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# Diode laser frequency stabilization to transient spectral holes and spectral diffusion in $\text{Er}^{3+} : \text{Y}_2\text{SiO}_5$ at 1536 nm

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## Abstract

Diode laser frequency stabilization to 500 Hz Allan deviation is demonstrated over 2 ms integration times with drift reduced to 7 kHz/min. This was achieved at 1536 nm in the technologically important communications band by stabilizing external cavity diode lasers to regenerative transient spectral holes in the inhomogeneously broadened  ${}^4\text{I}_{15/2}(1) \rightarrow {}^4\text{I}_{13/2}(1)$  optical absorption of  $\text{Er}^{3+} : \text{Y}_2\text{SiO}_5$ . Spectral diffusion, which currently limits the achievable stabilization performance, has been studied using stimulated photon echoes. Due to spectral diffusion, significant broadening of the homogeneous linewidth at low magnetic fields from a few kHz to tens of kHz develops as the waiting time  $T$  between pulses two and three was increased from microseconds up to the  $T_1 \sim 10$  ms lifetime of the excited state. This evolution of the homogeneous linewidth has been mapped out as a function of magnetic field. The classic spectral diffusion can be reduced to negligible levels upon application of a magnetic field in a 0.02 atomic percent  $\text{Er}^{3+} : \text{Y}_2\text{SiO}_5$  crystal. © 2001 Elsevier Science B.V. All rights reserved.

*Keywords:* Laser stabilization; Rare earths; Spectral hole burning; Coherent transients; 1.5  $\mu\text{m}$  wavelength

## 1. Spectral hole burning as a frequency reference

As recently reported, persistent [1,2] and transient spectral hole burning (SHB) [3,4] in  $\text{Tm}^{3+}$  and  $\text{Er}^{3+}$  doped crystals provides a new method to frequency stabilize lasers. SHB-stabilized lasers already play an important role in a number of SHB-based applications that require extreme frequency stability [5] and will greatly improve prototype devices [6]. For SHB devices using materials optimized for signal correlators or memories and for coherent transient spectroscopy

experiments, references at specific wavelengths and with stability over specific time scales are required. We have demonstrated that these requirements are naturally met by using a spectral hole in a separate piece of the same SHB material to provide the frequency reference for laser stabilization [3,4]. Spectral holes can also be chosen at arbitrary locations within the inhomogeneously broadened absorption profile allowing user specific tailoring of reference frequencies.

## 2. Experimental

The laser frequency locking experiments have been performed on a 0.005%  $\text{Er}^{3+} : \text{Y}_2\text{SiO}_5$  crystal

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in a moderate magnetic field of  $B = 0.2$  T also readily obtainable using permanent magnets. Transient SHB on the  ${}^4I_{15/2}(1) \rightarrow {}^4I_{13/2}(1)$  transitions at 1536.14 (site 1) and 1538.57 nm (site 2) arises from population storage in the excited state of the optically active ion with a lifetime  $T_1 \sim 10$  ms [7]. The inhomogeneous linewidth is  $\Gamma_{\text{inh}} = 500$  MHz, and the homogeneous linewidth  $\Gamma_{\text{h}} \sim 5$  kHz was determined from photon echo decays [8].

Two lasers were independently stabilized to separate SHB frequency reference crystals having a linear absorption of  $\sim 50\%$  at line center. Crystal dimensions were 5 mm along  $D_1$ , 6 mm along  $D_2$  and 1 mm along  $b$ . Each crystal was oriented with  $D_1$  parallel to the magnetic field, the lasers'  $k$ -vectors parallel to  $b$ , and  $E$  polarized along  $D_2$  [7]. Both were immersed in superfluid helium at 1.6 K. Each crystal was exposed to only one laser, with irradiances of  $100 \mu\text{W}/\text{cm}^2$  using  $\sim 3$  mm beam diameters.

The experimental setup has been published elsewhere [1–4]. Two custom-made external cavity diode lasers have been stabilized using the conventional Pound–Drever–Hall technique [9]. The spectral holes are used in much the same way as traditional frequency references such as a mode of a Fabry–Perot cavity. The lasers were externally phase-modulated with modulation index  $M \sim 0.4$  at 27 and 30 MHz. The error signal derived from the spectral hole was applied to a fast servo loop modulating the injection current of the laser diode whereas a slow servo loop made corrections to the piezo-driven feedback prism plate of the laser cavity.

During active stabilization each laser burned a transient spectral hole at a different frequency in its reference crystal. Error signal feedback to the laser led to a continuous regeneration of the transient spectral hole. The transient hole and consequent locking dynamics have been addressed in a separate paper of this volume [10]. No commercial system is able to measure laser frequency with the precision required to characterize these lasers. Thus, the two lasers were beat with each other and the Allan deviation [11] of the beat frequency was determined [1–4].

