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Efficient solid state memories for quantum cryptography

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ABSTRACT

Long distance quantum cryptography requires quantum repeaters which use quantum memories. The latter are designed to store and retrieve photon quantum states on demand. Although quantum memories have been demonstrated in atomic vapors and ultra cold gases, a solid state alternative may better fulfill quantum memories requirements. Rare earth based crystals, which exhibit long coherence lifetimes, are actively studied for this purpose. Memory efficiency, i.e. the probability to retrieve a photon after storage, should be close to unity for practical applications. This can be achieved in highly doped crystals. Although Pr–Pr interactions could be detrimental in this case, we show that in a 3% Pr^{3+} doped $La_2(WO_4)_3$ crystal ground state hyperfine coherence lifetime is still close to that measured at low Pr concentration. Since the latter determines the memory storage time, this result suggests that highly doped crystals may be useful for efficient quantum memories.

1. Introduction

Quantum cryptography [1] is the most advanced application in the field of quantum information, being commercially available. It is based on two fundamental principles of quantum mechanics: (i) a system can exist in a superposition state; (ii) a measurement cannot gives full information on the system state. From these properties, protocols, like BB84 [2], have been proposed to exchange data encoded in quantum systems in a highly secured way. Indeed, if any spying occurs during data exchange, i.e. measurement is performed on the quantum system, it will be detected by the parties in communication. This highly secured communication is used to exchange a secret key with is then used to code messages. Quantum cryptography is therefore more precisely called quantum key distribution (QKD).

Quantum cryptography uses photons as quantum systems for their high velocity and robustness against electromagnetic perturbations. Data can be encoded in various quantum states like polarization or arrival time in the case of single photons. As in classical optical communications, a major limitation is due to light attenuation. For example, telecom optical fibers have an attenuation coefficient of 0.2 dB/km, so that optical amplifiers are needed after $\approx$100 km propagation. In quantum cryptography, these amplifiers are useless since they could not preserve or regenerate photons quantum states. As a result, QKD is limited to 250 km for useful data exchange rates [3]. To overcome light attenuation, quantum repeaters have been proposed [4]. These devices rely on entanglement swapping between distant sources of entangled photon pairs. A long distance is broken into smaller links and repeated entanglement swapping is performed until two entangled photons are shared by the communicating parties. Then, teleportation can be used to exchange data. However, entanglement generation and swapping are highly probabilistic and quantum repeaters have to include storage and on demand retrieval of photons quantum states. A quantum memory (QM) is designed for this task [5,6].

2. Quantum memories

Storage in a QM is achieved by absorbing the photon into a material system, which creates an atomic superposition state. As long as the coherence lifetime (T_2) of the corresponding transition is not exceeded, photon quantum state can be faithfully retrieved by using specially designed protocols (see below).

Requirements for QM are the following:

- high efficiency, i.e. a ratio between absorbed and retrieved photons close to unity;
- high fidelity of the quantum state of the outgoing photon compared to that of the incoming photon;
- long storage time, which translates in long coherence lifetime for the transition mapping photon quantum state;
- large bandwidth for storing short photon pulses generated by the high rate photon pair sources;
- multimode storage or ability to separately store several photons.

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[1] In memoriam.

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The last two points are especially important for quantum cryptography which requires reasonably high QKD exchange rate for practical applications [7].

Quantum memories have been demonstrated in atomic vapors and ultra cold clouds [8–10]. These systems exhibit long coherence lifetimes, even at room temperature in some cases, but do not entirely fulfill the QM requirements for large bandwidth and multimode storage. Moreover, coherence lifetimes, limited by atom movements, do not exceed about 1 ms. This limit could be overcome in the solid state. Crystals based on rare earth ions represent a very promising approach for longer coherence lifetimes and could even better match QM requirements of bandwidth and multimode storage [11].

