High Power Solid-State Sapphire Whispering Gallery Mode Maser

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***Abstract*— We present new results on a cryogenic solid-state maser frequency standard, which relies on the excitation of Whispering Gallery (WG) modes within a doped monocrystalline sapphire resonator (α-Al_2O_3). Included substitutively within the highest purity HEMEX-grade sapphire crystal lattice are Fe^{2+} impurities at a concentration of parts per million, an unavoidable result of the manufacturing process. Mass conversion of Fe^{2+} to Fe^{3+} ions was achieved by thermally annealing the sapphire in air. Above-threshold maser oscillation was then excited in the resonator at zero applied DC magnetic field by pumping high-Q WG modes coincident in frequency with the Electron Spin Resonance (ESR) energy levels of the Fe^{3+} spin population.**

A two stage annealing process was undertaken, resulting in an improvement of six orders of magnitude in output power over the previous best implementation of this scheme, and giving an output seven orders of magnitude more powerful than a typical commercial hydrogen maser. We estimate the limit on the frequency stability due to the Schawlow-Townes fundamental thermal noise limit on the order of $1 \times 10^{-17}/\sqrt{\tau}$.

I. Introduction

A recent collaboration between French and Australian laboratories has seen the development of a novel extension of the classic Cryogenic Sapphire Oscillator (CSO), the so-called "Whispering Gallery Maser Oscillator" (WHIGMO). The scheme, an active resonator-oscillator, is implemented in a very similar experimental package to the CSO but relies on the presence of paramagnetic Fe^{3+} ions in the sapphire crystal lattice. These ions exhibit an Electron Spin Resonance (ESR) at microwave frequencies, forming a three level system at zero applied DC magnetic field from which maser oscillation may be obtained. Both the CSO and the WHIGMO consist of a cylindrical HEMEX grade sapphire resonator placed in a cavity under vacuum, cooled to $\sim$4.2K on a cryogenic insert in a liquid helium dewar, and injected with microwave radiation to excite high-Q Whispering Gallery (WG) modes. The WHIGMO differs from the CSO in that it not coupled to an external loop oscillator and controlled using the well known Pound frequency stabilization technique. The WHIGMO also operates without requiring a magnetic field to be applied, unlike other solid-state masers such as the Superconducting Cavity Maser Oscillator based on ruby described by Dick et al. in [1].

An in-depth discussion of the principle of operation of the WHIGMO has been given in several previous publications [2]–[6]. Briefly, we excite above-threshold maser action in the sapphire resonator due to a coincidence in frequency between the Fe^{3+} ESR, and a very high Q whispering gallery mode ($Q > 10^9$) which acts as the sustaining amplifier in the stimulated emission scheme. All that is required to excite the maser at 12.04 GHz in the WHIGMO is to inject a signal at the ESR 'pump' frequency of 31.3 GHz. The system can be thought of essentially as a free-running loop oscillator with the "loop" being the path taken by the microwave energy as it totally internally reflects around the sapphire boundary [2]. The gain medium is the Fe^{3+} spin population which is continuously distributed along the length of the loop, and whose effect is strongly enhanced by the WG mode ($WGH_{17,0,0}$) lying within the ESR bandwidth. Previously the maser output power of a particular WHIGMO resonator has been on the order of -94 dBm. This paper reports the results after a recent annealing process on a the sapphire resonator which increased the maser output power to -37 dBm measured at the output of the cryogenic insert. Accounting for transmission line losses, this represents an output power of -30.4 dBm at the output of the resonator which is the highest power achieved to date for a WHIGMO implementation.

II. Paramagnetic Impurities in Sapphire

It is known that residual paramagnetic impurities are present in HEMEX grade sapphire at a level of parts-per-million to parts-per-billion. The influence of these impurities

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on the electromagnetic properties of sapphire at cryogenic temperatures is well characterized. For example, the presence of ions such as Ti^{3+}, Cr^{3+}, Mo^{3+}, V^{3+}, Mn^{3+}, and Ni^{3+} leads to a frequency-temperature turning point due to opposite sign effects of temperature-dependent Curie law paramagnetic susceptibility, and temperature dependence of permittivity [7]–[10]. Operating at the frequency-temperature turning point allows frequency fluctuations due to temperature instability to be nullified to first order, and has been crucial to achieve state-of-the-art short term fractional frequency stability in CSOs in the past [11], [12].

