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Interference of spontaneous emission of light from two solid-state atomic ensembles

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Abstract. We report an interference experiment of spontaneous emission of light from two distant solid-state ensembles of atoms that are coherently excited by a short laser pulse. The ensembles are erbium ions doped into two $LiNbO_3$ crystals with channel waveguides, which are placed in the two arms of a Mach–Zehnder interferometer. The light that is spontaneously emitted after the excitation pulse shows first-order interference. By a strong collective enhancement of the emission, the atoms behave as ideal two-level quantum systems and no which-path information is left in the atomic ensembles after emission of a photon. This results in a high fringe visibility of 95%, which implies that the observed spontaneous emission is highly coherent.

Contents

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1. Introduction

Spontaneous emission from atoms is one of the most commonly observed quantum effects in physics [1, 2]. Inherent to the emission is the randomness of the spontaneous process. Therefore, one may think that the spontaneous emission cannot be phase coherent with respect to an excitation laser, which is a point of view often repeated in textbooks on optics and lasers. However, the coherence properties of spontaneous emission have been thoroughly discussed theoretically, i.e. in the context of resonance fluorescence [3]–[5], superradiance [6], or optical free-induction decay (FID) [1, 7, 8]. In the case of resonance fluorescence experiments, for instance, subnatural linewidths have been observed using heterodyne measurements [9, 10], which demonstrates that resonance fluorescence emission can be highly coherent.

Another way of exploring the phase coherence of spontaneous emission is by performing interference experiments. Yet few reports on interference of spontaneous emission from atoms have been published. A pioneering interference experiment in this context was performed by Eichmann *et al* [11], where two trapped $^{198}Hg^{+}$ ions played the role of slits in a Young's double-slit experiment. At low laser intensities, they observed interference fringes in the resonance fluorescence from the two ions. This experiment has been thoroughly discussed [2], [12]–[15] and an interesting which-path interpretation has been given [12]. There it was argued that if excitation and emission take place in a closed two-level system, then the information about which path the photon took is erased from the atoms (quantum erasure [16]), and as a result interference is observed. However, if the emission leaves the atom in a state different than the initial state one could, in principle, know by which path the photon passed, and the interference pattern disappears. These two cases were explored in [11] by detecting either π- or σ-polarized light, where interference was observed in the former case but not in the latter. However, the visibility when observing π-polarized light was limited by a number of factors, including spontaneous Raman scattering to other states than the initial one.

Here, we present an experiment where erbium ions doped into two $LiNbO_3$ crystals, i.e. solid-state atomic ensembles, placed in the two paths of a Mach–Zehnder interferometer, are excited by a coherent laser pulse. We show that the spontaneous emission following the pulsed excitation, detected at the output of the interferometer, exhibits first-order interference with high visibility. The use of macroscopic atomic ensembles collectively enhances the spontaneous emission in the forward direction on the transition connected by the coherent excitation laser [1, 17]. This type of emission is also known as optical FID emission [1, 7], which has a N^2 intensity dependence on the number of atoms N since all atoms are initially spontaneously radiating in phase. The collective N^2 enhancement of the emission probability means that the spontaneous emission on the excited transition will dominate over emissions on other transitions. The ensembles can then be considered as being composed of ideal two-level atoms, as required for observing high-visibility interference from the which-path argument mentioned above. Due to the long coherence time of the optical transition we used, the collective spontaneous emission can be clearly separated in time from the excitation pulse making it possible to detect it. The resulting interference fringe visibilities are excellent ($V = 95\%$), clearly demonstrating that spontaneous emission of light can be coherent.

Our experiment relates closely to an experiment proposed by Mandel [18]. There he supposed that two-level atoms in two independent ensembles were prepared in two coherent superposition states with relative phase $\Delta\phi$. This could be done by exciting the ensembles with two coherent laser pulses having a phase difference $\Delta\phi$, as in the experiment discussed

