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# Precise spectroscopy of $^{167}\text{Er}:\text{Y}_2\text{SiO}_5$ based on laser frequency stabilization using a fiber laser comb

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**Abstract:** Precise spectroscopy of the hyperfine level system of  $^{167}\text{Er}$ -doped  $\text{Y}_2\text{SiO}_5$  was achieved in the frequency domain. By using an optical frequency comb to stabilize the light source frequency to an accuracy on the order of hertz on a long-term scale, Allan deviation  $< 10$  Hz was achieved for an integration time of 180 s. As a result, spectral hole-burning experiments yielded a more accurate hole spectrum with a narrow homogeneous linewidth. The method opens the way to the straightforward exploration of relaxation mechanisms in the frequency domain by simple steady-state measurements.

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## 1. Introduction

In recent years, quantum repeaters and quantum memories, which are indispensable for the construction of global quantum information networks and for long preservation of quantum information, have been studied intensively [1–3]. Rare-earth ion-doped crystals are now attracting attention to realize light-matter interaction with high efficiency and fidelity, which is required for the development of quantum memories [4–6]. Even though they are solids, rare-earth ion-doped crystals have very narrow homogeneous linewidths  $\Gamma_h$  ( $< 10$  kHz), comparable to that of monoatomic gases such as  $^{87}\text{Rb}$  [7]. For example, the observed  $\Gamma_h$  approached 100 Hz in  $\text{Eu}^{3+}:\text{Y}_2\text{SiO}_5$  (YSO) [8,9] and even 73 Hz for the  $\text{Er}^{3+}$  Kramers ion in  $\text{Er}^{3+}:\text{YSO}$  under strong magnetic field of 7 T [10]. The reason for the narrow  $\Gamma_h$  is that the optical transitions in rare-earth ions are intra- $4f$  electron transitions, and Coulomb shielding by  $5s$  and  $5p$  shell electrons suppresses interactions with other atoms in the crystal [11]. Because of the extremely narrow  $\Gamma_h$ , the system can be described as a solid-state atomic system.

Among the rare-earth ion-doped crystals,  $\text{Er}^{3+}:\text{YSO}$  is a particularly promising material for quantum memories [12–15]. It has an optical transition level in the telecommunications wavelength band ( $\sim 195$  THz), where low-loss photon transport by optical fibers is possible. In addition, the host crystal (YSO) is a nonmagnetic crystal with an extremely small total spin density. Hence,  $\text{Er}^{3+}:\text{YSO}$  has a long population lifetime and optical coherence time. Because of the above advantages, quantum memory using an atomic frequency comb (AFC) [16,17] and quantum state manipulation using electromagnetically induced transparency (EIT) [18] have been studied in the telecommunications band. More recently,  $^{167}\text{Er}^{3+}$  ions have attracted much attention. This is because among Er isotopes, the  $^{167}\text{Er}^{3+}$  ion is the only one with a nuclear spin ( $I = 7/2$ ), which gives rise to a rich hyperfine (HF) structure in the  $\text{Er}^{3+}$  energy levels. While this HF structure of  $^{167}\text{Er}^{3+}$  is expected to provide even longer coherence time and efficient

spin pumping [6,15], demonstrating these will also require very high precision spectroscopic techniques to access the fine energy structure.

In such precision spectroscopy, improving laser frequency stability, in particular, is essential to promote such studies using rare earth ions. For instance, frequency fluctuation of the light source is a bottleneck to realizing quantum memory protocols using AFCs and investigating the spectral diffusion mechanisms in the frequency domain [19–21]. Several methods have been proposed for laser frequency stabilization, including the use of atomic or molecular absorption lines as reference frequencies and the use of stable optical resonators [22–24]. Recently, optical frequency combs (OFCs) have been rapidly developed in a wide range of fields such as high-resolution coherent Raman spectro-imaging [25] and absolute frequency measurement [26]. If OFCs can be used to stabilize a light source, it will be possible to stabilize the frequency of the light source over a wide bandwidth at any frequency, which will be useful for various types of laser spectroscopy.

In this paper, we report the first successful use of OFCs to stabilize the frequency of a light source for spectroscopy on the order of hertz. To demonstrate the impact of laser frequency stabilization, we evaluated the homogeneous linewidth by spectral hole burning (SHB) spectroscopy. In addition, a coherence time obtained from SHB was compared with those obtained from two-pulse photon echo (PE) measurements. As a result, it was found that the fast relaxation process that is considered to be caused by superhyperfine interaction and had previously been observed only in time-domain measurements, could be obtained with good accuracy in the frequency domain. Therefore, frequency stabilization is very useful for studying relaxation processes such as spectral diffusion more precisely.

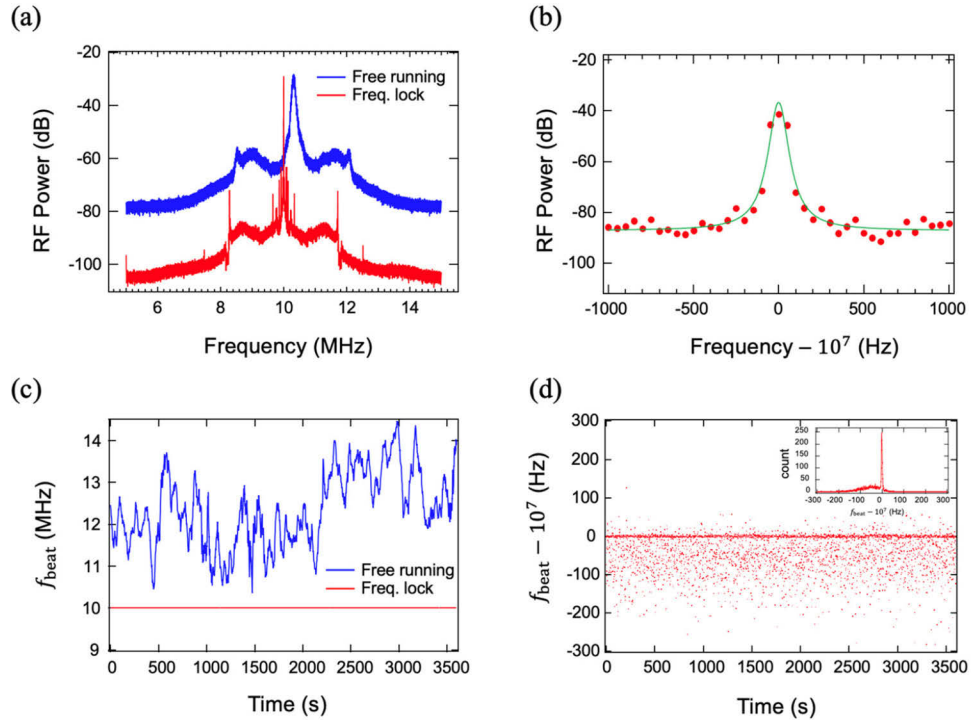
## 2. Evaluation of long-term frequency stability of a light source

