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MEASUREMENT OF IODINE CELL PURITY AND ABSOLUTE FREQUENCY SHIFTS FOR LASER STABILIZATION

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Abstract – This contribution deals with the investigation of uncertainties achievable through stabilization of lasers using saturated absorption in iodine vapour. The main focus is on the iodine cell technology and evaluation.

We present results from two independent methods for the measurement of the purity of molecular iodine in absorption cells. The purity was tested by improved method based on measurement of induced fluorescence and evaluation by the Stern-Volmer formula. Original measurement setup was upgraded with a system for compensation of unwanted effects of back-reflected and scattered light and multimode regime of the pumping laser. The modified arrangement results in a significant improvement of reproducibility.

The evaluation of absolute frequency shifts caused by iodine cells was measured by direct frequency comparison of iodine-stabilized frequency doubled Nd:YAG lasers. The absolute frequency shift of a reference laser system was calibrated by frequency comparison with a stabilized optical comb locked to the radiofrequency etalon, the measuring system consists of a commercial laser and our stabilization system with 3f technique. Correct function of the stabilization setup was verified through the evaluation of the Allan standard deviations.

A series of measurements with a special refillable iodine cell was conducted on both experimental setups for three various levels of impurities. Results of both methods are in a very good correlation and show very high purity of iodine in measured cells. The results also help for definition of procedures for improving iodine cell manufacturing technology at the Institute of Scientific Instruments.

Keywords: laser stabilization, iodine, spectroscopy

1. INTRODUCTION

Laser frequency stabilization to hyperfine components of iodine has been and still is a cornerstone of the primary metrology of length even when optical frequency synthesis in laser metrology is becoming mainstream in these days due to the great advances in femtosecond laser technology [1]. Frequency locking of comb generators may be done in the radiofrequency domain to atomic clocks extending their stability into the optical spectral range or in the reverse direction to high stability lasers and to generate rf optical clock [2]. Compared to He-Ne iodine stabilized lasers or frequency doubled Nd:YAG lasers it is still a complex and sensitive technology while practical calibration of laser sources for displacement interferometry relies primarily on He-Ne-I_2 system.

Molecular iodine with its dense spectrum of components stretching over a significant part of the visible range of optical spectrum is the absorbing medium of choice for design of lasers operating on optical frequencies in coincidence with iodine transitions. The technique of saturated subdoppler spectroscopy either with low-frequency derivative or frequency-modulation detection methods allows to isolate a single narrow spectral component and derive a discrimination signal which can be used for the control of laser optical frequency. Stability of such a laser system is limited by the linewidth of the selected component and signal-to-noise ratio of its detection.

Absolute frequency shifts of iodine based laser etalons are determined by the absorption iodine cell itself when other technical sources of shifts are eliminated. The iodine cell once filled carries the shift with it and an independent method able to evaluate the cell quality helps a lot when the source of laser shift is searched. The purity of iodine determines the value of resulting shift and depends on the care which is taken to the cell preparation and filling. Stabilized laser systems with the 633 nm He-Ne laser source are limited by poor signal-to-noise ratio of the hyperfine components detection because the coincidence of laser optical frequency is only with a weak R(127)11-5 transition. Limits of resolution of absolute laser frequency shift are on the level of several kHz of optical frequency. Introduction of frequency-doubled Nd:YAG lasers operating at 532 nm brought a narrow-linewidth and low-noise laser source with a frequency in coincidence with much stronger iodine hyperfine components. The detection of iodine transitions may be optimized to get the best relative stability through compromising signal-to-noise ratio and linewidth which can be reduced by lower pressure of iodine vapour through cooling the cold finger. Relative stability of Nd:YAG iodine stabilized lasers are approaching the 10^{-14} limit which is quite close to the long term stability of primary radiofrequency etalons. Absolute frequency shift associated with the iodine cell becomes more critical here as well as the cell manufacturing process. This demands improvements also in iodine cell quality evaluation and generates questions about the limits of the preparation – filling – evaluation chain.

