Stationary Inversion of a Two Level System Coupled to an Off-Resonant Cavity with Strong Dissipation

S. Hughes[1,2,*] and H. J. Carmichael[2]

[1]*Department of Physics, Queen's University, Kingston, Ontario, Canada K7L 3N6*
[2]*Department of Physics, University of Auckland, Private Bag 92019, Auckland, New Zealand*

We present an off-resonant excitation scheme that realizes pronounced stationary inversion in a two level system. The created inversion exploits a cavity-assisted two-photon resonance to enhance the multiphoton regime of nonlinear cavity QED and survives even in a semiconductor environment, where the cavity decay rate is comparable to the cavity-dot coupling rate. Exciton populations of greater than 0.75 are obtained in the presence of realistic decays and pure dephasing. Quantum trajectory simulations and master equation calculations help elucidate the underlying physics and delineate the limitations of a simplified rate equation model. Experimental signatures of inversion and multiphoton cavity QED are predicted in the fluorescence intensity and second-order correlation function measured as a function of drive power.

The term "population inversion" refers to a system in which an excited state population exceeds that of a lower level. Its use is of broad interest to laser science. Semiclassical arguments readily show that a steady-state drive, e.g., a continuous wave (cw) laser, cannot create population inversion in a two level system (TLS) because of stimulated emission; thus, standard atomic and molecular lasers typically exploit a cascaded scheme through three, four, or more levels to achieve inversion [1]. In 1988, an important limitation of the semiclassical argument was pointed out by Savage [2], who demonstrated that the multiquanta regime of an atom-based cavity QED system can achieve stationary population inversion in a TLS. Specifically, by including the effects of quantum noise, and with the pump resonant with the degenerate atom-cavity resonance, Savage [2]—and later Lindberg and Savage [3]—predicted achievable inversions of around 0.01–0.03. The unanticipated result was explained in terms of a 4-state two-quanta model of an on-resonance cavity-atom system in the bad-to-intermediate cavity regime. Lindberg and Savage [3] suggest that the inversion might be measured in high pump resonance fluorescence, as a small excess (a few %) over the saturation fluorescence intensity. To the best of our knowledge this prediction has not been verified by experiment on either atomic or semiconductor systems.

There have been a growing number of recent experiments in semiconductor cavity systems using coherent excitation, most exploiting a deliberate detuning between the cavity and quantum dot (QD). For example, Majumdar *et al.* [4] studied the coupling between a photonic crystal cavity and an off-resonant QD under coherent excitation of either the cavity or the QD exciton; they investigated exciton power broadening under coherent excitation in the presence of a detuned cavity. Ulhaq *et al.* [5] investigated linewidth broadening of a resonantly excited QD in a micropillar cavity, again with QD-cavity detuning. Both groups demonstrate significant power broadening of the target QD exciton, and, importantly, the experimental capacity to explore off-resonant QD-cavity systems using nonlinear coherent excitation in a controlled way. There have also been remarkable developments in circuit QED [6–8], where anharmonic cavity QED effects are realized at dipole coupling strengths 2–3 orders of magnitude larger than in atomic or QD systems.

In this Letter, we introduce a scheme to create pronounced population inversion in a dissipative TLS coupled to a off-resonant cavity. Supported by quantum trajectory (QT) simulations [9,10] and master equation (ME) calculations, we show that off-resonant excitation of a detuned TLS-cavity system, with intermediate to strong coupling, results in population inversions that can easily exceed 0.25. These stationary inversions are enabled through two-photon excitation of the dressed states, and, strategically, only occur for a detuned TLS and cavity. In fact, significant prototype inversions have been seen in a circuit QED device excited via so-called sideband transitions [11], using an excitation scheme similar to ours; however, the unanticipated inversion is discussed only briefly in terms of a simplified rate equation model. We use QT simulations to, first, connect to, then extrapolate beyond rate equations. We show that they have a limited range of validity, and focus on the possibility of achieving significant inversion far outside the rate equation regime. We show that such inversion exists for practical semiconductor QD systems where it is potentially important for microlasers and few photon quantum light sources. Other nonstandard routes to gain include lasing without inversion [12], but here we are dealing with conventional inversion-driven gain, although with an unfamiliar pumping scheme. We also suggest

experiments for measuring the inversion, via the fluorescence intensity measured as a function of drive power [4,5], or through the second-order correlation function; both exhibit clear signatures of the proposed off-resonant stationary inversion regime.

