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PAPER

Coherent population trapping with a controlled dissipation: applications in optical metrology

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Abstract

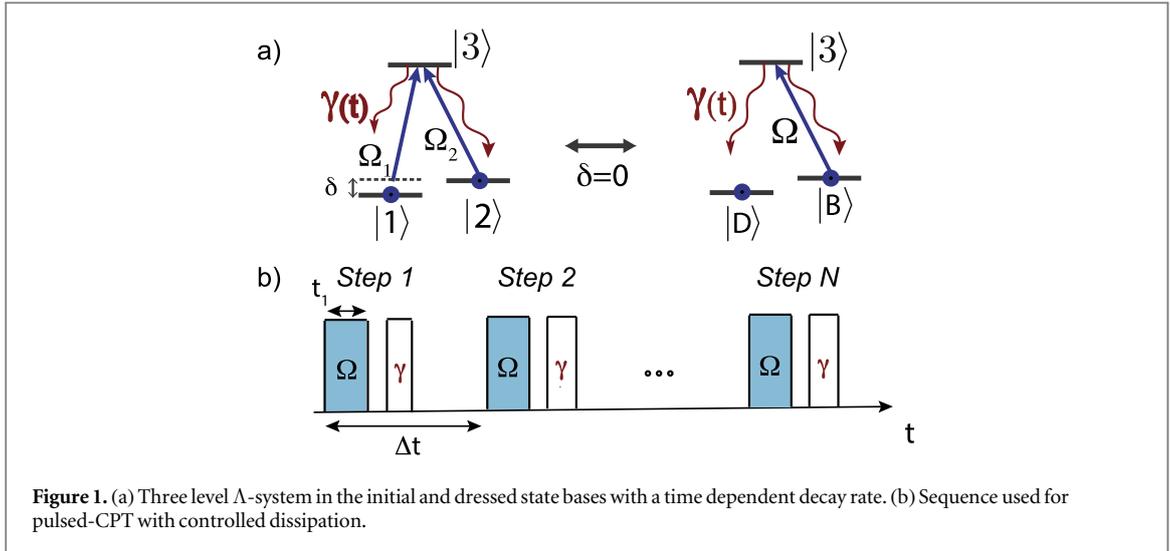
We analyze the properties of a pulsed coherent population trapping protocol that uses a controlled decay from the excited state in a Λ -level scheme. We study this problem analytically and numerically and find regimes where narrow transmission, absorption, or fluorescence spectral lines occur. We then look for optimal frequency measurements using these spectral features by computing the Allan deviation in the presence of ground state decoherence and show that the protocol is on a par with Ramsey-CPT. We discuss possible implementations with ensembles of alkali atoms and single ions and demonstrate that typical pulsed-CPT experiments that are realized on femto-second timescales can be implemented on micro-seconds timescales using this scheme.

Since its observation [1], coherent-population-trapping (CPT) and its counterpart electromagnetically-induced-transparency (EIT) [2] have enabled a wide range of experimental achievements. Using a three-level Λ -scheme and exploiting a quantum interference effect in the excited state, coherent transfer of population (STIRAP) between vibrational states [3], efficient cooling of atoms [4], precise atomic clocks [5, 6], or light storage [7, 8] have been realized. Ramsey-CPT schemes have also been shown to improve the sensitivity of frequency measurements by removing power broadening issues [6]. Further, pulsed-CPT schemes have been investigated theoretically [9–14] and experimentally using femto-second lasers [15–17] with implications for multimode quantum memories [18].

Recently, a novel pulsed-CPT scheme was realized by engineering a Λ -system in the microwave domain and exploiting the hyperfine interaction between the electron spin of a nitrogen-vacancy (NV) defect in diamond and a nearby ¹³C nuclear spin [19]. The originality of the experiment is that relaxation was externally controlled through optical pumping by a far detuned laser that couples the excited state to the ground state in the Λ -system via a metastable state. By externally controlled dissipation, we mean that the decay from the excited state to the two ground states in the Λ scheme can be triggered at will. In general such a scheme with controlled relaxation is useful for atomic systems where the excited state population lifetime is too long compared to the decoherence mechanisms. Further, compared to schemes where spontaneous emission takes place during the excitation, this method can be used to measure the excited state population while the dark state is being prepared.

In this work, we analyze such a pulsed-CPT scheme with a controlled decay from the excited state, both analytically and numerically. We show that interleaving sequences of unitary and fully dissipative steps gives rise to narrow dark transmission and photoluminescence spectral lines whose widths do not depend upon the spontaneous emission rate but solely upon the control pulse area and number of steps. Then, we discuss implications of this scheme for metrology and estimate its precision compared to Ramsey-CPT. Finally, we present possible experimental implementations using neutral alkali atoms or trapped ions, and show how pulsed-EIT experiments that are typically realized using femto-second lasers can be implemented on micro-seconds timescales.

We study the response of a three-level Λ -system excited by two near-resonant pulsed fields with Rabi frequencies Ω_1 and Ω_2 in the presence of a controlled relaxation rate $\gamma(t)$, as sketched in figure 1(a). The time



dependence of γ means that a scheme where the population in the excited can be transferred to the ground states at will is implemented using an external tuning knob. In [19], this was realized using a laser field. In the last section of this manuscript, we present another means to do so using optical pumping via a single excited state. Throughout the manuscript the field 2 will be resonant and we look for the change in transmission of the field 1 or of the excited state population as the frequency detuning of the field 1, $\delta = \omega_1 - \omega_{13}$ is varied. Here ω_1 denotes the frequency of the field 1 and ω_{13} the resonance frequency of the transition $|1\rangle - |3\rangle$. The sequence is composed of a periodically pulsed simultaneous excitation of the two transitions without decay interleaved by controlled total relaxation, as depicted figure 1(b). One sequence is composed of N pulses separated by a time interval Δt . t_1 is the length of the excitation pulses.