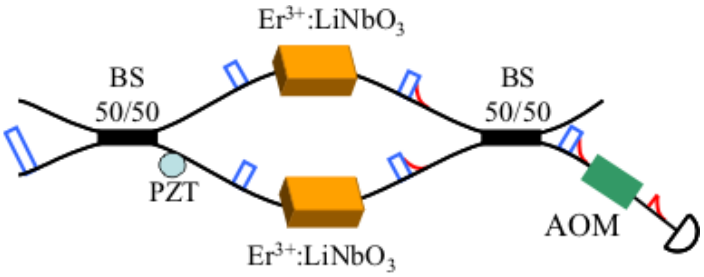

Figure 1. Experimental set-up for observing interference of collective spontaneous emission from two solid-state atomic ensembles. The excitation light pulse is created by intensity modulation of a cw external-cavity laser diode using a combination of acousto- and electro-optic modulators (not shown). The laser pulse is split into two pulses at a fiber 50/50 beam splitter (BS), which coherently excites the erbium ions doped into two $LiNbO_3$ waveguides. These are placed inside a pulse-tube cooler at a temperature of 3 K and separated by 7 cm. The collective spontaneous emission from the erbium ensembles is then combined at another 50/50 fiber BS, forming a balanced Mach–Zehnder interferometer. A piezo-electric transducer (PZT) is used to control the phase of the interferometer. In front of the detector, the acousto-optic modulator (AOM) serves as an optical gate to suppress the excitation pulse, in order to avoid saturating the detector.

in this paper. Mandel [18] then found that the spontaneous emission from the two ensembles detected on a screen would show first-order interference, provided that the phase difference $\Delta\phi$ remained sufficiently stable. The main difference as compared to our experiment is that we detect the emission in a single spatial mode and we instead observe first-order interference by slowly scanning the phase difference $\Delta\phi$.

In comparison with the interference experiment of [11], a main novelty of this experiment is the use of macroscopic solid-state ensembles having long optical coherence times and the resulting collective enhancement of the spontaneous emission. These features allow us to observe much higher fringe visibilities. We also have a significantly larger spatial distance between the ensembles ($\sim$7 cm compared to $\sim$5 μm). Another important difference is the pulsed excitation in our experiment, as compared to the continuous excitation in resonance fluorescence experiment [11]. This results in a clear separation in time of the excitation pulse and the detection, which means that the atoms evolve freely after excitation until spontaneous emission takes place. This also avoids some additional complications related to resonance fluorescence experiments, where frequency side bands appear in the emission at high laser intensities (the Mollow triplet) [3]–[5].

2. The interference experiment

An excitation pulse created by intensity-modulating the cw-light from an external-cavity diode laser excited erbium ions doped into two $LiNbO_3$ inorganic crystals placed in the arms of an Mach–Zehnder interferometer, see figure 1. The erbium ions absorbing within the frequency bandwidth of the laser pulse were coherently excited, creating a macroscopic

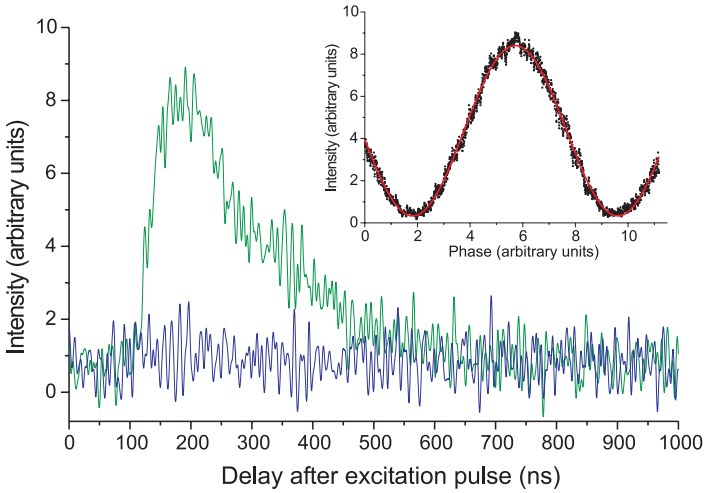

Figure 2. Interference of collective spontaneous emission in the high excitation regime. The graph shows the constructive (green) and destructive (blue) interference signals as a function of time after the end of the 2 μs long excitation pulse. The signals were detected by a classical detector. The optical detection gate was opened 130 ns after the excitation pulse, such that the signal the first 130 ns represents the detector noise level. Inset: the area under the signal (detector noise subtracted) as a function of phase difference. In this case, the measured interference visibility is $93 \pm 1.5\%$.