3. Quantum memories based on rare earth doped crystals

QM protocols rely on an atomic three level Λ system (Fig. 1). It consists of two ground state levels connected to a single excited one by two optical transitions of similar strengths. A possible storage scheme is the following: the incoming photon is absorbed along the $s \rightarrow e$ transition, creating a coherence between these levels. Before this coherence vanishes, a second light pulse is sent along the $c \rightarrow e$ transition in order to completely exchange populations between c and e levels. In this way, the original coherence, and therefore the quantum state of the photon, has been mapped to the $s \rightarrow c$ transition. This allows one to overcome the excited state population lifetime limitation and to take advantage of the long coherence lifetimes found in nuclear spin transitions for example. As long as the ground state $s \rightarrow c$ transition dephasing is negligible, the photon quantum state is stored. For retrieval, a pulse is again applied to the $c \rightarrow e$ transition, converting back the ground state coherence to the optical one and causing the emission of a photon in a quantum state identical to the initial one.

In this scheme, QM storage time equals the coherence lifetime of the s→c transition, which is therefore a fundamental parameter for a QM material. On the other hand, the coherence lifetimes of the optical transitions should only be long enough to enable the transfer to the s→c transition. In some QM protocols, like electromagnetically induced transparency [12,13], the optical transition T_2 plays no direct role.

Suitable three level Λ systems for QM can be found in rare earth doped crystals. Indeed, these ions exhibit many optical transitions and, in several cases, isotopes with non-zero nuclear spins like ^{141}Pr ($I=5/2$, 100% abundance) or ^{151}Eu and ^{153}Eu ($I=5/2$, respective abundances 47.8% and 52.8%). The ground state levels can therefore be chosen among hyperfine levels appearing as a substructure of electronic (crystal field) levels. Moreover, at temperatures around 4 K, hyperfine coherence lifetimes can reach 15 ms [14] and up to several tens seconds under an external magnetic field and RF pulses [15]. Optical coherence lifetimes can also be long, up to 4 ms [16]. The two transitions of the Λ can be allowed in low site symmetry or by applying a suitable external magnetic field [17,18]. This is because the latter mixes hyperfine levels with different nuclear spin projections with weights depending on the electronic levels considered.

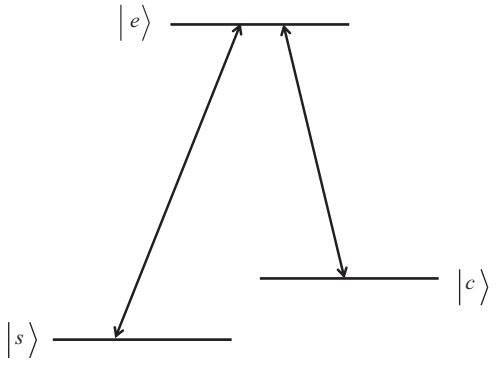

Fig. 1. A three level Λ system. *s* signal ground state level, *c* control ground state level, *e* excited level.

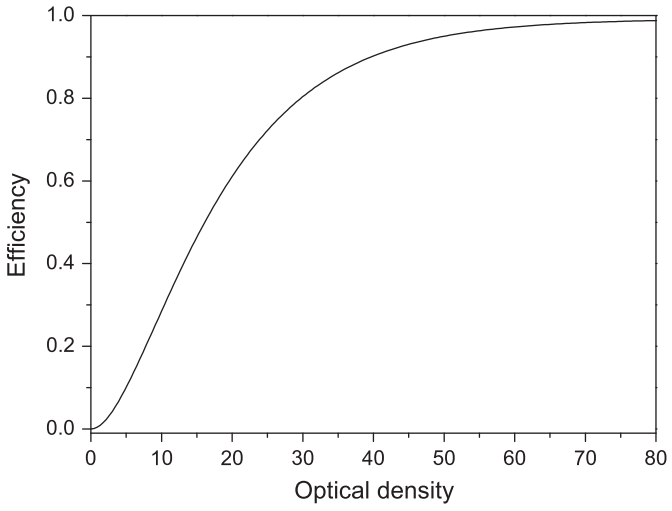

Fig. 2. Efficiency of the AFC protocol as a function of the optical density.

