Quantum storage in rare-earth-doped crystals for secure networks

O. Guillot-Noël\textsuperscript{a,}*, Ph. Goldner\textsuperscript{a}, E. Antic-Fidancev\textsuperscript{a}, A. Louchet\textsuperscript{b}, J.L. Le Gouët\textsuperscript{b}, F. Bretenaker\textsuperscript{b}, I. Lorgère\textsuperscript{b}

\textsuperscript{a}Laboratoire de Chimie Appliquée de l’Etat Solide, CNRS-UMR 7574, ENSCP, 11, rue Pierre et Marie Curie F-75231 Paris Cedex, France
\textsuperscript{b}Laboratoire Aimé Cotton, CNRS-UPR 3321, Bât. 505, 91405 Orsay Cedex, France

Available online 15 March 2006

Abstract

Quantum storage of photons in an atomic ensemble can be obtained by using three-level Λ systems. In these systems, two levels are coupled by optical transitions to a third one. Rare-earth ion-doped crystals are attractive materials for quantum storage because their hyperfine levels can have coherence lifetimes longer than 100 μs and thus can be used to build Λ systems. Tm\textsuperscript{3+} ions are especially interesting since they can be excited by ultra-stable laser diodes. The influence of an external magnetic field has been studied in order to obtain an efficient three-level Λ system with the hyperfine levels of the rare earth. The particular case of the Tm\textsuperscript{3+} ion in the Y\textsubscript{3}Al\textsubscript{5}O\textsubscript{12} host is discussed.

© 2006 Elsevier B.V. All rights reserved.

Keywords: Quantum storage; Rare-earth ion-doped crystals; Three-level Λ systems

1. Introduction

Quantum cryptography has recently emerged as a fully operating technique for extremely secure transmission of information in short-distance networks. The quantum keys are carried by photons and are distributed through optical fibers. However, long-distance communication is still challenging because of attenuation in the fibers. The needed quantum repeaters [1] rely on quantum information storage, a function that has not been implemented yet.

Several groups have been pursuing the ambitious objective of storing optically carried quantum information into an atomic ensemble entangled state and then returning quantum data to the optical carrier.

Fig. 1 represents a typical three-level system where Ω\textsubscript{1} and Ω\textsubscript{2} are the Rabi frequencies which characterize the strength of the atom–laser interaction. By adequate coherent combination of the two resonantly excited optical transitions, the atomic Raman coherence could be combined with the radiation in a single quantum state. So far, ensemble entanglement has been demonstrated only in atomic vapors [2] or in laser-cooled atom clouds [3]. However rare-earth ion-doped crystals (REIC) also appear as promising candidates in the quest for macroscopic quantum effects. They offer properties similar to atomic vapors with the advantage of no atomic diffusion. Hyperfine levels of rare-earth ions are commonly used to perform coherent spectroscopy experiments, like hole burning and photon echo. At low temperature (< 4 K) the optical coherence lifetime may reach several ms in these materials and a hyperfine coherence lifetime of 82 ms has been reported in Pr\textsuperscript{3+}:Y\textsubscript{2}SiO\textsubscript{5} [4].

Further investigation of systems in REIC is hampered by two main problems: (i) the use of hyperfine levels of rare-earth ions for Λ systems is limited by the ΔM\textsubscript{F} = 0 selection rule where M\textsubscript{F} is the nuclear spin projection, (ii) the lack of adequate laser sources in the spectral range of the most attractive rare-earth ions.