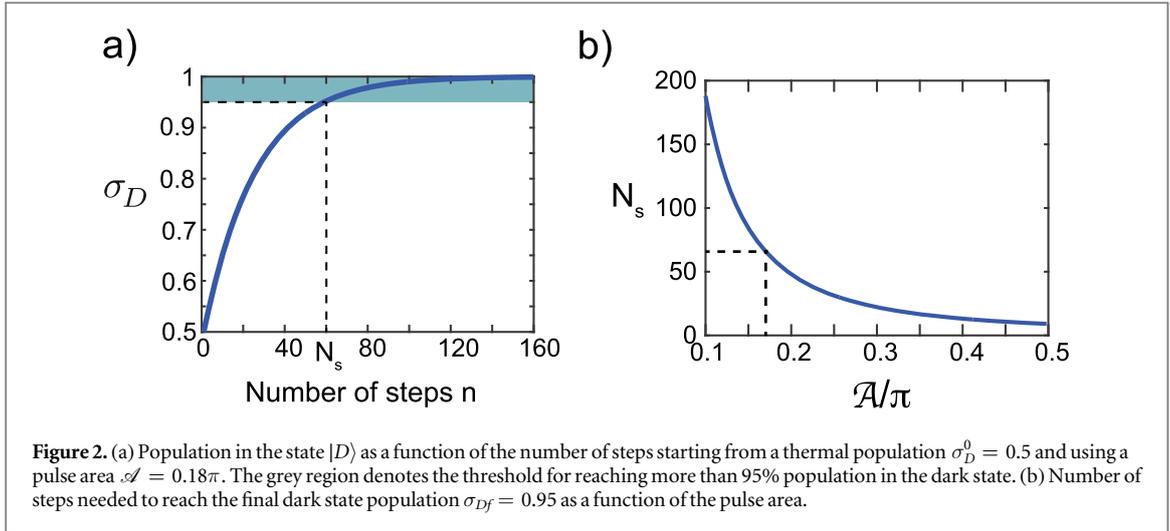
1. Pulsed-CPT under resonant excitation

At two-photon resonance ($\delta = 0$), one can write the Λ -system using a basis consisting of a dark state $|D\rangle = \frac{1}{\Omega}(\Omega_2|1\rangle - \Omega_1|2\rangle)$ which is an eigenstate of the coupled Λ -system [2], a bright state $|B\rangle = \frac{1}{\Omega}(\Omega_1|1\rangle + \Omega_2|2\rangle)$ where $\Omega = \sqrt{\Omega_1^2 + \Omega_2^2}$, and the excited state, as sketched in figure 1(a). The dark state $|D\rangle$ is not coupled to the excited state whereas the bright state $|B\rangle$ couples to the excited state with a Rabi pulsation Ω , and enables optical pumping to $|D\rangle$.

Neglecting decoherence between the two ground states ($\gamma_0 = 0$) and given that there is no population in the excited state at the beginning of each excitation pulse, the n th coherent excitation pulse transfers a population $(\sin \frac{\mathcal{A}}{2})^2 \sigma_B^{n-1}$ from the bright state to the excited state, where $\mathcal{A} = \Omega t_1$ is the pulse area and σ_B^{n-1} is the population in the bright state just before the n th pulse. This expression results from Rabi oscillations within the subspace $|B\rangle-|3\rangle$. Then, the induced decay transfers half of this quantity from the excited state population to the dark state and the other half to the bright state. Thus, given that $\sigma_B^n + \sigma_D^n = 1$ the population in the dark state at the n th steps is described by the equation $\sigma_D^n = \frac{1}{2}(\sin \frac{\mathcal{A}}{2})^2 (1 - \sigma_D^{n-1}) + \sigma_D^{n-1}$ which can be solved to give $\sigma_D^n = 1 - \left(1 - \left(\sin \frac{\mathcal{A}}{2}\right)^2 / 2\right)^{n-1} (1 - \sigma_D^0)$.

The dark state population is plotted in figure 2(a) as a function of n starting from $\sigma_D^0 = \sigma_B^0 = 0.5$ and using a pulse area $\mathcal{A} = 0.18\pi$. Here $N_s = 60$ steps are needed to reach a final dark state occupation above 0.95. We then plot the dependence of N_s on the pulse area in figure 2(b). We find that, as expected, using shorter pulses (smaller \mathcal{A}) means that more pulses are required to reach the same dark state population.

In practice, such a study of the dark state population can be realized by monitoring the population in the excited state. In contrast with the dark state population, the excited state population decays with the number of pulses [19]. This can be estimated by monitoring the photoluminescence which is proportional to the excited state population.



2. Pulsed-transmission with controlled decay

We now proceed with the study of the frequency response of such a pulsed-CPT scheme, with a focus on transmission measurements instead of photoluminescence read-out for simplicity. Experimentally, the imaginary part of the dipole amplitude $\sigma_{13} = \langle \hat{\sigma}_{13} \rangle$ is relevant for transmission experiments because it relates to the imaginary part of the linear susceptibility for the field 1. Here we assume that the field 1 is much weaker than field 2, $\Omega_1 \ll \Omega_2$, which greatly simplifies the analysis. Since it is much weaker, the field 1 will hereafter be called the probe and the field 2 the control. In order for the probe transmission to be modified, a macroscopic number of atoms, an optical cavity, or a quasi-1D geometry must be used. With many atoms, all dipole mean values $\sigma_{ij} = \langle \hat{\sigma}_{ij} \rangle$ would be locally averaged [20]⁵. In the case of a single atom in a cavity or with a quasi-1D geometry, the dipole coupling strength would be rescaled by the cavity [21] or waveguide or lens spatial mode structure [22].

To study this problem, we solve the time-dependent Bloch equations. The evolution of the mean value of the σ_{ij} is ruled by the six optical Bloch equations that are written in the appendix (A1). Assuming that $\Omega_1 \ll \Omega_2$, the population remains in the state $|1\rangle$ and the other states are not significantly populated. Under these hypotheses we find

$$\begin{aligned} \dot{\sigma}_{13} &= \left(-\frac{\gamma(t)}{2} + i\delta \right) \sigma_{13} + i\frac{\Omega_1(t)}{2} + i\frac{\Omega_2(t)}{2} \sigma_{12} \\ \dot{\sigma}_{12} &= (i\delta + \gamma_0) \sigma_{12} + i\frac{\Omega_2(t)}{2} \sigma_{13}, \end{aligned} \quad (1)$$

where δ is the detuning of the field Ω_1 , which can be treated as real or complex numbers in this approximate regime. γ_0 is the dephasing rate of the ground state coherence. Ω_1 and Ω_2 are non-zero during the excitation pulses whereas γ is non-zero between the excitation pulses. We can solve analytically this system of equations under the assumption of full relaxation, which implies that $\sigma_{13} = 0$ at the beginning of each excitation pulse.

2.1. Multiple interferences

We study $\text{Im}(\sigma_{13})$ as a function of the two-photon detuning δ in the absence of ground state dephasing γ_0 . Let us first note that the evolution of σ_{13} during the n th excitation pulse is directly related to the value of σ_{12} at the beginning of the n th pulse $\sigma_{12}^n = \sigma_{12}((n-1)\Delta t)$. Solving equation (1) yields a simple formula for σ_{12}^{n-1} . Assuming that $\sigma_{12}^0 = 0$, we get, for $n > 1$:

$$\sigma_{12}^n(\delta) = f(\delta) \sum_{l=0}^{n-1} e^{i\delta l \Delta t} \cos^l \left(\frac{\mathcal{A}}{2} \right), \quad (2)$$

⁵ If the medium is optically thick, coupled Maxwell-Bloch equations must be used, as in [11]. Retardation effects would take place, but the main conclusions of the paper remain unchanged.