The cavity QED system is described through a ME for the reduced density matrix of the TLS (exciton in a semiconductor dot) and cavity. Working in the interaction picture, i.e., in a frame rotating at the pump frequency, ω_L, the ME is [13]

$$\frac{d\rho}{dt} = \frac{-i}{\hbar}[H_s, \rho] + \left(\kappa \mathcal{L}[a] + \frac{\gamma}{2}\mathcal{L}[\sigma_-] + \frac{\gamma'}{8}\mathcal{L}[\sigma_z]\right)\rho, \quad (1)$$

with system Hamiltonian

$$H_s = \hbar\delta_{xL}\sigma^+\sigma^- + \hbar\delta_{cL}a^\dagger a + \hbar g(\sigma^- a^\dagger + \sigma^+ a)$$
$$+ \hbar\eta_x(\sigma^+ + \sigma^-), \quad (2)$$

where η_x is the coherent pump rate of the TLS, g is the TLS-cavity coupling strength, a is the cavity mode annihilation operator, σ^+, σ^-, and σ_z are Pauli operators, and $\delta_{\alpha L} = \omega_\alpha - \omega_L$ are pump laser detunings; dissipation is described by the Lindblad superoperator, $\mathcal{L}[\xi]\rho = 2\xi\rho\xi^\dagger - \xi^\dagger\xi\rho - \rho\xi^\dagger\xi$, with separate terms to account for decay of the cavity mode (rate 2κ), radiative decay (rate γ), and pure dephasing (rate γ') of the TLS.

Figure 1(a) depicts the dressed quasienergy levels of Hamiltonian (2), truncated to 5 states. The lowest four levels are the main ones responsible for cavity-assisted inversion, where decay of $|1, +\rangle$ populates $|0, +\rangle$ through the emission of a cavity photon. Importantly, two-photon resonance to $|1, +\rangle$, with negligible excitation of $|1, -\rangle$, is
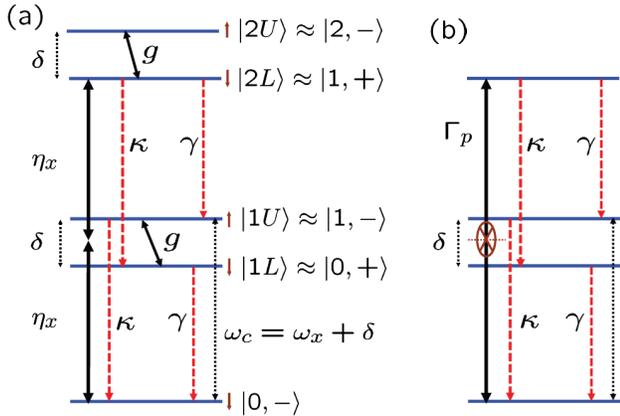

FIG. 1 (color online). (a) Quasienergy levels truncated to 5 states ($\delta \equiv \delta_{cx} = \omega_c - \omega_x$). $|1, +\rangle$ refers to one created cavity photon with one exciton (excited TLS) created; for a far-detuned cavity and exciton, this doubly excited state relates to the Jaynes-Cummings dressed states through $|2L\rangle \approx |1, +\rangle$. Bold arrows illustrate two-photon resonance, with the pump detuned from the cavity by about $-\delta/2$. Small arrows show the direction of the Stark shifts. (b) Simplified rate equation model, where $\Gamma_p \equiv \Omega_{tp}^2/2\kappa$ is an effective two-photon pump rate, with Ω_{tp} the two-photon Rabi frequency (see text).

achieved by setting $2\delta_{Lx} \approx \delta \equiv \delta_{cx} = \omega_c - \omega_x \gg g$, κ, γ, γ'. For increasing pump strengths, Stark shifts lower the resonance frequency. For comparison, a simplified *rate equation* model, with adiabatic elimination of intermediate states, is shown in Fig. 1(b).

