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Long-time-storage mechanism for Tm:YAG in a magnetic field

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We obtained a long-time-storage mechanism for spectral features in thulium ions doped into YAG by applying a magnetic field that splits the electronic ground state. We show experimentally that the storage time can be more than 30 s, which is 3 orders of magnitude longer than that of the metastable state that normally is used for information storage in this material. Level splitting and storage lifetimes for various magnetic field strengths of as much as 5 T were investigated. This storage mechanism will be relevant in the many coherent transient-based signal-processing schemes in which Tm:YAG is being used, and we demonstrate long-time storage in a basic data storage application. © 2003 Optical Society of America

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Tm:Y₃Al₅O₁₂ (Tm:YAG) crystal has been widely used in the field of coherent transient-based all-optical signal processing. In particular, it has been utilized in applications such as broadband radio-frequency spectrum analysis,¹ true time-delay generation for rf array processing,² spatial routing of temporal beams,^{3,4} and short-time data storage.⁵ Tm:YAG has the advantage, compared with other rare-earth-ion-doped materials, of absorbing light at 793 nm, a wavelength for which diode lasers are readily available. A metastable state of Tm, which has a lifetime of ~10 ms,⁶ has been utilized for repetitive programming to continuously accumulate identical waveforms to maintain the stored spectral features.⁷ In this Letter we demonstrate orders-of-magnitude improvement in the storage time of spectral information in Tm:YAG. The crystal becomes suitable for certain memory architectures functioning at high capacity, with low latency and at high and sustained data rates, that are currently based on Pr- and Eu-doped materials. Long persistence of the spectral recording can also mitigate the need for repetitive programming and constant refreshing of information.

The investigations described here were performed at 793.16 nm on the ³H₆(1)–³H₄(1) transition of Tm ions doped into YAG. Tm replaces Y in this crystal host. There are 8 formula units per cubic unit cell in YAG, which gives 24 different sites for the Tm ions but only 6 different orientations of the sites.^{8,9} Tm has a nuclear spin of 1/2, which gives no zero-field hyperfine splitting of the levels. When a magnetic field is applied, both the ground and the excited states are split into two Zeeman-type levels through the coupling of the nuclear magnetic moment to the field. These levels will be denoted hyperfine levels in this Letter.

Rare-earth ions with nuclear spin larger than 1/2, when they are doped into inorganic crystals, can have several hyperfine levels of the ground and excited states even when no magnetic field is applied owing to (pseudo) quadrupole interactions.¹⁰ The lifetimes of the ground-state hyperfine levels of these ions can

be very long (as much as several hours) and have been studied extensively for, e.g., Eu and Pr doped into different crystal hosts.^{11,12} Information can then be stored as spectral features created by changing the population distribution between the hyperfine levels. In the case of Tm:YAG, for which the hyperfine levels are degenerate at zero magnetic field, we show that splitting the ground state makes it possible to store such features during a time that is much longer than the lifetime of the metastable state mentioned above. These spectral features could be of the form of spectral holes (used in spectral hole-burning applications) or of more-complex spectral structures that normally arise in photon-echo based data storage or processing.

The application of a magnetic field often leads to an increase of dephasing time T_2 for optical transitions of rare-earth ions doped into crystals by reducing nuclear spin flips in the host material. This action was studied earlier in Tm:YAG,⁶ and we can confirm a significant increase in T_2 with the application of a magnetic field. In the research reported in Ref. 6, some indirect evidence of splitting of the electronic levels was also observed and briefly commented on.

A Tm:YAG crystal with 0.5 at. % doping of Tm and with a thickness of 1 mm was used for the experimental investigations. The experiments were performed with an Oxford Instruments Spectromag cryostat. The crystal was kept in liquid He at 1.7–1.8 K. The magnetic field was applied along the crystal's c axis, which was also the direction of light propagation in the crystal.

Spectral hole-burning experiments were performed with an external-cavity diode laser designed to give fast, accurate, and mode-hop-free frequency tuning over several laser cavity mode separations.¹³ Two acousto-optic modulators in series were used to control the power of the light before the crystal. The frequency of the laser light was controlled by the voltage applied to the intracavity electro-optical crystal of the external-cavity diode laser, permitting mode-hop-free frequency scans of several gigahertz. The laser beam

was then focused onto the crystal with a lens with a 20-cm focal length. The laser power at the cryostat was ~ 10 mW. The signal was monitored with a photodiode.