In this section, we show good long-term stability of a frequency-locked external cavity laser diode (ECLD). First, to evaluate the temporal change of the frequency fluctuation of the ECLD, the frequency fluctuation of the beat signal between the ECLD and the OFC was examined with the measurement system shown in Appendix A1.

The light source in this study, an ECLD, is a narrow linewidth CW laser (RIO, Grande). It has low phase noise and an output power of 2 W. The laser linewidth is  $< 1$  kHz, and the output frequency is in the C-band ( $\sim 195.10$  THz), which corresponds to a telecommunications wavelength. The frequency can be tuned within a range of  $\sim 2$  THz by adjusting the cavity temperature. For frequency stabilization of this ECLD using an OFC, a passively mode-locked Er-doped fiber laser comb was frequency-locked to another ultra-narrow linewidth laser used as a master laser [27], and then the ECLD was frequency-locked to the stabilized fiber laser comb (see Appendix A1). The repetition rate of the comb is 100 MHz, and the frequency of the ECLD can be locked to any comb tooth in the band of  $\sim 2$  THz centered at  $\sim 195$  THz.

Figure 1(a) shows the beat signal acquired by a signal analyzer (Agilent N9010A) with the resolution bandwidth (RBW) of 120 Hz, the video bandwidth (VBW) of 1 Hz, and the sweep time of 20 s. The large peak observed at around 10 MHz is the beat signal. In the free-running case, the full width at half maximum (FWHM) of the beat signal  $\gamma_{\text{free}}$  was about 300 kHz. The broad spectrum  $\pm 1.5$  MHz away from the beat signal is a servo bump which comes from the PID circuit which is passed in both cases, free-running and frequency-locking. In the case of frequency stabilization, the sharp, small peaks around  $10 \pm 1.8$  and  $10 \pm 2.7$  MHz are noise from the PID control circuit. Figure 1(b) is an enlarged signal view around 10 MHz during frequency stabilization. In this case, the RBW, VBW, and sweep time were set to 51 Hz, 1 Hz, and 700 ms, respectively. The linewidth of the beat signal  $\gamma_{\text{lock}}$  is about 150 Hz, and owing to frequency stabilization, the beat spectrum width is reduced by about three orders of magnitude.

Figure 1(c) shows the fluctuation of the center frequency of the beat signal ( $\Delta f$ ) obtained by using a frequency counter (Agilent 53230A) with an internal clock of 10 MHz as a reference signal and the integration time of 1 s. Figure 1(d) shows a magnified image of the temporal deviation



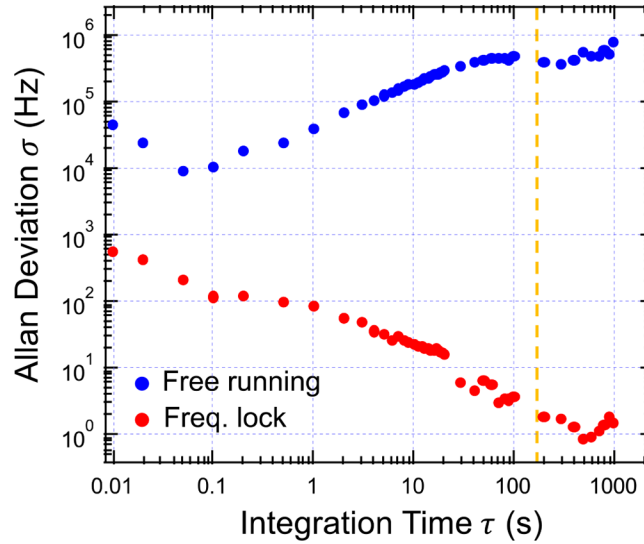
**Fig. 1.** (a) Detected beat signals during free running (blue) and frequency stabilization (red). 20-dB offset is added to the free running signal for the sake of clarity. (b) Enlarged image of the beat signal during frequency stabilization in (a). The center frequency is 10 MHz, and the frequency span is 2000 Hz. The green solid line represents the Lorentzian fitting curve. (c) Temporal fluctuation of the center frequency in each condition when the beat signal is measured for 1 h with the integration time of 1 s. (d) Magnified image of the temporal fluctuation of the beat signal during frequency stabilization in (c), where the center frequency is 10 MHz. Inset: distribution of frequency fluctuation with the bin width of 1 Hz.

of the beat frequency from 10 MHz in the frequency-stabilized case in (c). As can be seen in (c) and (d), during frequency stabilization, the fluctuations were suppressed to within about  $\Delta f_{\text{lock}} = 300$  Hz, whereas with free running, the temporal fluctuations exceeded  $\Delta f_{\text{free}} = 4$  MHz.

Even though the frequency fluctuation of the OFC is  $< 1$  Hz, the fluctuation of the beat signal,  $\Delta f_{\text{lock}}$ , is relatively large in comparison. As shown by the inset in Fig. 1(d), most of the fluctuations are around 5 Hz, but there is a broad component over a band of  $\sim 110$  Hz centered at  $-50$  Hz. This is because the feedback given to the ECLD does not include the temperature control of the cavity. The laser frequency tends to drift in the decreasing (increasing) direction due to the cavity temperature change following the slow rise (fall) of the room temperature. Since the repeated feedback is given to such unidirectional frequency drift, the fluctuation to the low (high) frequency side becomes larger, and therefore the broad component up to the order of 100 Hz was formed. In addition, since the OFC is transported to the optical coupler using a 15-m polarization-maintaining fiber, the linewidth of the OFC itself widens from  $< 1$  Hz to 5 Hz due to fiber noise.

