Hole burning study of Tm$^{3+}$:YAG hyperfine structure for quantum storage applications

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Available online 8 February 2006

Abstract

Quantum storage, which aims at transferring photon quantum states into matter, can be obtained by using an ensemble of atoms whose levels form a three level $A$ system. In these systems, two optical transitions couple two levels to a third one. This quantum storage scheme could be obtained with rare-earth ions in single crystals, since their optical and ground-state hyperfine transitions can exhibit long coherence lifetimes and their hyperfine structures could be used to build a three level $A$ system. Tm$^{3+}$ ions in Y$_3$Al$_5$O$_{12}$ are especially interesting since the $^3H_6$-$^3H_4$ transition can be driven by ultra-stable laser diodes. However, the selection rules on the nuclear spin projection can forbid the simultaneous coupling of two levels to a third one. In this paper, the hyperfine structure of Tm$^{3+}$ is investigated by hole burning spectroscopy under a magnetic field and compared to theoretical calculations based on crystal field calculations. The experimental results are found to be in good agreement with theory and show that some magnetic field orientations are able to relax the selection rules on the nuclear spin projection.

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Keywords: Quantum storage; Hyperfine structure; Hole burning spectroscopy

1. Introduction

Quantum storage consists in the transfer of the quantum state of a photon to an atom or an atomic ensemble and has been demonstrated in gases [1]. This process raises interest both in the fundamental and applied fields, since a quantum storage device could be used to build quantum repeaters. Such systems could increase the distance over which quantum cryptography through optical fibers can be used [2]. To obtain quantum storage, atoms in which a three-level $A$ system can be found are very useful [3]. Such a system includes three levels which can be connected by two optical
transitions, as shown in Fig. 1. An efficient system will have optical and ground-state hyperfine transitions with long coherence lifetimes, similar intensities for both optical transitions and will also be controlled by an ultra-stable laser source. These criteria can be well fulfilled by rare-earth ions in single crystals at liquid helium temperature. Indeed, slow and stopped light has been demonstrated with a three-level \( L \) system in Pr\(^{3+} \) using the hyperfine levels of the \( ^3H_4(0) \) and \( ^3H_6(0) \) crystal field levels (\( I = \frac{1}{2} \)). The arrows correspond to the optical transitions. If the nuclear spin projection is a good quantum number, the 2–3 transition is forbidden.

![Fig. 1. A three-level \( L \) system in Tm\(^{3+} \) using the hyperfine levels of the \( ^3H_4(0) \) and \( ^3H_6(0) \) crystal field levels (\( I = \frac{1}{2} \)). The arrows correspond to the optical transitions. If the nuclear spin projection is a good quantum number, the 2–3 transition is forbidden.](image)

(see Fig. 1) can be impossible if the nuclear spin projection \( M_I \) is a good quantum number. Since an optical transition cannot flip the nuclear spin, the selection rule \( \Delta M_I = 0 \) will forbid the 2–3 transition. We investigated this problem for Pr\(^{3+} \) and Tm\(^{3+} \) ions and showed theoretically that an external magnetic field can induce very different nuclear spin projections mixings in ground and excited levels and thus can relax the \( \Delta M_I = 0 \) selection rule \([7–9]\). In the case of Tm\(^{3+} \) in YAG, calculated branching ratio \( R \) between the 2–3 and 1–3 transitions can have values as high as 0.24 \([7]\) with an magnetic field oriented slightly away from the [1 1 1\] crystallographic axis.

Hole burning experiments have been performed in a Tm\(^{3+} \):YAG sample to confirm the calculations and preliminary results have been previously reported \([10]\). In this paper, a new experimental spectrum is presented and discussed.