To get clearly visible spectral structures, we burned the spectral holes by applying five optical pulses, each with a duration of $100 \mu\text{s}$. The pulses were separated by several hundreds of milliseconds, allowing all the excited ions to decay back from the metastable state between the pulses. After this sequence of burn pulses, we monitored the resultant spectral structures by scanning the laser over a frequency interval of 400 MHz at the frequency at which the spectral hole was burned. The duration of the scan was $190 \mu\text{s}$, and the power of the laser beam was attenuated so it would not affect the structure with the readout pulse. This hole-burning sequence was performed for a number of magnetic fields from 0.1 to 5 T. In Fig. 1 the spectral hole and the side structures that were due to the splitting of the ground and excited states can be seen for several magnetic field strengths.

The distance between the main hole and the side holes (seen as peaks in transmission in Fig. 1) is interpreted as the splitting of the excited state, and the distance to the antiholes is the splitting of the ground state. Thus the splitting of the ground state is substantially larger than that of the excited state. We interpret the absence of a third antihole in the group of antiholes as an indication of selection rules for transitions between the ground- and excited-state hyperfine levels. The weaker structures that can be discerned are interpreted as a splitting for ions in sites where the coupling of the magnetic moment to the magnetic field is different from that of the sites that mainly interact with laser light.

Lifetime measurements were performed with a cw Ti:sapphire laser (Microlase MBR-110). The narrow linewidth of this laser made it possible to burn spectral holes at lower magnetic fields (and thereby smaller frequency splitting between the hyperfine levels), as the smallest splitting that can be used for long-time storage by use of spectral hole burning is limited by the laser's linewidth. Spectral hole burning without focusing the laser beam onto the crystal reduced the sensitivity to vibrations of the sample with respect to the laser beam, leading to more-reliable lifetime measurements. The holes were burned with pulses of 100-ms duration and then repeatedly read out by chirping of the rf signal that drove the acousto-optic modulators. In this way, the decay of the spectral hole could be monitored for different magnetic fields, enabling the relaxation between the hyperfine levels of the ground state to be determined. For a magnetic field of 5 T the spectral hole decayed exponentially with a lifetime of a few seconds. For lower magnetic fields the decay was not a single exponential. For example, the decay at 0.5 T can be described by two components of approximately equal magnitude. The short-lived component had a lifetime of ~ 30 s, and the other component had a lifetime of at least several minutes.

In general, longer lifetimes were observed for smaller magnetic fields. At magnetic fields smaller than 10 mT the frequency jitter of the laser during

the burn pulse created holes with spectral widths comparable to the splitting of the hyperfine levels. It was then not possible to burn and read out the spectral holes, and the lifetimes of the ground-state hyperfine levels could not be determined. At 10 mT, the holes were monitored for minutes without any apparent decrease in hole amplitude.

Contributions to the relaxation can come from different processes involving phonons, such as the direct (one-phonon) process, a resonant two-phonon (Orbach) process, and a nonresonant Raman process.¹⁴ There are also temperature-independent processes, such as mutual Tm–Tm spin flip-flops and Tm–Al cross relaxation, which are of importance at low temperatures (a few degrees Kelvin). The former process increases with increasing concentration of Tm dopant ions, and the latter can be enhanced in case of resonance between the Zeeman splitting in Tm and the spin flip-flop energy of the host nuclei. Some processes, e.g., the one-phonon process, increase in strength with increasing Zeeman splitting of the ground state and hence with increasing magnetic field. These processes might therefore explain our observations.

To verify that the long-lived hyperfine states of the ground state of the Tm ions could be used for storing more-complex spectral structures than spectral holes, we performed photon echo experiments in which the hyperfine levels were used to store the spectral content of the applied pulses. In these experiments the external-cavity diode laser was again used as the light source. A photomultiplier tube was used for detecting the echoes, and a third acousto-optic modulator was used after the crystal to prevent the excitation pulses from saturating the detector.

