Tests of Sapphire Crystals Manufactured with Different Growth Processes for Ultra-stable Microwave Oscillators

Vincent Giordano, Christophe Fluhr, Serge Grop and Benoît Dubois

***Abstract*—We present the characterization of $8-12$ GHz whispering gallery mode resonators machined in high-quality sapphire crystals manufactured with different growth techniques. These microwave resonators are intended to constitute the frequency reference of ultra-stable Cryogenic Sapphire Oscillators. We conducted systematic tests near 4 K to determine the unloaded Q-factor and the turnover temperature for whispering gallery modes in the $8-12$ GHz frequency range. We have shown that high quality sapphire crystals manufactured with the Heat Exchange or the Kyropoulos growth technique are both suitable to meet a fractional frequency stability better than 1×10^{-15} for 1 s to 10,000 s integration times.**

I. Introduction

State-of-the-art ultra-stable microwave oscillators are currently based on a whispering gallery mode sapphire resonator operated in the range $8-12$ GHz. For room temperature oscillators, the best phase noise over 1 Hz to 1 kHz Fourier frequencies is achieved with a sapphire resonator oscillator incorporating a sophisticated electronic circuit degenerating the noise of the sustaining oscillator stage [1]. On the other hand, the Cryogenic Sapphire Oscillator (CSO), in which the sapphire resonator is cooled near 6 K, provides a fractional frequency stability better than 1×10^{-15} for integration times lower than 10,000 s [2], [3]. The recent demonstration of a low maintenance CSO based on a pulse-tube cryocooler paves the way for its deployment in real field applications [4]–[6]. Such oscillators are required for the operation of laser-cooled atomic clocks [7]. The CSO can provide the means to improve the resolution of space vehicles ranging and Doppler tracking provided by Deep Space Networks as well as those of Very Long Baseline Interferometry (VLBI) Observatories [8]–[13]. The CSO can also enhance the calibration capability of Metrological Institutes or help the qualification of high performance clocks or oscillators [7], [14], [15].

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This work has been realized in the frame of the ANR projects: Equipex Oscillator-Imp and Emergence ULISS-2G. The authors would like to thank the Council of the Région de Franche-Comté for its support to the Projets d'Investissements d'Avenir and the FEDER (Fonds Européen de Développement Economique et Régional) for funding one CSO.

Manuscript received XX, YY 2015; revised XX, YY 2015.

The CSO exceptional performances result from the intrinsic properties of the resonator material. A high Q-factor requires a monocrystal with a low structural defects density. Typically Q-factors of 1×10^9 are obtained for modes around 10 GHz with a high quality sapphire crystal cooled down to 4 K. However a pure sapphire crystal will not provide good frequency stability. Indeed the thermal compensation induced by accidental paramagnetic impurities that substitute to Al^{3+} is essential for the achievement of the highest frequency stability. The main issue is that the impurities concentration giving a suitable turnover temperature between 5-8 K is very low (typically less than 1 ppm) and cannot be warranted by the manufacturer. To build a CSO the designer has no other choice than to buy expensive crystals without knowing if they are suitable for his application. A cryogenic test is the only way to reveal the temperature dependence of the resonator frequency and thus to fix the operating temperature.

This paper presents a compilation of all the measurements we conducted on different sapphire crystals since the development of our first liquid-Helium cooled CSO [16]. We complete here our previous report on crystal characterizations presented in [17], adding recent results and giving a deeper analysis.

In section II, we clarify the characteristics of the sapphire crystal needed to reach a fractional frequency stability of 1×10^{-15}. In section III, we describe the different sapphire samples we tested. The preliminary measurements at room temperature are reported in section IV. The low temperature characterizations are presented in section V. Eventually, in section VI, we demonstrate that state-of-the-art frequency stability can be obtained with several kinds of sapphire cristals.

II. The quest for a 1×10^{-15} frequency stability

The CSO incorporates a cylindrical sapphire resonator as a frequency reference. The resonator axis is parallel to the crystal C-axis within ± 0.5 degree. It is placed in the center of a copper cavity that can be cooled down to 4 K. The whispering gallery modes of this structure are characterized by a high energy confinement in the sapphire cylinder due the total reflection at the vacuum-dielectric interface. The different resonators we designed operate on quasi-transverse magnetic whispering gallery modes as $WGH_{m,0,0}$ where m is the number of wavelengths in the resonator along the azimuth. To operate in the $8-12$ GHz frequency range, the resonator diameter is $\Phi = 30$-50 mm and its thickness $H = 20$-30 mm. If the azimuthal index is sufficiently high ($m \geq 13$ typically) the Q-factor can achieve 1×10^9 at the liquid-He temperature.