In this work, we show that Tm\textsuperscript{3+}-doped compounds can be very good candidates for three-level systems [5]. By applying an external magnetic field, it is possible to relax the ΔM\textsubscript{F} selection rule and then to control the relative strengths of the |1⟩→|3⟩ and |2⟩→|3⟩ optical transitions. Moreover, this ion can be excited by ultra-stable semiconductor lasers in the $^3H_6(0) \rightarrow ^3H_4(0)$ transition (around 790 nm) [6]. Such lasers can be stabilized easily to sub-kHz line width and jitter. The particular case of the Tm\textsuperscript{3+} ion in the Y\textsubscript{3}Al\textsubscript{5}O\textsubscript{12} host (YAG) is discussed.
2. Theoretical background

Tm$^{3+}$ is a non-Kramers’ ion with 12 f electrons and a single natural isotope ($^{169}$Tm) with I = 1/2 nuclear spin. For low point site symmetry, Tm$^{3+}$ electronic levels are singlets. Application of an external magnetic field removes the nuclear spin degeneracy and splits the electronic levels. A three-level Λ system with Tm$^{3+}$ ions would involve the two hyperfine levels of the ground state $^3$H_d(0) and one hyperfine level of the excited state $^3$H_d(0).

Levels and wave functions calculations are performed using the following Hamiltonian: $H = H_{\text{FI}} + H_{\text{CF}} + H_{\text{EZ}} + H_{\text{HF}} + H_{\text{NZ}}$ where $H_{\text{FI}}$ is the free-ion Hamiltonian. $H_{\text{FI}}$ depends on eight parameters, the $E^2$ Racah parameters ($k = 0,1,2,3$), the $x$, $\beta$, and $\gamma$ Trees parameters and the spin–orbit coupling constant $\zeta_{\text{4f}}$. $H_{\text{CF}}$ is the crystal-field Hamiltonian. Tm$^{3+}$ ion substitutes Y$^{3+}$ ion in YAG in a D2 point site symmetry which involves eight non-zero real $B_g^k$ crystal-field parameters, namely $B_0^g$, $B_2^g$, $B_4^g$, $B_6^g$, $B_8^g$, $B_{10}^g$, $B_{12}^g$, and $B_{14}^g$. $H_{\text{EZ}}$ and $H_{\text{NZ}}$ are the electronic and nuclear Hamiltonian, respectively. $H_{\text{HF}}$ is the hyperfine Hamiltonian.

The splittings between the hyperfine levels of the $^3$H_d(0) and $^3$H_d(0) states as well as their wave function as a function of the $B_0$ external magnetic-field direction are calculated by the complete diagonalization of $H$. For each orientation of the magnetic field, the nuclear part of the wave function is separated from its electronic part and the transition strengths ratio $R = |\langle 2|3\rangle_{\text{nucel}}|^2 / |\langle 1|3\rangle_{\text{nucel}}|^2$ between the $|1\rangle \rightarrow |3\rangle$ and $|2\rangle \rightarrow |3\rangle$ transitions is calculated (nucel means nuclear part of the wave function).

3. Results and discussion

In the case of Tm:YAG, the energy levels of Ref. [7] are used to calculate the free-ion and the crystal-field parameters. A rms standard deviation of 20.8 cm$^{-1}$ is found by using 71 experimental levels among the 91 levels of the 4f$^{13}$ configuration. We obtain: $E^0 = 17443(1)$ cm$^{-1}$, $E^1 = 7018(2)$ cm$^{-1}$, $E^2 = 33.38(2)$ cm$^{-1}$, and $E^3 = 671.5(1)$ cm$^{-1}$ for the Racah parameters; $x = 17.06(6)$ cm$^{-1}$, $\beta = -658(7)$ cm$^{-1}$, and $\gamma = 0$ cm$^{-1}$ for the Trees parameters and $\zeta_{\text{4f}} = 2619.7(9)$ cm$^{-1}$ for the spin–orbit coupling constant. The crystal-field parameters are: $B_0^g = 580(12)$ cm$^{-1}$, $B_2^g = -59(29)$ cm$^{-1}$, $B_4^g = -1223(36)$ cm$^{-1}$, $B_6^g = 29(9)$ cm$^{-1}$, $B_8^g = -1403(16)$ cm$^{-1}$, $B_{10}^g = -716(19)$ cm$^{-1}$, $B_{12}^g = -331(22)$ cm$^{-1}$, $B_{14}^g = 460(28)$ cm$^{-1}$, and $B_{16}^g = -369(22)$ cm$^{-1}$.

