Absolute Frequency Shifts of Stabilized Nd:YAG Lasers – a Question of Iodine Cell Technology

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***Abstract*— We present a technology of iodine cell preparation and an arrangement for the fluorescence purity investigation of the iodine following the cell manufacturing process, iodine purification and filling. The quality of the cells is evaluated by means of Stern-Vollmer formula from the fluorescence measurement and compared to direct frequency shift comparison using Nd:YAG lasers.**

I. INTRODUCTION

Optical frequency synthesis in the field of laser metrology is becoming mainstream in these days due to the great advances in femtosecond laser technology. Frequency locking of such comb generators may be done in the readiofrequency 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. Nd:YAG stabilized lasers seem to be a suitable option thanks to the relative simplicity of the stabilization scheme and stabilities that can be achieved are not far from those of the rf cesium clocks.

Stabilization on the basis of saturated absorption in molecular iodine may be called a traditional method and due to good signal-to-noise ratio at the 532 nm of frequency doubled Nd:YAG laser gives very good results. Good relative stability can be easily achieved with a low-noise laser source and properly designed stabilization scheme and control electronics but the absolute value of the optical frequency is given by center frequencies of hyperfine components of the iodine transitions. Only near-ideal purity of the iodine in the absorption cell can result in optical frequencies corresponding to theoretical values.

We present a technology of iodine cell preparation that has been a result of a long time development and verification by and independent method based on measurement of iduced fluorescence and evaluation by the Stern-Volmer formula [1,2] which has been used by the Bureau International des Poids et Mesures (BIPM) so there is a chance to compare our results [3]. The level of induced fluorescence intensity here is 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 main goal of this effort is to recognize the limits of the absolute precision of the optical frequency of iodine transitions and look for further improvements in the iodine cell manufacturing technology that may lead to even smaller frequency shifts.

This technique was introduced into the metrology practice with relation to He-Ne iodine stabilized lasers. With the stability that can be reached with iodine stabilized frequency-doubled Nd:YAG lasers the chance to compare the purity investigation with measured frequency shifts could go even below the kHz level [4,5,6].

II. 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 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.

Iodine cell filling process starts with distillation of the iodine from a commercial form into vacuum ampoules. The first distillation is done while the tubing is vacuum pumped simultaneously. This first degree of distillation is a crucial

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one, when the iodine is freed from impurities adsorbed from its surface. The next distillation 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 distilled into the evacuated and degassed cell via a break-seal and the cell is sealed.

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. The photo of the cell is in Fig. 1.

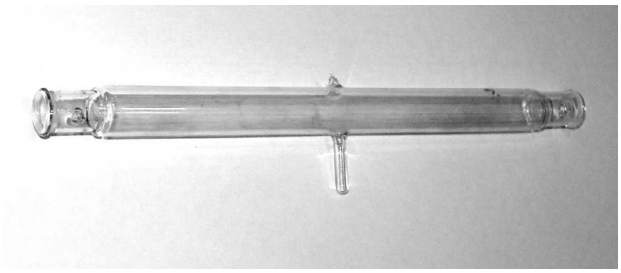

Figure 1. Flat windows iodine cell 500 mm long designed for operation in stabilized Nd:YAG laser.

III. MEASUREMENT OF INDUCED FLUORESCENCE

Our experimental arrangement is based on measurement of the level of fluorescence in the irradiated iodine cell with an Argon-ion laser for excitation of iodine. The most suitable seems 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 [3]. 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. Besides the overall power monitoring we decided to add one more level of 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. 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, Fig. 2.

The presence of a stray light arising from the iodine cell proved to be very difficult to suppress completely. This problem depended strongly on the configuration of the iodine cell itself and was small in case of longer and large diameter cells. 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 could be detected. At this moment the photomultiplier detects only the stray light. Later, during the measurement the level of background signal due to stray light varies as well but varies corresponding to the overall output power of the laser. The compensation of the effect of stray light is based on monitoring of the laser power by a photodetector. 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.