Next, to estimate the long-term frequency stability of the ECLD, we measured the Allan deviation  $\sigma(\tau)$  of the beat signal. As shown in Fig. 2, the increase in the Allan deviation due to random walk ( $\sigma_{\text{free}} \propto \sqrt{\tau}$ ) is dominant in the free-running condition, while the effect of white noise ( $\sigma_{\text{lock}} \propto 1/\sqrt{\tau}$ ) is dominant when frequency stabilization is used. By stabilizing the

frequency, the effect of the random walk is suppressed in the integration time range up to 500 s. Based on these results, for subsequent measurements described in the next section, we set the integration time to 180 s, at which the frequency of the ECLD is sufficiently stable. As is clear from Fig. 2, when we focus on the integration time of 180 s, we can see that the frequency fluctuation  $\sigma_{\text{lock}}$  is suppressed by about five orders of magnitude compared to the free-running condition ( $\sigma_{\text{free}}$ ). In this case, the Allan deviation during the frequency stabilization  $\sigma_{\text{lock}}$  is about 2 Hz. Consequently, by stabilizing the frequency using a fiber laser comb with very small frequency fluctuations, precision spectroscopy with a light source having a few hertz linewidth becomes possible.

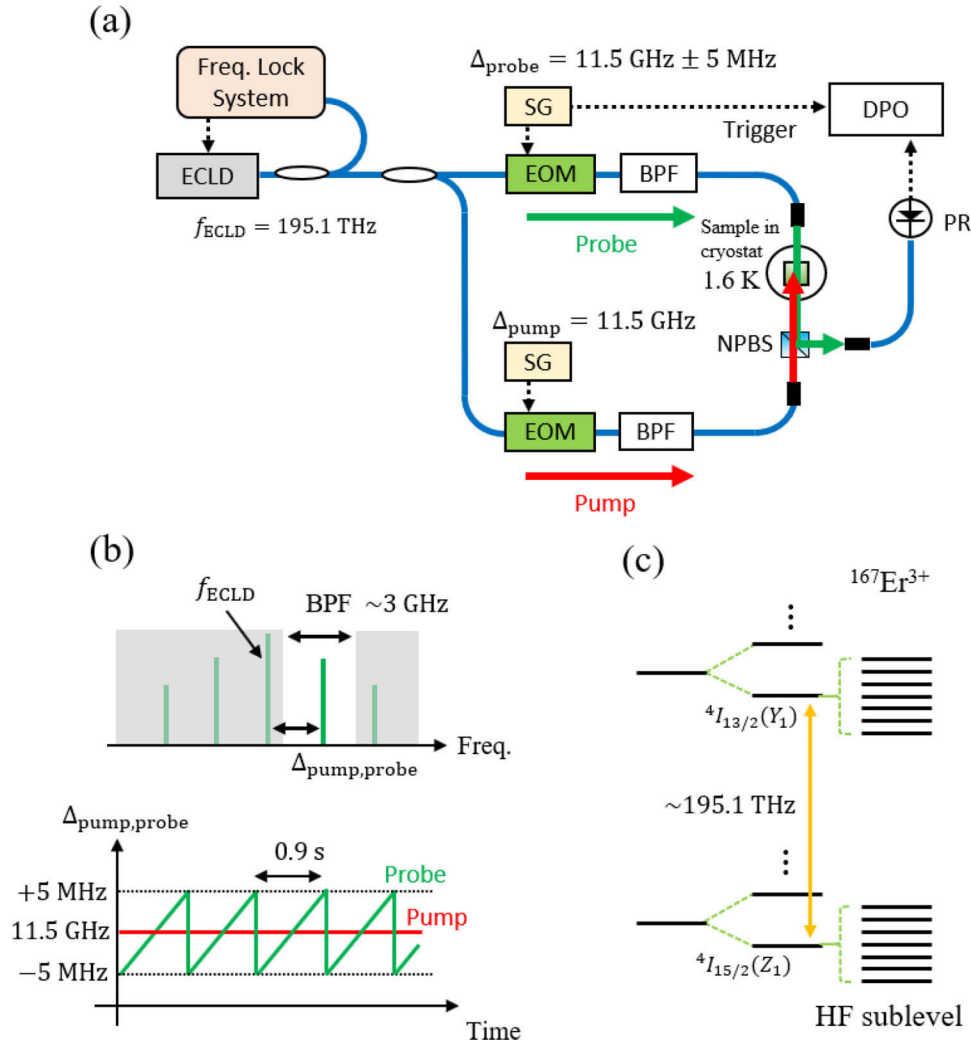


**Fig. 2.** Allan deviation versus integration time. The blue plot shows free running, and the red plot shows frequency stabilization. The yellow dashed line represents the integration time of 180 s, whose condition was used in the SHB experiments shown in the next section.

The Pound-Drever-Hall technique [28], which can achieve frequency stability on the order of millihertz and is often used, requires a Fabry-Perot resonator with high finesse. However, the cavity length cannot be changed to ensure frequency stability, and the frequency to be stabilized cannot be easily changed. In addition, frequency stabilization using narrow atomic absorption lines is limited to sub-kilohertz stability in the telecommunications wavelength band, although the stabilized frequency is tunable within an inhomogeneously broadened spectrum of several gigahertz [29]. On the other hand, frequency stabilization using OFCs shown here can stabilize the frequency of the ECLD to an arbitrary OFC tooth existing within a frequency band of several terahertz, below 5 Hz of Allan deviation even on the time scale of 100 s. Therefore, frequency stabilization using OFCs is very simple and robust.