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To get a chance to compare the iodine cell quality and evaluate the manufacturing process we assembled an experimental setup for measurement of induced fluorescence and evaluation by the Stern-Volmer formula [7, 8] which has been used by the Bureau International des Poids et Mesures (BIPM) [9]. The aim here was to compare the results from fluorescence measurement and calculation of the Stern-Volmer coefficient with direct beat frequency measurements made with iodine-stabilized Nd:YAG lasers. Better relative stability of Nd:YAG based laser etalons seemed to be a suitable source of absolute frequency shifts evaluation with higher precision [10]. The chance to get closer to the limit of Stern-Volmer coefficient associated with zero-level of contamination in cells seemed obvious. The setup for fluorescence measurement with a noisy and mode-unstable 502 nm Argon-ion laser needed to improve significantly as well to get a reproducibility of the Stern-Volmer coefficient good enough for comparison with the absolute shift measurements.

Our first experiments with the fluorescence measurements were done with the small He-Ne-I_2 cells with Brewster windows and were compared to the absolute laser frequency comparison data measured previously during international laser comparisons. The results were summarized in [11]. It can be seen that the uncertainties of the absolute frequency shift measurements are on the level of several kHz. The reproducibility of the Stern-Volmer coefficient evaluation remains poor predominantly due to high level of stray light in a small-diameter and short glass tube. This motivated our effort to eliminate this influences not only through passive measures but also with quantification, active monitoring of the unwanted effects and their elimination in data post-processing. Further modification of the arrangement to incorporate longer and larger diameter cells for Nd:YAG iodine-stabilized lasers even reduced the problems with stray light and the reproducibility was significantly improved.

2. IODINE CELL TECHNOLOGY

Cells made at our institute are made of fused silica glass. This material allows perfect vacuum processing at a high temperature and thus additional releasing of gasses from the walls of the cell is efficiently eliminated. Joints between the cell tube and optical windows are a critical problem. We used either welding or soldering at a high temperature over 1000° C with a special solder. This technology was preferred for the cells with Brewster angle windows.

Iodine cells designed to operate in an extracavity arrangement for stabilization of frequency doubled Nd:YAG lasers are made with plane windows equipped with antireflection coatings on both sides of each window. The coatings are a traditional multilayer structure of TiO_2 and SiO_2, while the top covering layer is SiO_2, the same material as the cell tube itself. This was intended to avoid any possible contamination of the cell. The iodine cells for Nd:YAG lasers we tested on impurities are 500 mm long to achieve acceptable signal-to-noise ratio even in a single-pass configuration. The cold finger is placed in the center and cell windows are antireflection coated, wedged and slightly tilted to further suppress reflections and etalon effects.

Preparation of the cell for filling starts with evacuation. Vacuum manifold we assembled for this purpose consists of three stage pumping – diaphragm, turbomolecular and ionization pump. The core including the attached cell is enclosed in a box with heating for degassing. The limit for the temperature is here the sensitivity of the AR coatings where 200 °C should not be exceeded. Evacuation and degassing lasts several days, sometimes up to one week and goes down to the level 10^{-9} torr.

Iodine cell filling process starts with sublimation of the iodine from a commercial form into vacuum ampoules. The first sublimation is done while the tubing is vacuum pumped simultaneously. This first degree of sublimation is a crucial one, when the iodine is freed from impurities adsorbed from its surface. The next sublimation steps of the multistage process are performed only at vacuum between sealed ampoules with iodine and the impurities are removed by several types of molecular sieves. Finally the iodine is filled into the evacuated and degassed cell via a break-seal and the cell is sealed.

3. MEASUREMENT OF INDUCED FLUORESCENCE

The assembly for measurement of induced fluorescence on iodine cells and evaluation by the Stern-Volmer formula was in the first step derived from the arrangement proposed by [9]. The measurement principle is based on detection of the level of induced fluorescence intensity which is here limited by several relaxation processes such as ionization, predissociation and collisional quenching. The quenching – nonradiative transitions – can be caused by collisions with either iodine molecules or by collisions with molecules or atoms of impurities (foreign-gas quenching). The last mentioned process is the one that reflects the purity by reduction of the lifetime of a state and can be evaluated by monitoring of the level of spontaneous emission from the irradiated cell.

The Stern-Volmer diagram shows a dependence of the reciprocal intensity of the detected fluorescence related to the intensity equivalent to 10 Pa iodine pressure on the inverted iodine pressure. It is based on the normalized Stern-Volmer formula that makes the measurement of the level of quenching possible and reproducible while the detection of the total value of fluorescence is technically impossible. The parameter from the normalized Stern-Volmer formula which can express the level of collisional quenching caused by impurities in the cell is the K_0 coefficient representing the slope of the line of the diagram.

