Electric control of collective atomic coherence in an erbium-doped solid

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Abstract. We demonstrate the fast and accurate control of the evolution of collective atomic coherences in an erbium-doped solid using external electric fields. This is achieved by controlling the inhomogeneous broadening of erbium ions emitting at 1536 nm using an electric field gradient, thanks to the linear Stark effect. The manipulation of atomic coherence is characterized with the collective spontaneous emission (optical free induction decay (FID)) emitted by the sample after an optical excitation, which does not require any previous preparation of the atoms. We show that controlled dephasing and rephasing of the atoms by the electric field result in collapses and revivals of the optical FID. Our results show that the use of external electric fields does not introduce any substantial decoherence and enables the manipulation of collective atomic coherence with a very high degree of precision on the timescale of tens of nanoseconds. This provides an interesting resource for photonic quantum state storage and quantum state manipulation.

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1. Introduction

The coherent control of quantum systems plays a central role in quantum information science and in quantum technology in general. In particular, the coherent manipulation of collective atomic coherences in material systems is crucial for applications in photonic quantum storage \cite{1, 2} and in ensemble based quantum computing \cite{3, 4}. A promising route towards these applications is to use solid state atomic ensembles implemented with rare-earth (RE) ion-doped solids.

In a solid state environment, the optical atomic lines of the RE impurities are inhomogeneously broadened \cite{5}. When atoms are excited, for example following the absorption of a light pulse, the atomic dipoles oscillate at different frequencies, leading to inhomogeneous dephasing. In order to enable a constructive interference between all the emitters that will lead to a collective re-emission of the stored light, this dephasing must be controlled. The rephasing of the dipoles can be triggered by optical pulses, as in traditional photon echo techniques. These techniques, while very successful to store classical light \cite{6, 7} and as a tool for high resolution spectroscopy \cite{5, 8}, suffer from strong limitations for the storage of single photons \cite{9}. Another possibility is to exploit the fact that some materials exhibit permanent dipole moments, which give rise to a linear Stark effect \cite{10}. The frequency of the atoms can then be controlled with a moderate external electric field. This effect has been used for the demonstration of Stark modulated photon echoes \cite{11}--\cite{15} as a tool for high resolution spectroscopy. The electric control of the resonance frequency of the atoms is also the key resource of a recently proposed modified photon echo protocol based on controlled and reversible inhomogeneous broadening (CRIB) \cite{16}--\cite{19}.

The great advantage of using electric fields to manipulate atomic coherences is that it does not change the population distribution in the ground and excited states, contrary to optical rephasing pulses. In combination with an optical transfer of population to a long lived ground state, this enables, in principle, the long-term storage and retrieval of single photon fields with unit efficiency and fidelity \cite{17, 19}. Quantum storage with unit efficiency can also be achieved without the transfer to the long lived ground state, using only electric fields for the light retrieval \cite{20, 21}. The first proof of principle experiment of the CRIB protocol was performed in a Eu-doped crystal \cite{22, 23} followed by another demonstration in a Pr-doped crystal \cite{20}. The maximal efficiency of the storage and retrieval is directly proportional to the quality of the manipulation of the atomic coherences. It is thus extremely important to have a good characterization of the rephasing of the dipoles.

A CRIB experiment requires sophisticated optical pumping techniques in order to first isolate a narrow absorption peak within a large transparency window. Moreover, it is difficult to characterize the quality of the rephasing directly from a CRIB experiment, since the efficiency of the storage also depends on other parameters, such as available optical depth or quality of the optical pumping for the preparation of the memory. In this paper, we use a much simpler method to characterize the electric manipulation of the atomic coherence. We propose to infer the dynamics of the atomic coherences by studying the collective spontaneous emission of light from the atoms (a phenomenon known as optical free induction decay (FID) \cite{24}). In particular, we show that controlled dephasings and rephasings of the atoms via the electric field result in collapses and revivals of the FID. The observation of FID does not require any optical preparation of the sample and can thus be done with a relatively simple experimental setup. In practice it is sufficient to excite the atoms with a single optical pulse in resonance with the...
atomic transition and to measure the collective emission of light after the pulse. The FID is, however, strongly nonlinear with respect to the excitation pulse intensity and vanishes in the limit of weak excitation pulses [24, 25].