We first derive the TLS population within the simplified model. With coherent drive, and neglecting pure dephasing, we obtain the steady-state population of the TLS excited state (population of excitons),

$$\bar{n}_x \equiv \langle\sigma^+\sigma^-\rangle_{ss} = \frac{4\Omega_{tp}^2}{(\gamma + 4\Omega_{tp}^2/2\kappa)(\gamma + 2\kappa)}, \quad (3)$$

with two-photon drive $\Omega_{tp} = (4\eta_x^2/\delta)^2/(g/\delta)^2$. Note that this expression differs from the one used by Leek *et al.* [11]; their formula, $\bar{n}_x = \Omega_{tp}/(\Omega_{tp} + \gamma/2\kappa + \gamma\Omega_{tp}/2\kappa)$, is obtained by treating Ω_{tp} as the rate of an incoherent drive.

Using QT simulations we identify a regime where the rate equation picture is approximately valid while also allowing for significant inversions. Broadly, we require $\delta/\kappa \gtrsim 10^4$, $g/\kappa \gtrsim 10^3$, and $\eta_x/\kappa \gtrsim 10^2$. Figure 2(a) shows a sample QT on the boundary of this region, with $\gamma/\kappa = 0.05$. Taking level shifts into account, the two-photon resonance is near $\delta_{xL} = -0.49g$. We highlight
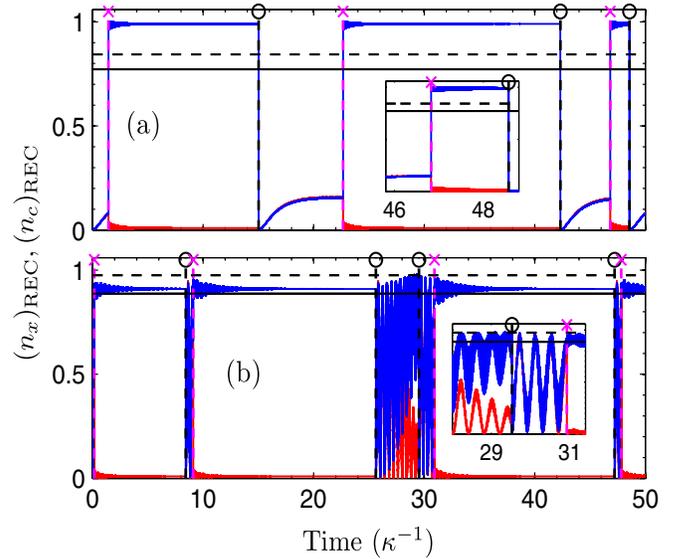

FIG. 2 (color online). Sample QTs near, (a) [$g = 1000\kappa$, $\eta_x = 100\kappa$, $\gamma = 0.05\kappa$], and outside, (b) [$g = 100\kappa$, $\eta_x = 150\kappa$, $\gamma = 0.05\kappa$], the rate equation regime. The blue (upper) solid curve plots the expectation of the TLS excited population (exciton population) conditioned upon the record of quantum jumps, while the red (lower) solid curve is the conditioned expectation of the cavity photon number. Quantum jumps are indicated by the crosses and circles (and vertical dashed lines) for photon emission via the cavity and TLS, respectively. The steady-state expectation of the exciton population is shown with the horizontal solid line, while the rate equation estimate is shown with the dashed horizontal line. The insets enlarge the regions near $t\kappa \approx 46$–48 and $t\kappa \approx 29$–31 in (a) and (b), respectively.

the following: (i) the TLS jumps to an excitation close to
unity with the cavity excitation close to zero; (ii) each such
jump is initiated by the emission of a photon by the cavity
corresponding to the transition $|1, +\rangle \rightarrow |0, +\rangle$ in Fig. 1(a);
(iii) all excitation periods terminate with the emission of a
photon by the TLS corresponding to the transition
$|0, +\rangle \rightarrow |0, -\rangle$ in Fig. 1(a); (iv) the intervals between
excitation periods (e.g., $t\kappa \approx 15\text{–}22$) see the cavity and
TLS populations grow continuously and in step, corre-
sponding to coherent excitation of the $|0, -\rangle \rightarrow |1, +\rangle$
transition. Solid and dashed horizontal lines show that
there is nevertheless a discrepancy between the exact
(solving the ME in a suitably large photon number basis
[14]) population ($\bar{n}_x \sim 0.77$) and that predicted by Eq. (3)
($\bar{n}_x \sim 0.84$); most likely this is due to our neglect of level
shifts when deriving Ω_{tp}.