In the QM process described above, several effects have been neglected like light propagation in the crystal and transitions inhomogeneous broadenings. These processes tend to reduce QM efficiency and or fidelity. This explains that QM protocols with high efficiency are more complex. Examples include controlled inhomogeneous reversible broadening (CRIB) [19], gradient echo memories (GEM) [20], atomic frequency comb (AFC) [21] and spectral hole burning [22]. All these protocols store one photon in an ensemble of atoms and can therefore store multiple photons, although efficiency may depend on the memory bandwidth, i.e. the number of photons stored. These protocols have been studied in Pr:Y_2SiO_5 [23], Eu:Y_2SiO_5 [20], Tm:$Y_3Al_5O_{12}$ [24], Er:Y_2SiO_5 [25], Nd:YVO_4 [21] and Nd:Y_2SiO_5 [26]. High efficiencies (up to 45%) have been demonstrated [11], as well as single photon level storage and recall [21,24,25], transfer to the spin coherence [27] and multimode storage [27]. However, a long storage, highly efficient and large bandwidth QM based on a rare earth doped crystal has not been yet demonstrated. One difficulty is that high efficiencies require high optical densities. As an example, efficiency of the AFC protocol [28] is plotted as a function of the optical density in Fig. 2. One way to increase optical density is to increase rare earth concentration in the crystal. However, this could results in a decrease of coherence lifetimes because or rare earth–rare earth interactions. In the next section, recent results on a highly doped Pr:$La_2(WO_4)_3$ sample are presented.

4. Coherence lifetimes in Pr:$La_2(WO_4)_3$

Experiments were performed at 3.2 K on a 3% Pr:$La_2(WO_4)_3$ sample (2×10^{20} ions/cm^3). Excitation was provided by a single mode dye laser (Coherent 899-21, 1 MHz bandwidth) at 602.74 nm (vacuum) in resonance with praseodymium $^3H_4(0)\rightarrow{}^1D_2(0)$ transition. Pulse frequencies and amplitudes were set by two independent AA acousto-optic modulators in double pass configuration. Light was detected by a 10 MHz bandwidth Hamamatsu C5460 avalanche photodiode.

In Pr:$La_2(WO_4)_3$, praseodymium ions substitute lanthanum ions. Since they have similar atomic radii (1.14 and 1.18 Å, respectively

[29]), doping related strains are reduced [30]. This is favorable to high peak absorption coefficients. In the 3% Pr doped sample, the optical inhomogeneous linewidth Γ_{inh} is 42 GHz, which is only 4.7 times the linewidth measured in a 0.02% Pr doped sample. In comparison, $\Gamma_{inh} = 30$ GHz in a Pr:Y_2SiO_5 crystal at 1.6×10^{19} ions/cm^3 doping level. In this case, the mismatch between Pr and Y atomic radii (r_Y=1.02 Å) is much larger. The peak absorption coefficient in the 3% Pr:$La_2(WO_4)_3$ sample is 23 times larger than that in the 0.02% sample but reaches only 28 cm^{-1} because of the low oscillator strength of the $^3H_4(0) \rightarrow {}^1D_2(0)$ transition.

As mentioned above, hyperfine transitions coherence lifetimes need to be long for a QM material. This was investigated in a Raman echo experiment, using two-color optical excitation and heterodyne detection [31]. The Raman echo decay corresponding to $\pm 1/2g \rightarrow \pm 3/2g$ ground state hyperfine transition is shown in Fig. 3. At times shorter than 100 μs, the Raman free induction decay adds to the echo signal. A fit by a two exponential law gives a hyperfine coherence lifetime of ≈ 250 μs, similar to the low concentration value. This result shows that Pr–Pr interactions are small on the time scale of the Raman coherence lifetime and suggests that such a highly doped material could be used as QM material.