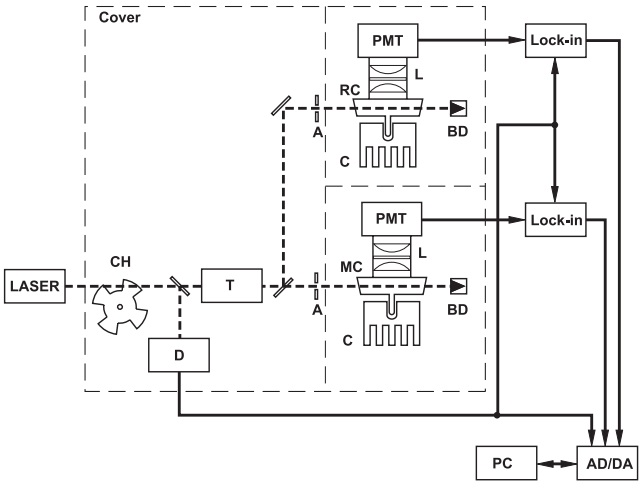

Figure 2. Experimental arrangement of the iodine excitation and fluorescence measuring apparatus. CH – chopper; T – telescope; D – photodetector; A – aperture; PMT – photomultiplier; BD – beam dump; C – Peltier cooler with a radiator.

Laser source envelope linewidth when the laser runs at maximum power is up to 5 GHz while the Doppler-broadened absorption lines of iodine namely the R(26) 62-0 does not exceed 0.6 GHz. Fluctuations of the spectral power density are a source of additional noise in the detection chain. They can be partially eliminated by a longer averaging time and filtering but there is also a long-time drift present which may influence the whole set of measurements lasting tens of minutes. We added another set of fluorescence measurement with a reference cell and identical optical and photodetector setup which monitors the amount of optical power corresponding to the absorption profile of our interest. The cell cold finger is kept at constant temperature and thus with constant pressure of gaseous iodine during the measurement process the detected intensity of fluorescence can be used to compensate the varying distribution of power spectral density.

The reference cell suffers from the effects of stray-light as well as the measured cell. To separate all these effects preparation of the measurement starts with cooling of both cold fingers with liquid nitrogen until all fluorescence fades away and the ratio of detected stray-light is recorded for the reference cell detection chain and for the measured cell as well

with respect to the power monitored by the photodetector. During the whole measurement process the normalized level of stray-light is subtracted in both detection channels. The level of detected fluorescence in the measuring channel is than compensated with respect to the reference one.

Whole assembly with the cell under investigation and with the photodetectors was enclosed in a black metal box to reduce the presence of any extra light. The laser beam was chopped by a mechanical rotating chopper running at 500 Hz. Both photomultipliers output signals are phase-sensitively demodulated to further suppress the presence of any external background light. Data acquisition and processing was performed by an 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 0.1 Hz and a further digital filtering (averaging) over a period of 1 minute. The whole experiment was controlled by a PC computer able to adjust also the temperature controller. This allowed us to let the whole measurement run without personal presence. We did about ten data acquisitions within an iodine temperature range from -10°C up to 20°C. Each time the temperature was changed at least a 5 minute delay was needed for the settling of the iodine pressure.

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 in log-log scale.

IV. MEASUREMENT OF FREQUENCY SHIFT

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 I_2 /532-3L from Time Base, Düsseldorf, Germany [7] with a prestabilization to a passive Fabry-Perot cavity through a frequency-modulation spectroscopy Pound-Drever technique [8] 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.

Our first comparison was performed with a set of two cells tested to their purity by the evaluation of the Stern-Volmer coefficient through induced fluorescence measurement. Conditions of the stability recording and frequency shift measurement were kept at the levels mentioned in the recommendations issued by CIPM[9,10].

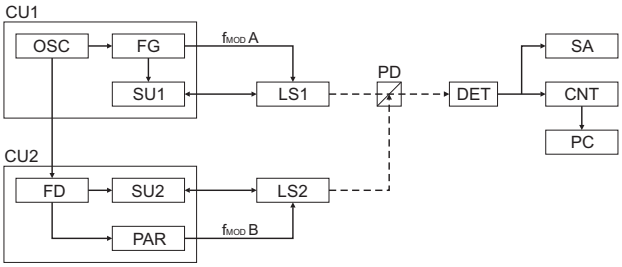

Figure 3. Simplified schematic of the arrangement for synchronization of amplitudes and phase of modulation signals of a pair of frequency doubled Nd:YAG iodine stabilized lasers and beat signal detection. CU1, CU2 - Control unit; OSC - Xtal oscillator; FG - Fmod generator; SU1, SU2 - Stabilization unit; FD - Frequency divider; PAR - phase and amplitude regulator; LS1, LS2 - Laser system; PD - polarization divider; DET - photodetector; SA - spectral analyzer; CNT - counter; PC - computer.