We next choose parameters that start to deviate away
from the rate equation picture. Figure 2(b) is plotted for
$\delta/\kappa = 10^3$, $g/\kappa = 10^2$, and $\eta_x/\kappa = 1.5 \times 10^2$, still with
$\gamma/\kappa = 0.05$. This places us close to the circuit QED re-
gime of Leek *et al.* [11]. Although a similar jump pattern is
seen, the following differences are noticed: (i) the jump
"up" reaches an excitation probability significantly less
than unity (a small piece of $|1, U\rangle \approx |1, -\rangle$ is mixed into
the state realized after the jump); (ii) the drive strength has
been increased, which induces Rabi oscillations on the
$|0, -\rangle \rightarrow |1, +\rangle$ transition (e.g., right before $t\kappa = 31$);
(iii) the TLS and cavity do not continue indefinitely to
Rabi oscillate in step, as for $t\kappa \approx 26 - 30$, where the TLS
shows a damped evolution toward a number near 0.9, while
the cavity photon number decays to zero. This behavior
illustrates a marked difference between QTs and rate-
equation-inspired quantum jumps. The final state of the
damped Rabi oscillation is $|1, L\rangle \approx |0, +\rangle$, but this state is
reached via a coherent evolution in response to the back
action of a null measurement—the system localizes to this
state because no photon happened to be scattered through
the cavity; the two-photon resonance still assists passage to
the inverted state $|1, L\rangle \approx |0, +\rangle$, but through the absence
rather than the presence of a quantum jump.

QT simulations have clarified two key points: (a) the rate
equation model is extremely restricted even for circuit QED
parameters, and (b) significant population inversion can be
achieved when dissipation rates increase relative to the
dipole coupling. This suggests that significant inversions
might be achieved using semiconductor QD parameters.

Turning now to semiconductor QED systems, we adopt
the following scaled parameters in the strong coupling
regime: $\kappa = g/2.5$, $\gamma = g/200$, and $\gamma' = g/25$. For cur-
rent strongly coupled QD systems, g is around 0.1 meV
(≈ 24 GHz), with similar values measured in a wide range
of devices, including examples comprised of photonic
crystals [15], micropillars [16], and microdisk cavities
[17]. To connect with measurements of intensity linewidth
broadening (e.g., Refs. [4,5]), we calculate the intensity of

the exciton mode, $I_x \propto \bar{n}_x$, or the cavity mode, $I_c \propto \bar{n}_c$. We
solve the full ME, Eq. (1), in a basis that can be truncated at
an arbitrary photon or exciton state, allowing us to compute
both weak-excitation-approximation results (truncation at
one quantum) and the regime of multiquanta cavity QED.
While weak excitation (semiclassical regime) is commonly
assumed when analyzing dissipative semiconductor cavity
systems, we will show that the approximation drastically
breaks down in the proposed excitation regime.

We display in Figs. 3(a) and 3(b) the exciton population
as a function of laser-exciton detuning, with $\delta = 10g$, for
two different photon-number truncations. All calculations
predict substantial power broadening and show the ex-
pected peaks at the bare cavity and exciton resonances,
in agreement with recent weak-coupling experiments
[4,5]. With multiphoton effects included, however, and
for sufficiently large drive, the two-photon resonance
emerges approximately half way between ω_x and ω_c.
The resonance yields pronounced exciton populations, ex-
ceeding $\bar{n}_x = 0.75$ for $\eta_x \approx 2g$ (inversions of $w = \bar{n}_x -
0.5 > 0.25$). The position of this intermediate "resonance"
changes with the pump strength, which gives a direct
signature of the Stark-shifted two-photon resonance.
Similar peaks appear in the cavity intensity (not shown).
As a strong contrast, on-resonance excitation ($\delta = 0$)
yields only very small inversions (a few %) [2,3].