In this paper, we use erbium ions doped into a Y\textsubscript{2}SiO\textsubscript{3} crystal. This is an interesting system since erbium ions have an optical transition at the telecommunication wavelength of 1536 nm. It could thus, in principle, enable the realization of a light matter quantum interface between photons that can be transmitted with low loss in optical fibres and stationary atoms in a solid. Such a quantum memory at telecommunication wavelength would be useful in the context of quantum repeaters [26]–[28]. The spectroscopic properties of Er\textsuperscript{3+} : Y\textsubscript{2}SiO\textsubscript{3} have been extensively studied, including optical coherence [29]–[32], spectral diffusion [30, 33], hyperfine structure [34], Zeeman relaxation lifetimes [35], Zeeman g factors [36] and erbium host interactions [37]. Slow light has also been achieved in this material using coherent population oscillation [38]. Optical pumping techniques have also been developed [39], and a proof of principle experiment of CRIB with weak light pulses at the single photon level has been recently demonstrated in an Er\textsuperscript{3+} : Y\textsubscript{2}SiO\textsubscript{3} crystal [40].

2. Control of collective atomic coherences using the linear Stark effect

We now explain in more detail how the dephasing and rephasing of atomic dipoles can be controlled via the linear Stark effect, and how this affects the collective emission of light from the sample. In the presence of an external dc electric field, the energy levels of an atom with a permanent dipole moment are shifted by an amount proportional to the electric field. This phenomenon is known as the linear dc Stark effect [10]. If the dipole moments are different for different electronic levels, this shift leads to a shift in the associated optical transition frequency. The linear dc Stark effect can be observed in RE-doped solids, since the ions possess a permanent dipole moment induced by local electric fields due to the crystal environment. The detuning of the atom transition \( \Delta \) due to the linear Stark effect can be described as [35]

\[
\Delta = \frac{\Delta \mu_e \chi \cos \theta}{\hbar} E,
\]

where \( \Delta \mu_e \) is the difference between the permanent dipole moments for the two states of the optical transition, \( E \) is the applied electric field amplitude, \( \chi = (\epsilon + 2)/3 \) is the Lorentz correction factor, \( \epsilon \) is the dielectric constant of the sample and \( \theta \) is the angle between \( \overrightarrow{\Delta \mu_e} \) and \( \overrightarrow{E} \). Since the crystal used in this experiment (Y\textsubscript{2}SiO\textsubscript{3}) has an inversion symmetry, there are two classes of erbium ions with dipole moments pointing in opposite directions [10]. This leads to the splitting of the resonance frequency of the atoms when a homogeneous electric field is applied. Each frequency is shifted by \( \Delta \) or \( -\Delta \) with respect to the unperturbed absorption frequency \( \omega^\text{at} \). If the electric field intensity varies with the position in the sample, each atom experiences a different Stark shift, which leads to an additional inhomogeneous broadening.

When a light pulse is absorbed in an inhomogeneously broadened sample, the atoms in resonance with the light will be excited. While the excited atoms are in phase after the pulse is turned off, they are in a superradiant state, and a strong collective emission takes place in the forward mode defined by the input pulse. This emission will then decay when the atoms dephase due to inhomogeneous dephasing. The decay rate of the FID depends on the spectral distribution of the excited atoms. In the real experiment, the spectral distribution is basically given by the spectrum of the excitation pulse and the spectral width of the laser (assuming that
the homogeneous line width is smaller than the pulse and laser width). It is thus possible to accelerate the decay in a controlled way by broadening the spectral distribution of the atoms with the linear Stark effect, using an electric field gradient. The atoms are then no more in a superradiant state and the collective emission is inhibited. However, if no random phase is acquired during the process, it is possible to undo the inhomogeneous dephasing due to the electric field and to obtain a revival of the emission [16]–[18]. This can be realized by reversing the polarity of the electric field, which will reverse the detuning of each atom. The phase evolution will now be reversed and after a given time, all the atoms will be in phase again, leading to a collective emission of light.

More formally, this process can be written as follows: suppose that the phase of each atom evolves in time with frequency $\omega$, i.e. we can write for the phase evolution of the $j$th atom: $e^{-i\omega t}$. An external electric field will shift the frequency of the $j$th atom by $\Delta_j$. Suppose that electric field is turned on at time $t = 0$ and turned off at time $t = \tau$. The phase acquired by the atom is then $e^{-i(\omega t + \Delta_j)\tau}$. Now, suppose that instead of turning off the electric field, it is switched to the opposite polarity at time $t = \tau$. The frequency shift $\Delta_j$ then becomes $-\Delta_j$ and the phase of the atom at time $t$ is given by

$$e^{-i(\omega^0_{\Delta_j} + \Delta_j)\tau} e^{-i(\omega_{\Delta_j} - \Delta_j)(t-\tau)}.$$  

(2)

It can be seen that, at a time $t = 2\tau$, the externally introduced phase shifts $\Delta_j$ cancel out. If the optical transition has a natural inhomogeneous broadening, the atomic evolution due to the controlled dephasing and rephasing is superposed with the natural evolution due to the inhomogeneous dephasing. Hence, in the case of FID, the intensity of the light after the rephasing should reach the intensity of the unperturbed FID signal.