The CSO is a Pound-Galani oscillator [18]. In short, the resonator is used in transmission mode in a regular oscillator loop, and in reflection mode as the discriminator of the classical Pound servo [19]. The sustaining stage is placed at room temperature. The choice of the sustaining amplifier is not critical as the CSO short term frequency stability will be limited by the Pound discriminator noise [20]. The results presented in the last section have been obtained with a CSO sustaining stage based on two cascaded commercial GaAs FETs low noise amplifiers (small signal gain 30 dB, -1 dB compression point: 18 dBm). Only the second one is saturated and the open loop gain is about 3 dB. In an autonomous cryocooled CSO, the sapphire resonator is placed into a cryostat and in thermal contact with the second stage of a pulse-tube cryocooler delivering typically 0.5 W of cooling power at 4 K. The resonator temperature can be stabilized above 4 K at ± 200 μK. Details on the design and on the techniques that have to be applied to get an ultimate frequency stability can be found in [9], [20]–[22].

Our goal is to provide an ultra-stable oscillator that meets the most stringent short-term frequency stability specifications, such as those for the Deep Space Network for satellites and space vehicles navigation. In this way, let us specify a short-term fractional frequency stability expressed in terms of Allan standard deviation as:

$$\sigma_y(\tau) \leq 1 \times 10^{-15} \text{ for } 1 \text{ s} \leq \tau \leq 10,000 \text{ s} \tag{1}$$

The questions we wish to address are: i) what are the required resonator properties and ii) what type of sapphire material is suitable to meet this specification.

A. Resonator design and adjustment

As explained in [9], in our most advanced CSO, a $\Phi = 54$ mm, $H = 30$ mm resonator is operated on the $WGH_{15,0,0}$ mode near 10 GHz. The input coupling coefficient β_1 has to be set near to unity to optimize the Pound discriminator. The output coupling coefficient β_2 should be low enough to avoid degrading the loaded Q-factor but high enough to stay compatible with the gain of the sustaining amplifier. Overall insertion losses through the cryostat of -30 dB or so is generally a good trade-off. With optimized coupling coefficients, i.e., $\beta_1 = 1$ and $\beta_2 \ll 1$, the loaded Q-factor is half the unloaded one: $Q_L \approx \frac{1}{2} Q_0$.

B. Short term frequency instability: The Line Splitting Factor

Noise in electronics is the main source of short-term instabilities. Without characterizing the exact nature of the oscillator individual noise sources, one would like to have a rough estimate of what the resonator bandwidth should be to achieve a given frequency stability. It is generally admitted that the attainable frequency stability $\sigma_y(\tau)$ at a given time interval (e.g., $\tau = 1$ s) cannot be lower than a given fraction of the resonator bandwidth $\Delta\nu$ [23]. We thus define an empirical figure of merit, namely the "Line Splitting Factor", or LSF, as the ratio of the frequency fluctuations $\delta\nu$ averaged over τ of the generated signal divided by the resonator bandwidth. If ν is the signal frequency, the LSF is:

$$LSF(1\text{s}) = \frac{\delta\nu}{\Delta\nu} = Q_L \times \frac{\delta\nu}{\nu} = Q_L \times \sigma_y(1s) \tag{2}$$

The LSF provides a way of quantifying the overall effect of the noise associated with the oscillator electronics. Considering the best experimental results obtained to date [3], [20], [24], we assume $LSF \approx 2.5 \times 10^{-7}$, which is a conservative value. Thus, to get a stability of 1×10^{-15}, the minimum resonator unloaded Q-factor is:

$$Q_0 = 2 \times Q_L = 2 \ \frac{LSF(1\text{s})}{\sigma_y(1\text{s})} \geq 500 \times 10^6 \tag{3}$$

C. Medium term frequency limitation

The resonator frequency is determined by its geometry and by the wave velocity inside the medium. These physical characteristics are in turn affected by the resonator temperature and the power of the injected signal. The resonator sensitivity to these environmental parameters limits the oscillator medium term frequency stability.