The WHIGMO exploits this frequency-temperature turnover phenomenon, in addition to the Electron Spin Resonance (ESR) associated with a parts-per-billion population of Fe^{3+} ions in the lattice. At zero applied DC magnetic field, the ESR forms a three level system corresponding to the spin-$|1/2\rangle$, $|3/2\rangle$, and $|5/2\rangle$ states. Maser oscillation in the resonator may be achieved by exciting the 'pump' transition $|1/2\rangle \rightarrow |5/2\rangle$, resulting in a non-radiative relaxation transition $|5/2\rangle \rightarrow |3/2\rangle$ (spin-spin decoherence), and finally a radiative 'maser' signal at 12.04 GHz corresponding to the $|3/2\rangle \rightarrow |1/2\rangle$ transition [13]. The concentration of Fe^{3+} ions in a typical HEMEX grade sapphire sample is on the order of only several parts per billion, however masing can normally be observed despite this small concentration by virtue of the extremely high Q factor of the sustaining Whispering Gallery mode.

III. Annealing of the Sapphire

It is desirable to increase the concentration of Fe^{3+} impurities in the resonator for several reasons. Of primary concern is that rather than being 'doped', the paramagnetic impurities in the resonator are an unintentional and unpredictable result of the manufacturing process. As such, some crystals with be 'too perfect' and contain a concentration of Fe^{3+} too small to excite above-threshold maser oscillation. In crystals with a sufficiently high concentration to excite maser action, the output power can be on the order of only -100 to -90 dBm, and as such the signal-to-noise ratio is very small. Amplification to countable levels can add noise and degrade frequency stability. Finally, the fractional frequency stability of the maser output signal is fundamentally limited by thermal noise, described by the Schawlow-Townes equation:

$$\sigma_y(\tau) = \frac{1}{Q_L}\sqrt{\frac{k_B T}{2P_{\text{maser}}\tau}} \tag{1}$$

Where Q_L is the loaded Q factor of the WG mode corresponding to the maser signal, k_B is the Boltzmann constant, T is the temperature of the resonator, P_{maser} is the output power of the maser in Watts, and τ is the integration time in seconds. From this, we see that potential fractional frequency stability improves with the square root of maser power. An improvement in the Fe^{3+} concentration provides more active ions for stimulated emission which can significantly boost maser output power. Large scale conversion of Fe^{2+} to Fe^{3+} ions can be achieved by thermally annealing the sapphire in an oxidizing environment (air). HEMEX grade sapphire does undergo an annealing process as part of the growth process, however the sapphire under test, dubbed *Gepetto*, had insufficient Fe^{3+} concentration upon manufacture to cause sustained masing in initial tests.

To improve the Fe^{2+} to Fe^{3+} ratio in *Gepetto*, a post-growth thermal annealing process was undertaken in two stages. The first annealing, carried out at Institut FEMTO-ST in Besançon France, was at 1600K for 20 hours discontinuously (5 days $\times$ 4 hours/day). The second annealing, undertaken at Crystal Systems in the USA, was at 1600K for 24 hours. In both cases, *Gepetto* was annealed in air. It is suspected that the second annealing process was longer than necessary, as some minor evaporation from the crystal surfaces was observed. This did not shift mode frequencies or degrade mode Q factors significantly. It is important to note that the annealing process does not create new Fe^{3+} sites in the sapphire lattice, it merely causes conversion of existing parts-per-million concentration of Fe^{2+} sites to Fe^{3+}. To assess the efficacy of the annealing processes, the Fe^{3+} concentration N was determined through measurements of anomalous bistability and magnetic susceptibility which appeared in the WHIGMO after annealing.

IV. Bistability Measurements

Prior to annealing, no maser oscillation could be excited in this sample, and only a very weak absorption effect at WG modes near the ESR center frequency was observed. After the first annealing stage, maser oscillation at an output power of -94 dBm was achieved. An anomalous bistability was measured at the $WGH_{17,0,0}$ frequency in which a different threshold power was observed depending on the direction of the power sweep. As has been reported previously [6], this bistability can only be explained by the saturation of the Fe^{3+} ESR at 12.04 GHz.

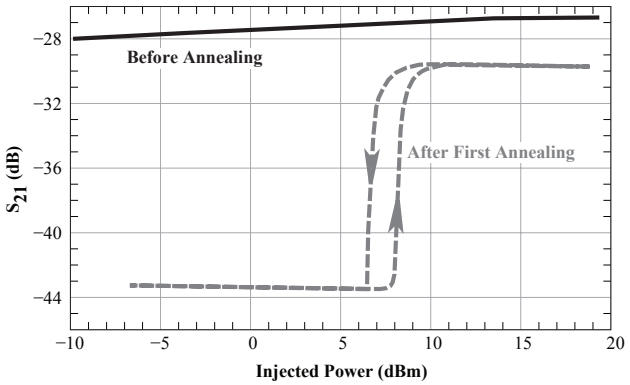

Fig. 1. Transmission coefficient for the $WGH_{17,0,0}$ mode as a function of injected power before and after the first annealing. The absorption effects are due to saturation of the Fe^{3+} ions whose ESR bandwidth encompasses the WG mode in question.

