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Thermal-noise Limits to the Frequency Stability of Burned Spectral Holes

M. T. Hartman,^{1, a)} N. Wagner,² S. Seidelin,³ and B. Fang^{1, b)}

¹⁾*LNE-SYRTE, Observatoire de Paris, Université PSL, CNRS, Sorbonne Université, 75014 Paris, France*

²⁾*Institut für Halbleitertechnik, Technische Universität Braunschweig, Hans-Sommer-Str. 66, Braunschweig, 38106 Germany*

³⁾*Univ. Grenoble Alpes, CNRS, Grenoble INP and Institut Néel, 38000 Grenoble, France*

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Techniques in frequency stabilization of lasers to fixed-spacer optical cavities have advanced to the point where the ultimate frequency stabilities are limited by thermal noise in the cavity materials for standard cavity configurations at room temperature. The use of spectral-hole burning (SHB) in laser stabilization has produced promising results in early experiments. In this letter we explore the thermal-noise limits to frequency stability in burned spectral holes. We compile known material parameters for a typical system used in SHB experiments (Eu³⁺-doped Y₂SiO₅) to make numerical estimates for the fundamental thermal-noise induced frequency instability in spectral-holes for the liquid-helium temperature and dilution temperature cases. These efforts can guide the design of future SHB experiments and clarify which important material parameters remain to be measured.

I. INTRODUCTION

Spectral-hole burning (SHB) is a technique for producing persistent narrow transmissive spectral lines, or ‘spectral holes’, within an inhomogeneously broadened absorption band¹. In practice, within a doped crystal, the broadened absorption band is provided by random spectral displacement of the narrow homogeneous absorption lines of the dopant ions or atoms caused by crystal matrix deformation at the doping-locations. Selecting a system for which the ground state has multiple hyperfine levels allows for transmissive lines to be ‘burned’ (or photo imprinted) into the absorption band by using a pump laser to deplete one of the hyperfine levels which is resonant with the pump laser frequency. The result is a powerful technique which allows the production of an arbitrary spectral pattern at the desired working frequencies. With this capability, SHB has been leveraged by researchers for diverse applications in fields such as quantum memory and information processing^{2–8}, ultra-wideband spectral analysis^{9,10}, quantum opto-mechanics^{11–13}, and laser frequency stabilization^{14–20}.

Regardless of their application, experiments involving SHB are concerned, to one degree or another, with the frequency noise in the spectroscopic measurement of the spectral hole. This is directly evident in application to laser frequency stabilization, for which SHB has been proposed as an alternative to optical cavities as a frequency reference. In this context, sources of frequency noise can be categorized into two groups: the first group, detection noise (shot noise, electronic noise, etc), limits

the ability to resolve the spectral hole frequency. While the requirement to use low probe laser power in SHB accentuates this group, it is a technical noise source which has been mitigated by innovative detection schemes^{20–22}. The second group consists of noise sources that shift the frequency of the spectral line itself. This group contains noise sources that produce a fundamental physical limit and are the focus of this letter.

Currently, state of the art ultra-stable lasers reference their frequency to linear optical cavities, which consist of two partially reflective mirrors spaced a distance, d , apart. An incident spatially mode-matched laser beam will produce a build-up of intracavity laser power when the resonance condition is met, that is, when the cavity length is an integer, N , multiple of the half wavelengths, λ , of incident light, $d = N\frac{\lambda}{2}$. Typically a feedback scheme^{23–25} uses the intracavity field to produce an error signal to control the incident laser frequency, ν_0 , locking the laser to one of the resonant modes which are spaced at intervals of the free spectral range, $\text{FSR} = \frac{c}{2d}$, where c is the speed of light in vacuum. The result is a reference where the fractional frequency fluctuations are proportional to cavity length changes: $\frac{\delta\nu}{\nu_0} = -\frac{\delta d}{d}$.