1) Thermal sensitivity: For an ideal resonator made of pure sapphire, although the frequency thermal sensibility would decrease significantly with temperature, it will never be low enough to achieve the target stability with a state-of-the-art temperature controller. Fortunately, it turns out that high-purity sapphire crystals always contain a small concentration of paramagnetic impurities, as Cr^{3+}, Fe^{3+}, Mo^{3+} or Ti^{3+}. At low temperatures, these residual impurities induce a temperature dependant magnetic susceptibility. In certain circumstances this temperature dependance compensates the intrinsic sensitivity of the pure sapphire. Following the notation introduced by Mann [25], for a high-order whispering gallery mode the thermal dependence of the resonator frequency ν can be written as:

$$\frac{\nu(T) - \nu_{0K}}{\nu_{0K}} = AT^4 + \frac{1}{2} \ \chi'(\nu, T) \tag{4}$$

where ν_{0K} would be the mode frequency at 0 K in a pure sapphire resonator. $A \approx -3 \times 10^{-12}$ K^{-4} combines the temperature dependence of the dielectric constant and the thermal expansion [26], [27]. χ' is the real part of the susceptibility induced by the paramagnetic dopants. If the sapphire crystal contains a density N of a paramagnetic ion characterized by its Electron Spin Resonance (ESR) frequency ν_j and a spin-to-spin relaxation time τ_2, $\chi'(\nu)$ is a dispersive Lorentzian function that nulls at ν_j [28]:

$$\chi'(\nu, T) = \chi_0(T) \frac{(2\pi\tau_2)^2(\nu - \nu_j)\nu_j}{1 + (2\pi\tau_2)^2(\nu - \nu_j)^2} \tag{5}$$

The thermal dependence is contained in the dc-susceptibility $\chi_0(T)$, which results from the distribution of the ions among their energy levels through the effect of thermal agitation. Assuming the ion in the crystal lattice behaves like a free spin S, $\chi_0(T)$ is expected to follow the Curie law [28]:

$$\chi_0(T) = N\frac{\mu_0 g^2 \mu_B^2}{3k_B T} S(S+1) \quad (6)$$

where μ_0 is the permeability of vacuum and k_B the Boltzmann constant. g is the Landé factor, μ_B is the Bohr Magneton. To show the $1/T$ dependence of the susceptibility, let us introduce $C(\nu)$ as:

$$\frac{C(\nu)}{T} = \frac{1}{2}\,\chi'(\nu, T) \quad (7)$$

Equation (4) can be rewritten as :

$$\frac{\nu(T) - \nu_{0K}}{\nu_{0K}} = AT^4 + \frac{C(\nu)}{T} \quad (8)$$

A thermal compensation can occur if the derivative of (8) nulls, which imposes $C(\nu) < 0$, and thus $\chi'(\nu, T) < 0$, which means $\nu < \nu_j$, i.e., the signal frequency is below the ESR frequency. In this case, the temperature T_0 at which the resonator thermal sensitivity nulls is:

$$T_0 = \left(\frac{C(\nu)}{4A}\right)^{1/5} \quad (9)$$

The ESR characteristics of ions commonly found in sapphire are given in table I. We also reported the value of T_0 calculated from (9) for $N = 1$ ppm and for a WGH mode at 10 GHz. The Landé g-factor of the Ti^{3+} ion in sapphire is highly anisotropic. In a direction perpendicular to the crystal C-axis, we have: $g < 0.1$ [29]. The thermal behavior of a quasi-transverse magnetique mode is only slightly impacted by the Ti^{3+} ion.

TABLE I
ESR OF THE IRON-GROUP PARAMAGNETIC IONS THAT CAN BE FOUND IN HIGH-PURITY SAPPHIRE CRYSTALS.

Ion	S	$\nu_j(GHz)$	τ_2 (ns)	$T_0(K)$ 1 ppm/10 GHz
Cr^{3+}	3/2	11.4	7	12.6
Fe^{3+}	5/2	12.0	20	13.9
Mo^{3+}	3/2	165.0	12	8.4
Ti^{3+}	1/2	1134.0	> 0.03	< 4

It is clear that an impurity density such as $N = 1$ ppm is too large to get a turnover point below 8 K, which is required to optimize the cryogenerator operation and to keep a high Q-factor. T_0 being proportional to $N^{1/5}$, the Cr^{3+} and Fe^{3+} concentrations should be very low as their ESR frequency is near 10 GHz. Figure 1 shows the behavior of two modes at each side of the Cr^{3+} ESR. These frequency-to-temperature evolutions have been calculated using the previous equations and assuming a Cr^{3+} concentration of 10 ppb only.