3. The experiment

In the experiment we used a Y$_2$SiO$_5$ crystal doped with erbium ions Er$^{3+}$ (with 10 ppm concentration). The atoms were excited on the transition $^4I_{15/2} \rightarrow ^4I_{13/2}$ at the telecom wavelength of 1536 nm [30]. The Y$_2$SiO$_5$ crystal has three mutually perpendicular optical-extinction axes labelled $D_1$, $D_2$ and $b$. The direction of light propagation $\vec{k}$ is along the $b$-axis. The dimensions of the Er$^{3+}$: Y$_2$SiO$_5$ crystal are 6 mm × 3.5 mm × 4 mm along the $b$-, $D_1$- and $D_2$-axis, respectively. The crystal was cooled to 2.6 K in a pulse tube cooler (Oxford Instruments). The inhomogeneous width is approximately 250 MHz and the optical peak absorption depth of the crystal is 2. In order to create the electric field gradient, we implemented a quadrupole scheme [22] using four electrodes attached directly to the crystal, perpendicular to the $D_2$-axis (see inset of figure 1). Such a configuration creates an electric field in the $D_2$ direction which changes linearly along the axis parallel to the light propagation. The electrodes were thin aluminium stripes, each of 1 mm width and spaced by 1.5 mm. To switch the electric field we used a fast electrical switch with a switching time of 10 ns and minimal/maximal voltage $-100/+100$ V.

The experimental setup is shown in figure 1. The light source was a free running external cavity diode laser (Toptica) at 1536 nm with laser bandwidth of approximately 2 MHz. The light was amplitude modulated by an AOM in order to create the excitation pulses at a repetition rate of 10 kHz, with duration 3 $\mu$s. The light was then coupled to a single mode optical fibre and passed through a variable fibre attenuator, before being focused in free space through the crystal.
Figure 1. Experimental scheme used to demonstrate the electric control of collective atomic coherences. Light pulses created with an acousto-optic modulator (AOM1) are attenuated with a fibre variable attenuator (VAR ATT) and focused through an Er$^{3+}$: Y$_2$SiO$_5$ crystal cooled at 2.6 K in a pulse tube cooler. The excitation pulses are then blocked by an optical gate implemented with a fibre AOM2 and the weak FID signal at the single photon level is detected with a superconducting single photon detector (SSPD). The inset shows the crystal with the quadrupole configuration of electrodes that produce the electric field gradient along the light propagation direction.

in the cryostat. The beam waist (diameter) in the crystal was 70 $\mu$m. After the crystal, the light was again coupled into a single mode fibre and sent through a fibre coupled AOM that served as optical gate in front of the detector to block the excitation pulses. With respect to the applications in quantum memories, where usually only few photons are absorbed, the measurements were made in the low excitation regime (with an excitation pulse area $\ll \pi$, typically with about $10^6$ photons in the excitation pulse). As the amplitude of the FID signal is strongly nonlinear with respect to the excitation intensity, the FID signal was extremely weak (about 50 photons) and was detected with a superconducting single photon detector [41].

4. Experimental collapses and revivals of collective spontaneous emission

We now present experimental results of the observation of collapse and revival of collective emission. In a first experiment, we apply a positive Stark pulse after the end of the optical pulse and we observe the decay rate of the FID for different values of electric fields. The results are shown in figure 2(a). We see that the decay becomes faster when the electric field increases, due to the applied broadening. For high value of electric fields, we also observe a small revival after the FID goes to zero. This is due to the spectral distribution of the induced broadening, which approaches a square shape when the field increases. In figure 2(b), we plot the
Figure 2. (a) Decay of the FID for different values of electric fields. The electric field is switched on after the pulse and kept constant afterwards. (b) Broadening of the excited atoms as a function of the voltage applied on the electrodes. The value plotted is the FWHM of the resonance frequency distribution given by the Fourier transform of the decay curves in (a).