### 3. Evaluation of homogeneous linewidth by spectral hole burning in $^{167}\text{Er}^{3+}:\text{YSO}$

In this section, we describe a homogeneous linewidth measurement by SHB as an application of precision spectroscopy using frequency stabilization. The homogeneous linewidth  $\Gamma_h$  is an important parameter that represents the optical coherence time  $T_2$  and the relaxation mechanism due to various interactions. However, it has been difficult to measure it in the frequency domain, such as by saturation absorption spectroscopy, due to the frequency fluctuation of laser sources. Instead, the PE method, a time-domain measurement, is often used to estimate narrow homogeneous linewidths. If we can obtain these parameters accurately in the frequency domain,



**Fig. 3.** (a) SHB measurement apparatus. The details of the frequency-lock system are described in Appendix A1. SG: signal generator. EOM: electro-optic modulator. BPF: band-pass filter. NPBS: nonpolarizing beam splitter. PR: photoreceiver. DPO: digital photo oscilloscope. (b) Frequency detuning and time sequence of modulation sidebands for the pump and probe beams. (c) Energy diagram of  $^{167}\text{Er}^{3+}$ .

we can reveal the relaxation mechanism as well as the energy level structure. In order to achieve this in the SHB measurement, it is necessary to pay attention to the spectral linewidth and frequency fluctuations of the laser source as well as the power broadening of spectral holes. If they are larger than the homogeneous linewidth, they will dominate the spectral hole width and overestimate the true homogeneous linewidth. Therefore, precision spectroscopy using a frequency-stabilized light source with a narrow linewidth is indispensable for SHB measurements.

### 3.1. Sample and experimental setup

The sample is  $^{167}\text{Er}^{3+}:\text{YSO}$ , prepared by mixing isotopically pure  $^{167}\text{Er}$  (Atox Co. LTD) with high-purity  $\text{SiO}_2$  and  $\text{Y}_2\text{O}_3$  powders followed by growth by the Czochralski method. The concentration of  $^{167}\text{Er}^{3+}$  is 10 ppm [30]. Because the host crystal YSO is a nonmagnetic crystal

with a small total spin density, it has been reported that the interaction between the electron spins of  $^{167}\text{Er}$  and the nuclear spins of the host crystal is suppressed and a narrow homogeneous linewidth is obtained [31]. The energy levels of  $^{167}\text{Er}^{3+}$  split due to various interactions (Fig. 3(c)). There are six naturally occurring isotopes of Er, but only  $^{167}\text{Er}^{3+}$  has the HF sublevels, which are splitting levels due to HF interaction with nonzero nuclear spin ( $I=7/2$ ).

The optical coherence time and nuclear spin coherence time of the HF sublevels are found to be much longer than the coherence time of the Stark interlevel transition ( $< 1 \mu\text{s}$ ) [15,30,32]. There are two magnetically nonequivalent sites in the Y of the host crystal YSO, and the zero-phonon transition wavelength between the Stark levels is different at each site. In this demonstration, the transition at site 1 with an optical lifetime of 10 ms was used, and the measurement was performed under zero magnetic field.

Figure 3(a) shows the setup for SHB experiments. The light source was a CW ECLD with frequency stabilization (Fig. 6 in Appendix A1). The laser beam was divided into pump and probe beams. Each beam was phase-modulated by an EOM connected to a signal generator (Anritsu, MG3693C), and variable sidebands could be produced. Figure 3(b) shows the optical sideband and frequency sweep range by phase modulation. The first-order sideband of each beam was used as the optical transition wavelength. Other sideband frequency components were removed by a band-pass filter (BPF: Yenista, XTM-50). The probe and pump beams were focused onto the sample in a coaxial opposing arrangement. The sample was cooled to 1.6 K in a cryostat. The sample size is  $5 \times 5 \times 6 \text{ mm}$  in the  $D_1 \times D_2 \times b$ -axis direction. The wavenumber vector  $\mathbf{k}$  of the laser beams is parallel to the  $b$ -axis direction of the crystal, and the laser spot diameter is  $60 \mu\text{m}$ . To suppress the power broadening effect, the laser power was set to  $8 \text{ mW/cm}^2$  for the pump light and  $< 1 \text{ mW/cm}^2$  for the probe light.

As shown in Fig. 3(b), the center frequency of the ECLD,  $f_{\text{Laser}}$ , was set to 195.105100 THz, and the detuning  $\Delta_{\text{pump}}$  from the center frequency of the first-order optical sideband of the pump light was set to 11.580 GHz, resulting in  $f_{\text{pump}}=195.116680 \text{ THz}$ . This corresponds to the center frequency of an inhomogeneously broadened  $^4I_{15/2}(Z_1) - ^4I_{13/2}(Y_1)$  transition. The probe light was frequency swept over a range of  $\pm 5 \text{ MHz}$  centered at  $f_{\text{pump}}$  for 0.9 s. The probe light transmitted through the sample was detected by an InGaAs photoreceiver (Newport, model 2153) and averaged 200 times by a digital oscilloscope (Tektronix, MSO64) to obtain the SHB spectrum.

### 3.2. Effect of laser frequency stabilization in SHB experiments

Figure 4(a) shows the inhomogeneously broadened spectrum  $^4I_{15/2}(Z_1) - ^4I_{13/2}(Y_1)$  transition, which spreads over a frequency region of  $\sim 2 \text{ GHz}$  and contains half of the  $16 \times 16$  HF inter-sublevel transitions. A sharp main hole spectrum appears at the pump frequency (11.580 GHz). The red solid line in Fig. 4(b) shows the hole spectrum with frequency locking, which has a hole width  $\Gamma_{\text{hole,lock}} = 358 \text{ kHz}$ , defined as the spectral FWHM. When the ECLD (RIO, Grande) used in section 2 (hereafter, ECLD1) and another ECLD with a larger frequency fluctuation (ECLD2) is used in the free-running condition, the hole spectra have different hole widths,  $\Gamma_{\text{hole,free}}^{\text{ECLD1}} = 487 \text{ kHz}$  and  $\Gamma_{\text{hole,free}}^{\text{ECLD2}} = 1.1 \text{ MHz}$ , respectively, reflecting their bandwidths and frequency fluctuations. By using ECLD1, the hole width can be narrowed by a factor of two or more even in the free-running condition, but in the frequency stabilized condition, the linewidth of both ECLDs can be narrowed to the same order. This is due to the suppression of frequency fluctuations to the order of hertz. The experimental results for ECLD1 are summarized in Table 1.