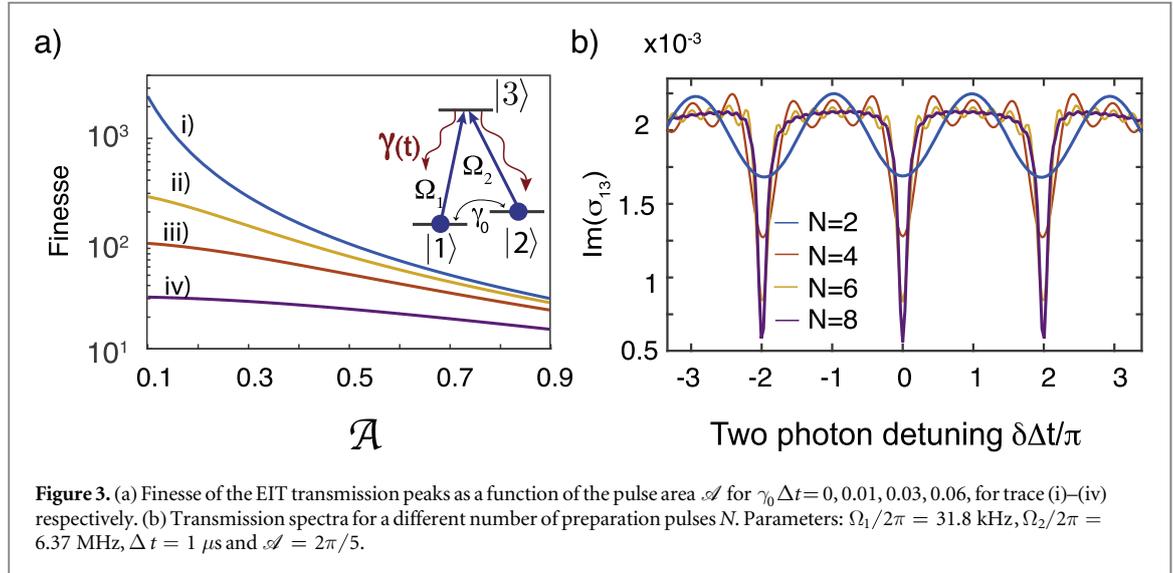


Figure 3. (a) Finesse of the EIT transmission peaks as a function of the pulse area \mathcal{A} for $\gamma_0 \Delta t = 0, 0.01, 0.03, 0.06$, for trace (i)–(iv) respectively. (b) Transmission spectra for a different number of preparation pulses N . Parameters: $\Omega_1/2\pi = 31.8$ kHz, $\Omega_2/2\pi = 6.37$ MHz, $\Delta t = 1 \mu\text{s}$ and $\mathcal{A} = 2\pi/5$.

with

$$f(\delta) = \frac{1}{2} \frac{\Omega_1 e^{i\delta \Delta t}}{\delta^2 - \left(\frac{\Omega_2}{2}\right)^2} \left(\frac{\Omega_2}{2} \left(\cos(\delta t_1) - \cos\left(\frac{\mathcal{A}}{2} t_1\right) \right) + i \left(\delta \sin\left(\frac{\mathcal{A}}{2} t_1\right) - \frac{\Omega_2}{2} \sin(\delta t_1) \right) \right).$$

We observe that σ_{12}^n is proportional to a geometric sum, analogous to the transmission of light in a Fabry–Pérot cavity. We thus expect to observe a periodic spectrum with interference peaks separated by the free spectral range defined as $\Delta \delta_{\text{FSR}} = \frac{2\pi}{\Delta t}$. The width of each peak should thus decrease with the number of steps until a steady state is reached. When N is high enough, a simple formula including γ_0 can be found, and the second term in the product appearing in equation (2) will show peaks with a width related to a finesse given by

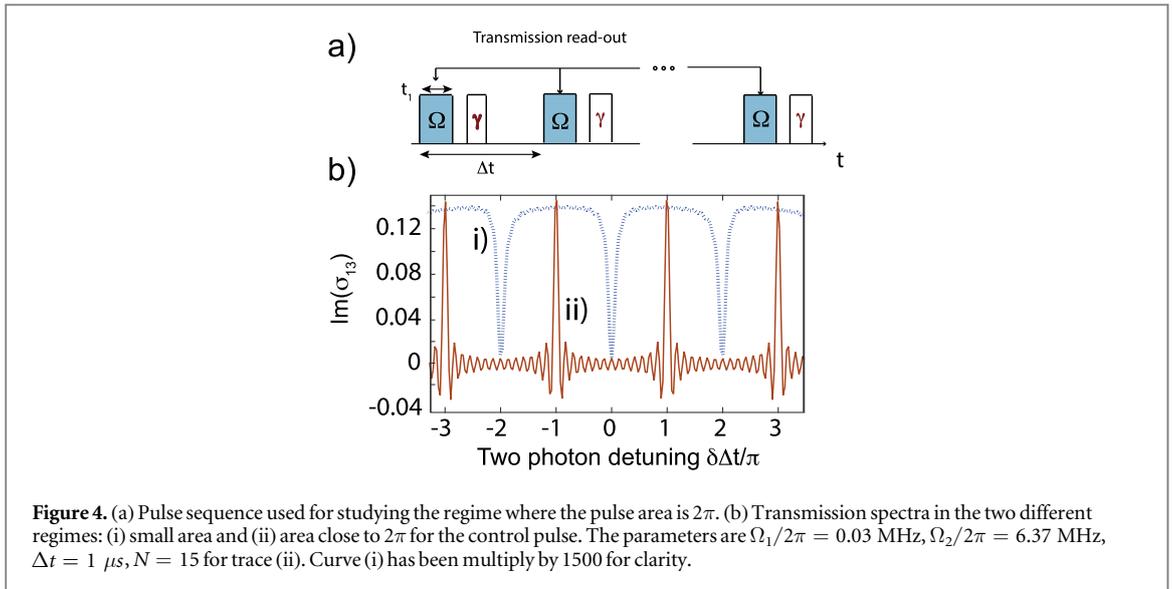
$$\mathcal{F} = \frac{\pi \sqrt{e^{-\gamma_0 \Delta t} \cos\left(\frac{\mathcal{A}}{2}\right)}}{1 - e^{-\gamma_0 \Delta t} \cos\left(\frac{\mathcal{A}}{2}\right)}.$$

This formula is valid for $\cos\left(\frac{\mathcal{A}}{2}\right) > 1/2$, i.e. for $\mathcal{A} < 2\pi/3$. The width of each peak is then $1/(\mathcal{F} \Delta t)$. We note that \mathcal{F} increases when \mathcal{A} decreases, as shown figure 3(a).