### 3. Stabilization results

Fig. 1 shows the Allan deviation for (a) the free running and (b) actively stabilized lasers. The free running lasers show frequency stability comparable to or even better than similar commercially available systems and are already sufficiently stable for many applications in spectroscopy. With the lasers locked to transient spectral holes in  $\text{Er}^{3+}:\text{Y}_2\text{SiO}_5$ , an improvement in the Allan deviation over the free running lasers of more than two orders of magnitude to 500 Hz has been achieved for integration times of 2 ms. Smaller Allan deviations of  $\sim 200$  Hz have been measured during quiet periods. Long-term linear laser drift, evidenced by an upturn of the Allan deviation at longer integration times, has been greatly reduced to about 7 kHz per minute by choosing an intermediate phase setting at the mixer between detector signal and local oscillator. This technique combines the excellent short-term stability of the spectral hole with the good long-term stability of the inhomogeneous line [3,4].

Even though the transient spectral hole provides a narrow several kHz wide reference, one might expect to achieve lower Allan deviation values over short integration times as measured in  $\text{Tm}^{3+}:\text{YAG}$  [3,4]. In this case, however, spectral diffusion limits the currently achieved performance as discussed in the following paragraph. Technical

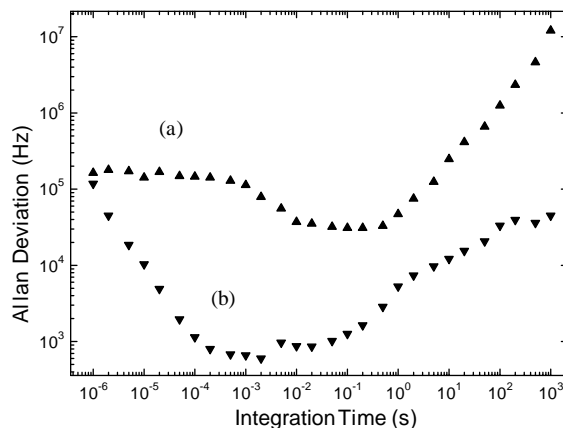


Fig. 1. Allan deviation for the heterodyne beat frequency between two diode lasers: (a) lasers free-running, (b) independently locked to separate transient spectral holes at  $B = 0.2$  T.

limitations are also set by temperature dependent residual amplitude modulation at the electro-optic modulators and voltage offsets created in the electronic servo loop. Since a transient spectral hole is a dynamic reference, any voltage offsets introduced to the error signal baseline cause the lock point to shift and consequently add long-term laser drift.

#### 4. Spectral diffusion in $\text{Er}^{3+}:\text{Y}_2\text{SiO}_5$

To investigate the evolution of the spectral hole width by spectral diffusion, stimulated photon echo spectroscopy has been exploited. A 0.02 atomic percent  $\text{Er}^{3+}:\text{Y}_2\text{SiO}_5$  crystal was used with the identical geometry described above. The diode laser output of 1.8 mW saturated the output of an Erbium fiber amplifier producing 35 mW for spectroscopy. An acousto optic modulator gated collinear echo excitation pulses that were focused into the crystal. Initial experiments were carried out with the diode laser SHB stabilized. However, using short pulses of  $\sim 500$  ns length, whose Fourier spectral width ( $\sim 2$  MHz) was large compared to the homogeneous broadening, relaxed the requirement for an ultrasharp laser linewidth. Photon echoes were strong enough for direct detection with an InGaS photodiode and were recorded on a digital oscilloscope.

Systematic measurements of the stimulated photon echo strength were made as first to second pulse separation  $t_{12}$  was swept at various waiting times  $T$  (second to third pulse separation). The first two pulses set up a population grating between the ground and excited state with a grating spacing  $1/t_{12}$ . This grating decays due to population decay ( $T_1$ ) and tends to smear out due to spectral diffusion. Pulse three probes the grating, creating the stimulated photon echo as a measure of population decay and spectral diffusion during the interval  $T$ . At small magnetic fields and for waiting times up to tens of microseconds, the stimulated echo decays were non-exponential, indicating the existence of spectral diffusion. The observed decays were fitted to a Mims expression of the form  $I(t) = I(0)\exp(-4t/T_M)^x$  [12], which allowed determining the homogeneous linewidth

using  $\Gamma = 1/\pi T_M$ . Detailed discussion of measured  $x$  values has been reported elsewhere [8]. Fig. 2 maps the evolution of the homogeneous linewidth as a function of the waiting time  $T$  for a variety of magnetic fields from  $B = 0.8$  up to 3 T at  $T = 1.6$  K. Spectral diffusion is evident at low magnetic fields since there is a significant broadening of the homogeneous linewidth as the waiting time  $T$  is increased. Clearly, larger magnetic fields suppress homogeneous line broadening and a plateau is reached after more than 100  $\mu\text{s}$ , where the homogeneous linewidth remains unchanged. Solid lines in Fig. 2 represent least square fits to the simple expression  $\Gamma(T) = \Gamma_0 + \Gamma_1 \times [1 - \exp(-RT)]$  proposed by Yano [13]; each case shows good agreement with the data. The parameter  $\Gamma_0$  is the homogeneous linewidth measured by two pulse echoes. Fitted parameters included  $\Gamma_1$ , the plateau value of the linewidth for large waiting times  $T$ , and  $R$ , which is related to the rate of spin flips causing the spectral diffusion.