When the frequency is stabilized, the actual laser linewidth of ECLD1, taking into account the fluctuations ( $\sigma_{\text{lock}} \sim 2 \text{ Hz}$ ), should be less than  $1 \text{ kHz}$  with an integration time of 180 s, and the hole width  $\Gamma_{\text{hole,lock}}$  should be able to be measured with a resolution of less than  $1 \text{ kHz}$ . Therefore, the measured  $\Gamma_{\text{hole,lock}}$  are not overestimated and has physical significance. Assuming that the observed  $\Gamma_{\text{hole,free}}$  corresponds to a convolution of  $\Gamma_{\text{hole,lock}}$  and  $\sigma_{\text{free}} (\sim 300 \text{ kHz})$ ,  $\Gamma_{\text{hole,free}}$  can

**Table 1. Laser fluctuation and results of SHB measured with ECLD1**

| Laser condition | $\sigma$ at $\tau = 180$ s | $\Gamma_{\text{hole}}$ | $\Gamma_{\text{h}}$ | $T_2$              |
|-----------------|----------------------------|------------------------|---------------------|--------------------|
| Freq.-locked    | 2 Hz                       | 358 kHz                | 179 kHz             | 1.77 $\mu\text{s}$ |
| Free running    | 300 kHz                    | 487 kHz                | 244 kHz             | 1.30 $\mu\text{s}$ |

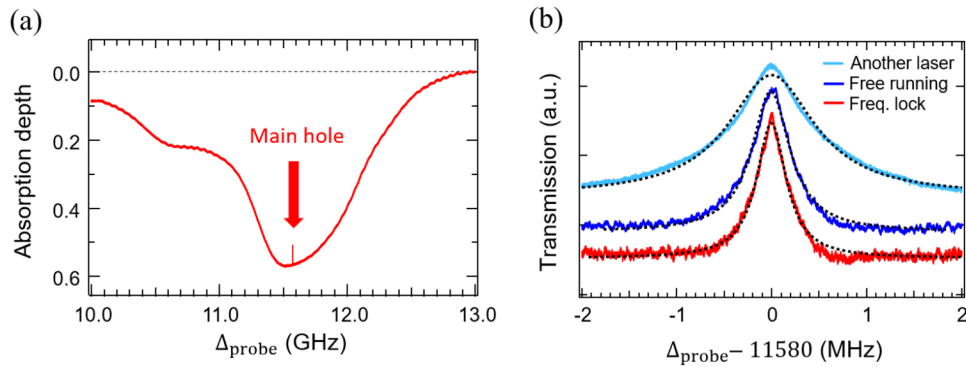
be actually reproduced by substituting to  $\sqrt{\sigma_{\text{free}}^2 + \Gamma_{\text{hole,lock}}^2} = \sqrt{300^2 + 358^2} \sim 467$  kHz. The value is in good agreement with the measurement.

From the obtained hole width  $\Gamma_{\text{hole}}$ , the homogeneous linewidth  $\Gamma_{\text{h}}$  and optical coherence time  $T_2$  can be evaluated by the following relation that takes into account the effect of power broadening [33].

$$\Gamma_{\text{hole}} = \Gamma_{\text{h}} \left( 1 + \sqrt{1 + \Omega^2 T_1 T_2} \right) = \begin{cases} 2\Gamma_{\text{h}} & (\Omega^2 T_1 T_2 \ll 1) \\ \Gamma_{\text{h}} \sqrt{\Omega^2 T_1 T_2} & (\Omega^2 T_1 T_2 \gg 1) \end{cases} \quad (1)$$

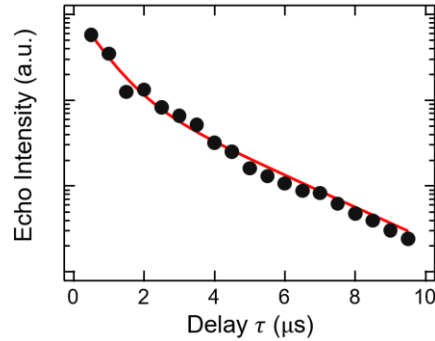
Here,  $\Omega^2 T_1 T_2$  is the dimensionless intensity,  $T_1$  is the population lifetime, and  $\Omega$  is Rabi frequency. The homogeneous linewidth is defined as  $\Gamma_{\text{h}} = 1/(\pi T_2)$ . When the intensity  $\Omega^2 T_1 T_2$  is sufficiently small, the homogeneous linewidth is equal to the hole width divided by 2. The calculated  $\Gamma_{\text{h}}$  values during free running and frequency stabilization are  $\Gamma_{\text{h, free}} = 244$  kHz and  $\Gamma_{\text{h, lock}} = 179$  kHz, respectively. The optical coherence times  $T_2$  derived from these  $\Gamma_{\text{h}}$ 's are 1.30 and 1.77  $\mu\text{s}$ , respectively, as shown in Table 1.