As manifest in equation (2), there are in fact two distinct multiple-interference regimes. A regime in which $\cos\left(\frac{\mathcal{A}}{2}\right) \approx 1$, where constructive interferences occur when $\delta \Delta t = 2m\pi$ ($m \in \mathbb{Z}$) and a regime in which $\cos\left(\frac{\mathcal{A}}{2}\right) \approx -1$, where constructive interferences take place at $\delta \Delta t = (2m + 1)\pi$. We will study the physics underlying these two situations in the next sections by evaluating σ_{13} when the area is smaller than 2π and when the area is close to 2π .

2.2. Pulsed-EIT regime

We first study the transmission spectra using the analytical solution of the Bloch equations (A1) derived in the appendix, in the limit where the control pulse area is below 2π . Transmission spectra measured at the end of the last pulse of the sequence are plotted as a function of the two-photon detuning for a different number of steps N in figure 3(b). The parameters are $\Omega_1/2\pi = 31.8$ kHz, $\Omega_2/2\pi = 6.37$ MHz, $\Delta t = 1 \mu\text{s}$ and $\mathcal{A} = 2\pi/5$. As anticipated, a periodic pattern appears in the transmission spectrum. Close to two-photon resonance, we observe a spectrum that is similar to EIT and we clearly see a step-by-step narrowing of the spectrum. One sees that the central peak amplitude gets closer and closer to zero after several pulses. The multiple interferences in the accumulated ground state coherence also implies that the width of the peaks narrows down as the number of pulses N increases, as per equation (2) and that other transmission peaks that are equally spaced at frequencies $\delta \Delta t = 2m\pi$ where $m \in \mathbb{Z}$ appear due to this frequency comb driving as expected. This confirms that there is a sequential pumping into the dark state $|D\rangle = \frac{1}{\Omega}(\Omega_2|1\rangle - \Omega_1|2\rangle)$. Indeed, adding detunings to both fields only changes the local phase in $|D\rangle$ to $\exp(i\delta \Delta t)$ which remains an eigenstate if $\delta \Delta t = 2m\pi$.



2.3. Pulsed Autler–Townes regime

Let us now investigate the regime where the pulse area is 2π . When $\mathcal{A} = 2\pi$, the Bloch vector undergoes a full rotation on the Bloch sphere related to the $|2\rangle$ – $|3\rangle$ transition. The probe measured right after the excitation pulses would be fully transmitted. In order to optimally measure the ground state coherence evolution in this regime, the probe transmission must thus be acquired in the middle of the coherent excitation, i.e. on the north pole of the Bloch sphere, as depicted in figure 4(a).

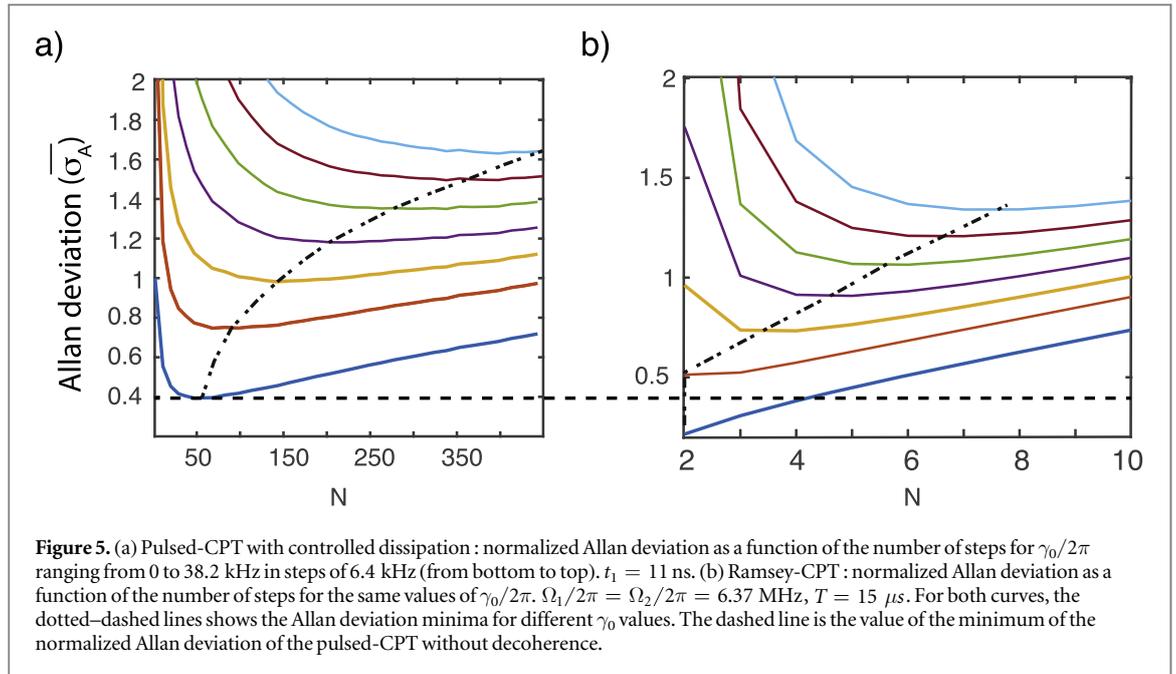
Figure 4(b), trace (i) shows the evolution of $\text{Im}(\sigma_{13})$ as a function of the two photon detuning, in the steady state and small area limit. The parameters are the same as for figure 2(b), that is we have a pulse area $2\pi/5$. 4(b), trace (ii) shows the same spectrum for $\Omega_1/2\pi = 31.8$ kHz, $\Omega_2/2\pi = 6.37$ MHz, $N = 15$ and $\Delta t = 1$ μ s, that is for a pulse area of 2π . The difference between the two excitation regimes shown in trace (i) and (ii) is striking: instead of transmission peaks at detunings such that $\delta\Delta t = 2m\pi$ with $m \in \mathbb{Z}$, absorption peaks occur for $\delta\Delta t = (2m + 1)\pi$.