Our resulting arrangement uses fibre light delivery to separate the exciting laser which is also a source of excess heat, noise and vibrations due to air cooling from the apparatus. The expanded and collimated light is chopped mechanically for the purpose of synchronous detection which can eliminate any background light. Light detection is ensured by photomultipliers with a long rectangular active area which seems useful for imaging the shining laser beam through a lens. A slit-shaped aperture in front of the imaging lens reduces the stray light from cell walls. Electronic

processing starts with analogue lock-in detection, than sampled with analog/digital module with a controller and digital signal processor communicating with a control computer via a CAN bus. To eliminate the laser intensity noise we applied analog low-pass filtering with a cut-off frequency 1 Hz and a further digital filtering (averaging) over a period of 10 seconds.

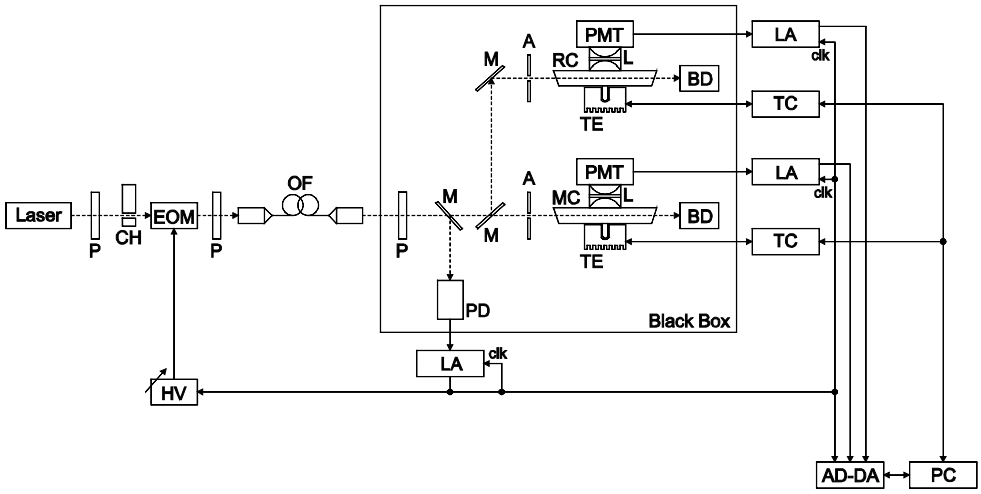

Fig. 1. Experimental arrangement of the iodine excitation and fluorescence measuring apparatus. P – polarizer; CH – chopper; EOM – electrooptic modulator; OF – single-mode optical fiber with collimators; PD – photodetector; M – mirrors; RC – reference cell; MC – measured cell; A – aperture; L – objective with two lens and color filter; PMT – photomultiplier; BD – beam dump; TE – Peltier cooler with a radiator; LA – lock-in amplifier; TC – temperature controller; AD-DA – board with analog/digital converters and digital signal processor; PC – computer; HV – high-voltage amplifier.

Laser source for iodine excitation is an Argon-ion laser operating at the 502 nm wavelength coinciding with absorption lines in iodine characterized by a high sensitivity to collisional quenching that is caused by a long lifetime of the excited level. The strongest absorption line within the spectral width of the laser is the R(26) 62-0 of the $^{127}I_2$ which contributes predominantly to the fluorescence measured. The laser operates mostly in a multimode regime, so the reproducibility of the measured data was strongly influenced by variations of the spectral component of the laser output power coinciding with the absorption profile of the R(26) 62-0 line. First, we tried to operate the laser at low level of output power where the single-longitudinal mode regime was possible to keep for a short period of time. Still the overall instability resulted in mode-hopping and the transition of desire could not be targeted precisely. On the contrary the full power where the laser covered approx. 5 GHz of total emission linewidth proved to be a better option while the resulting noise arising from varying power spectral distribution profile was smaller. The fluorescence measurement was influenced significantly by a presence of stray light arising from the iodine cell. It proved to be difficult to suppress by covers and introduction of a set of apertures. This problem depended strongly on the configuration of the iodine cell itself and was small in case of longer and large diameter cells. In case of those with antireflection (AR) coated windows designed for different wavelength, those without AR coatings or small cells the stray light reflected from the windows incident on the cell walls caused a significant portion of useful signal going in some cases up to one third of the detected fluorescence.