The $WGH_{15,0,0}$ mode at 10 GHz shows a turnover temperature $T_0 \approx 6$ K. Around T_0 the mode frequency can be approximated by a quadratic function of the temperature. When the resonator is stabilized at T_0, its frequency is no longer sensitive to temperature fluctuations. A high frequency

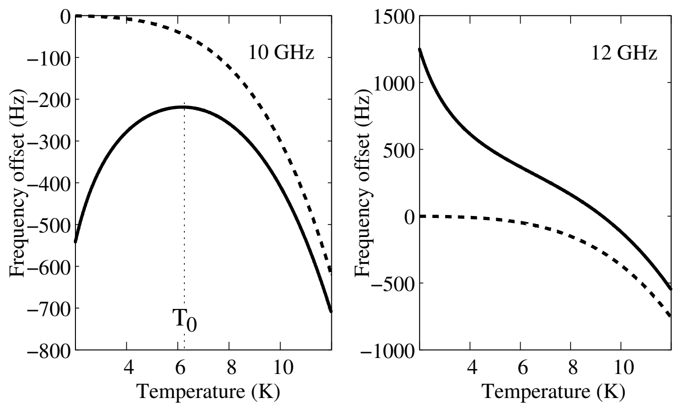

Fig. 1. Calculated frequency-to-temperature evolution of the $WGH_{15,0,0}$(10 GHz) and $WGH_{19,0,0}$ (12 GHz) of a 54×30 mm sapphire resonator. Dashed lines: pure Al_2O_3, Bold lines: Al_2O_3 with Cr^{3+} (10 ppb in weight)

stability can be achieved. Conversely the frequency of the mode $WGH_{19,0,0}$ is above the Cr^{3+} ESR frequency and no turnover can be observed, making it not suitable to build an ultra-stable oscillator.

2) Power sensitivity: The CSO sensitivity to the injected power results mainly from the thermal effect and from the radiation pressure, which becomes noticeable when the Q-factor is high. At a signal power higher than typically 100 μW, the resonator sensitivity is about 1×10^{-8}/W [30], [31]. At lower powers this sensitivity is affected by the paramagnetic impurities for modes laying nearby or in the ESR bandwidth [24]. For a mode near 10 GHz there exists a value P_0 of the injected power specific to each resonator at which the mode sensitivity nulls to the first order. This effect greatly relaxes the specification on the signal power stabilization.

D. Aging

Besides pure fluctuations, the properties of the resonator, or an environmental parameter, may *drift* with time. For example, it has been observed that the mechanical stress induced during the resonator assembly might relax with a long time constant. Should the change in such a stress cause a corresponding change in the wave velocity of the resonator medium, the oscillator frequency will also drift with time [32]. Such phenomena will induce degradations in the CSO frequency stability over the longer term. This "aging" is generally characterized in the time domain by an Allan standard deviation increasing proportionally with the integration time τ. As illustrated in figure 2, two types of mounts have been tested. Some resonators have a protruding spindle that permits to attach the resonator without perturbing the effective volume in which the electromagnetic wave is confined. Some others are simply held in place by a brass screw passing through a 5 mm hole along their axis. In this last case the stress induced by the screw should affect the effective volume, and consequently a larger drift is expected.

III. SAPPHIRE SAMPLES

Since our earlier works more than 15 years ago, we have tested different crystals and geometries. Those that have been

TABLE II
CHARACTERISTICS OF THE TESTED RESONATORS

Resonator Designation	Year of Delivery	$\Phi \times H$ (mm $\times$ mm)	Growth method	Manufacturer	Features
HEM-CS	1995	50 x 20	Heat Exchange	Crystal System	Open cavity experiments
HEM-CS	2007	54 x 30	Heat Exchange	Crystal System	Optimized 10 GHz CSO, spindle mounting
HEM-CS	2012	54 x 30	Heat Exchange	Crystal System	Optimized 10 GHz CSO, spindle mounting
CZ-PST	2011	54 x 30	Czochlrasky	Precision Sapphir Technology	Preliminar samples, screw mounting
KY-PST	2012	54 x 30	Kyropoulos	Precision Sapphir Technology	Preliminar samples, screw mounting
KY-PST	2013	54 x 30	Kyropoulos	Precision Sapphir Technology	Optimized 10 GHz CSO, screw mounting
BAG-CI	2013	30 x 17	Bagdasarov	Cristal Innov	Very preliminar sample
KY-CI	2013	30 x 17	Kyropoulos	Cristal Innov	Very preliminar sample

used for this comparison are listed in the table II. Some of them are shown in figure 2.

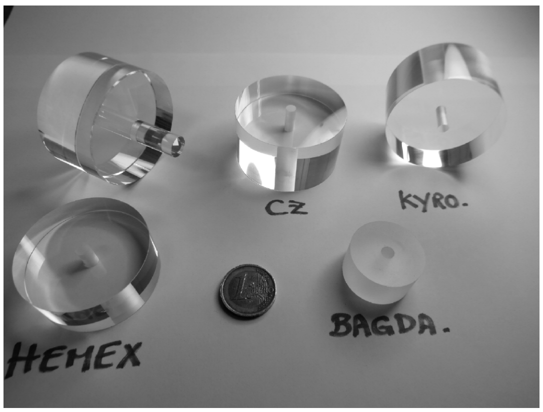

Fig. 2. Picture showing different types of sapphire resonators. Up-right, a HEMEX resonator with a spindle. The Bagdasarov crystal is not polished.