This $\mathcal{A} = 2\pi$ regime is in fact reminiscent of the Autler–Townes effect where, under continuous wave excitation (CW) and under saturation ($\Omega_2 \gg \gamma$), an absorption doublet appears at $\delta = \pm\Omega_2/2$ [23]. Here, when $\mathcal{A} = 2\pi$, $\sigma_{13} = 0$ at the end of each excitation pulse, so the induced decay has no effect on the coherences. To understand why the absorption peaks occur at $\delta\Delta t = (2m + 1)\pi$ then, it is instructive to define an effective Rabi frequency Ω_{eff} for the whole duration Δt , so as to tend and to eventually compare to the quasi-continuous driving (CW). Defining a Rabi frequency for a duration Δt is possible here because controlled decay has no influence when $\mathcal{A} = 2\pi$. We define Ω_{eff} as the Rabi frequency that would populate the excited state during a time Δt with the same probability as a shorter excitation at a Rabi frequency Ω_2 for a time t_1 . Equating the pulse areas, we obtain $\Omega_{\text{eff}}\Delta t = \Omega_2 t_1$. Since $\mathcal{A} = 2\pi$ here, we get $\Omega_{\text{eff}} = 2\pi/\Delta t$. The two first absorption peaks appear at $\delta = \pm\pi/\Delta t$ which means $\delta = \pm\Omega_{\text{eff}}/2$. This analysis shows that the doublets appear at the same frequencies as the CW Autler–Townes doublets. This also shows that the transition from EIT to Autler–Townes, also investigated in [24, 25], can be observed using a Λ -scheme with controlled dissipation.

3. Prospects for metrology

The narrow transmission lines in this stroboscopic state preparation have implications both in atomic clocks or magnetometry. CPT in fact already found use in precision measurements of atomic transitions [5]. Here we are interested in the performance of the pulsed-CPT for metrology, such as magnetic field or time measurements. Atomic clocks or magnetometers often use Ramsey sequences, where two $\pi/2$ pulses separated by a time T are used to drive a two-level system. In such a pulsed regime, scanning the laser frequency gives rise to a sinusoidal spectrum with periodicity $1/T$, providing a means to precisely estimate the atomic transition without power broadening. The observed fringe contrast will be given by the coherence time of the transition and so is the precision of the frequency measurement [5, 6].

Ramsey-CPT not only minimizes power broadening but it uses sub-natural lines [5]. The principle is to first prepare a dark state in the three-level system, to let the ground state coherence evolve ‘in the dark’ and to measure the evolution of the ground state by applying a second pair of identical read-out pulses. If the lasers are two-photon detuned, the initial dark state population is transferred to the bright state so the fluorescence (or the



absorption) is increased. This provides a means to read-out the atomic frequency close to the two-photon resonance condition. Here, due to the multiple interferences, the presented pulsed-CPT scheme will show even narrower lines so one could expect a high metrological performance. We show that this is not the case.

The frequency stability of clocks are typically characterized by the Allan deviation [26] which, in the presence of white frequency noise, reads

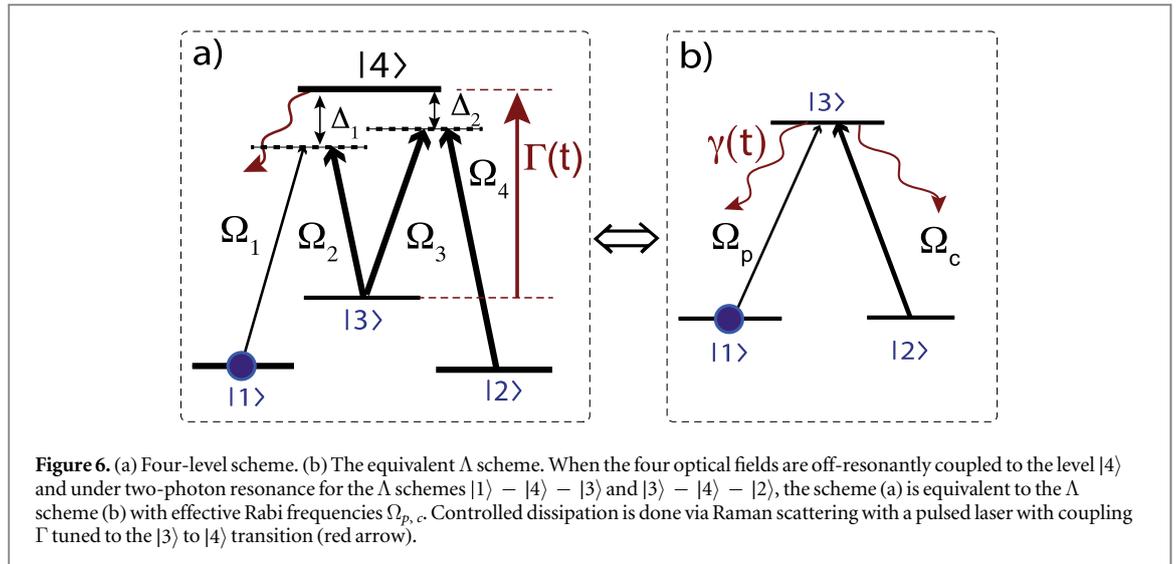
$$\sigma_A(\tau) = \frac{1}{\nu_{\text{at}}} \frac{\sigma_S}{\left(\frac{\partial S(\nu)}{\partial \nu}\right)_{\nu_m}} \sqrt{\frac{T_c}{\tau}},$$

where T_c is the time of an interrogation cycle, τ is the averaging time, ν_{at} is the atomic resonance frequency, σ_S is the noise of the measured signal S and ν_m is the frequency at which the signal is measured. A small Allan deviation indicates a high precision in the frequency estimation. Magnetometers can be characterized in the very same way, the signal S being proportional to the magnetic moment [27].

Here, we compute $\sigma_A(\tau)$ for the pulsed-CPT and Ramsey-CPT schemes and quantify their performances as a function of the number of steps and for various decoherence rates in the ground state. To avoid optimization complications related to the choice of probe intensity, we chose to use fluorescence instead of transmission read-out for both schemes and use $\Omega_1 = \Omega_2$ as in [19]. Further, we use a pulse area far below 2π to be in the EIT regime. The analytical solutions presented earlier cannot be used here since we need to compute the excited state population instead of the coherence. We thus resort to numerical simulations and use the XMDS package to solve the full Bloch equations [28].

The fluorescence signal is acquired during each de-excitation pulse (that is when γ is on), except from the first one. The signal acquired during the n th decay process is directly proportional to the population at the end of the n th excitation pulse $\sigma_{33}(n\Delta t + t_1)$. In order to make it independent on the actual experimental apparatus (collection efficiency, photodetector gain, number of atoms, ect.) the Allan deviation σ_A is normalized to that of a two-pulse pulsed-CPT experiment without ground state decoherence (we write it $\sigma_A^{N=2}$). For a fair comparison between the pulsed-CPT and Ramsey-CPT schemes, we use the same total sequence time T_c . Increasing the number of steps within T_c means that the signal will increase and that the precession time between each pulse will decrease. Note that, even with the Ramsey-CPT scheme, more than 2 pulses are usually required in order to gather signal statistics and thus to optimize the Allan deviation.