An equation of state was derived [14], given by:

$$Y^2 = 2X^2\left[\left(1+\frac{2C}{1+\Delta^2+X^2}\right)^2+\left(\xi+\frac{2\Delta^2 C}{1+\Delta^2+X^2}\right)^2\right] \tag{2}$$

where Y^2 is the input power, X^2 is the output power, the cooperativity C is given by $C = \eta\chi_0 {Q_L}^{\rm WG} {Q_L}^{\rm ESR}$ and $\xi \approx \Delta \approx 0$. The filling factor (close to unity) is represented by η, and the loaded Q factors of the WG mode $WGH_{17,0,0}$ and the ESR are given by ${Q_L}^{\rm WG}$ and ${Q_L}^{\rm ESR}$ respectively. Fitting to the measured absorption curves gives Fe^{3+} concentrations of $N = 0.017$ ppb before annealing, and $N = 1.7$ ppb after the first annealing, representing a 100 fold increase in the Fe^{3+} to Fe^{2+} ratio. The ion concentration is deduced from the cooperativity C which is a function of N through the static susceptibility χ_0.

V. Magnetic Susceptibility Measurements

A second annealing procedure was undertaken to further improve the output power of the maser. After this second annealing, the concentration of active paramagnetic ions was large enough that a strong magnetic susceptibility effect could be observed. WG modes around the ESR $|5/2\rangle \rightarrow |3/2\rangle$ transition frequency of 19.3 GHz were observed with a vector network analyzer at varying power levels. The mode frequency was first measured using the minimum power required such that the system was not purely absorptive (~ -20 dBm), and then at the maximum power available at the output of the network analyzer ($\sim$8 dBm). Shifts in the WG modes were several tens of kHz, and can be related to the real part of the complex magnetic susceptibility through the well-known equation

$$\chi' = 2\frac{\delta\nu}{p_{m\perp}\nu} \tag{3}$$

where $p_{m\perp}$ is the perpendicular magnetic filling factor, and $\delta\nu$ is the frequency shift of the WG mode at frequency ν. A Gaussian fit was made to the real part of the susceptibility data, as it is best suited to describing inhomogeneously broadened systems such as this. An appropriate form for the lineshape of the fit is given in [15] as:

$$\chi' = \left(\frac{2}{\pi}\int_0^{\frac{\tau_2}{\pi}(\omega_{23}-\omega)} e^{-y^2}\,dy\right)\chi_0\omega_{23}\tau_2 e^{-\frac{{\tau_2}^2}{\pi}(\omega_{23}-\omega)^2} \tag{4}$$

where χ_0 is the static or DC magnetic susceptibility, ω_{23} represents the angular frequency of the $|5/2\rangle \rightarrow |3/2\rangle$ transition, and τ_2 is the spin-spin relaxation time. The Gaussian fit to the data is related to the ion concentration through χ_0 which is a function of N. Performing a fit using the Levenberg-Marquardt method with two free parameters (N, and τ_2), we obtain the results $N = 55$ ppb, $\tau_2 = 5.9$ ns. The fit is shown in Figure 2. It should be clearly noted that the fit gives only approximate values for N and τ_2 for several reasons. The lack of data points in the most critical area of the curve, near the center of the ESR, affects the closeness of the fit significantly. In addition, the relaxation time should not be considered precise as the real part of the susceptibility is not particularly sensitive to this parameter. However, the fit does give (at worst) an order of magnitude estimate of ion concentration in the sapphire which is sufficient to use as a 'figure of merit' for the efficacy of the annealing process. Another estimate for ion concentration may be found by assuming the pump transition is saturated and calculating the concentration from the maximum maser output power. Using this method, we deduce the Fe^{3+} concentration in the resonator to be on the order of $\sim$150 ppb. We conclude thus that the second annealing process resulted in an improvement in ion concentration on the order of 2900-5900 times that of the original, unannealed sapphire.

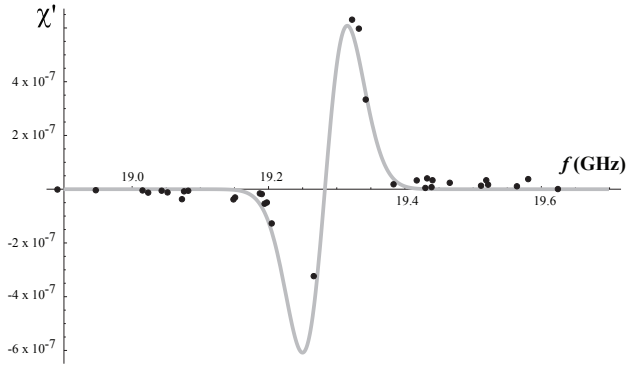

Fig. 2. Gaussian fit to the real part of the complex magnetic susceptibility. Data points were calculated from the power dependent fractional frequency shifts of WG modes lying within $\Delta f_{ESR,|5/2\rangle\rightarrow|3/2\rangle}$