By contrast, frequency fluctuations in spectral holes can result from phenomena which produce energy shifts in the transition. The significance of externally driven effects on the frequency stability in SHB has motivated the characterization of spectral-hole line shifts due to several types of environmental perturbations. These characterizations have focused on a particularly promising medium, Eu³⁺ doped into a Y₂SiO₅ crystal matrix (Eu:YSO), whose long lifetime at cryogenic temperatures and long coherence time present desirable spectroscopic properties. One such important effect is the line shift resulting from a change in the electric field interacting with the dipole moment of the ion transition. This can result from either an applied external field (Stark effect)²⁶ or from external mechanical stress causing a deformation of the

^{a)}Currently at: Laboratoire Astroparticule et Cosmologie (APC) 75013 Paris, France

hartman@apc.in2p3.fr

^{b)}Electronic mail: bess.fang@obspm.fr

crystal lattice around the ion^{17,27,28}. A second significant effect is a change in phonon density which results in an energy shift in the transition as predicted in a two-phonon scattering process²⁹. The result is manifest as temperature dependent line shift^{16,17,29–32}. Making use of these previous studies' characterizations of spectral-hole response to external forces, as well as new material property measurements, this letter estimates the fundamental frequency-stability of spectral-holes due to intrinsic thermal-noise.

Thermal noise presents a fundamental physical limit in numerous precision measurement experiments. Broadly speaking, thermal noise describes the fluctuation of a measurable coordinate as a result of internal thermal-energy driven motion. In the context of the development of cavity-based ultra-stable lasers, thermal noise is manifest as a fluctuation in the apparent surface position of cavity mirrors, as calculated by application of the fluctuation-dissipation theorem^{33–36}, and is already well known as the limiting frequency-noise source in standard optical-cavity-referenced lasers³⁷. This limitation has driven the exploration of SHB as an alternative frequency reference for the development of ultra-stable lasers^{14–17,19,20}, yet, to our knowledge, there currently exist no estimates of thermal-noise in SHB. As the mechanisms producing frequency shifts in spectral holes are fundamentally different from those in optical cavities, we here calculate thermal-noise in SHB. In this work, we derive expressions for thermal-noise induced frequency-fluctuations via two mechanisms, thermomechanical and thermodynamical noise, for a typical sample geometry used in SHB: a rectangular parallelepiped crystal doped with rare-earth ions. We then apply known material and spectroscopic parameters to provide representative numerical estimates of thermal-noise driven fractional frequency instability in SHB.

II. THERMOMECHANICAL NOISE

Thermomechanical noise, or Brownian thermal noise, describes a displacement noise that is the result of mechanical energy dissipation, driven by random thermal motion, through the internal mechanical loss angle, ϕ , due to internal friction. A coupling mechanism of thermomechanical noise to spectral-hole frequency-noise is through stress induced deformation of the crystal lattice. An important material parameter in this coupling, the frequency sensitivity to mechanical-stress, ($K_D = \frac{\delta\nu}{\delta P} [\frac{\text{Hz}}{\text{Pa}}]$), has been previously characterized in Eu:YSO along two dielectric axes ($D = 1, 2$) using externally applied uni-axial pressures²⁸.

To aid in our calculation of thermal noise we make use of the fluctuation dissipation theorem (FDT). Developed by Callen et al.³⁸, the FDT describes systems in which a perturbing force F , which displaces coordinate x , is irreversibly dissipated into thermal energy. In general, the FDT gives the power spectral density (PSD) of fluctua-

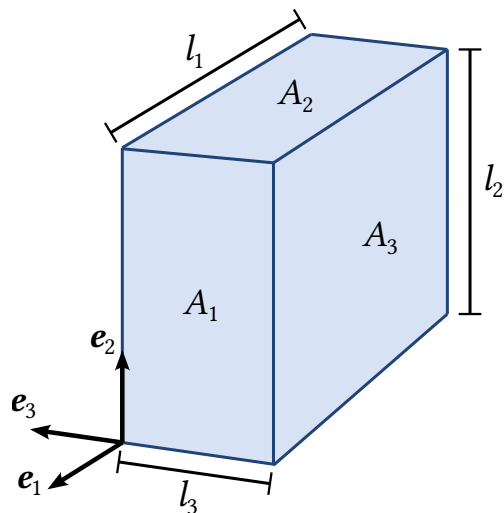


FIG. 1. Dimensional diagram of a crystal used in SHB.