The populations are first prepared in the dark state. The Allan deviation (measured for the value of ν_m which minimizes it) is plotted in figure 5(a) as a function of the number of steps for different γ_0 . The results for $\overline{\sigma}_A = \sigma_A/\sigma_A^{N=2}$ are plotted for $\gamma_0/2\pi$ varying from 0 to 38.2 kHz in steps of 6.4 kHz. We observe that for each decoherence rate, an optimal number of steps N_{opt} is found that minimizes $\overline{\sigma}_A$. This can be understood as follows: for $N < N_{\text{opt}}$, the spectral lines are not optimally narrow because the multiple-interferences are not at play. For $N = N_{\text{opt}}$ however, we are in the multiple-interferences regime. As it can be expected also, the minimum of the Allan deviation decreases when γ_0 increases. As the decoherence rate increases, more pulses are indeed needed to preserve the dark state so the optimal number of steps increases with γ_0 . When N increases



again above N_{opt} , the precession time is reduced, therefore the linewidth increases and so does the Allan deviation (see figure A2).

It is instructive to compare this scheme with the well-established Ramsey-CPT protocol. Simulations for Ramsey-CPT are presented in figure 5(b), where the number of steps and decoherence rates in the ground state are also varied. The main difference with pulsed-CPT with controlled dissipation is that the decay from the excited state to the ground state takes place at all times and that, for the Ramsey-CPT sequence, the photoluminescence signal is accumulated over all the excitation pulses [5]. We chose an area \mathcal{A} which totally prepares the dark state at the end of each pulse. The results for the Allan deviation are also shown for $\gamma_0/(2\pi)$ varying from 0 to 38.2 kHz in steps of 6.4 kHz (from bottom to top). The general trend for the Allan deviation is similar than for pulsed-CPT: when γ_0 increases, more pulses are needed to optimize σ_A as a result of a compromise between signal strength and ground state coherence amplitude. The optimum Allan deviation is on the same order of magnitude as for the pulsed-CPT with controlled decay. The dotted-dashed line in figures 5(a) and (b), shows the Allan deviation minima. As discussed, the minima increase with γ_0 and require a larger and larger number of steps as γ_0 increase for both Ramsey-CPT and pulsed-CPT protocols. The horizontal dashed line shows that the precision is slightly better when using the Ramsey-CPT for $\gamma_0 = 0$, which in fact holds for all ground state decoherence rates.

4. Experimental implementation

The presented scheme was already realized in the microwave regime using NV centers in diamonds using coupled electronic and nuclear spins [19]. It might be beneficial to use optical fields and enlarge the range of applications to, for instance, ultra-cold atoms. We now discuss a possible way to implement such a pulsed-CPT with controlled dissipation using neutral alkali atoms and trapped ions.

Alkalies. In order to realize this scheme with alkali atoms, one possibility is to engineer an effective three level system using four atomic levels and two Raman transitions as depicted in figure 6(a). This can for instance be realized with all isotopes of rubidium or cesium on the $F \rightarrow F - 1$ transitions of the D_1 and D_2 lines. Provided the four lasers are detuned by more than several excited state linewidths, we can adiabatically eliminate the state $|4\rangle$ and write $|\frac{\partial \sigma_{i4}}{\partial t}| \ll |\Delta \sigma_{i4}|$, where $i = 1, 2, 3$ for all the optical transitions. This means that two Raman transitions $|1\rangle - |4\rangle - |3\rangle$ and $|3\rangle - |4\rangle - |2\rangle$ coherently drive the long-lived transitions $|1\rangle - |3\rangle$ and $|2\rangle - |3\rangle$. The resulting effective Rabi frequencies on transition $|1\rangle - |3\rangle$ and $|2\rangle - |3\rangle$ are given by $\Omega_p = \Omega_1 \Omega_2 / \Delta_1$ and $\Omega_c = \Omega_3 \Omega_4 / \Delta_2$ respectively. We also require the difference between the detunings Δ_1 and Δ_2 to be greater than the width of these two Raman transitions. This means that $\Delta_1 - \Delta_2 \gg (\Omega_p, \Omega_c)$. If this condition is not satisfied, ground state coherence between the ground state $|1\rangle$ and $|2\rangle$ generated solely by fields 1 and 4 would take place.

This four-level scheme is then equivalent to the three-level system shown figure 6(b). The lifetime of the decay from the state $|3\rangle$ can effectively be shortened via a spontaneous Raman scattering using a laser driving the transition between $|3\rangle$ and $|4\rangle$. The detection can be done by measuring the Raman scattered light intensity, or

the laser transmission in an EIT-like experiment. Combined with this pulsed-CPT technique, efficient clocks or magnetometers can thus be realized.

The coherence time in the two ground states $|1\rangle$ and $|2\rangle$ can be several milliseconds with trapped alkali atoms. Typical pulsed-EIT experiments require pulses that are shorter than the excited state decay time require femto-second lasers [13, 15, 17]. In comparison, the proposed pulsed-EIT can easily be observed on micro-second timescales so that light pulses can be generated simply using acousto-optic modulators. This in fact bears similarity with the experiment done in a rubidium cell in [29], where cavity-like features were observed using multiple interference on long-lived spin waves in a gradient echo memory [30]. Operating on microseconds time scale may have important practical consequences. For instance, it could be experimentally feasible to shape the relative phase between the laser pulses after each step in order to generate amplification of the probe [31] or to observe the step-by-step growth of the collective ground state spin wave amplitude via EIT.

Trapped ions. Another possible experimental implementation of the pulsed-CPT scheme is using trapped ions on a quadrupolar transition. For instance, using $^{40}\text{Ca}^+$, this transition is driven by a 729 nm laser tuned to the $S_{1/2}$ to $D_{5/2}$. A Λ -system can be realized using the Zeeman sublevels $S_{1/2}(m = 1/2)$ and $S_{1/2}(m = -1/2)$ and a single excited Zeeman levels in the $D_{5/2}$ manifold [32]. A laser at 854nm, tuned to the $D_{5/2}$ to $P_{3/2}$ transition can then induce spontaneous Raman scattering back to the two ground states of $S_{1/2}$. Repeating sequences of 729 + 854 nm excitations thus realizes the pulsed-CPT protocol with controlled dissipation using the long-lived $D_{5/2}$ excited state.