dipole moment in the two samples. Owing to the long optical coherence time of the transition (see below), a strong collective spontaneous emission (or FID emission) was observed after the excitation pulse (see figure 2). By collective, we mean that the spontaneous emission is enhanced by constructive interference in the forward direction along the spatial mode of the excitation laser, leading to an emission probability proportional to N^2, where N is the number of atoms in the excitation volume [1, 17]. In general, the FID emission decays due to inhomogeneous or homogeneous dephasing processes, as seen in figure 2. The collective enhancement only takes place in the forward direction on the excited transition, where an optical coherence has been induced. The spontaneous emission into other spatial modes and on other transitions is non-collective, therefore leading to an emission probability only proportional to the number of atoms N. We emphasize that, while non-collective spontaneous emission on the excited transition can be coherent, emission on other transitions is entirely incoherent.

The erbium ions were excited on the near-infrared transition $^4I_{15/2}$–$^4I_{13/2}$ at 1532 nm [19]. In general, rare-earth-metal-ion-doped solid-state materials have spectrally narrow absorption lines and excellent optical coherence properties at low temperatures (<4 K) [20]. The erbium ions can then be considered as a frozen gas naturally trapped in the crystalline host. In $Er^{3+}:LiNbO_3$ the $^4I_{15/2}$–$^4I_{13/2}$ absorption spectrum is inhomogeneously broadened to about 250 GHz by site-to-site variations in the static interaction between Er^{3+} ions and the $LiNbO_3$ host [21]. The homogeneous linewidth, however, is of the order of 30 kHz at the experimental temperature of $\sim$3 K [22], which corresponds to an optical coherence time of $T_2 \sim 10\,\mu$s. To obtain this coherence time, a small magnetic field (>0.1 Tesla) must be applied along the crystal

c-axis to reduce magnetic spin interactions in the material, which otherwise lead to fast optical decoherence [20, 21].

In our experiment, we used two Er^{3+}-doped $LiNbO_3$ waveguides (20 mm long and 10 mm wide). The $LiNbO_3$ crystal surfaces were doped with erbium ions by indiffusion, and optical channel waveguides (Ti-indiffused) were integrated on the surface [22, 23], allowing single-mode waveguiding of the 1.5 μm light through the entire interferometer. The waveguides were not identical because waveguide II had two times higher erbium doping concentration than waveguide I (waveguide I: $4 \times 10^{19}\,cm^{-3}$ surface concentration before indiffusion), resulting in a higher absorption in waveguide II. The fiber in one of the arms of the interferometer was partly coiled around a piezo element, which allowed control of the phase difference of the interferometer. The entire interferometer was installed in a pulse-tube-refrigerator. The $Er^{3+}:LiNbO_3$ waveguides were placed on the low-temperature level for cooling to about 3 K, whereas the 50/50 fiber BSs were placed at ambient temperature to ensure proper functioning. As a result, the arms of the interferometer were 2.63 m long. Note that there was then a temperature gradient of about 300 K across the interferometer. Since the fibers in the interferometer were not polarization maintaining, it was necessary to project the axis of polarization of the emission from the two ensembles on to a common axis. This was done by placing a fiber polarization controller (FPC) and a fiber polarizer (FP) in front of the detector (all outside the pulse-tube cooler). The total loss in each arm of the interferometer was roughly 14 dB, mostly due to input and output couplings of light between the single-mode fibers and waveguides. The AOM serving as optical gate and the FPC + FP introduced another 8 dB loss between the output of the interferometer and the detector.

To characterize the maximum visibility of the interferometer, we performed an interference experiment using a cw laser tuned off the erbium resonance (to 1550 nm). With the cooling system turned on, we obtained a maximum visibility of about 92%, whereas with the cooling system turned off about 100% was obtained (the absolute error estimated from several measurements were about 1% in both cases). In the former case, the visibility was clearly limited by phase noise introduced by vibrations in the pulse-tube cooler. The experiment was performed at a repetition rate of 13 Hz, which was found to limit the effect of vibrations on the phase noise. The long-term passive stability of the interferometer was then good enough to perform interferometric measurements over tens of minutes.