Many techniques are now used for sapphire bulk-crystal growth [33]. The Heat Exchange Method (HEM) has been commercially developed by the company Crystal Systems (Salem, MA, USA), now part of GT Advance Technology (USA). HEM is known to produce high quality sapphire crystals with low dislocation and impurity densities. In the pioneering work at the University of Western Australia (UWA), the superior properties of the HEMEX grad sapphire microwave resonator were demonstrated [34]. Since then, all the CSOs that have been built in different laboratories around the world [35]–[38] incorporate an HEMEX sapphire resonator. The Kyropoulos technique is also able to produce large, high quality sapphire monocrystals. Nevertheless its use in a CSO has never been reported. Czochralski crystals are expected to have a higher defect density although Q-factors of about 1 billion at 4 K have been already reported [39]. We tested Kyropoulos and Czochralski crystals that have been manufactured by Precision Sapphire Technologies (PST- Lithuania). The last crystals we characterized are very preliminary Kyropoulos and Bagdasarov samples provided by the French technology institute Cristal Innov. These samples are smaller than the others and are not polished. The Bagdasarov method is similar to the Horizontal Bridgman technique, which is widely used to grow plates of large surface area. This method produces crystals with a moderate quality.

The structural defect density will be affected by the growth rate and the thermal gradient experienced by the crystal during the process. It is thus expected that the Q-factor is dependent on the growth technology and on the skill of the manufacturer. The dopants content is certainly more impacted by the quality of the raw material used. The refractory metal in which the crucible is made can also contaminate the crystal.

IV. PRELIMINARY MEASUREMENTS AT ROOM TEMPERATURE

Figure 3 shows the room temperature $Q \times f$ product for some of the tested crystals.

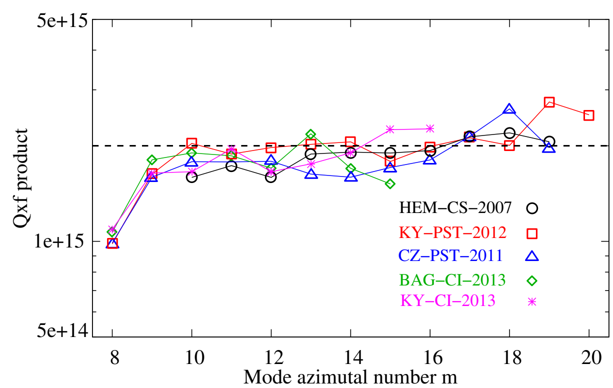

Fig. 3. Room temperature $Q \times f$ factor as a function of the mode azimuthal number.

For all the 54×30 mm resonators the mode $WGH_{15,0,0}$ at 10 GHz presents $Q \times f = 2 \times 10^{15}$ consistent with other measurements [40]. The dotted line in figure 3 is the expected $Q \times f$ product assuming the sapphire dielectric losses are predominant and proportional to the signal frequency. Modes with low azimuthal index show extra radiation losses and thus are not limited by the resonator material. For $m > 10$ all the tested crystals present a room temperature unloaded Q-factor limited by the sapphire dielectric losses: no impact of the growth method has been observed at 300 K.

V. LOW TEMPERATURE MEASUREMENTS

A. Q-factor

The resonators were mounted inside a copper cavity designed for a $54{\times}30$ mm resonator operating on the $WGH_{15,0,0}$ mode at 10 GHz. This assembly is cooled down to 4 K. Figure 4 shows the cavity mounted in the cryostat. The resonator unloaded Q-factor is determined from the measurements made with a Vector Network Analyzer (V.N.A.). The power injected into the resonator is always below 50 μW to avoid any saturation of the paramagnetic ions.

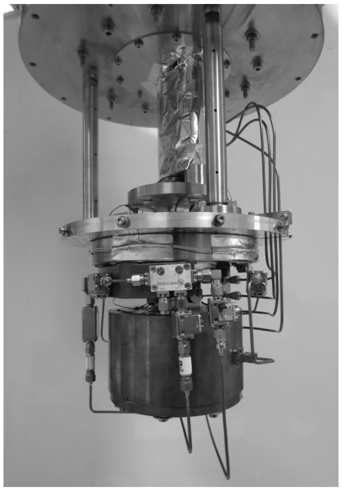

Fig. 4. Cavity mounted inside the cryostat. The microwave components placed around the resonator are needed for the Pound-Galani oscillator.