tions in x , S_x (or, conversely, the PSD of fluctuations in F , S_F), over a spectrum of angular frequencies ω ,

$$S_x(\omega) = \frac{4k_B T_0}{\omega} \Im[\chi(\omega)], \quad (1)$$

$$S_F(\omega) = \frac{4k_B T_0}{\omega} \Im\left[\frac{1}{\chi(\omega)}\right], \quad (2)$$

as a function of the internal thermal energy (as given by the Boltzmann constant, k_B , and temperature, T_0) and the imaginary part of the complex mechanical susceptibility of x to F ,

$$\chi(\omega) = \frac{x(\omega)}{F(\omega)}, \quad (3)$$

which contains resistive and dissipative components.

We examine the case of a rectangular parallelepiped crystal matrix of volume $V_x = l_1 l_2 l_3$, illustrated in FIG. 1, where the lengths, l_j , along axes e_j , and orthogonal crystal cross-sectional areas, A_j , are given by the corresponding dimensional matrices,

$$\mathbf{M}_l = \begin{bmatrix} l_1 & 0 & 0 \\ 0 & l_2 & 0 \\ 0 & 0 & l_3 \end{bmatrix}, \quad \mathbf{M}_A = \begin{bmatrix} l_2 l_3 & 0 & 0 \\ 0 & l_1 l_3 & 0 \\ 0 & 0 & l_1 l_2 \end{bmatrix}. \quad (4)$$

Given a material with a complex Young's modulus $\tilde{E} = E(1 - i\phi)$ and Poisson's ratio σ , we can write the mechanical compliance matrix,

$$\mathbf{M}_c = \frac{1}{\tilde{E}} \begin{bmatrix} 1 & -\sigma & -\sigma \\ -\sigma & 1 & -\sigma \\ -\sigma & -\sigma & 1 \end{bmatrix}, \quad (5)$$

which allows us to express the complex mechanical susceptibility of the crystal,

$$\chi = \mathbf{M}_l \mathbf{M}_c \mathbf{M}_A^{-1}. \quad (6)$$

We can use the fractional-frequency stress sensitivity,

$$\vec{k} = \begin{bmatrix} k_1 \\ k_2 \\ k_3 \end{bmatrix} = \frac{1}{\nu_0} \begin{bmatrix} K_1 \\ K_2 \\ K_3 \end{bmatrix}, \quad (7)$$

with (2) calculate the PSD of fractional-frequency noise contribution along each dielectric axis,

$$\begin{aligned} \vec{S}_{\frac{\delta\nu}{\nu_0}}(\omega) &= \frac{4k_B T_0}{\omega} (\mathbf{M}_A^{-1})^2 \Im(\chi^{-1}) \left(\vec{k} \odot \vec{k} \right) \\ &= \frac{4k_B T_0}{\omega} \frac{E\phi}{(1-2\sigma)(1+\sigma)} \frac{1}{l_1 l_2 l_3} \\ &\quad \times \begin{bmatrix} \frac{1}{l_2 l_3} (k_1^2 l_2 l_3 (1-\sigma) + k_2^2 l_1 l_3 \sigma + k_3^2 l_1 l_2 \sigma) \\ \frac{1}{l_1 l_3} (k_1^2 l_2 l_3 \sigma + k_2^2 l_1 l_3 (1-\sigma) + k_3^2 l_1 l_2 \sigma) \\ \frac{1}{l_1 l_2} (k_1^2 l_2 l_3 \sigma + k_2^2 l_1 l_3 \sigma + k_3^2 l_1 l_2 (1-\sigma)) \end{bmatrix}. \end{aligned} \quad (8)$$

where \odot denotes a Hadamard product, an element-wise multiplication.

III. THERMO-SPECTRAL NOISE

A second category of thermal noise, thermodynamic noise, describes the measurable effects from intrinsic temporal variations of temperature over a probed volume known from statistical thermodynamics³⁹. Such a class of fundamental noise has been identified and characterized in laser cavity experiments, where temperature fluctuations give rise to cavity-length fluctuations via the thermoelastic and thermorefractive effects in the cavity mirror substrates^{34,40,41} and coatings⁴¹⁻⁴³. By contrast, spectral-hole temperature dependence is the result of phonon-scattering driven spectral line shifts with a general T^4 trend²⁹. This mechanism, aside from anomalous localized deviations³², produces a general T^3 trend in frequency sensitivity to temperature $\left(\Gamma = \frac{d\nu}{dT} \Big|_{T_0} \left[\frac{\text{Hz}}{\text{K}} \right] \right)$ driving experiments to lower temperatures for performance gains.