VI. ESR Linewidth Broadening

Being inhomogeneously broadened, the bandwidth of the Fe^{3+} ESR in strongly dependent on the distribution and number of active Fe^{3+} spin packets in the resonator. After the first annealing process (100$\times$ improvement in ion concentration), two masers could be excited: one at 12.04 GHz corresponding to the $WGH_{17,0,0}$ mode, and another (weaker) maser at 12.029 GHz corresponding to an unidentified WG mode. Both WG modes fell within $\Delta f_{ESR,|3/2\rangle\rightarrow|1/2\rangle}$. After the second annealing process, the up to 5900$\times$ increase in Fe^{3+} population caused the ESR linewidth to be broadened significantly enough to encompass a third WG mode of appropriate characteristics at 11.96 GHz. This resulted in the observation of a third maser signal. In reality, the ESR encompasses many more than three WG modes, however only these three modes have sufficiently high Q factor and perpendicular magnetic filling factor to support maser oscillation. It is possible to excite all three masers simultaneously for several choices of pump frequency. Figures 3 and 4 show how the masers saturate, and the way in which they 'switch on' and 'switch off' at different pump frequency due to the differing Q factors of their corresponding WG modes at the maser frequency.

VII. Conclusion

In conclusion we have described how thermally annealing HEMEX grade sapphire in an oxidizing environment can cause mass conversion of Fe^{2+} impurities to Fe^{3+}. The conversion was confirmed by fitting to the bistability and susceptibility curves measured after the annealing process, and deducing the ion concentration N from the cooperativity and static susceptibility. Broadening of the ESR linewidth was observed

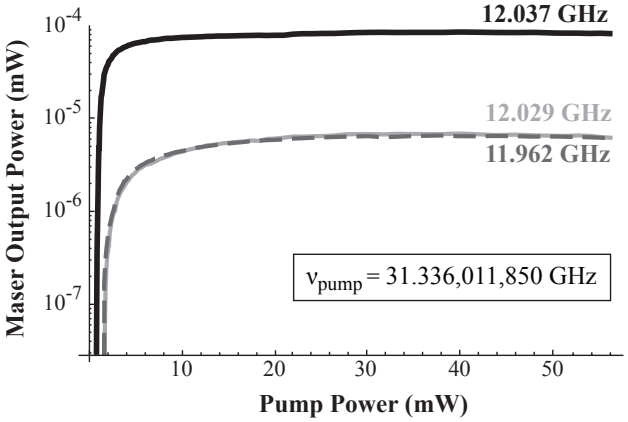

Fig. 3. Output power for the three masers as a function of the input pump power. A particular choice of pump frequency was made such that all three masers were excited simultaneously. All three masers become saturated for an input power of ~ 10 dBm.

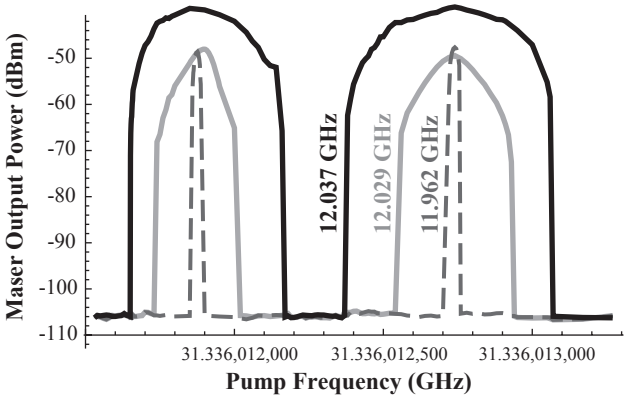

Fig. 4. Output power for the three masers as a function of the input pump frequency. The plot shows a sweep covering two 'pump' WG modes near 31.3 GHz, although the masers may be excited by several more pump modes within $\Delta f_{ESR,|1/2\rangle\rightarrow|5/2\rangle}$. The differing span for which each maser is excitable is related to the Q factor of its corresponding WG mode

after the second annealing, confirming the improvement in ion concentration and allowing a third maser to oscillate. The increase of active ions in the maser due to the annealing process resulted in an unprecedented output power of -30.4 dBm at the crystal, an improvement of six orders of magnitude in output power over the previous best implementation of this scheme. This high-power WHIGMO implementation is seven orders of magnitude more powerful than a typical commercial hydrogen maser and represents a very promising new type of frequency standard. Using Equation 1 with typical values for frequency-temperature turnover point and WG mode Q-factor, we estimate a new Schawlow-Townes thermally limited fundamental frequency stability on the order of $1 \times 10^{-17}/\sqrt{\tau}$ for the high-power WHIGMO. Tests of fractional frequency stability are underway at the time of writing.

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