Figure 3. Collapse and revival of collective emission for different electric field sequences. The unperturbed FID signal is represented by the dashed line. The dashed–dotted curve represents the damped FID signal when the electric field is not switched. The voltage applied on the electrodes is ±95 V. (a) Temporary revival obtained when the polarity of the electric field is reversed at time $\tau$ and the field remains constant afterwards. (b) Revival obtained when the polarity of the electric field is reversed at time $\tau$ and the field is turned off at time $2\tau$. The revived signal then follows the unperturbed FID signal.
Figure 4. Multiple collapse and revival obtained when the polarity of the electric field is reversed repeatedly. The voltage applied on the electrodes is $\pm 95\,V$.

full-width at half-maximum (FWHM) of spectral distribution, given by the Fourier transform of the FID decay. The linear dependence confirms that the spectral distribution of the excited atoms is proportional to the applied electric field. We then show that the controlled dephasing is reversible, by switching the polarity of the Stark pulse after a time $\tau$ and observing the revival of collective emission due to the rephasing of the atoms. Different sequences of electrical pulses can be used in order to manipulate the atomic coherence in the desired manner. In figure 3, we present two examples of such sequences. If the electric field is kept constant after the switching, the atoms are in phase again after a time $2\tau$ and we observe a temporary revival of the collective emission at this time (figure 3(a)). However, if the field is switched off to zero at the time $2\tau$, the controlled phase evolution is frozen and the natural inhomogeneous dephasing governs the evolution. In that case, the revived signal follows the unperturbed FID signal (figure 3(b)). The dashed curve represents the original unperturbed FID signal. If the electric field is switched repeatedly, it is also possible to induce multiple revivals, as shown in figure 4.

In all cases it is clearly observed that the quality of rephasing is excellent, i.e. the revived FID signal reaches almost the unperturbed signal. This suggests that the process of manipulating atomic coherence using electric fields does not introduce any substantial noise which would cause additional decoherence. In order to have a more quantitative estimation of the quality
of the rephasing, we induced temporary revivals at different times. We varied the duration of
the first Stark pulse, and hence the moment of the switching $\tau$. We measured the visibility $V$
of the revival as a function of $\tau$. $V$ is defined as $V = I_{\text{revival}}/I_{\text{FID}}$, where $I_{\text{revival}}$
is the maximal intensity of the revival and $I_{\text{FID}}$ is the intensity of the unperturbed FID at the
corresponding time. In order to have an accurate estimation of the visibility, it is important that the revival
and the reference measurement (unperturbed) FID are taken in the same experimental conditions.
Since the FID intensity is very dependent on the laser intensity and to a lesser extent on the laser
frequency, and that the typical measurement times are a few hundred seconds, it is difficult to
ensure the same experimental conditions for the two measurements. Particularly, the drift of the
laser frequency inside the inhomogeneous line can yield different results for two consecutive
measurements. In order to overcome this problem, we implemented a measurement sequence
with two subsequent optical excitation pulses within 10 $\mu$s. The electric field was applied for
one pulse, while the other pulse served as reference. The result is shown in figure 5. We observe
that the visibility stays constant above 0.95 within the error bars for all delays. In figure 5, we
also plot the time of the revivals as a function of $\tau$. We measure a slope of 1.99 $\pm$ 0.02, which
confirms that the revivals happen after a time $2\tau$.

These results show that the dephasing and rephasing of the collective atomic coherence
can be controlled to a very high degree. This is a crucial capability for applications in photonic
quantum storage based on CRIB. More generally, this ability to switch on and off at will
the collective emission of light from the sample is an interesting resource for quantum state
engineering and quantum state manipulation.

Figure 5. Visibility of the revival as a function of the first Stark pulse duration $\tau$. In order to obtain an estimation of the visibility free of intensity fluctuations, the measurement sequence is composed of two optical pulses. The electric field sequence is applied to one pulse, while the other pulse serves as reference (unperturbed FID). For long Stark pulses durations, the visibility is more difficult to estimate due to the low count rate. The error bars correspond to the statistical uncertainty of photons counts. The time of revival as a function of $\tau$ with a slope of 1.99 $\pm$ 0.02 is also plotted.
5. Conclusion

In conclusion, we have shown that the collective atomic coherence of an ensemble of erbium ions embedded in a solid state matrix can be controlled to a high degree on the timescale of tens of nanoseconds using external electric fields. Controlled inhomogeneous dephasing and rephasing was implemented using a reversible linear gradient of electric field on the crystal. We used optical FID to test our capacity to manipulate the atomic coherence. In particular, we showed that the controlled dephasing and rephasing of the atomic dipoles results in collapses and revivals of the collective emission of light from the sample. The experimental results show that the use of the electric field does not introduce any substantial decoherence and enables us to manipulate the atomic coherence efficiently on the timescale of tens of nanoseconds. It thus provides a useful resource in quantum information science, in particular for quantum storage applications.