In the case of SHB, we are interested in the temperature variations in the volume of crystal being probed by a laser beam of radius r_b . Historically, for most cases (≈ 10 cm optical cavities at room temperature), the time scales of interest for temperature fluctuations are much shorter than the system's thermal time constant, $\tau_{\text{th}} = \frac{r_b^2 \rho c_p}{\kappa}$, set by the radius of the cylindrical probed area, r_b , and the density, ρ , specific heat capacity, c_p , and thermal conductivity, κ , of the probed material; in these cases, an adiabatic approximation, which assumes that temperature variations occur within confines of the probed region, is valid³⁴. However, in the case of small beam sizes or high thermal diffusivity, defined as $\alpha = \frac{\kappa}{\rho c_p}$, temperature fluctuations diffuse over a distance larger than the probe beam during the measurement times of interest, requiring a more general expression for the PSD of temperature fluctuations within a volume has been solved for the case of a beam reflecting from a surface⁴⁰, and, in a case more applicable to

SHB, in transmission through an optic, where we follow Braginsky & Vyatchanin⁴⁴:

$$S_T(\omega) = \frac{k_B T_0^2}{\pi l_3 \kappa} \int_0^\infty \frac{u e^{-u}}{(\omega \tau_{\text{th}})^2 + u^2} du. \quad (9)$$

From (9), we can write the PSD of fractional-frequency noise due to temperature fluctuation driven spectral-hole line shifts, which we refer to as thermo-spectral noise,

$$S_{\frac{\delta\nu}{\nu_0}}(\omega) = \gamma^2 S_T(\omega), \quad (10)$$

in terms of the fractional-frequency temperature sensitivity, $\gamma = \frac{1}{\nu_0} \Gamma$.

IV. NUMERICAL ESTIMATES

To provide a representative estimate of the fundamental stability limit in current SHB experiments, here we make numerical estimates for thermal-noise induced frequency instability using the typical material and experimental parameters of current and past SHB laser stabilization experiments. Namely, we follow the experimental setup as presented by the SHB group at LNE-SYRTE^{18,20}, which utilizes a rectangular cuboid YSO crystal of dimension $8 \times 8 \times 4$ mm²⁸ doped with Eu^{3+} , however other SHB laser stabilization experiments use similarly dimensioned samples (for example a YSO cuboid of $5 \times 6 \times 1$ mm at Montana State University⁴⁸; a YSO cylinder of length 5 mm and 10 mm in diameter at NIST¹⁷). While characterizations of material and spectroscopic properties of $\text{Eu}:\text{YSO}$ at cryogenic temperatures are incomplete, we have compiled some known property values at various temperatures in TABLE I, which are used in all numerical estimates made in this section.

For thermomechanical noise, we take the interaction between the orthogonal axes to be coupled via Poisson's ratio, and, in the case described in TABLE I (i.e., the LNE-SYRTE SHB experiment, where $l_1 = l_2 = L$ and $l_3 = l$), (8) becomes:

$$\begin{aligned} S_{\frac{\delta\nu}{\nu_0}}(\omega) &= \sum_j \vec{S}_{\frac{\delta\nu}{\nu_0}}(\omega) \cdot \vec{e}_j \\ &= \frac{4k_B T_0}{\omega} \frac{E\phi}{(1-2\sigma)(1+\sigma)} \frac{1}{V_x} \left((k_1^2 + k_2^2) \left(1 + \sigma \frac{l}{L} \right) \right. \\ &\quad \left. + k_3^2 \left(1 + \sigma \frac{2L-l}{l} \right) \right). \end{aligned} \quad (11)$$