Figure 5 shows the unloaded Q-factors measured at 4 K.

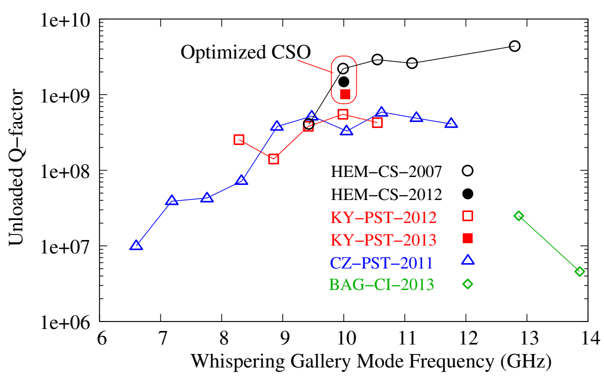

Fig. 5. Whispering gallery mode unloaded Q-factor at 4 K.

For high azimuthal numbers, the unloaded Q-factor is ultimately limited by the resonator material dielectric losses. However, at low temperatures, it can be affected by some other detrimental effects:

- Cavity wall losses due to non-optimized geometry
- Residual contamination of the resonator surface
- Extra-losses induced by nearby spurious modes
- Extra-losses due to the coupling probes
- Structural resonator defects (dislocations, inclusions, paramagnetic impurities,...)

In fact once the operating mode has been chosen for a given resonator, the cavity has to be designed to limit the spurious modes nearby. Moreover the geometrical imperfections lift the mode degeneracy of the cylindrical resonator. Each resonance splits in two twin modes [41]. For all tested crystals the whispering gallery mode splitting is about 1 to 10 kHz. The mode splitting is thus greater than the width of the resonance line. Nevertheless the coupling factors of the twin modes obtained at low temperature are hardly predictable and have to be adjusted to favor only one resonance. This fine tuning requires multiple cool down cycles and ideally a careful cleaning at each run. These time consuming procedures have not been strictly followed for all the measurements presented here. Thus this is only for few of them, i.e., those that were used to build an optimized oscillator, that the measured Q-factor can be considered as approaching the true material limitation. For the others and following our experience we expect that after careful optimization and cleaning the Q-factor value can gain up to 20% of its preliminary value.

The HEMEX resonator shows superior performances reaching $Q_0 = 2$ billions for the operating mode at 10 GHz. $Q_0 \geq 5 \times 10^8$ is achieved for some modes of the Czochralski and Kyropoulos resonators from PST. These crystals are thus suitable to meet the short-term frequency stability of 1×10^{-15}. The cavity geometry is not adapted for the resonators provided by Cristal Innov. For these crystals the spurious mode density was too high to find high-order whispering gallery modes. Here we only report the modes $WGH_{10,0,0}$ (12.87 GHz) and $WGH_{11,0,0}$ (13.86 GHz) we observed in the Bagdasarov resonator, which obviously presents detrimental radiation losses. Further experimental investigations have to be conducted on these crystals to determine the actual material losses.

B. Turnover temperature

To determine the turnover temperature, the resonator temperature set-point is increased step by step. At each step and after waiting for the system stabilization the modes resonance frequencies are measured with the V.N.A. referenced to a Hydrogen Maser to ensure long term stability. For all the samples the V.N.A. output power was set to 100 μW. The cable losses, including the room temperature cable liking the cryostat input to the V.N.A., are typically 6–8 dB. Thus the power injected in the resonator is no more than 25 μW. We assume that in this setting the turnover temperature is nearly insensitive to the power. Eventually for each mode presenting a turnover the collected data are fitted with a second order polynomial to calculate T_0. Figure 6 gives the turnover temperature as a function of the mode frequency for all the crystals we tested.

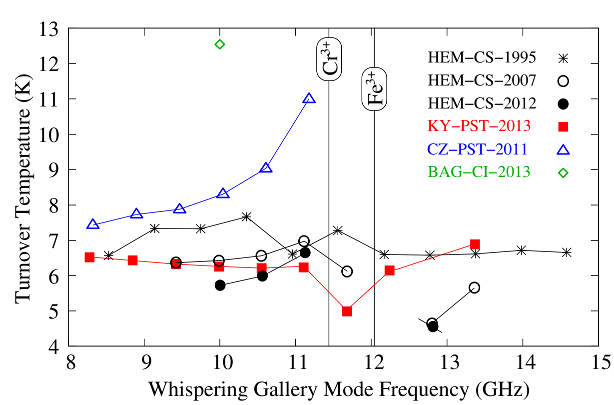

Fig. 6. Turnover temperature as a function of the mode frequency.