2. Hyperfine structure of Tm\(^{3+} \) ions

Tm\(^{3+} \) has an even number of electrons which results in a complete quenching of the total angular momentum in low symmetry sites and a zero hyperfine interaction at first order. The nuclear spin of thulium single natural isotope \(^{169}\text{Tm} \) is \( \frac{1}{2} \), leading to vanishing second-order hyperfine and quadrupole interactions. This explains the lack of hyperfine structure for Tm\(^{3+} \) ions in \( Y_2Al_5O_{12} \) crystal, since they occupy a site of \( D_2 \) symmetry. However, if an external magnetic field is applied, the combination of the electronic Zeeman and second-order hyperfine interactions can lead to large hyperfine splittings. The effect of the nuclear Zeeman interaction is often much smaller. In addition, for some orientations of the magnetic field, the hyperfine levels of different crystal fields levels can exhibit different mixings of the pure \( M_I \) levels. This effect can be discussed by using a spin Hamiltonian which has the form

\[
H_{SH} = \sum_{i=x,y,z} \gamma_i B_0 I_i,
\]

where \( B_0 \) is the magnetic field and \( \gamma_i \) the principal values of the effective enhanced nuclear gyromagnetic
tensor. It depends on the CF level because of the electronic Zeeman and second-order hyperfine interactions

$$\gamma_i = -g_n\beta_n - 2g\beta A_{ii},$$

with

$$A_{ii} = \sum_{n \neq 0} A \frac{|\langle 0|J|n\rangle|^2}{E_n - E_0},$$

where $g_n(g)$ is the nuclear (electronic) $g$-factor and $\beta_n(\beta)$ the nuclear (electronic) Bohr magneton. $A$ is the hyperfine constant and $J$ the total angular momentum. The index 0 denotes the first (lowest energy) CF level ($^3H_6(0)$ or $^3H_4(0)$), $n$ the other CF-split levels of the considered multiplet and $E_i$ is the energy of level $i$. If the gyromagnetic tensor is different enough in the $^3H_6(0)$ or $^3H_4(0)$ levels, it is possible to relax the selection rules on the nuclear spin projections and induce simultaneously the 1–3 and 2–3 transitions. Calculations of the $\gamma$ tensor has been performed using a complete Hamiltonian which included free ion and CF parameters as well as the magnetic effects: electronic and nuclear Zeeman interactions and the hyperfine interaction. In the case of the $^3H_6(0)$ and $^3H_4(0)$ levels, the calculated intensity ratios between the 1–3 and 2–3 transitions can reach 0.24. To confirm this very promising values, the $\gamma$ tensor has been investigated experimentally by using hole burning spectroscopy.

3. Experimental results

Measurements were performed on a 5 mm thick 0.1 at% Tm$^{3+}$:YAG sample. The crystal was cut perpendicular to the [1 1 0] axis. Hole burning spectra were obtained between 1.5 and 4.2 K using a stabilized laser diode operating at 793 nm and a 4500 G magnetic field. The laser spot diameter on the sample was 800 μm. The holes were burnt at center frequency in 10 shots of 50 μs and the transition probed in 750 μs over a 20 MHz interval. To ensure a nearly complete relaxation to the ground-state after burning, the probe sequence was started 10 ms after the last burning pulse. This value was chosen to correspond to the lifetime of the intermediate $^3F_4$ multiplet.

Tm$^{3+}$ ions can occupy six crystallographic sites in YAG. All these sites are identical but have different orientations with respect to the crystal axes (Fig. 2). This brings an additional complexity since, in general, a magnetic field will produce six different hyperfine splittings and branching ratios. However, if $B_0$ is oriented along some directions of the crystal cubic symmetry axes, some sites will be magnetically equivalent. Moreover, the electric dipole moment of the $^3H_6(0)$–$^3H_4(0)$ transition is oriented along the $y$ direction of the local site axis. This allows one to excite only some sites by choosing appropriate laser polarizations. As an example, if $B_0$ is along the [1 1 1] direction, the six sites are divided into two equivalent groups including, respectively, sites 1, 3, 5 and 2, 4, 6. If the laser is polarized along the same direction, sites 2, 4 and 6 are not excited and finally the system behaves as if there were only one site. Another interesting experimental set-up is obtained when the magnetic field is along [1 1 1] and the laser polarization along [1/3 1 1]. In this case, only sites 1, 4, 6 are excited and sites 4 and 6 are equivalent. The hyperfine splittings of the ground and excited CF levels ($\delta_g$ and $\delta_e$) for these sites have been calculated for a magnetic field magnitude of 4500 G (Table 1). It appears that in the range of...
probed around the burning frequency, only the hyperfine structure of sites 4 and 6 can be observed. The calculated large splittings for site 1 have been confirmed by hole burning experiments with weaker magnetic fields. The spectrum of Fig. 3 can therefore be interpreted by considering a single site. Hole C corresponds therefore to a transition to the excited state from the pumped ground-state level and is located at \(d_e\) from the central hole A. In the same way, antiholes B and D are, respectively, located at \((\delta_g - \delta_e)\) and \(\delta_g\) from hole A. As seen from Table 1, the agreement between experimental and calculated values of the hyperfine splittings is good.