The mechanical loss angle is known to be a function of temperature⁴⁹, though different materials show significantly different, to even inverse, proportionalities with temperature⁵⁰. Measurements of ϕ in YSO⁴⁶ have indicated decreasing internal mechanical losses with cooling, while not reaching the temperatures concerned here. As

Parameter	Value (Room Temp)	Value (4K)	Value (300 mK)	Units	Reference
r_b	3.5	-	-	mm	-
$l_1 \times l_2 \times l_3$	$8 \times 8 \times 4$	†	†	mm ³	28
ρ	4400	†	†	kg m ⁻³	45
E	150	158	158	GPa	45,46
σ	0.26	†	†	-	45
c_p		2×10^{-2}	$1 \times 10^{-5} \Delta$	J kg ⁻¹ K ⁻¹	47
κ		40	0.02 Δ	W m ⁻¹ K ⁻¹	47
ν_0 *	516.85	-	-	THz	28
K_1 *	N/A	46	‡	Hz Pa ⁻¹	28
K_2 *	N/A	-19	‡	Hz Pa ⁻¹	28
K_3 *	N/A	◇	‡ ◇	Hz Pa ⁻¹	28
Γ *	N/A	23 000	5	Hz K ⁻¹	32

TABLE I. Parameter values used in the estimation of thermal noise in SHB

* Value at spectroscopic site 1, the site typically used in laser stabilization

† Value at room temperature was used for estimates, it is not expected to vary greatly going to cryogenic temperatures

‡ Value unknown at 300 mK. For calculations at 300 mK we use 4 K values.

◇ Value unknown for dielectric axis 3, we set this value to the larger of other two axis values (46 Hz Pa⁻¹) for the numerical estimates.

Δ Values at 4 K are measured, whereas values at 300 mK are model projected.

such, in FIG. 2, we plot the Allan deviation (ADEV),

$$\sigma_{\frac{\delta\nu}{\nu_0}}(\tau) = \sqrt{\frac{1}{\pi} \int_0^\infty S_{\frac{\delta\nu}{\nu_0}}(\omega) \frac{\sin^4(\frac{1}{2}\tau\omega)}{(\frac{1}{2}\tau\omega)^2} d\omega}, \quad (12)$$

of relative frequency instability due to thermomechanical (Brownian) noise over three orders of magnitude of ϕ ; other physical parameter values used for this plot can be found in TABLE I. Additionally, while previous SHB experiments operated near liquid helium temperatures^{17,20} (≈ 4 K), a change to operating at dilution refrigerator temperatures (< 1 K) has been strongly motivated³². As such, for each value of ϕ , we plot the Brownian noise levels for temperatures of both $T = 4$ K and $T = 0.3$ K.

For thermo-spectral noise we find the PSD of fractional-frequency fluctuations using Eq. (10) where we compute the integral in Eq. (9) numerically. Using the relationship given in Eq. (12), we plot the resulting Allan deviation of thermo-spectral noise induced frequency instability in black in FIG. 2. Similarly, we perform this calculation for the two mentioned temperatures.

V. DISCUSSION AND CONCLUSIONS

In this letter we have presented key figures of merit in the computation of the thermal-noise limits to spectral-hole frequency stability. We find the thermal noise in Eu:YSO is dominated by thermomechanically-driven Brownian noise at time scales above 10^{-4} s. This term dominates over thermodynamically driven thermo-spectral noise, which is suppressed by a combination of the high thermal diffusivity of YSO at cryogenic temperatures as well as the low temperature sensitivity (Γ) of spectral-holes at dilution temperatures, particularly near

a zero crossing found at spectroscopic site 1 in Eu:YSO around 300 mK³².

While the fundamental frequency stability of spectral holes is important in various experiments, we highlight the direct importance to laser frequency stabilization via SHB. In this domain, ultra-stable lasers find a direct application in time and frequency metrology, where they are used in the probing of atomic transitions in optical lattice clocks. To this end, the goal of reaching the fundamental performance limit of these optical clocks, the quantum projection noise, sets a requirement of fractional-frequency instability of order 10^{-18} for 0.1 to 10 s averaging time in the probe laser⁵¹. In this context, when assuming a mechanical loss angle of order 10^{-4} or lower, SHB in Eu:YSO is a compatible frequency reference for this application. This assumption is realistic in the view of the first mechanical quality factor measurements in YSO⁴⁶.