Figure 7 presents the experimental frequency-to-temperature $\nu(T)$ dependences for the HEM-CS-2007 resonator and for modes placed on each sides of the Cr^{3+} and Fe^{3+} ESR frequencies. The simulated behaviors are reported in bold lines.

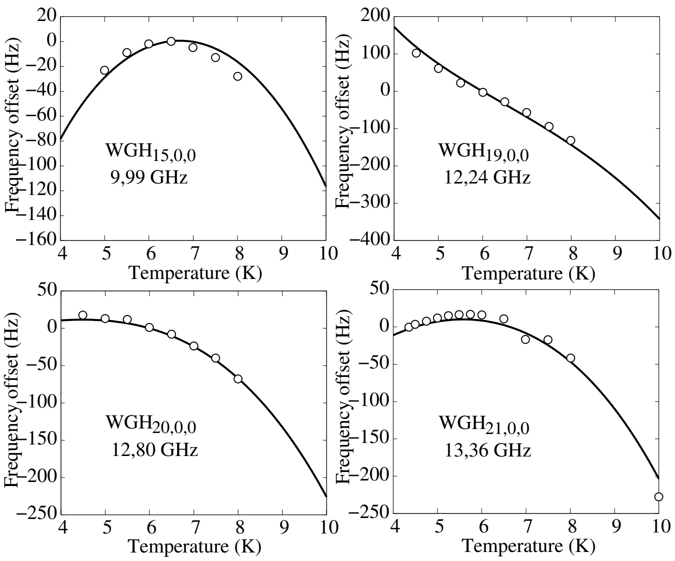

Fig. 7. HEM-CS-2007 resonator frequency-to-temperature $\nu(T)$ for the modes $WGH\ m = 15, 19, 20$ and 21. Open circles: experimental data. Bold lines: simulated thermal behaviors assuming 150 ppb of Mo^{3+}, 3 ppb of Cr^{3+} and 3 ppb of Fe^{3+}. The origin of the frequency offset has been arbitrarly chosen.

Equation (4) was used to compute $\nu(T)$, the susceptibility $\chi'(\nu)$ being the summation of the contribution of all ionic species. As explained in [24], the individual dc-susceptibilities χ_0 have been evaluated with the Van Vleck equation [42], [43]. Indeed the Curie law has been derived for a free system of spin S, which consists in $2S + 1$ levels equally spaced. In a real crystal, the ground state is split by the crystal field in multiple degenerated Kramers doublets separated by a Zero Field Splitting. Strictly speaking, the dc-susceptibility should therfore be calculated by using the Van Vleck equation. Nevertheless the Curie law deserves to be introduced as it explicitely shows the $1/T$ dependence of the susceptibility. Moreover the difference in the χ_0 values calculated with the two models is less than 20% in the temperature range of interest.

In old samples provided in the 90s (HEM-CS-1995), all whispering gallery modes in a large frequency range present a turnover temperature almost independent of the mode order (see figure 6). Luiten [26] demonstrated that it is due to the predominance of the Mo^{3+} ion, whose ESR frequency is 165 GHz. The spread in turnover temperatures observed for modes with $\nu < 12$ GHz could result from Cr^{3+} or/and Fe^{3+} residuals. The concentration of these residuals should be very low as the turnover temperature imposed by the Mo^{3+} ions is not greatly affected.
In most recent HEMEX crystals, the relative concentrations of Cr^{3+} and Fe^{3+} are higher. Starting from the lowest frequencies, the turnover temperature increases as the mode approaches the Cr^{3+} ESR. HEM-CS-2007 resonators keep a turnover temperature between $\nu_{Cr} = 11.44$ GHz and $\nu_{Fe} = 12.04$ GHz but not for the $WGH_{19,0,0}$ at 12.24 GHz just above the Fe^{3+} ESR. The $WGH_{20,0,0}$ mode at 12.80 GHz recovers a turning point but at a lower temperature, i.e., 4.6 K. For the HEM-CS-2007 resonators the impact of the Cr^{3+} and Fe^{3+} ESR are clearly visible: T_0 follows a dispersive-like curve resulting from the summation of the two dispersive Lorentzians centered on ν_{Cr} and ν_{Fe}. In HEM-CS-2012, the Cr^{3+} density seems to be higher as no turnover is observed between ν_{Cr} and ν_{Fe}.
For the Kyropoulos resonator (KY-PST-2013) all the modes between 8 and 13 GHz present a turnover temperature near 6 K apart from the $WGH_{18,0,0}$ (11.72 GHz) mode, for which $T_0 = 5.4$ K. The behavior of this resonator is very close to those of the HEM resonators, certainly with a lower Fe^{3+} density.
The Czochralski crystal shows turnover temperatures only for the WG modes below the Cr^{3+} ESR, indicating that this ion is the predominant paramagnetic impurity.