Monitoring of the laser power helped to eliminate overall power fluctuations but was not enough to suppress the spectral instability. We arranged a two-level compensation of the laser instability with the help of a reference iodine cell. The reference cell with an identical arrangement of a photomultiplier and optics is held at a constant cold finger temperature a little below the laboratory environment temperature (15 °C). The output signal from the reference photomultiplier and reference detection chain with identical phase-sensitive detection relates to the fluorescence detected at the line of interest and thus to the laser power within the absorption line profile.

To separate the value of the stray light level from the total signal we used cooling of the iodine cell cold finger with liquid nitrogen. This reduces the pressure of the iodine vapor down to the negligible level where no fluorescence arising from the iodine could be detected. At this moment the photomultiplier detects only the stray light. The source of the stray light to be compensated this way is the input window of the cell and the cell tube close to the input. To avoid saturation effects it is useful to place the sensing photodetector close to the input window. In this case the level of stray light arising from the glass body of the cell is not affected by linear absorption in iodine and stays constant during the measurement process. The level of background signal due to stray light varies only corresponding to the overall output power of the laser. The whole compensation system relies on monitoring the laser power by a photodetector and on the setup with the reference cell monitoring the spectral component of the laser output power coinciding with the absorption line. In our configuration we used the output from photodetector also as a source of

reference signal for phase-sensitive detection with lock-in amplifiers to avoid unwanted phase jitter caused by phase-lock loop control of the laser beam chopper. The background signal due to the stray light is present in the photomultiplier signal from the measured as well as from the reference cell. The measurement of this background with cooling to the liquid nitrogen level has to be done at the same moment both with the reference and measured cell.

The recordings of Stern-Volmer diagram showed even after all these compensations fluctuations that were correlated with the output power variations of the excitation laser. It became clear that some of the effects are non-linear and proportional compensations were not enough to get rid of them. We introduced a stabilization of the laser optical power together with fiber light delivery. The laser light coupled into the fiber was fed through polarizer and electrooptic amplitude modulator and another polarizer was placed at the output collimator. Linearly polarized light was needed for measurements of cells with Brewster-angle windows. The closing of the power stabilization feedback over the whole fiber eliminated also power and polarization fluctuations within the fiber.

The whole experiment was controlled by a PC computer with the control software programmed in the LabView environment. The temperature of the cell cold finger was set through the software due to communication with the temperature controller. All measurements were performed at least twice with ascending and descending temperature to eliminate possible hysteresis due to delayed settling of iodine pressure.

4. MEASUREMENT OF FREQUENCY SHIFTS

We assembled a beat-frequency arrangement with two frequency doubled 532 nm Nd:YAG lasers stabilized by the saturated absorption spectroscopy technique to detect frequency shifts caused by iodine cell impurities. The systems used were the iodine luminescent optical frequency standard ILP I2 /532-3L from Time Base, Düsseldorf, Germany [12] with a prestabilization to a passive Fabry-Perot cavity through a frequency-modulation spectroscopy using the Pound-Drever technique [13] and single Nd:YAG laser Prometheus from Innolight, Germany with the in-line configuration of the saturated absorption spectroscopy and third-harmonic detection chain and stabilization system of our design. The optical setup was designed to allow simple exchange of the iodine cells for frequency shift measurements. The laser frequency standard ILP I2 /532-3L was calibrated by comparison with the optical frequency comb synthesizer with repetition frequency derived from GPS synchronized Rubidium clock operated in the Laboratory of primary metrology of the Czech metrology institute in Prague, Czech republic.

Conditions of the stability recording and frequency shift measurement were kept at the levels described in the recommendations issued by CIPM [14]. To eliminate the influence of residual Doppler background that may cause some frequency shift we arranged the comparison with both lasers locked to the same hyperfine component of molecular iodine, the a_{10} of the R(56) 32-0 transition. The component is also close to the center of the Doppler broadened transition so it can be expected that the flat background slope should produce minimal shifts. To resolve the direction of the frequency shift an additional acousto-optic modulator producing a specific and stable shift of 80 MHz was added. The conditions of the laser arrangement operation of the reference laser were not precisely those recommended by CIPM, while it is a commercial system but it was calibrated as a whole. The assembly for testing of the cell under investigation was kept at -15 °C of the cell cold finger, modulation depth 1 MHz and beam intensity 17 mW/ cm^2. Signal-to-noise ratio in the detection chain was 1:50 at 100 Hz bandwith.