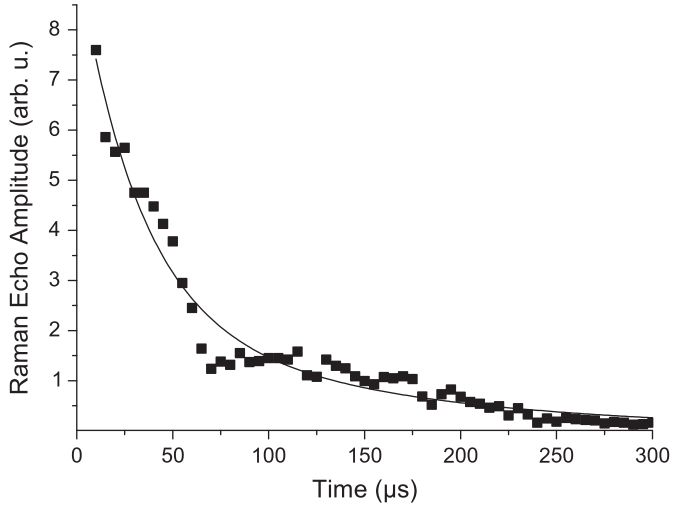

Fig. 3. Raman echo amplitude for the $\pm 1/2g \rightarrow \pm 3/2g$ transition as a function of the pulse separation in a 3% Pr:$La_2(WO_3)_4$ sample (squares) and fit by a two exponential function (solid line).

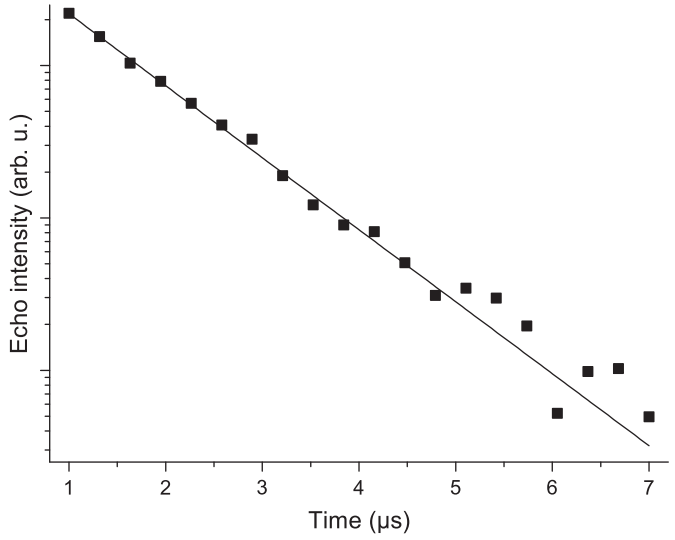

Fig. 4. Photon echo decay ($^3H_4(0) \rightarrow {}^1D_2(0)$ transition) as a function of the pulse separation in a 3% Pr:$La_2(WO_3)_4$ sample.

Optical transition ($^3H_4(0) \rightarrow {}^1D_2(0)$) coherence lifetime was also measured using two pulse photon echoes (Fig. 4). For the 3% doped sample, we found $T_2 = 4.5$ μs, much shorter than the value found in the 0.02 % sample where $T_2 = 20$ μs. In this case, Pr–Pr interactions are much larger and mainly due to energy transfer by a cross-relaxation involving ground and excited states : $[^1D_2, {}^3H_4] \rightarrow [^1G_4, {}^3F_4]$ [32]. As a result, $^3H_4(0) \rightarrow {}^1D_2(0)$ coherence lifetime is limited by the population lifetime of the $^1D_2$ level (6 μs). Although the T_2 value is quite small, it still larger than a typical optical π pulse which duration is about 1 μs. Transfer of the optical coherence to the spin one should therefore still be possible.

5. Conclusion

Quantum cryptography allows one to transmit data in a highly secure way. It is currently limited to a few hundreds kilometers because of light attenuation. To extend it over larger distances, quantum repeaters are necessary. These devices use quantum memories which are able to store and retrieve a photon quantum state. Several stringent requirements have to be fulfilled by quantum memories in order to achieve usable data exchange rates. They could be obtained in solid state memories using rare earth doped crystals. Indeed, several memory protocols have been successfully investigated in these systems. Several problems still need to be solved including improving the memory efficiency, i.e. the probability to retrieve a photon after storage. This can be obtained in materials with very high optical densities. For this purpose, we studied a crystal with a low inhomogeneous linewidth even at high doping level, Pr:$La_2(WO_4)_3$. We found that hyperfine coherence lifetime is about 250 μs in a 3% Pr doped sample, similar to what was measured in a low concentration sample. Since this value determines the quantum memory storage time, this result suggests that concentrated crystals can be useful for this application.

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