons in the ground state [22,23]. Implementing such a protocol would be an important proof of principle of controlled dynamics in the USC regime.

2 Dynamical detection of USC

It has been proposed to detect ground-state photons by converting them into real ones letting them decay to an ancillary uncoupled atomic level $|u\rangle$ [24], a channel active if and only if $c_{02} \neq 0$, marking thus USC. The Hamiltonian of this system is $H = \epsilon_u |u\rangle \langle u| + \omega_c a^{\dagger} a \otimes |u\rangle \langle u| + H_R$. For moderate g/ω_c this process, which is called stimulated emission pumping (SEP) [2], has a low yield, as it depends on $|c_{02}|^2 \propto g^4$ [23]. We can amplify this pair production channel by driving the system coherently with a two-tone field [22], $W(t) = \mathscr{W}_s(t) \cos(\omega_s t) + \mathscr{W}_p(t) \cos(\omega_p t)$ acting on the atom, $H_c(t) = W(t) [|u\rangle \langle g| + |g\rangle \langle u|]$, which implements a Λ configuration (see Fig. 1b) for suitable choice of the frequencies ω_k . For not too large $g \lesssim \omega_c = \epsilon$, the dynamics is captured by a simplified three-level model [22] described by the following

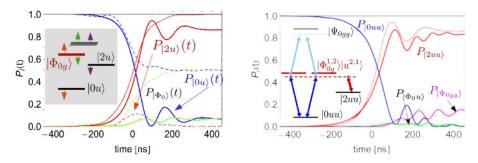


Fig. 2. Population histories for systems up to 40 levels, with pulses coupled to all the transitions. Paramenters: $\epsilon = \omega_c$ and $\epsilon_u = -4\omega_c$; pulse amplitudes and time scales are suited for superconducting qubits [11,13,23]. (a) Single atom with $g/\omega_c = 0.2$. Dynamical Stark shifts suppressing population transfer in a three-level system (dashed line) are partially self-compensated in a multilevel structure (full thick lines) and can be eliminated by properly crafted pulses (thin line). Inset: Stokes-induced Stark shifts of different sign tend to cancel in multilevel structures. (b) Coherent population transfer for two-atoms Rabi model with $g/\omega_c = 0.2/\sqrt{2}$ (thick lines) is qualitatively equivalent to population transfer for a single atom with coupling $g/\omega_c = 0.2$ (thin line), yielding already $\gtrsim 90\%$ efficiency with no phase control. In the inset: level scheme of the effective Hamiltonian H_5 ; the pump pulse may address unwanted transitions to the dressed $|0gg\rangle$ state.

Hamiltonian in the rotating frame

$$H_3(t) = \left[\frac{\mathscr{W}_p(t)}{2} |0u\rangle \langle \Phi_0| + \frac{c_{02}(g)\mathscr{W}_s(t)}{2} |2u\rangle \langle \Phi_0|\right] + \text{h.c.}$$
(2)

Shining the two pulses in the "counterintuitive" sequence (the Stokes before the pump, Fig. 1c), this Hamiltonian implements STIRAP [2] which yields complete and faithful population transfer $|0u\rangle \rightarrow |2u\rangle$ via the intermediate virtual state $|\Phi_0\rangle$. The necessary conditions are that $c_{02}(g) \neq 0$ and that $\max[\mathscr{W}_k(t)]$ are large enough to guarantee adiabatic dynamics, ~100% efficiency being attainable for $g/\omega_c \gtrsim 0.2$ in typical devices (see Fig. 1d). The implementation of Λ -STIRAP in superconducting AAs has been proposed in the last decade [25–28], demonstrated only recently [29,30], and may be a promising tool for processing in complex architectures [31,32]. In our case, detection of a two-photon state in the cavity is a "smoking gun" for USC, STIRAP providing coherent amplification of the USC pair production channel.

2.1 Advanced dynamics in multilevel systems

The simple Hamiltonian equation (2) is valid for small enough field amplitudes \mathscr{W}_k . This is not the case if g is small since attaining adiabaticity would require a large \mathscr{W}_s . The main effect is a dynamical Stark shift $\Delta E_0 - \Delta \epsilon_u =: 2\delta(t)$ of the $|0u\rangle \leftrightarrow |\Phi_0\rangle$ splitting, which translates in stray nonvanishing detunings during the protocol. In particular it produces a two-photon detuning $\delta(t)$ comparable to $c_{02}\mathscr{W}_s$ which may suppress STIRAP (dashed curves in Fig. 2a).

Actually the three-level analysis must be generalized to account for the multilevel nature of the system [23]. Fortunately, this turns out to mitigate the detrimental effect of dynamical Stark shifts. We studied this problem by considering up to 40 levels of the composite system and a control field which in the laboratory frame has the complete structure $H_c(t) = W(t)[(|u\rangle\langle g| + (1/\eta)|g\rangle\langle e|) + h.c.]$. It describes the

experimentally relevant case of a ladder type "dipole" coupling of the field to the AA, η being the ratio between the corresponding matrix elements. Results in Figure 2a (red curves) show that experimentally detectable population transfer is also achieved in this non-ideal case. This is due to the fact that the large Stokes field also couples to transitions between higher energy levels, and produces dynamical Stark shifts with the same time dependence of the stray $\delta(t)$, which it may be partially compensated (see the inset of Fig. 2a).