To compare these results with those from time-domain measurements, we performed two-pulse PE measurements by using the same stabilized ECLD1. The experimental system is the same as in Ref.32, where the CW source is shaped into a pulse using an AOM. Figure 5 shows the measurement results, showing that there are two components in the observed signal. The coherence time of each component was 2.5  $\mu\text{s}$  and 9.3  $\mu\text{s}$  by fitting with a double exponential function. Since this measurement was performed under zero magnetic field, the faster relaxation component may be due to the modulation by spectral diffusion effects such as the superhyperfine



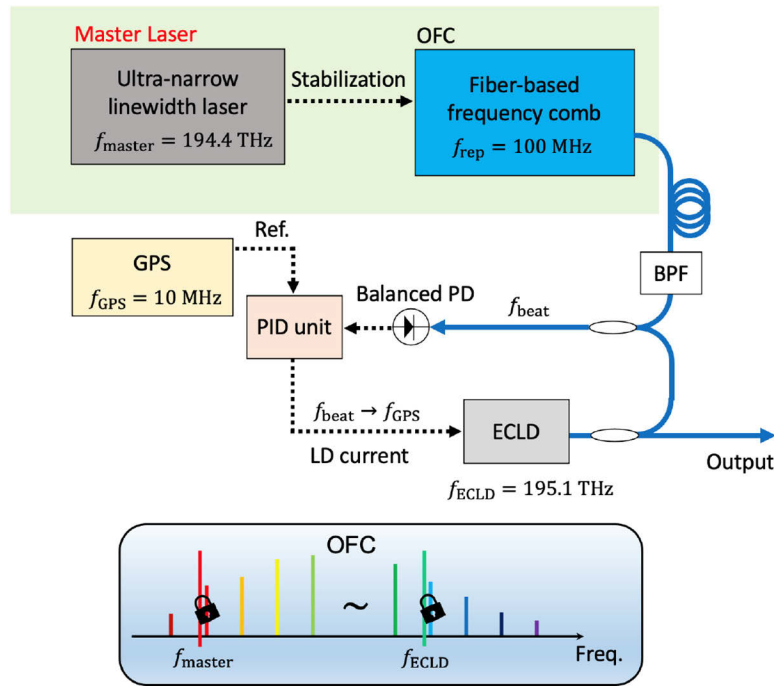
**Fig. 4.** (a) Inhomogeneous broadened transmission spectrum of the  $^4I_{15/2}(Z_1) - ^4I_{13/2}(Y_1)$  transition and main hole. (b) Normalized hole spectra during frequency stabilization (red) and free running (blue) under the use of ECLD in Figs. 1 and 3. The light blue solid line is the hole spectrum in free running when another ECLD with larger frequency fluctuation was used. The center frequency is the peak frequency of each hole spectrum. The thick black dotted lines represent the fitted Lorentzian curves, and the free running hole spectra are shown with an offset for easy viewing. The difference between the peak and edge absorption depth of the hole spectrum during frequency stabilization was 0.062.

(SHF) interaction, which is the interaction between the electrons of  $^{167}\text{Er}^{3+}$  and the nuclear spins of the Y component of the host crystal YSO [34,35]. Considering this result, the obtained hole width is mainly determined by the faster relaxation component with a large magnitude because of the Fourier transform relationship between the two measurements. Furthermore, considering the effect of power broadening, the two results agree even better. Therefore, we can conclude that the results obtained in the SHB measurement are consistent with the PE measurement results. In the sense, SHB measurement is suitable for the investigation of the fast decay component like due to SHF interaction.



**Fig. 5.** Decay envelope of the observed echos (black circles) in two-pulse PE measurement. The double exponential fit of the signal is shown by the red line. Details of the measurement system and pulse sequence are seen in Ref. 32.

In addition, the generation of spectral holes that accurately reflect the homogeneous linewidth has a significant impact on the realization of the AFC memory protocols. It is known that the efficiency is reduced due to the finite homogeneous linewidth [36]. Efficient AFC echo and spin-wave storage, as well as the storage of large numbers of time-position modes, require narrowing the homogeneous linewidth as much as possible. Since AFCs are created using SHB-based methods, they are affected by laser frequency fluctuations. Therefore, the elimination of laser frequency fluctuations is also essential for efficient AFC quantum memory. By stabilizing the frequency, it is possible to create high-resolution spectral holes. In the future, further narrowing of the spectral hole by suppressing the SHF interaction is expected by applying a magnetic field toward the realization of efficient AFC echo.



**Fig. 6.** Schematic diagram of laser frequency stabilization system. BPF: band-pass filter. ECLD: external cavity laser diode. GPS: global positioning system. Details of the stabilization of the optical frequency comb (OFC) in the green shaded area are reported in Ref. 27.

#### 4. Conclusion

In conclusion, frequency stabilization using a fiber laser comb is an excellent method for precision spectroscopy and application to AFC quantum memory protocols. We obtained an Allan deviation of  $< 10$  Hz with an integration time of 180 s by stabilization based on this method. In addition to being more than five orders of magnitude more stable than when running free, the effect of frequency fluctuations is eliminated even during long measurement times. Furthermore, since the light source can be locked to any frequency comb tooth, the frequency to stabilize can be selected every 100 MHz within the range of 2 THz. To demonstrate the impact of this technique on the spectroscopy of rare-earth ions, homogeneous linewidth measurements of HF inter-sublevel optical transitions in  $^{167}\text{Er}^{3+}:\text{YSO}$  were performed by SHB. As a result, the homogeneous linewidths were found to be in good agreement with those obtained from PE measurements, although there was an effect of power broadening. The homogeneous linewidth of the optical transitions between HF sublevels can be measured accurately in the frequency domain, which was previously limited by frequency fluctuations. In addition, it is now possible to create high-resolution AFCs. This paves the way for the realization of AFC quantum memory and further understanding of the relaxation mechanism.

#### Appendix

##### A1. Frequency stabilization system using a fiber laser comb

A schematic of the frequency stabilization system is shown in Fig. 6. In this study, an ultra-narrow linewidth laser (Menlo Systems, ORS) was used as a master laser to stabilize the frequency of the ECLD. The master laser cavity is made of ultra-low-expansion glass with a finesse of

over 400,000, and the linewidth and fluctuation are both  $< 1$  Hz. Since the frequency of the master laser is not tunable, the frequency of the ECLD was stabilized indirectly via an OFC. We first frequency-locked the passively mode-locked Er-doped fiber laser comb (NEOARK CO.) to the master laser and then frequency-locked the ECLD to the fiber laser comb to achieve arbitrary selection of the stabilized frequency. The repetition rate of the OFC is 100 MHz, and the frequency of the ECLD can be locked to any frequency comb tooth in the band of approximately 2 THz. Therefore, the frequency of the ECLD is indirectly locked to the master laser through the OFC. The frequency locking mechanism of the OFC to the master laser is similar to that reported by A. Ishizawa *et al.* [27]. Note that we used a signal from an RF signal generator as a reference to stabilize the carrier envelope offset in this study. The Allan deviation of the stabilized OFC is approximately 5 mHz with an integration time of 1 s. The output from the stabilized OFC passes through a BPF with a bandwidth of 3 GHz to select only the OFC close to the frequency of the ECLD. Then, the beams from the OFC and the ECLD are combined using an optical coupler, and the resulting beat signal is detected by a balanced photodetector. The frequency difference in the beat signal against the reference signal of 10 MHz from the global positioning system (GPS) is fed back to the LD current of the ECLD by a PID control unit (NEOARK CO.). By closed-loop control consisting of this sequence, the frequency of the ECLD is finally stabilized. During free-running, the reference signal from GPS is blocked and there is no feedback to the LD current.