Significant improvement of signal-to-noise ratio of the beat signal and reduction of the influence of the phase jitter due to beat generated by the close-frequency modulation signals was performed by a synchronous modulation of both laser systems which reduced interference of the modulation signals. Precise equalization of both amplitude and phase was adjusted by monitoring of the beat signal with a radiofrequency spectrum analyzer and reducing the signal linewidth down to the level of a beat signal of free running unmodulated lasers. Synchronization of the modulation signals was derived directly from a single quartz oscillator and following set of dividers generating the modulation signals with adjustable attenuator and phase shifter.

Result of the stability recording confirms the stabilities that can be achieved with frequency doubled Nd:YAG stabilized laser standards of optical frequencies and approaches the 10^{-13} level for integration time 100 s. The recording of Allan standard deviation in Fig. 2. shows that frequency noise of both systems is dominated by random noise fluctuations and the mean value of the detected frequency shift is not influenced by drifts. Together with careful elimination of DC offsets in the key components of the detection chain and servo loop we can consider the relative frequency shift of the pair of lasers given predominantly by the difference between absolute frequency shifts of hyperfine components of the iodine spectrum of the two cells.

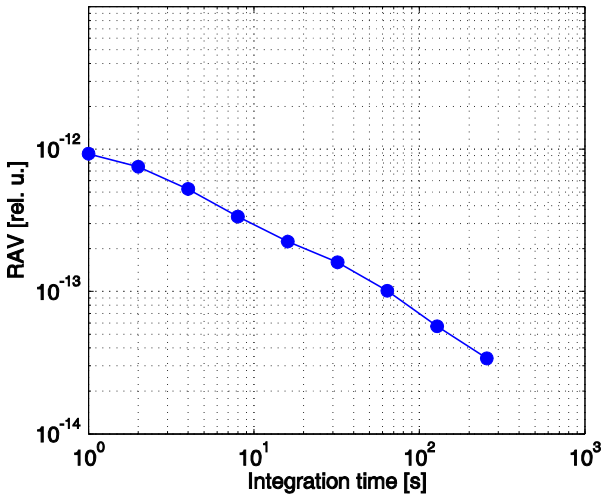

Fig. 2. Recording of the Allan standard deviation of the beat signal measurement between the two iodine stabilized frequency doubled Nd:YAG lasers.

5. RESULTS AND CONCLUSIONS

The experimental tools for Stern-Volmer coefficient evaluation and absolute frequency shift measurement were intended primarily to get a useful feedback for the development of the iodine cell manufacturing technology. The process currently in use includes triple vacuum sublimation of the solid iodine into ampoules and finally into the iodine cell. We tried to test the sense of the triple sublimation by making a testing iodine cell with several break-seal valves allowing refilling through repeated welding to the vacuum manifold. The cell was 500 mm long with wedged AR coated windows nearly perpendicular to the beam axis. The cell was designed to operate in our frequency doubled Nd:YAG stabilized laser etalon.

Measurements of the level of induced fluorescence on the cell refilled three-times under different conditions are summarized in Fig. 3. in a Stern-Volmer diagram with calculated coefficients K_0. Recordings of the dependence of normalized reciprocal intensity of induced fluorescence on the reciprocal pressure are performed several times under ascending, resp. descending pressure to express the reproducibility of the measurement. Resulting coefficients together with the expressed standard deviations are in Tab. 1.

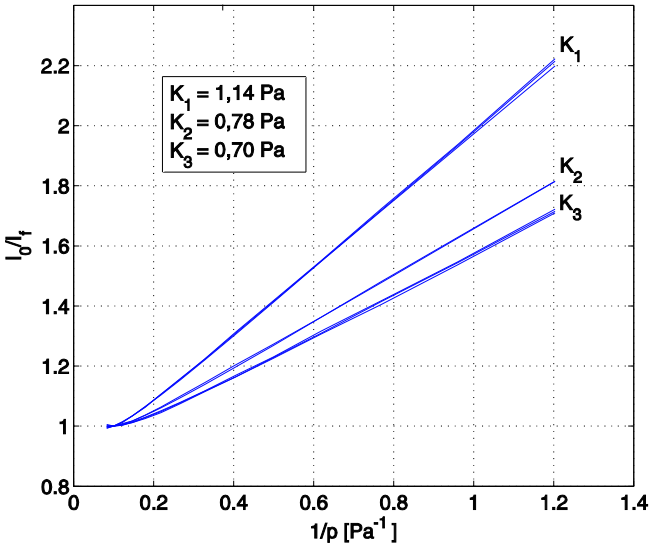

Fig. 3. Stern-Volmer diagrams of the three measurements performed on a testing iodine cell repeatedly filled through single, double and triple sublimation of iodine.