In Fig. 2 the result of a basic data storage experiment, performed at a magnetic field of 50 mT, is shown. A 16-bit pattern was stored by use of frequency-chirped pulses with a bandwidth of 40 MHz

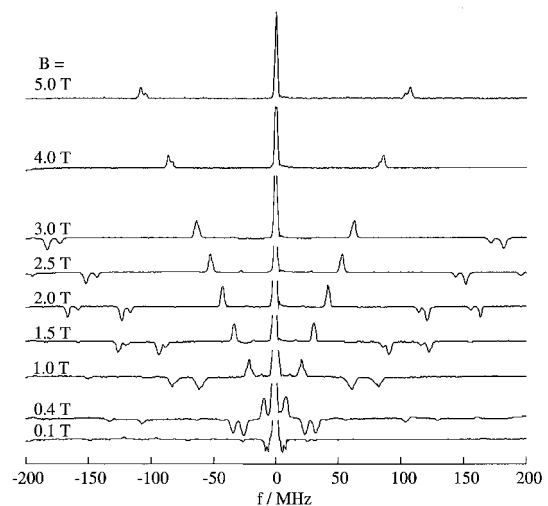


Fig. 1. Spectral structure that arises as a result of hole burning at magnetic field strengths of 0.1–5.0 T. The traces are single-shot transmission measurements for which the laser frequency has been scanned over 400 MHz in $190 \mu\text{s}$. For clarity, the traces have been offset by an amount proportional to the applied magnetic fields.

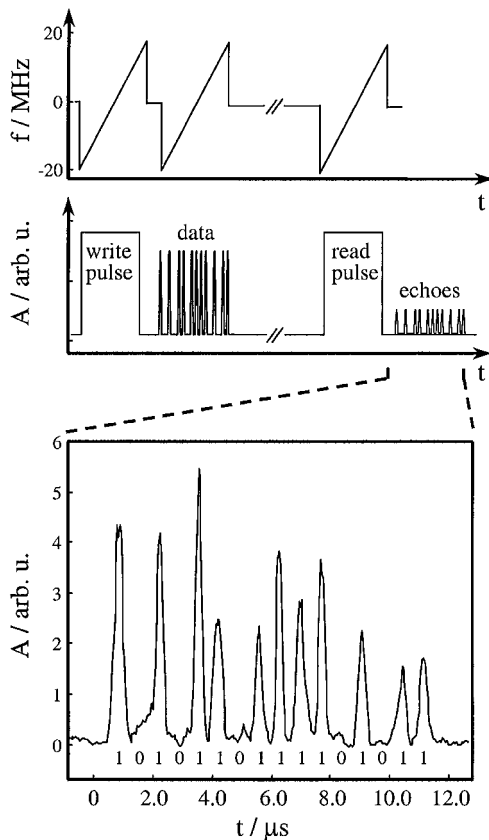


Fig. 2. Experimental demonstration of storage of a 16-bit pattern for 200 ms by use of photon echoes. Top, frequency (f) and amplitude (A) of the light versus time; bottom, a single-shot trace of the recalled data.

and read out after 200 ms. To show that photon echo storage or processing on even longer time scales is possible, we stored a single pulse, using pulses that were chirped to give a strong signal. The stored pulse could be read out after 30 s with a signal-to-noise ratio of better than 20. We believe that it would be straightforward to apply other data storage schemes, e.g., the swept carrier technique,⁵ to store larger amounts of data.

In this Letter we have shown that the storage time for spectral features in Tm:YAG can be increased by at least 3 orders of magnitude by application of a magnetic field. Long-time storage proved to be possible even for moderate magnetic fields (less than 50 mT). This fact makes it possible to use permanent magnets, rather than superconducting coils, which significantly reduce the complexity of the setup. If a particular data storage time is desired in a specific

application, a magnetic field strength should be chosen such that the level splitting is sufficient for the application, because the lifetime varies inversely with the splitting. We believe that the possibility of long-time storage demonstrated in this Letter could simplify and improve several of the existing schemes that are being considered in this crystal and also open possibilities for implementations of new schemes.

It would be interesting to study further the transition rules and coherence times between the hyperfine levels of the ground and excited states, as doing so could lead to ways of spin polarizing the sample. Spin polarization might be of interest for applications related to quantum state storage, quantum teleportation, and quantum information processing.¹⁵

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