Differential AC-Stark shifts. One well-known issue with optical metrology is the differential AC-stark shift induced by other nearby electronic levels here. This effect comes from the two optical fields that drive the Λ scheme, which can off-resonantly couple to these extra levels and generate a fictitious magnetic field proportional to the degree of circular polarization. This induces systematic shifts that are equivalent to a two photon detuning. Estimating precisely differential light shifts in the case of pulsed-CPT is, in general, complicated since they depend crucially on the level structure. However, other schemes, such as hyper-Ramsey interferometry [33] or Ramsey-comb spectroscopy technique [34, 35] have been proposed to mitigate parasitic effects of the light shifts. Using these proposals in combination with pulsed-CPT and controlled dissipation may enhance the metrological precision and will be left to further studies.

5. Conclusion

We studied analytically the dynamics of a three-level system driven by a pulsed train of coherent fields in the presence of a controlled decay from the excited state. Formulas for the width of the transmission window were found and Autler–Townes doublets were recovered for pulse areas that are multiple of 2π . We compare the Allan deviation of the protocol to the Ramsey-CPT scheme with metrological applications in mind and demonstrate that they perform almost equally well. We have also shown that it is possible to observe the step-by-step growth of the dark-state amplitude using several experimental platforms. In general, this work adds a new dimension to CPT and EIT. Using more evolved pulse protocols, it may be possible to create ultra-narrow lines and use it for more efficient atomic clocks, magnetometers or for new light storage protocols [36].

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Appendix. Analytical solution

We list here the full Bloch equations that are used to describe the pulsed-CPT scheme with a controlled dissipation $\gamma(t)$ from the excited state. They read :

$$\begin{cases} \dot{\sigma}_{13} = -\left(\frac{\gamma(t)}{2} + i\delta\right)\sigma_{13} + i\frac{\Omega_1}{2}(\sigma_{11} - \sigma_{33}) + i\frac{\Omega_2}{2}\sigma_{12} \\ \dot{\sigma}_{12} = -(\gamma_0 + i\delta)\sigma_{12} + i\frac{\Omega_2}{2}\sigma_{13} - i\frac{\Omega_1}{2}\sigma_{23}^* \\ \dot{\sigma}_{23} = -\frac{\gamma(t)}{2}\sigma_{23} + i\frac{\Omega_2}{2}(\sigma_{22} - \sigma_{33}) + i\frac{\Omega_1}{2}\sigma_{13}^* \\ \dot{\sigma}_{11} = \gamma(t)\sigma_{33} + i\frac{\Omega_1}{2}\sigma_{13} - i\frac{\Omega_1}{2}\sigma_{13}^* \\ \dot{\sigma}_{22} = \gamma(t)\sigma_{33} + i\frac{\Omega_2}{2}\sigma_{23} - i\frac{\Omega_2}{2}\sigma_{23}^* \\ \dot{\sigma}_{33} = -\dot{\sigma}_{11} - \dot{\sigma}_{22}. \end{cases} \quad (\text{A1})$$

In the following, we set $\gamma_0 = 0$. If the Rabi frequency of the field 1 is much smaller than the Rabi frequency of the field 2, we have $\sigma_{11} \approx 1$, $\sigma_{22} \approx 0$ and $\sigma_{23} \approx 0$, so that

$$\begin{cases} \dot{\sigma}_{13} = i\delta\sigma_{13} + i\frac{\Omega_1}{2} + i\frac{\Omega_2}{2}\sigma_{12} - \frac{\gamma(t)}{2}\sigma_{13} \\ \dot{\sigma}_{12} = i\delta\sigma_{12} + i\frac{\Omega_2}{2}\sigma_{13}. \end{cases} \quad (\text{A2})$$

The solution for the n th step of the sequence is given by the equations

$$\sigma_{12}(t) = \begin{cases} e^{i\delta t}(A_n e^{i\frac{\Omega_2}{2}t} + B_n e^{-i\frac{\Omega_2}{2}t}) + \alpha & \text{if } t \in [(n-1)\Delta t, (n-1)\Delta t + t_1] \\ e^{i\delta(t - ((n-1)\Delta t + t_1))}\sigma_{12}((n-1)\Delta t + t_1) & \text{if } t \in](n-1)\Delta t + t_1, n\Delta t], \end{cases} \quad (\text{A3})$$

and

$$\sigma_{13}(t) = \begin{cases} e^{i\delta t}(A_n e^{i\frac{\Omega_2}{2}t} - B_n e^{-i\frac{\Omega_2}{2}t}) - 2\frac{\delta}{\Omega_2}\alpha & \text{si } t \in [(n-1)\Delta t, (n-1)\Delta t + t_1] \\ e^{i(\delta - \gamma)(t - ((n-1)\Delta t + t_1))}\sigma_{13}((n-1)\Delta t + t_1) & \text{si } t \in](n-1)\Delta t + t_1, n\Delta t]. \end{cases} \quad (\text{A4})$$

Where $\alpha = \frac{1}{2} \frac{\Omega_1 \Omega_2}{\delta^2 - \left(\frac{\Omega_2}{2}\right)^2}$. The initial conditions $\sigma_{12}(0) = 0$ and $\sigma_{13}(0) = 0$ give $A_1 = \alpha \frac{\delta - \frac{\Omega_2}{2}}{\Omega_2}$ and $B_1 = -A_1 - \alpha$.

The continuity of σ_{12} and the hypothesis $\sigma_{13}(n\Delta t) = 0$ for all n is valid for full de-excitation and give the following expression for A_n and B_n ⁶

$$\begin{aligned} A_{n+1} &= c^n \left(A_1 + k \frac{d - \left(\frac{d}{c}\right)^{n+1}}{1 - \frac{d}{c}} \right) \\ B_{n+1} &= A_{n+1} e^{i\Omega_2 n \Delta t} - 2\alpha \frac{\delta}{\Omega_2} e^{i\left(\frac{\Omega_2}{2} - \delta\right)n \Delta t}. \end{aligned} \quad (\text{A5})$$

In the equation (A5), $c = e^{-i\frac{\Omega_2}{2}\Delta t} \cos\left(\frac{\Omega_2}{2}t_1\right)$,