Fig. 3 shows a stimulated echo decay in a magnetic field of  $B = 2$  T and temperature of 1.6 K measured as a function of the delay  $T$  between the second and third pulses, with the delay  $t_{12}$  fixed at 4  $\mu\text{s}$ . Here, spectral diffusion, being faster than population decay, dominates the rapid initial non-exponential part, followed by a single exponential decay with a time constant

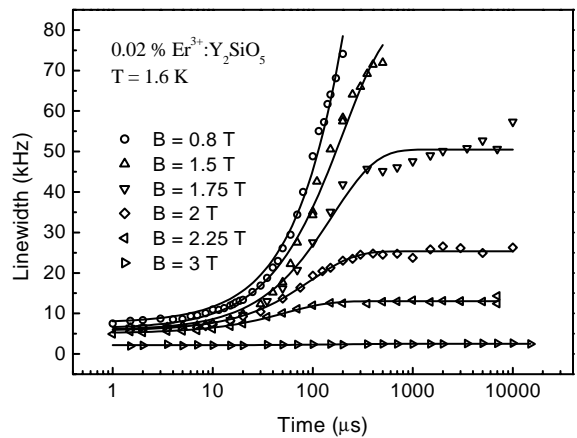


Fig. 2. Stimulated photon echo measurement of the dependence of the linewidth on the waiting time  $T$  between pulses two and three for magnetic fields between  $B = 0.8$  and 3 T, showing the presence of spectral diffusion.

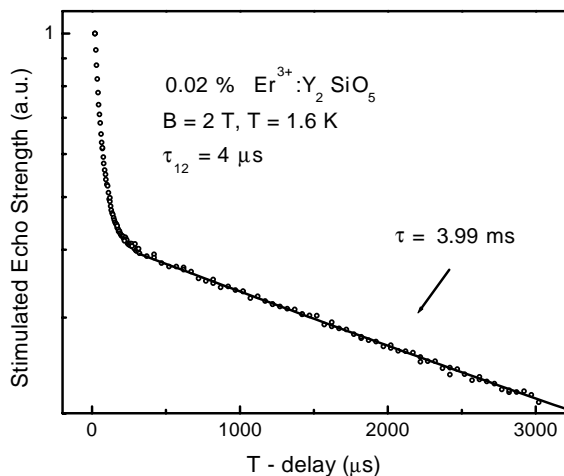


Fig. 3. Stimulated photon echo decay as a function of time delay  $T$  with pulse one to pulse two delay  $t_{12} = 4 \mu\text{s}$  fixed. The solid line represents a single exponential fit yielding a time constant of  $\tau = 3.99 \text{ ms}$ .

$\tau = 3.99 \text{ ms}$  approaching the theoretical limit set by population decay  $2\tau \leq T_1 \cong 10 \text{ ms}$  [7].

Working at low temperature minimizes the significance of phonon broadening by decreasing available phonon energies leaving electronic spin flip–flop transitions as the primary mechanism for spectral diffusion. Mutual  $\text{Er}^{3+}$ – $\text{Er}^{3+}$  spin flip–flop transitions involving nearby  $\text{Er}^{3+}$  ions change the local magnetic field. Applying a magnetic field in the direction of maximum  $\text{Er}^{3+}$   $g$  value makes the ground Zeeman splitting large compared to the thermal energy and freezes out the population of the upper Zeeman level of the ground state, thus inhibiting  $\text{Er}^{3+}$  spin flip–flops. Indeed, applying the magnetic field along  $D_1$  is a preferential direction for maximizing the  $g$  value as shown by angle dependent measurements that we will report elsewhere. Nuclear spin flips of host crystal ions can contribute to a lesser degree to fluctuating local fields; those effects have been minimized here by choosing the low-nuclear-spin host  $\text{Y}_2\text{SiO}_5$ .

Together these are important results showing how to optimize the  $\text{Er}^{3+}:\text{Y}_2\text{SiO}_5$  material for laser frequency references and SHB-based signal processing applications. A more detailed paper on the spectral diffusion in  $\text{Er}^{3+}:\text{Y}_2\text{SiO}_5$  with several Erbium concentrations will be published elsewhere.

## 5. Summary

In conclusion, we demonstrated sub-kilohertz laser frequency stabilization to regenerative transient holes in  $\text{Er}^{3+}:\text{Y}_2\text{SiO}_5$  at the important  $1.5 \mu\text{m}$  optical communications wavelength. Spectral diffusion in this material, currently limiting the locking performance, can be overcome upon application of a stronger magnetic field. This newly demonstrated behavior also makes  $\text{Er}^{3+}$  materials more attractive for SHB signal processing devices.

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