The experiment was carried out both in a high and low excitation regime. In the former, a strong excitation pulse was used, such that a classical detector could be used to detect the emission at the output of the interferometer. This resulted in a good signal-to-noise ratio and shorter integration times for each point on the interference curve. In the latter, we reduced the excitation pulse energy such that a single-photon detector could be used for detection. Although longer integration times were needed to obtain good signal-to-noise ratios, this experiment more clearly emphasizes the quantum nature of the spontaneous emission. In both cases, however, the experiment can be explained in terms of coherent states of light (bright or weak). In order not to saturate the detector, we used an AOM as an optical gate before the detector. In the high excitation experiment, the optical gate of 1 μs was opened 130 ns after the excitation pulse. In the low excitation experiment, the optical gate was opened 700 ns after the excitation pulse, whereas the 100 ns detection window of the single-photon detector was opened 1 μs after the excitation pulse.

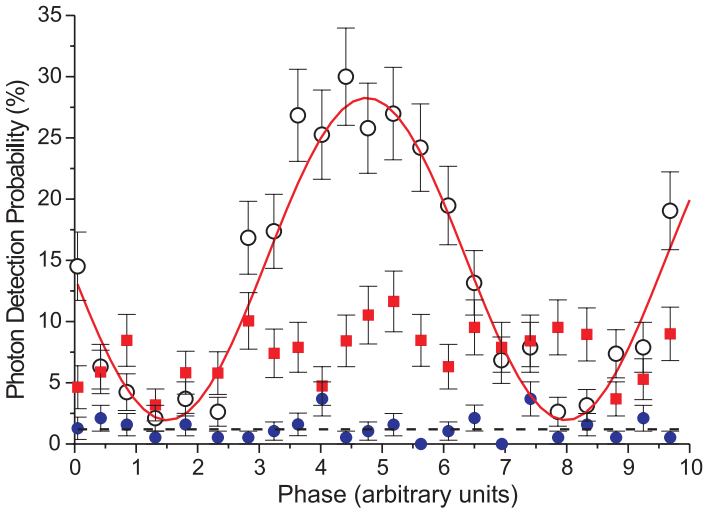

Figure 3. Interference in the low excitation regime. The points represent the measured detection probability as a function of the phase of the interferometer for three different experimental situations. For black open circles, the excitation laser is on resonance (1532 nm) and the light emitted by both ensembles is detected and shows clear interference. The solid line is a sinusoidal fit, leading to a net visibility of $95 \pm 5\%$. For blue filled circles, light from both ensembles is detected, but the excitation laser is far out of resonance (1550 nm). In this case, the detection probability drops to the level of dark noise of the single-photon detector (represented by the dashed line). For red filled squares, the laser is on resonance, but only the light emitted by one ensemble is detected, and as expected no interference fringes are observed.

3. Results and discussion

In the high excitation regime, the optical pulse had a duration of $2\,\mu$s and a peak power of 2 mW (4×10^{10} photons per pulse) at the entrance of the interferometer (see above concerning losses in the interferometer). In figure 2, the collective spontaneous emission is shown with the phase difference of the interferometer tuned to constructive and destructive interference. The maximum spontaneous emission signal (in front of the detector) was about 90 nW at constructive interference, corresponding to 7×10^4 photons/100 ns. The decay of the signal in this case is approximately $\sim$150 ns, which corresponds rather well to that obtained by numerically solving the Maxwell–Bloch equations using parameters corresponding to the current experiment. By measuring the area of the signal as a function of phase difference we obtained clear interference fringes, as shown in the inset of figure 2.

In the low excitation regime, the peak power of the pulse was reduced to $10\,\mu$W (2×10^8 photons per pulse). Since free-induction decay emission is a third-order non-linear process [7], this reduction was sufficient to make it possible to detect the emission using a single-photon detector. By recording the detection probability while scanning the phase difference of the interferometer, we observed interference fringes of $95 \pm 5\%$ visibilities (detector noise subtracted), as shown in figure 3. Note that this result is within the technical limit of 92% set by phase noise in the interferometer (see previous section). The detection probability at

constructive interference was 30%, which translates to 3 photons per 100 ns detection window in front of the detector (taking into account the 10% detection efficiency). The detector noise level was 1.2% due to dark counts. We verified that the photons detected were indeed emitted from the ensembles, and not laser light leaking through the intensity modulators. This was done by tuning the laser wavelength outside the optical resonance (to 1550 nm), such that no atoms were excited. As expected, the detection probability then dropped to the noise level of the detector, see figure 3, which proves that the intensity modulators provided good enough extinction to observe the few-photon spontaneous emission. To show that the interference is due to emission from both ensembles in the two arms, we 'turned off' the collective emission from one of the arms by removing the magnetic field on the corresponding sample. This reduces the optical coherence time by several orders of magnitude, which in turn drastically shortens the decay of the collective signal. The emission from this arm was then at a non-detectable level at the time of the single-photon detection window. As expected, the photon detection probability then showed no interference as a function of phase difference (see figure 3), and it dropped to about one-fourth of the constructive interference signal observed with the collective emission 'turned on' in both arms.

