Frequency shift of an optical frequency standard as a function of probe modulation of the radiation frequency, pressure, and gas temperature in an absorbing cell

S.M. Ignatovich, N.L. Kvashnin, M.N. Skvortsov

Abstract. Frequency shifts of an optical $Nd:YAG/I_2$-standard are measured in a pressure range of molecular iodine 5 – 22 mTorr and deviation of probe modulation frequency 480 kHz – 4 MHz for the hyperfine structure component a1 of the molecular iodine absorption line R(56) 32 – 0. The frequency shift of the optical standard is estimated under varied temperatures of a cell and cell finger, which determines the pressure of the molecular iodine vapour. The requirements are defined to temperature stability of the cell and its finger for obtaining high long-term frequency stability of the optical frequency standard.

1. Introduction

The most widespread optical frequency standards in the instrument implementation are based on a diode-pumped solid-state $Nd:YAG$-laser with an intracavity frequency doubler stabilised at a wavelength of 532 nm by saturated absorption resonances in molecular iodine I_2. The employment of diode-pumped $Nd:YAG$-lasers gives a chance to develop reliable, small laser systems. Actually reachable total efficiency of a $Nd:YAG$ laser with respect to the consumed power is 10 % and more. A compact size of the device and relatively low heat loads related to the high efficiency allow one to design stable optical resonators with a very rigid structure, which, in turn, provides a high stability of radiation spectral characteristics. A high potential of the standard is determined by a wide tuning range of the $Nd:YAG$-laser. The radiation frequency of the $Nd:YAG/I_2$ system developed at the Institute of Laser Physics SB RAS can be stabilised by nonlinear absorption resonances in iodine in the frequency range $18775 – 18796$ cm^{-1}.

There are many methods for detecting saturated absorption resonances. Basov and Letokhov [1] suggested detection of luminescence intensity resonances in the case of saturated absorption in a standing wave. In certain cases, this method has many advantages, which provide obtaining a high long-term stability and reproducibility of the laser radiation frequency. We used the method of observing resonances in lumi-

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nescence and the stabilisation technique by third harmonic zero in a response to a probe frequency modulation of laser radiation.

In this method of resonance detection, one of the greatest frequency shifts, which determine the frequency reproducibility and long-term stability of an optical $Nd:YAG/I_2$ standard, is the shift related to the amplitude and quality of the probe frequency modulation of the laser radiation. Methods of digital signal processing are capable of solving the problems of precision laser frequency modulation and signal synchronous demodulation in a principally new way. This may have a positive result and minimise the instability of the frequency standard due to this kind of the frequency shift.

2. Experimental setup

An optical frequency standard comprises two tunable $Nd:YAG$-lasers with ring cavities and intracavity frequency doubling, a Fabry – Perot interferometer, luminescence iodine cell, and electronic system for automatic frequency tuning. A more detailed information about the employed ILP1064/532-30/80-3S laser is available in [2]. A block diagram of the setup is shown in Fig. 1. The precision probe modulation of laser radiation was provided by a system of two $Nd:YAG$-lasers. For obtaining a narrow line, the radiation frequency of the first laser was stabilised by a resonance frequency of the interferometer according to the Pound – Drever – Hall method. Emission of the second laser is tied to that of the first laser by phase self-tuning. The reference oscillator for the phase tuning system was a digital synthesiser operating at a carrier frequency of 8 MHz with a precision frequency modulation at the frequency of 487 Hz and programmable frequency deviation in the range 0 – 4 MHz. The modulation accuracy is provided by tuning the carrier frequency with a step of 0.23 Hz and using a modulation table depth of 4096 points. Thus, the first laser operated without frequency modulation, and the radiation of the second laser was precisely modulated. This radiation passes to the absorbing iodine luminescence cell. For obtaining resonances of saturated absorption, two oncoming waves were produced in the cell. For this purpose, an incident wave was backward reflected from a mirror. The radiation frequencies of the lasers are stabilised by a signal of the third harmonic of saturated absorption resonance of the iodine hyperfine structure using the self-tuning system, which adjusts the interferometer length. A luminescence signal is recorded by a photomultiplier and detected in the third harmonic of the modulation frequency.