Each new filling of the testing cell was tested in a beat-frequency recording experiment of the two iodine-stabilized frequency doubled 532 nm Nd:YAG lasers. The uncertainty of the frequency difference measurements is expressed through Allan standard deviation recorded during each beat-frequency recording (Fig. 4.). This cannot cover any possible systematic frequency shifts due to changes in the many parameters of the whole stabilized laser systems such as iodine cell cold finger temperature (iodine pressure), modulation effects, electronic offsets, non-harmonic distortion of the modulation signal, etc. but all of them were adjusted according to the recommendations [14] or suppressed with great care and – most importantly – were kept constant during all the three measurements.

Tab. 1. Values of Stern-Volmer coefficients with expressed standard deviation and related absolute frequency shifts measured by a beat frequency recording with a reference laser.

No. of sublimations	1	2	3
S-V coef. [Pa]	K1=1,136 ±0,0144	K2=0,777 ±0,007	K3=0,6967 ±0,0058
Abs. freq. shift [kHz]	z e = -7,0	z e = -0,7	z e = +0,9
Allan std. dev. [r.u., for 100 s]	2,11e-12	1,41e-12	5,25e-13

The main aim of this work was to assemble and to put into operation a laboratory setup which would allow us to monitor the iodine purity of the manufactured cells with an independent method with reproducible results that would correspond with the uncertainties achievable by iodine-stabilized frequency doubled Nd:YAG lasers. The measurements of the level of induced fluorescence and evaluated through Stern-Volmer normalized formula can help us as a feedback to improve the cell manufacturing, preparation and filling technology.

From the metrological point of view not only relative stability matters but also the absolute frequency shift is a question. With the present-day iodine cell technology we need to be able to resolve the iodine impurity caused shifts well below the kHz limit. Frequency doubled Nd:YAG lasers stabilized to saturation spectroscopy in iodine are able to reach or even overcome the 10^{-13} limit of relative stability due to much better signal-to-noise ratio achievable at the 532 nm wavelength compared to the traditional He-Ne laser etalons operating at 633 nm. This gives the chance to extend the precision of previous investigations of iodine cell purity and related frequency shifts performed with intracavity cells of He-Ne lasers and the estimation of the resolution and sensitivity limit of the Stern-Volmer coefficient based evaluation. It would be useful to get some comparison but when we try to operate at the level of tens of the K_0 coefficient there seems to be no chance to approach this resolution and level of uncertainty with the He-Ne system. At least the limiting value of K_0 corresponding to no contamination which was estimated at 0,8 Pa [9] seems to correspond to our investigations.