## A2. Allan deviation

The average of the instantaneous frequency  $f(t)$  over the integration time  $\tau$  is given by

$$\bar{f}_k(\tau) = \frac{1}{\tau} \int_{t_k}^{t_k+\tau} f(t) dt. \quad (2)$$

The deviation of the frequency fluctuation,  $\bar{f}_{k+1} - \bar{f}_k$ , over the integration time  $\tau$  is the Allan deviation  $\sigma(\tau)$ , which is defined as

$$\sigma^2(\tau) = \frac{1}{N-1} \sum_{k=1}^{N-1} \frac{1}{2} (\bar{f}_{k+1} - \bar{f}_k)^2. \quad (3)$$

Since frequency fluctuations vary on various time scales depending on their type, the Allan deviation against the integration time shown in Fig. 2 allows us to evaluate the frequency fluctuations on the time scale of the measurement, and is often used to evaluate the frequency stability [37,38].

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**Data availability.** Data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon reasonable request.

## References

1. S. Wehner, D. Elkouss, and R. Hanson, "Quantum internet: A vision for the road ahead," *Science* **362**(6412), eaam9288 (2018).
2. M. K. Bhaskar, R. Riedinger, B. Machielse, D. S. Levonian, C. T. Nguyen, E. N. Knall, H. Park, D. Englund, M. Lončar, D. D. Sukachev, and M. D. Lukin, "Experimental demonstration of memory-enhanced quantum communication," *Nature* **580**(7801), 60–64 (2020).
3. H. J. Kimble, "The quantum internet," *Nature* **453**(7198), 1023–1030 (2008).
4. M. Zhong, M. P. Hedges, R. L. Ahlefeldt, J. G. Bartholomew, S. E. Beavan, S. M. Wittig, J. J. Longdell, and M. J. Sellars, "Optically addressable nuclear spins in a solid with a six-hour coherence time," *Nature* **517**(7533), 177–180 (2015).