To achieve good resolution of the beat signal to be measured by a counter (HP 53132A) we have chosen a pair of hyperfine components a_{10} and a_{11} of the R(56) 32-0 transition that falls within a frequency range of 250 MHz of the counter input which allows adjustments of comparator level and hysteresis. According to [10,11] the frequency difference is 126.513 MHz with an uncertainty 1.5 kHz. Our estimation of the frequency shift is related to this value. Further improvement of signal-to-noise ratio of the beat signal 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 linewith 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 (Fig. 3).

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 10-14 level for integration time 100 s. The recording of Allan variances in Fig. 4 shows that frequency noise of both systems is dominated by random noise fluctuations and the mean value of the 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 shits of hyperfine components of the iodine spectrum of the two cells.

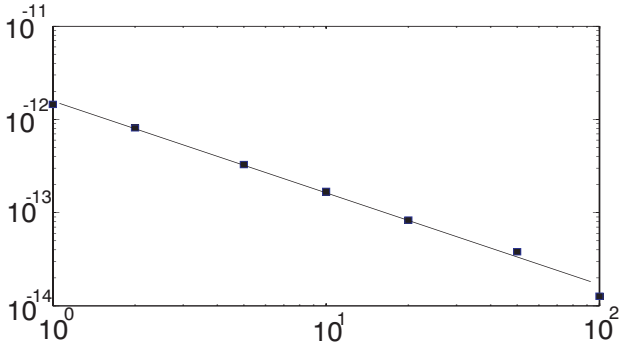

Figure 4. Recording of the Allan variances of the beat signal measurement between the two iodine stabilized frequency doubled Nd:YAG lasers.

V. CONCLUSION

The aim of this work was to further improve the technology of iodine cell manufacturing and filling leading to not only to good signal-to-noise ratio of the detected signal but also to reduction of frequency offsets of the stabilized lasers caused by frequency shits determined by the iodine purity and quality of the cell. We assembled and to put into operation a setup which allows us to monitor the iodine purity in the cells by the method based on evaluation of Stern-Vollmer coefficient derived from normalized relative changes of induced fluorescence related to iodine pressure. We decided for this method mainly due to comparability of our results with previous studies performed at BIPM. The purity measurements are one of the ways how to get feedback evaluating the quality from an independent source.

Final absolute value of laser frequency stabilized by means of subdoppler spectroscopy of iodine vapour is influenced by various other factors and effects, physical as well as technical but the quality (purity) of the cell is the most crucial one. Once the cell is sealed there is no way to change it and this technique of monitoring of the level of impurities in iodine cells seems to be the only chance how to distinguish the factors influencing the frequency precision. The other parameters with direct relation to the absolute laser frequency shift are technical and can be measured and eliminated relatively easily, such as electronic offsets in the detection chain, iodine cell cold finger temperature, modulation amplitude, non-harmonic distortion of the modulation signal, etc.

Frequency doubled Nd:YAG lasers stabilized to saturation spectroscopy in iodine are able to reach or even overcome the 10^{-14} 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.

Our first iodine cell comparison performed with two cells with Stern-Volmer coefficients $K_0 = 0.85$ Pa, resp. $K_0 = 1.7$ Pa show a frequency difference $\Delta f = 3$ kHz with a precision expressed by the Allan variances in Fig. 4. The value K_0 of the first cell is at the level extrapolated in [3] which is assumed to have a zero frequency shift. Relative frequency shift 3 kHz measured here fits into the relation observed here. It can be expected that with more iodine cells to come and more values of their frequency shifts will give a more complex picture of the limits of not only relative stability but of the absolute precision that can be achieved by iodine stabilized Nd:YAG lasers below the kHz level.

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