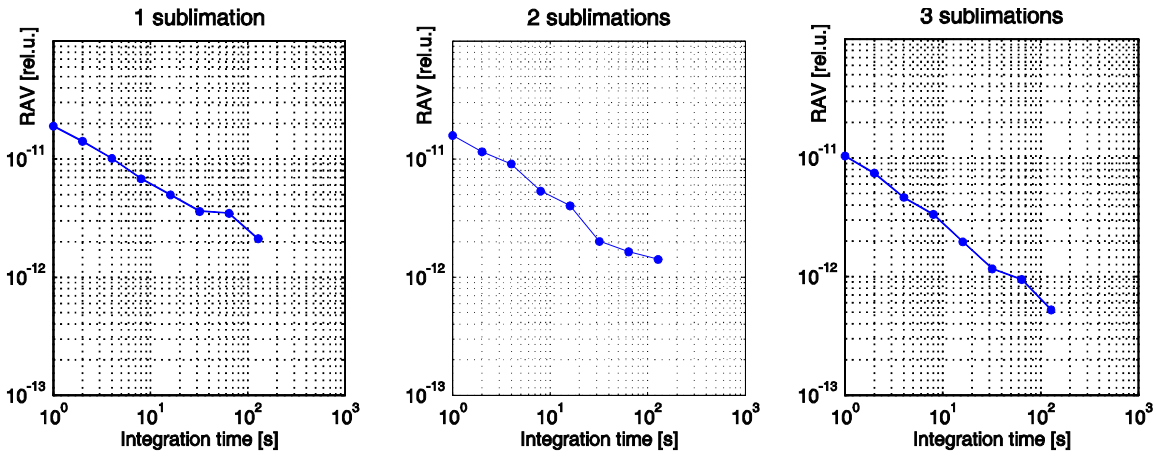

Fig. 4. Recordings of the Allan standard deviation of the beat signal measurement between the reference Nd:YAG laser and the laser equipped with the testing cell refilled under different conditions.

It is clear that the results correspond to each other where the smaller K_0 coefficient means smaller frequency shift. All these also follow the common sense where more iodine purification steps represent better results. It can be questioned whether the shift on the 0,9 kHz level achieved with the triple-purified iodine through sublimation is a systematic shift caused by other effects not related to the iodine purity. We suppose that at least the differences of the K_0 on the several 0,01 Pa and frequency shift below 1 kHz levels are most likely the "noise floor" of the method. From the technology point of view the more than single step iodine purification gives a great sense while the difference between two and three step method is still possible to distinguish but very small.

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REFERENCES

[1] Cundiff S T and Ye J 2003 Colloquium: femtosecond optical frequency combs *Rev. of Mod. Phys.* **75** 325-342

[2] Rovera G D, Ducos F, Zondy J-J, Acef O, Wallerand J-P, Knight J C and Russell P St J 2002 Absolute frequency measurement of an I_2 stabilized Nd:YAG optical frequency standard *Meas. Sci. Technol.* **13** 918-922

[3] Petrů F, Popela B and Veselá Z 1992 Iodine-stabilized He-Ne lasers at lambda=633 nm of a compact construction *Metrologia* **29** 4 301-307

[4] Petrů F, Popela B and Veselá Z 1993 Design and performance of compact iodine stabilized He-Ne lasers at lambda=633 nm with a short optical-resonator *Meas. Sci. Technol.* 4 **4** 506-512

[5] Chartier J M, Robertsson L, Sommer M, Tschirnich J, Navrátil V, Gata R, Pucek B, Blabla J, Smydke J, Ziegler M, Zelený V, Petrů F, Veselá Z, Tomanyiczka K, Banreti E, Zakharenko Y G and Vitushkin L F 1991 International comparison of iodine-stabilized helium-neon lasers at lambda=633 nm involving 7 laboratories *Metrologia* vol.28 **1** 19-25

[6] Petrů F, Lazar J, Číp O, Popescu G and Chartier J M 1997 Frequency comparison of He-Ne/iodine lasers at lambda=633 nm between the NILPRP and the ISI, and traceability through the BIPM *Metrologia* Vol. 34 **6** 515-518

[7] Stern O and Volmer M 1919 *Phys. Z.* **20** 183-188

[8] Spieweck F 1985 Influence of small impurities in absorption cells of I_2 stabilized lasers upon their frequency *IEEE Trans. on Instrum. and Meas.* IM-**34** 246-248

[9] Fredin-Picard S 1989 A study of Contamination in $^{127}I_2$ Cells Using Laser-Induced Fluorescence *Metrologia* **26** 235-244

[10] Picard S et al. 2003 Comparison of $^{127}I_2$-stabilized frequency-doubled Nd:YAG lasers at the Bureau International des Poids et Mesures *Appl. Opt.* **42** 1019-1028

[11] Hrabina J, Petrů F, Jedlička P, Číp O and Lazar J 2007 Purity of iodine cells and optical frequency shift of iodine-stabilized He-Ne lasers *Optoelectronics and advanced materials-Rapid communications* Vol. 1 **5** 202-206

[12] Holzwarth R et al. 2001 Absolute frequency measurement of iodine lines with a femtosecond optical synthesizer *Appl. Phys. B* **73** 269-271

[13] Drever R W P, Hall J L, Kowalski F V, Hough J, Ford G M, Munley A J and Ward H 1983 Laser phase and frequency stabilization using an optical-resonator *Appl. Phys. B* **31** 97-105

[14] Quinn T J 1999 Practical realization of the metre *Metrologia* **36** 211-244