5. G. Heinze, C. Hubrich, and T. Halfmann, "Coherence time extension in  $\text{Pr}^{3+}:\text{Y}_2\text{SiO}_5$  by self-optimized magnetic fields and dynamical decoupling," *Phys. Rev. A* **89**(5), 053825 (2014).
6. M. Rančić, M. P. Hedges, R. L. Ahlefeldt, and M. J. Sellars, "Coherence time of over a second in a telecom-compatible quantum memory storage material," *Nat. Phys.* **14**(1), 50–54 (2018).
7. S. Ritter, C. Nölleke, C. Hahn, A. Reiserer, A. Neuzner, M. Uphoff, M. Mücke, E. Figueroa, J. Bochmann, and G. Rempe, "An elementary quantum network of single atoms in optical cavities," *Nature* **484**(7393), 195–200 (2012).
8. R. W. Equall, Y. Sun, R. L. Cone, and R. M. Macfarlane, "Ultralow optical dephasing in  $\text{Eu}^{3+}:\text{Y}_2\text{SiO}_5$ ," *Phys. Rev. Lett.* **72**(14), 2179–2182 (1994).
9. R. Yano, M. Mitsunaga, and N. Uesugi, "Ultralong optical dephasing time in  $\text{Eu}^{3+}:\text{Y}_2\text{SiO}_5$ ," *Opt. Lett.* **16**(23), 1884–1886 (1991).
10. T. Böttger, C. W. Thiel, R. L. Cone, and Y. Sun, "Effects of magnetic field orientation on optical decoherence in  $\text{Er}^{3+}:\text{Y}_2\text{SiO}_5$ ," *Phys. Rev. B* **79**(11), 115104 (2009).
11. W. Tittel, M. Afzelius, T. Chanelière, R. Cone, S. Kröll, S. Moiseev, and M. Sellars, "Photon-echo quantum memory in solid state systems," *Laser & Photon. Rev.* **4**(2), 244–267 (2009).
12. B. Lauritzen, S. R. Hastings-Simon, H. de Riedmatten, M. Afzelius, and N. Gisin, "State preparation by optical pumping in erbium-doped solids using stimulated emission and spin mixing," *Phys. Rev. A* **78**(4), 043402 (2008).
13. B. Lauritzen, J. Minář, H. de Riedmatten, M. Afzelius, N. Sangouard, C. Simon, and N. Gisin, "Telecommunication-wavelength solid-state memory at the single photon level," *Phys. Rev. Lett.* **104**(8), 080502 (2010).
14. D. Hashimoto and K. Shimizu, "Coherent Raman beat analysis of the hyperfine sublevel coherence properties of  $^{167}\text{Er}^{3+}$  ions doped in an  $\text{Y}_2\text{SiO}_5$  crystal," *J. Lumin.* **171**, 183–190 (2016).
15. J. V. Rakonjac, Y.-H. Chen, S. P. Horvath, and J. J. Longdell, "Long spin coherence times in the ground state and in an optically excited state of  $^{167}\text{Er}^{3+}:\text{Y}_2\text{SiO}_5$  at zero magnetic field," *Phys. Rev. B* **101**(18), 184430 (2020).
16. E. Saglamyurek, J. Jin, V. B. Verma, M. D. Shaw, F. Marsili, S. W. Nam, D. Oblak, and W. Tittel, "Quantum storage of entangled telecom-wavelength photons in an erbium-doped optical fibre," *Nat. Photonics* **9**(2), 83–87 (2015).
17. I. Craiciu, M. Lei, J. Rochman, J. M. Kindem, J. G. Bartholomew, E. Miyazono, T. Zhong, N. Sinclair, and A. Faraon, "Nanophotonic quantum storage at telecommunication wavelength," *Phys. Rev. Appl.* **12**(2), 024062 (2019).
18. E. Baldit, K. Bencheikh, P. Monnier, S. Briaudeau, J. A. Levenson, V. Crozatier, I. Lorgère, F. Bretenaker, J.-L. Le Gouët, O. Guillot-Nöel, and Ph. Goldner, "Identification of  $\Lambda$ -like systems in  $\text{Er}^{3+}:\text{Y}_2\text{SiO}_5$  and observation of electromagnetically induced transparency," *Phys. Rev. B* **81**(14), 144303 (2010).
19. R. M. Macfarlane, "High-resolution laser spectroscopy of rare-earth doped insulators: a personal perspective," *J. Lumin.* **100**(1–4), 1–20 (2002).
20. M. Nicolle, J. N. Becker, C. Weinzettl, I. A. Walmsley, and P. M. Ledingham, "Gigahertz-Bandwidth Optical Memory in  $\text{Pr}^{3+}:\text{Y}_2\text{SiO}_5$ ," arXiv:2102.13113
21. J. Stuart, M. Hedges, R. Ahlefeldt, and M. Sellars, "Initialisation protocol for efficient quantum memories using resolved hyperfine structure," arXiv:2103.04581
22. M. Aldous, J. Woods, A. Dragomir, R. Roy, and M. Himsworth, "Carrier frequency modulation of an acousto-optic modulator for laser stabilization," *Opt. Express* **25**(11), 12830–12838 (2017).
23. P. B. Sellin, N. M. Strickland, T. Böttger, J. L. Carlsten, and R. L. Cone, "Laser stabilization at 1536 nm using regenerative spectral hole burning," *Phys. Rev. B* **63**(15), 155111 (2001).
24. K. Ikeda, Y. Hisai, K. Yoshii, H. Kosaka, F.-L. Hong, and T. Horikiri, "Compact frequency-stabilized pump laser for wavelength conversion in long-distance quantum communication," *J. Opt. Soc. Am. B* **35**(8), 2023–2028 (2018).
25. T. Ideguchi, S. Holzner, B. Bernhardt, G. Guelachvili, N. Picqué, and T. W. Hänsch, "Coherent Raman spectro-imaging with laser frequency combs," *Nature* **502**(7471), 355–358 (2013).
26. Th. Udem, J. Reichert, R. Holzwarth, and T. W. Hänsch, "Absolute optical frequency measurement of the Cesium  $D_1$  line with a mode-locked laser," *Phys. Rev. Lett.* **82**(18), 3568–3571 (1999).
27. A. Ishizawa, K. Hitomi, K. Hara, K. Hitachi, T. Nishikawa, T. Sogawa, and H. Gotoh, "Simple method for stabilizing an optical frequency comb to an optical reference without an RF signal generator," *OSA Continuum* **2**(5), 1706–1712 (2019).
28. R. W. P. Drever, J. L. Hall, F. V. Kowalski, J. Hough, G. M. Ford, A. J. Munley, and H. Ward, "Laser phase and frequency stabilization using an optical resonator," *Appl. Phys. B* **31**(2), 97–105 (1983).
29. G. J. Pryde, T. Böttger, R. L. Cone, and R. C. C. Ward, "Semiconductor lasers stabilized to spectral holes in rare earth crystals to a part in  $10^{13}$  and their application to devices and spectroscopy," *J. Lumin.* **98**(1–4), 309–315 (2002).
30. T. Tawara, G. Mariani, K. Shimizu, H. Omi, S. Adachi, and H. Gotoh, "Effect of isotopic purification on spectral-hole narrowing in  $^{167}\text{Er}^{3+}$  hyperfine transitions," *Appl. Phys. Express* **10**(4), 042801 (2017).
31. T. Böttger, C. W. Thiel, Y. Sun, and R. L. Cone, "Optical decoherence and spectral diffusion at 1.5  $\mu\text{s}$  in  $\text{Er}^{3+}:\text{Y}_2\text{SiO}_5$  versus magnetic field, temperature, and  $\text{Er}^{3+}$  concentration," *Phys. Rev. B* **73**(7), 075101 (2006).
32. M. Hiraishi, M. Ijspeert, T. Tawara, S. Adachi, R. Kaji, H. Omi, and H. Gotoh, "Optical coherent transients in  $^{167}\text{Er}^{3+}$  at telecom-band wavelength," *Opt. Lett.* **44**(20), 4933–4936 (2019).
33. H. Vries and D. A. Wiersma, "Photophysical and photochemical molecular hole burning theory," *J. Chem. Phys.* **72**(3), 1851–1863 (1980).
34. T. Zhong, J. M. Kindem, E. Miyazono, and A. Faraon, "Nanophotonic coherent light-matter interfaces based on rare-earth-doped crystals," *Nat. commun.* **6**(1), 8206 (2015).

35. B. Car, J.-L. Le Gouët, and T. Chanelière, "Superhyperfine induced photon-echo collapse of erbium in  $\text{Y}_2\text{SiO}_5$ ," *Phys. Rev. B* **102**(11), 115119 (2020).
36. M. Afzelius, C. Simon, H. de Riedmatten, and N. Gisin, "Multimode quantum memory based on atomic frequency combs," *Phys. Rev. A* **79**(5), 052329 (2009).
37. D. W. Allan, "Time and frequency (time-domain) characterization, estimation, and prediction of precision clocks and oscillators," *IEEE Trans. UFFC* **34**(6), 647–654 (1987).
38. D. W. Allan, M. A. Weiss, and J. L. Jespersen, "A Frequency-domain view of time-domain characterization of clocks and time and frequency distribution systems," *Proceedings of the 45th annual symposium on frequency control* 667–678 (1991).