What is surprising about these predictions is that
they show a pronounced influence from higher lying
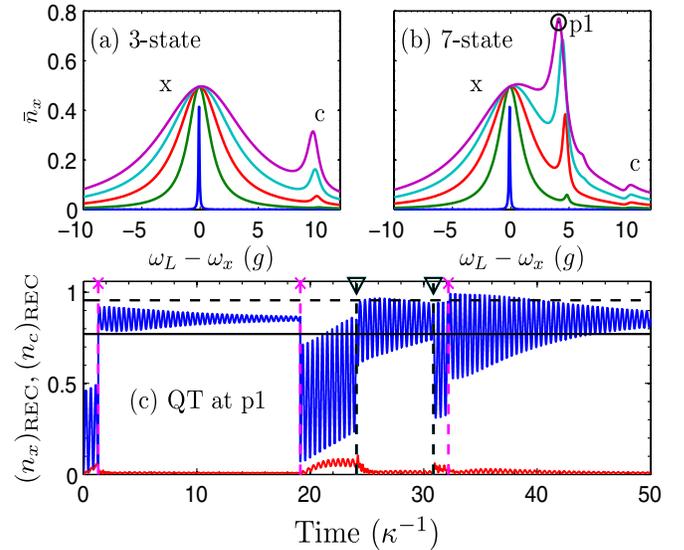ladder states, which are notoriously difficult to see in a

FIG. 3 (color online). Semiconductor quantum-dot QED sys-
tem. (a),(b) Pump-dependent steady-state exciton number versus
detuning calculated with one-photon and three-photon trunca-
tions; for $\eta_x = 0.02g\text{–}2g$ (blue to purple, lower to higher) and
$\delta = 10g$. Note the clear peak in the three-photon case near
$\omega_L - \omega_x \approx \delta/2$. (c) Sample QT simulation on the peak labeled
p1; symbols are defined as in Fig. 2, with the additional pure
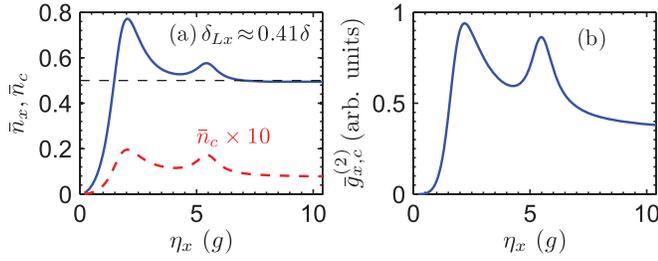dephasing event (*phase glitch*) indicated by an inverted triangle.

FIG. 4 (color online). (a) Exciton (blue) and cavity (red) population, and (b) second-order correlation function versus drive strength, η_x, for the semiconductor case of Fig. 3.

semiconductor system because of the relatively large cavity broadening. Recent experiments exploring QD anharmonicities have been reported by Kasprzak *et al.* [18], using advanced coherent nonlinear spectroscopy. The signatures observed are very weak. On the other hand, the scenario we investigate shows a profound impact from the second rung of the Jaynes-Cummings ladder, specifically the state $|2-\rangle \approx |1, +\rangle$, where a quantized level of the radiation field serves as a rung in a multilevel pumping scheme. Indeed, we stress that the two-photon resonance and resulting inversion would not exist except for the influence of the higher lying Jaymes-Cummings levels. This is an important finding for assessing the nonlinear cavity regime. Even in atomic cavity QED systems, signatures of the two-photon resonance with resonant atom-cavity coupling are weak [19].