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References

[1] Hammerer K, Sørensen A and Polzik E 2008 arXiv:0807.3358
[2] Tittel W et al 2008 arXiv:0810.0172
[3] Tordrup K, Negretti A and Molmer K 2008 Phys. Rev. Lett. 101 040501
[4] Wesenberg J et al 2009 Phys. Rev. Lett. 103 070502
[5] Macfarlane R M 2002 J. Lumin. 100 1
[6] Mossberg T W 1982 Opt. Lett. 7 77
[7] Lin H, Wang T and Mossberg T W 1995 Opt. Lett. 20 1658
[8] Macfarlane R and Shelby R 1987 Coherent Transients and Holeburning Spectroscopy in Rare Earth Ions in Solids: Spectroscopy of Crystals Containing Rare Earth Ions ed A Kaplyankii and R Macfarlane (Amsterdam: Elsevier)
[9] Ruggiero J, Gouet J-L, Simon C and Cheliere T 2009 Phys. Rev. A 79 053851
[10] Macfarlane R M 2007 J. Lumin. 125 156
[11] Wang Y P and Meltzer R S 1992 Phys. Rev. B 45 10119
[12] Meixner A J, Jefferson C M and Macfarlane R M 1992 Phys. Rev. B 46 5912
[13] Graf F R, Renn A, Wild U P and Mitsunaga M 1997 Phys. Rev. B 55 11225
[14] Graf F R et al 1997 Opt. Lett. 22 181
[15] Chelieire T et al 2008 Phys. Rev. B 77 245127
[16] Moiseev S A and Kröll S 2001 Phys. Rev. Lett. 87 173601
[17] Kraus B et al 2006 Phys. Rev. A 73 020302
[18] Nilsson M and Kröll S 2005 Opt. Commun. 247 393
[19] Sangouard N, Simon C, Afzelius M and Gisin N 2007 Phys. Rev. A 75 032327
[20] Hétet G et al 2008 Phys. Rev. Lett. 100 023601
[21] Longdell J J, Hetet G, Lam P K and Sellars M J 2008 Phys. Rev. A 78 032337
[22] Alexander A L, Longdell J J, Sellars M J and Manson N B 2006 Phys. Rev. Lett. 96 043602
[23] Alexander A L, Longdell J J and Sellars M J 2007 J. Opt. Soc. Am. B 24 2479
[24] Brewer R G and Shoemaker R L 1972 Phys. Rev. A 6 2001

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[25] Afzelius M et al 2007 New J. Phys. 9 413
[26] Duan L-M, Lukin M D, Cirac J I and Zoller P 2001 Nature 414 413
[27] Sangouard N et al 2007 Phys. Rev. A 76 050301
[28] Sangouard N et al 2008 Phys. Rev. A 77 062301
[29] Sun Y et al 2002 J. Lumin. 98 281
[30] Böttger T, Sun Y, Thiel C W and Cone R L 2006 Phys. Rev. B 74 075107
[31] Böttger T, Thiel C W, Sun Y and Cone R L 2006 Phys. Rev. B 73 075101
[32] Bottger T, Thiel C W, Cone R L and Sun Y 2009 Phys. Rev. B 79 115104
[33] Crozatier V et al 2007 J. Lumin. 127 65
[34] Guillot-Noël O et al 2006 Phys. Rev. B 74 214409
[35] Hastings-Simon S R et al 2006 Phys. Rev. B 78 085410
[36] Sun Y, Böttger T, Thiel C W and Cone R L 2008 Phys. Rev. B 77 085124
[37] Guillot-Noel O et al 2007 Phys. Rev. B 76 180408(R)
[38] Baldit E et al 2005 Phys. Rev. Lett. 95 143601
[39] Lauritzen B et al 2008 Phys. Rev. A 78 043402
[40] Lauritzen B et al 2009 arXiv:0908.2348v1
[41] Gol’tsman G N et al 2001 Appl. Phys. Lett. 79 705