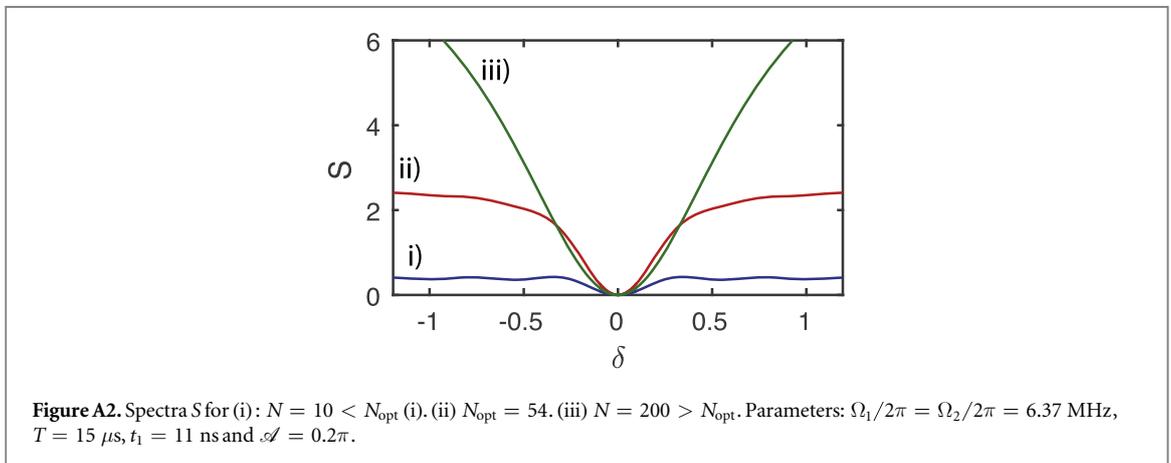
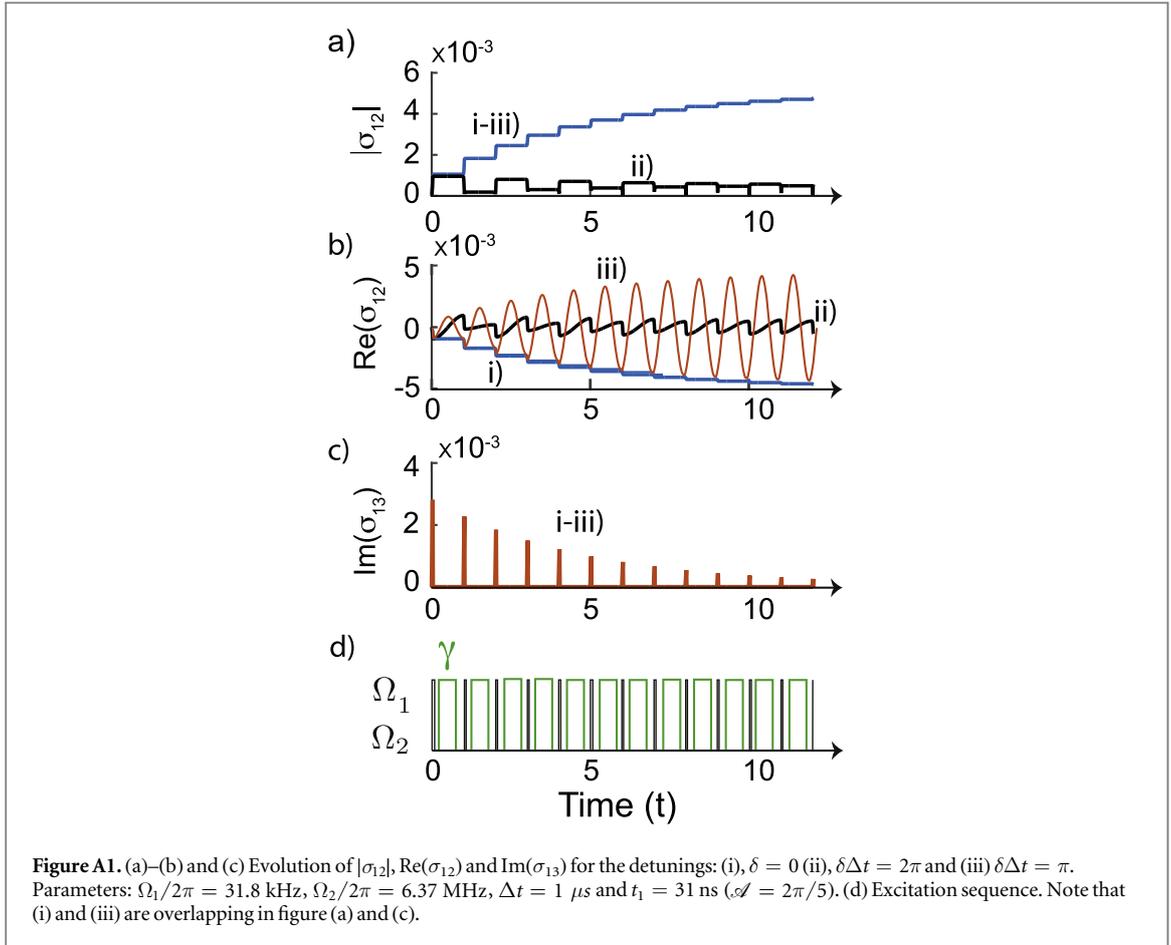
$$k = \frac{\alpha}{2} \left(e^{i\delta(\Delta t - t_1)} - 1 + 2\frac{\delta}{\Omega_2} (1 - e^{-i\frac{\Omega_2}{2}t_1} e^{i\delta\Delta t}) \right)$$

and $d = e^{-i\left(\frac{\Omega_2}{2} + \delta\right)\Delta t}$.

The time evolution of the coherences is plotted in figures A1(a)–(c) for a small pulse area and for three different detunings of Ω_1 . Trace (i) corresponds to the resonance case $\delta = 0$, trace (ii), $\delta = \pi/\Delta t$ and trace (iii) $\delta = 2\pi/\Delta t$. For $\delta = 0$ and $\delta = 2\pi/\Delta t$, we observe that the coherences σ_{13} tend to 0 when N increases, which corresponds to the preparation of the dark state: after enough pulses Ω_1 does not interact anymore with the atoms. We also see that for those two δ , the atomic coherence reaches a steady state. We note that the evolutions of $|\sigma_{12}|$ are exactly the same for the two first detunings because they have the same phase and amplitude at the beginning of each pulse.

Three fluorescence spectra used to estimate the Allan deviation for the pulsed-CPT are plotted in figure A2. They correspond to different sequences that have the same total length and pulse area but for different number

⁶ The recursive A_n is of the form $u_{n+1} = cu_n + kd^n$



of pulses. The signal is accumulated over all the de-excitation pulses except for the first one. Trace (i), (ii) and (iii) show spectra for $N = 10$, $N = 54$ and $N = 200$ respectively. For $N < N_{\text{opt}}$, the signal is not accumulated enough and so the photoluminescence rate drop is not steep yet because of the low number of excitation pulses. For $N > N_{\text{opt}}$, Δt decreases as the number of pulses increases, which leads to a dilatation of the spectrum and thus reduces the slope of the spectrum close to ν_m (see figure A2).

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