VI. CSO FREQUENCY STABILITY

Until now, due to its superior properties the HEMEX crystal was always chosen in the realization of an ultra stable microwave cryogenic oscillator. The previous results show that the Kyropoulos crystal can also constitute a good material for a cryogenic resonator. We thus incorporated the KY-PST-2013 resonator in one of our CSO. Its fractional frequency stability has been determined by beating its output with another CSO equipped with a HEMEX grad resonator. Figure 8 shows two CSOs running in the laboratory.

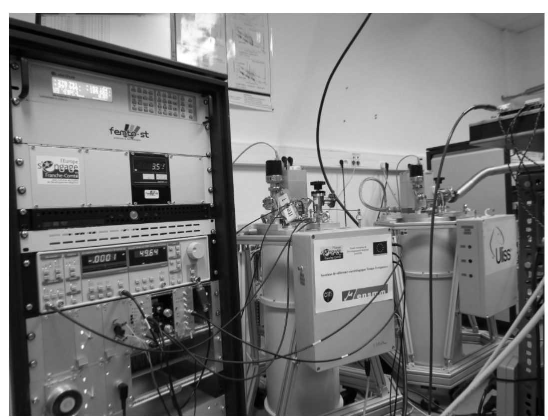

Fig. 8. Two CSOs in test at the FEMTO-ST Institute.

The resulting Allan standard deviation is given the figure 9.

The apparent flicker floor obtained with the Kyropoulos resonator is $\sim 5 \times 10^{-16}$ whereas it was 3×10^{-16} with an optimized HEMEX CSO [24]. Such a difference can be attributed to the lower Q-factor of the Kyropoulos resonator. As expected the long term drift of Kyropoulos CSO is higher than those observed with the HEMEX CSO. As already

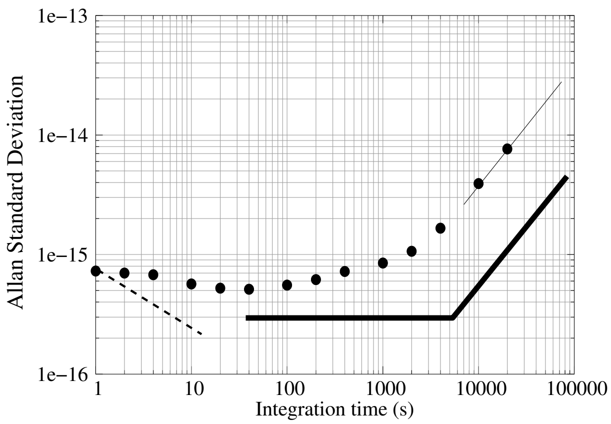

Fig. 9. Allan standard deviation calculated from the beatnote between two 10 GHz CSOs. Black bullets: one of the CSOs is equiped with a Kyropoulos crystal from PST, the other one incorporates an HEMEX sapphire crystal. Bold line: the best stability we obtained with a HEMEX resonator [24]. Dashed line: instrument noise floor.

mentioned, the Kyropoulos crystal is simply maintained by a brass screw passing through the hole along its axis. The resulting stress induced into the effective resonator volume relaxes with time, leading to a drift 10 times larger than the drift observed when the resonator is equipped with a spindle.

VII. Conclusion

We tested sapphire crystals manufactured with different growth methods in order to determine which type is suitable to build an ultra-stable cryogenic oscillator. At room temperature the Q-factor of all the tested crystals is limited by the sapphire loss tangent, i.e., $\tan\delta = 5 \times 10^{-6}$ at 10 GHz. At low temperatures the HEMEX and the Kyropoulos resonators both present a Q-factor higher than 5×10^{8} and a turnover temperature compatible with an operation in a cryocooler. These resonators have been used as the frequency determining element of a cryogenic oscillator and a fractional frequency stability better than 1×10^{-15} at short term has been obtained in both cases. Furthermore the analysis of the modes thermal behavior between 4 K and 12 K allows us to approximately determine the impurities contents of each crystal.

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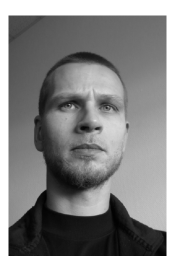

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