Moreover, the full signal can be recovered if appropriately crafted control is used (thin red curve in Fig. 2a). One option is to use a phase modulation of the Stokes pulse, as explained in [33], designed to compensate the effect of the Stokes field coupled to the u-g transition only. The fact that this is the relevant source of noise is suggested by success of the strategy even when the drive couples to all the ladder transitions of the AA.

3 Amplification by many atoms

Using an ensemble of N atoms is expected to amplify interaction effects, yielding an effective $g \to \sqrt{Ng}$, at least for very weakly coupled atoms. The spectrum of the N atoms Rabi Hamiltonian shows this equivalence even for nonperturbative values of g. This is the mechanism for obtaining USC in semiconductors subband polaritons [10], behaving as a system with very large N, but it is also interesting to exploit the limit of few more strongly coupled atoms, which is likely to be implemented in the near future in superconducting architectures [8]. In this limit, STIRAP is operated by the control Hamiltonian $H_c = W(t) \sum_{k=1}^{N} (|u^k\rangle \langle g^k| + h.c.)$. Figure 2b shows the protocol for two identical atoms coupled to a mode with a rather small $g/\omega_c = 0.2/\sqrt{2}$ compared with the result for a single atom with $g/\omega_c = 0.2$, showing the expected qualitative agreement. To illustrate the main features we analyze the simple situation of resonant and not too strong drives. In this limit, the relevant dynamics reduces to five levels (see inset in Fig. 2b) with Hamiltonian

$$H_{5}(t) = \left\{ \left(\frac{\mathscr{W}_{p}(t)}{2} |0uu\rangle + \frac{c_{02}(g)\mathscr{W}_{s}(t)}{2} |2uu\rangle \right) \left[\langle \Phi_{0}^{1} | \langle u^{2} | + \langle \Phi_{0}^{2} | \langle u^{1} | \right] \right. \\ \left. + \frac{\mathscr{W}_{p}(t)}{2} \left[|\Phi_{0}^{1}\rangle |u^{2}\rangle + |\Phi_{0}^{2}\rangle |u^{1}\rangle \right] \langle \Psi_{0gg} | \right\} + \text{h.c.} + (E_{0gg} - 2E_{0}) |\Psi_{0gg}\rangle \langle \Psi_{0gg} |$$

$$(3)$$

where $|\Phi_0^{\rm c}\rangle$ are single-atom Rabi ground states with the k-atom, whereas $|\Psi_{0\rm gg}\rangle$ is the two-atoms Rabi ground state. It is apparent that STIRAP may occur via the entangled intermediate state $[|\Phi_0^1\rangle|u^2\rangle + |\Phi_0^2\rangle|u^1\rangle]/\sqrt{2}$ with rescaled peak Rabi frequencies. In particular the Stokes effective field strength is larger since $c_{02}(g) \rightarrow \sqrt{2}c_{02}(g)$. However, the pump field may be almost resonant with other transitions, and equation (3) contains stray linkages with a competing target state $|\Psi_{0\rm gg}\rangle$ (magenta curve in Fig. 2b), which may spoil population transfer to $|2uu\rangle$. The difference between the single and the two-atom case, emphasized in Figure 2b, is due to this effect. It turns out that both the nonvanishing $E_{0gg} - 2E_0$ due to the interaction and to the dynamical Stark shifts in the multilevel structure detune the two-photon "ladder" $|0uu\rangle \rightarrow |\Psi_{0\rm gg}\rangle$ (see inset in Fig. 2b). We mention that the stray process can be fully suppressed by using a slight detuning $\delta_p = \delta_s$ of ω_p and ω_s , which preserves the two-photon resonance for the $|0uu\rangle \rightarrow |2uu\rangle$ transition.

4 Conclusions

We have proposed a dynamical detection method of USC based on two-pulse interference, which uses an ancillary level $|u\rangle$ to convert virtual photons of the Rabi ground state to real ones [19,20,24]. By driving adiabatically the system with a STIRAP protocol this channel is amplified by coherence, and ideally guarantees 100% efficiency. Three-level AAs could be in principle implemented both in semiconductor devices, where the harmonic mode may be coupled to an intersubband transition [10] with the valence band playing the role of the ancillary $|u\rangle$, and in superconducting AAs, which are by themselves multilevel systems [8,11,13]. However, hardware may pose limitations: in semiconductors the detection of THz photons is difficult [14,15], whereas in superconducting architectures the ancillary level $|u\rangle$ used in the Λ scheme, implicit in all the proposals [22,24,34], is not really uncoupled to the cavity mode thus providing a new channel for pair production which does not require USC [23]. This problem is bypassed using a Vee-STIRAP [23] technique, which may provide a unique dynamical tool for detecting virtual photons dressing excited states [20] in available flux-based superconducting devices. Finally, the STIRAP protocol is robust against imperfections of the drives being sensitive essentially to solid-state decoherence [35] in the trapped subspace [36] which in our case is spanned by the n = 0, 2 Fock states of the cavity. Decoherence decreases the efficiency but this is not a severe problem since in the worst case it is larger than 30% [36], and the very detection of photon pairs at the end of the protocol is already a "smoking gun" for USC.

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