From the complete diagonalization of $H$, the calculated wave functions allow us to determine the transition strengths ratio $R$ between the hyperfine levels of the $^3$H_d(0) and of the $^3$H_d(0) states as a function of the external magnetic field direction (Fig. 2). The branching ratio is strongly dependant on the orientation of the external magnetic field. This orientation is expressed in the local (x, y, z) crystal-field axes. A maximum value of $R = 0.24$ ($\sim 1:4$), showing that both transitions are allowed, is found in the x–y plane with $\theta = 90^\circ$ and $\phi = 6^\circ$ (Fig. 2). This non-vanishing value is due to the coupling effect of the electronic Zeeman and the hyperfine interactions. This coupling effect is strongly dependent on the crystal-field splitting of the J multiplet and thus leads to different hyperfine splittings for the ground and excited states which in turn induces different $M_I$ mixing in the $^3$H_d(0) and $^3$H_d(0) states. This effect releases the nuclear spin selection rules $\Delta M_I = 0$ making the Λ system effective.
Moreover, we have to prepare the crystal in such a way that absorption at a given frequency be ascribed to a single class of ions, exhibiting the same three-level system, with the same level spacing and the same transition strength on each one of the two lines. There are six identical sites for Tm\(^{3+}\) in YAG which are related to each other by cubic group symmetry operations. These sites have the same point symmetry, D\(_{2}\), but are oriented in different directions, as shown in Fig. 3. When an external magnetic field is applied, the six sites generally become non-equivalent, which means they are characterized by different hyperfine splittings and different values of the transition strengths ratio \(R\). We can take advantage of the different splitting values to select ions in a specific site, with optimized transition strength ratio. For magnetic fields directed along some cubic symmetry axes, the number of inequivalent sites is reduced. For example, when \(B_0\) is along the [111] direction, sites 1, 3, 5 as well as sites 2, 4, 6 are equivalent. For the \(^3\)H\(_4\)(0)\(\rightarrow\)^3\)H\(_0\)(0) transition, the electric dipole moment is oriented along the \(y\) axis ([110] direction) [8]. This means that when carefully chosen the external magnetic field direction and light polarization allows one to study a restricted number of sites.

Our YAG sample is a 5 mm thick 0.1% Tm-doped compound which is cut perpendicular to the [1\(\bar{1}\)0] axis. We have thus to find with this sample geometry, a convenient experimental combination of magnetic field and laser polarization orientations which gives a high branching ratio and a restricted number of magnetically inequivalent sites.

For a magnetic field along the [111] direction and by polarizing the laser along the [111] direction, only sites 1, 3, 5 are excited since the electric field has no \(y\)-component for sites 2, 4, 6. Unfortunately, the calculated branching ratio \(R = 5 \times 10^{-4}\) is very small. For a magnetic field in the ([110], [001]) plane and for a laser polarized along the [111] direction, sites 1, 3, 5 are not excited and sites 4 and 6 are magnetically equivalent. From the previous calculations, with a magnetic field around the [111] direction in the ([110], [001]) plane, large values of \(R\) are expected. The maximum branching ratio of 0.24 is found at 49° away from the [001] axis.

4. Conclusion

The magnetic interactions in Tm:YAG crystals have been studied under an external magnetic field in order to obtain an efficient three-level \(\Lambda\) system. Tm\(^{3+}\) ions appear to be a good candidate for such purpose. A complete calculation including free-ion, crystal-field and magnetic interactions has been performed as function of the external magnetic field direction. An efficient three-level \(\Lambda\) system is obtained with a maximum transition strengths ratio \(R = 0.24\) in the \(\theta = 90^\circ\) and \(\phi = 6^\circ\) direction. This study allows us to find a convenient combination of magnetic field and laser polarization orientations well-adapted to the crystal geometry. With a Tm:YAG sample cut perpendicular to the [1\(\bar{1}\)0] axis, an optimized configuration with a high branching ratio for the optically active sites can be found with the magnetic field in the ([110], [001]) plane and the laser polarized along the [111] direction.

Acknowledgment

This work was carried out under the ESQUIRE project supported by the IST-FET program of the EC.

References