Another important point is the fact that the experimental observation of side holes proves that the selection rules on the nuclear spin projection does not strictly apply. Considering the scheme of Fig. 1 and assuming that only the 1–3 and 2–4 transition are allowed (i.e. \(M_I\) is a good quantum number), burning on the 1–3 (respect. 2–4) transition will create a single antihole when probing the 2–4 (respect. 1–3) transition and nothing else. This simple spectrum consisting of a central hole and two side antiholes is actually observed when the magnetic field lies along the [1 1 1] direction and sites 1, 3, 5 are excited by a laser also polarized along [1 1 1]. This is in agreement with the low calculated branching ratio \(R\) in this configuration \((5 \times 10^{-4})\). On the other hand, if transitions 1–4 and 2–3 are partially allowed, a more complex structure can appear. This is the case of the spectrum shown in Fig. 3. However, the calculated \(R\) branching ratio is still quite low \((10^{-3})\) and it is possible that the partially allowed transitions are burnt but not probed. It should be also noted that the branching ratios have a very strong dependence on the magnetic field orientation, which explains the low values observed compared to the theoretically maximum value 0.24 [7].

The holes and antiholes positions can be used to determine the gyromagnetic tensor of Eq. (1) by using the equation

\[
\delta = \sqrt{\gamma_x B_0^2 + \gamma_y B_0^2 + \gamma_z B_0^2},
\]

which relates the \(\gamma\) parameters to the experimental hyperfine splittings. Using the \(\delta_g\) and \(\delta_e\) values deduced from Fig. 3, Eq. (2) gives \(\gamma_{x,g} < 0.368\) MHz/T, \(\gamma_{y,g} < 0.265\) MHz/T, \(\gamma_{z,e} < 0.347\) MHz/T and \(\gamma_{x,e} < 0.245\) MHz/T. The calculated values \((\gamma_{x,g} = 0.112\) MHz/T, \(\gamma_{x,g} = 0.189\) MHz/T, \(\gamma_{z,e} = 0.063\) MHz/T and \(\gamma_{x,e} = 0.223\) MHz/T) are within these limits. However, more experiments have to be performed to determine completely the gyromagnetic tensor and to have a more meaningful comparison with calculations.

### Table 1

<table>
<thead>
<tr>
<th>Site number</th>
<th>(\delta_g) (MHz)</th>
<th>(\delta_e) (MHz)</th>
<th>(R)</th>
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</thead>
<tbody>
<tr>
<td>1,3,5</td>
<td>203.06</td>
<td>26.44</td>
<td>0.0005</td>
</tr>
<tr>
<td>2,4,6</td>
<td>7.5</td>
<td>7.88</td>
<td>0.0099</td>
</tr>
<tr>
<td>(6.9 ± 0.03)</td>
<td>(6.5 ± 0.03)</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Experimental values (precision ± 0.03 MHz) are given in bold characters inside brackets. The field amplitude is 4500 G. The \(R\) parameter is the branching ratio between the 2–3 and 1–3 transitions (see Fig. 1).

### 4. Conclusion

The hyperfine structure of the first crystal field levels of the \(^3\)H\(_6\) and \(^3\)H\(_4\) levels has been investigated...
by hole burning spectroscopy. With a magnetic field oriented along the [1 1 1] direction and the laser polarization along the [1 1 1] direction, a complex spectrum appears which is ascribed to the burning and probing of two magnetically equivalent sites. A third excited site has larger splittings which lie outside the probed region. Antiholes and holes are observed, which shows that the 2–3 and 1–4 transitions are partially allowed in this configuration. This result is in agreement with theoretical calculations based on crystal field wavefunctions. The latter also reproduce well the experimental hyperfine splittings of the ground and excited states. This gives good hope to achieve the theoretical branching ratios of 0.24 with a magnetic field close to the [1 1 1] crystallographic axis.

References