In order to understand this experiment, one may follow a single photon going through the interferometer. After the first BS, the photon is in a state of superposition of being in the two arms. The photon is then absorbed by the two ensembles, which are ideally ensembles of two-level quantum systems in resonance with the photon. The photon is now stored in both ensembles as a delocalized single excitation. After some time the photon is spontaneously emitted, the two modes are combined on the second BS, and the photon is thereafter detected by the single-photon detector. Only if the emitted photon is phase coherent with the absorbed photon can one observe perfect interference visibility. Hence the experiment presented here clearly and directly demonstrates the coherent nature of the observed spontaneous emission.

As discussed above, an important condition for observing interference is that only emission on the excited transition is observed. The detection of a photon emitted on another transition implies that the atom is left in another state than the initial one, and which-path information is left in the erbium-ion ensembles. In the case of Er^{3+}:$LiNbO_3$, the energy structure is very rich due to the different crystal-field (CF), Zeeman and hyperfine levels [19]. In this experiment, the atoms were excited from the lowest CF level in the electronic ground state $^4I_{15/2}(0)$ to the lowest CF level in the first electronically excited state $^4I_{13/2}(0)$. It is the collective enhancement on the excited $^4I_{13/2}(0)$–$^4I_{15/2}(0)$ transition that allows us to discriminate against emission to other states (particularly to other CF levels in the ground state). In this way, the atoms act as ideal two-level quantum systems, and no information about the previous excitation is left within the atoms. Note that no spectral filtering was used in the experiment.

In the introduction, we mentioned that a theoretical calculation of a thought experiment closely related to this experiment has been published by Mandel [18]. In particular, he calculates the expected visibility as a function of the number of atoms in each ensemble and the degree of excitation of the atoms. In the case when the number of atoms in each ensemble is the same, N, the theoretical visibility is [18]

$$V = \frac{N\cos^2(\theta/2)}{1+(N-1)\cos^2(\theta/2)},$$

where θ is the normal pulse area. If $N \gg 1$, as in the experiment presented here, the visibility is close to 1, almost independently of the excitation θ, except when all atoms are excited ($\theta = \pi$).

If $N = 1$, however, as in the case of the two trapped ions in the experiment presented in [11], the visibility becomes strongly dependent on the degree of excitation θ, and only at low excitation $\theta \approx 0$ does one observe perfect visibility [2, 18]. The use of large atomic ensembles presents an advantage also from this point of view.

Interference of light emitted by atoms has also been studied from a more applied perspective, because it plays a central role in quantum information research. In quantum networks, for instance, quantum states of light stored and retrieved from independent atomic memories would need to interfere with very high fringe visibilities [24]. In this context conditional first-order quantum interference of Raman photons produced by four-wave mixing in two three-level ensembles of cold atoms has been reported [25, 26]. There the emission of the interfering photons is also collectively enhanced, but simultaneous with the excitation laser (which is at a different frequency). The observation of interference is conditional on the detection of a first photon which projects the ensembles in a state with a delocalized collective atomic spin excitation. Note also that the fundamental effect of collective spontaneous emission observed in this paper is at the heart of photon echo techniques [1], which are being studied in the context of photonic quantum storage [22], [27]–[29].

4. Conclusions

To conclude, we have demonstrated high-visibility interference of the spontaneous emission of light from two spatially separated solid-state atomic ensembles. The high contrast observed has been made possible by the strong collective enhancement of the spontaneous emission which causes the multi-level erbium ions to behave as ensembles of ideal two-level systems. This clearly demonstrates that light spontaneously emitted from separated atomic systems can be highly coherent, provided that the initial excitation is coherent and that no which-path information is left in the atoms.

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