Having demonstrated how to achieve large population inversion through coherent off-resonance excitation in a dissipative semiconductor system, we now consider some common experimental techniques that might detect it. Savage [2] suggests that a maximum inversion created by driving at saturation would yield a direct signature in fluorescence. A method more specifically sensitive to two-photon resonance involves carrying out a photon cross-correlation, i.e., measuring the correlation function $\bar{g}^{(2)}_{x,c} \equiv \langle \sigma^+ \sigma^- a^\dagger a \rangle_{ss} = \bar{g}^{(2)}_{c,x}$, which allows one to access information about the quantum statistics and penetrate the pumping cycle that creates inversion. Figures 4(a) and 4(b) display the dependence of the populations and second-order correlation function on the coherent drive strength for a semiconductor system. Two regions of maximum inversion are seen in the populations, with maxima in the cross-correlation function occurring in one to one correspondence.

Finally, it is worth emphasizing that the predicted inversions survive over a very wide range of experimental parameters—e.g., for the semiconductor system, when γ, γ', and δ all increase by a factor of 5, we still obtain peak populations ranging from 0.65–0.85. In fact, larger detunings can result in larger inversions since this avoids exciting the intermediate states, though, of course, more pump power is required. A more detailed coverage of the parameter dependence, including the influence from electron-phonon coupling with nonlinear coherent driving [20,21], will be described elsewhere.

In conclusion, we have proposed a cavity QED scheme with off-resonance excitation that achieves stationary population inversions exceeding 0.25 in semiconductor QDs. We also explored the connection to previous measurements in circuit QED and uncovered the limitations of a simplified rate equation model: quantum trajectories clarify the dynamic leading to quantum-noise-induced inversion for dipole couplings ranging over 3 orders of magnitude in units of cavity loss. More generally, we demonstrated the existence of a cavity-assisted two-photon resonance that significantly enhances the regime of multiphoton cavity QED, in particular, for QDs. This can benefit the continued worldwide search for genuine quantum signatures of cavity QED in a number of interesting material systems, along with the development of semiconductor-based single QD lasers [22].

*shughes@physics.queensu.ca
[1] See, e.g., P. W. Milloni and J. H. Eberly, *Laser Physics* (Wiley, New York, 2009).
[2] C. M. Savage, Phys. Rev. Lett. **60**, 1828 (1988).
[3] M. Lindberg and C. M. Savage, Phys. Rev. A **38**, 5182 (1988).
[4] A. Majumdar *et al.*, Phys. Rev. B **82**, 045306 (2010).
[5] A. Ulhaq *et al.*, Phys. Rev. B **82**, 045307 (2010).
[6] J. M. Fink *et al.*, Nature (London) **454**, 315 (2008).
[7] L. V. Bishop *et al.*, Nature Phys. **5**, 105 (2008).
[8] B. R. Johnson *et al.*, Nature Phys. **6**, 663 (2010).
[9] L. Tian and H. J. Carmichael, Phys. Rev. A **46**, R6801 (1992).
[10] J. Dalibard, Y. Castin, and K. Mølmer, Phys. Rev. Lett. **68**, 580 (1992).
[11] P. J. Leek *et al.*, Phys. Rev. B **79**, 180511(R) (2009).
[12] See, e.g., H. J. Carmichael, Phys. Rev. A **56**, 5065 (1997).
[13] See, e.g., H. J. Carmichael, *Statistical Methods in Quantum Optics 1* (Springer, New York, 1998).
[14] To check for numerical convergence in the basis size, we verify that the highest lying moments, $\langle (a^\dagger)^n a^n \rangle_{ss}$ for an n-photon basis, are negligible, and that increasing the basis size does not influence the results.
[15] J. P. Reithmaier *et al.*, Nature (London) **432**, 197 (2004).
[16] T. Yoshie *et al.*, Nature (London) **432**, 200 (2004).
[17] E. Peter *et al.*, Phys. Rev. Lett. **95**, 067401 (2005).
[18] J. Kasprzak *et al.*, Nature Mater. **9**, 304 (2010).
[19] I. Schuster *et al.*, Nature Phys. **4**, 382 (2008).
[20] C. Roy and S. Hughes, Phys. Rev. Lett. **106**, 247403 (2011).
[21] S. Ulrich *et al.*, Phys. Rev. Lett. **106**, 247402 (2011).
[22] M. Nomura *et al.*, Nature Phys. **6**, 279 (2010).