Here, we point out there are notable limitations to the estimates provided, namely missing material parameters. In the area of thermo-spectral noise, we have measurements of c_p and κ down to 2 K, and below we rely on a model fit to the data, however this provides a reasonable value and is, anyway, the significantly smaller contribution to the total thermal noise. On the subject of thermo-mechanical noise, the sensitivity of spectral holes to stress along dielectric axis 3, K_3 , has not been yet been measured, and will require modifications of our crystal and setup to be able to do so²⁸. Instead, in this letter, we substituted this value with the higher measured sensitivity of the other two axes for a conservative estimate, but the possibility of an exceptionally high sensitivity along this axis cannot be entirely excluded. Furthermore, the other dielectric axes, K_1 and K_2 , have only been measured around 4 K. The mechanism however, is deformation of the crystal lattice around the dopant ion due to external

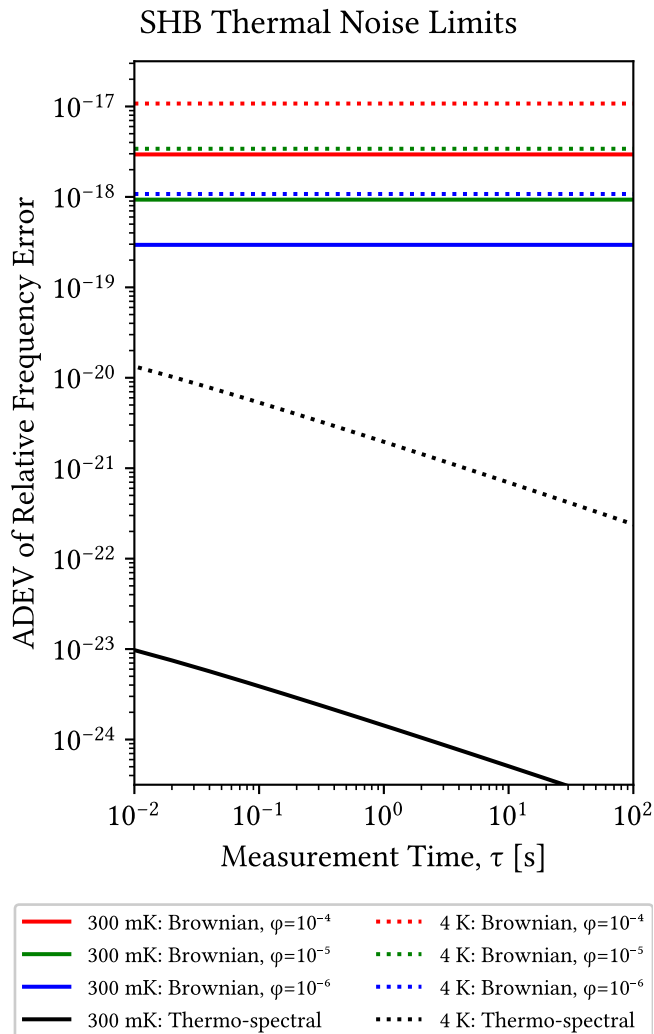


FIG. 2. Allan deviation (ADEV) as function of measurement time for temperatures $T = 300$ mK (solid lines) and $T = 4$ K (dotted lines). The colored lines correspond to the Brownian noise level for loss angles of $\phi = 10^{-4}$ (red), $\phi = 10^{-5}$ (green) and $\phi = 10^{-6}$ (blue), whereas the black lines describe the thermo-spectral noise limit.

stress. Since this mechanical deformation resulting from physical stress is governed by the rigidity of the crystal, which is not expected to vary significantly at these temperatures, we consequently do not expect this parameter to change significantly from 4 K to 300 mK.

Even though there are still missing material properties which limit the accuracy to which we can calculate thermal-noise driven spectral-hole frequency instability, we believe that it does not drastically alter the numerical estimation of SHB stability presented here. Furthermore, the framework presented here provides key elements in the design of future Eu:YSO SHB experiments as well as needs for further YSO material characterizations.

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DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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