We used this frequency stabilisation of laser radiation in [3, 4] except for the employment of precision probe modulation of the laser frequency. An advantage of the new sche-
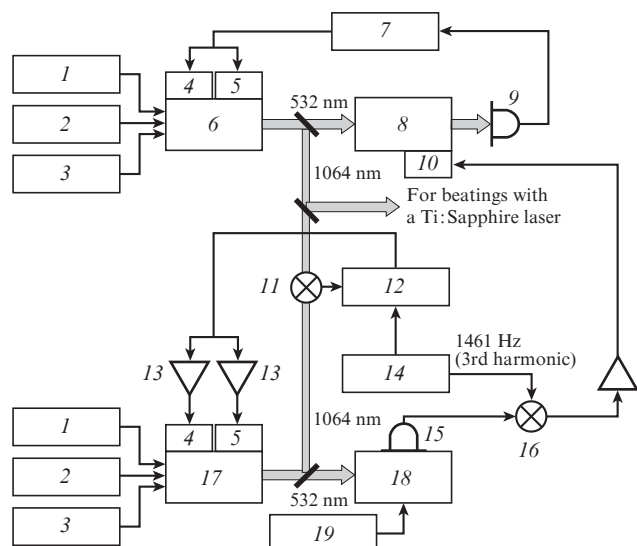
Figure 1. Block diagram of the setup:
(*1*) current source of the diode laser; (*2*) thermostabiliser of the diode laser; (*3*) thermostabiliser of the Nd : YAG laser; (*4*) piezoceramic PZT F translation stage; (*5*) piezoceramic PZT S translation stage; (*6*) Nd : YAG laser (1064 nm); (*7*) frequency self-tuning; (*8*) interferometer; (*9*) photodetector; (*10*) piezoceramic PZT translation stage; (*11*) photodetector of beatings; (*12*) phase detector (8 MHz @ 487 Hz); (*13*) amplifiers for piezoceramics; (*14*) frequency-modulated synthesiser (8 MHz @ 487 Hz); (*15*) PEM; (*16*) synchronous detector; (*17*) Nd : YAG laser (532 nm); (*18*) I$_2$ cell; (*19*) thermostabiliser for iodine cell.

matic, in addition to the precision probe modulation, is that the frequency of the first laser is not modulated. It is only shifted by the carrier frequency of the digital synthesiser from the optical resonance frequency of saturated absorption; thus, it can be used as an output frequency-stabilised optical signal.

A comparison of two optical standards designed by the block diagram shown in Fig. 1 and stabilised by the frequency of the hyperfine component a1 of the molecular iodine absorption line R(56) 32−0 shows that the Allan variance is approximately 10^{-14} for the integration time of above 100 s.

By varying either the deviation of probe modulation in one of the standards or the pressure of molecular iodine vapour controlled by a temperature change of the iodine cell's cooled finger, we observed a frequency shift in one of the optical standards relative to the other. The frequency shifts of the optical standards were measured in the ranges of molecular iodine pressures 5−22 mTorr and amplitudes of probe modulation frequency deviation 480 kHz−4 MHz for the hyperfine structure component a1 of the absorption line R(56) 32−0 in molecular iodine.

Dependences of optical standard frequency shifts measured at various deviations of the probe modulation and molecular iodine vapour pressures for the hyperfine structure component a1 of the molecular iodine absorption line are shown in Fig. 2. One can see that at elevated pressures, the frequency of the optical resonance reduces.

Measurements of the frequency shift as a function of the deviation of probe modulation were also taken for the hyperfine structure components a10 and a15 of the absorption line R(56) 32−0 of molecular iodine. The corresponding results are presented in Fig. 3.
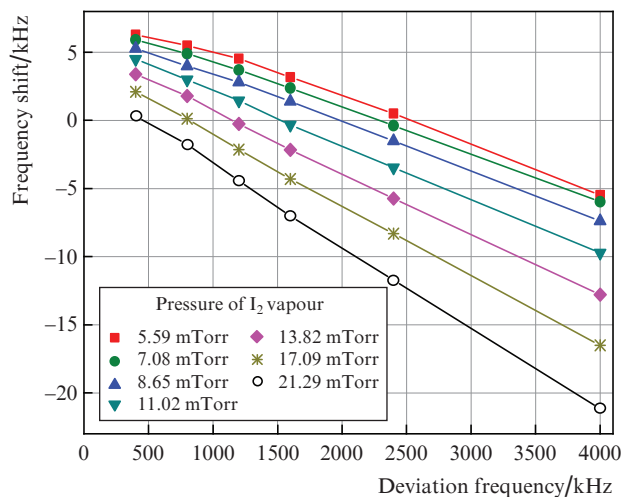

Figure 2. Frequency shifts of the optical standard vs. deviation of probe modulation frequency at various pressures of the molecular iodine vapour for the hyperfine structure component a1 of the absorption line R(56) 32−0 of molecular iodine.

The dependence of the frequency shift on the value of probe modulation is related to asymmetry of the resonance profile. At a small probe modulation, the frequency corresponding to zero third harmonic signal pertains to the point, where the third derivative of the resonance line profile is zero. In this case, with an increase in the probe modulation this frequency shifts due to the asymmetry of the resonance profile caused by the total absorption Doppler profile. From Fig. 3, one can see that the frequency shift for various hyperfine structure components related to the changed probe modulation depends on the component positions in the total Doppler absorption line profile.
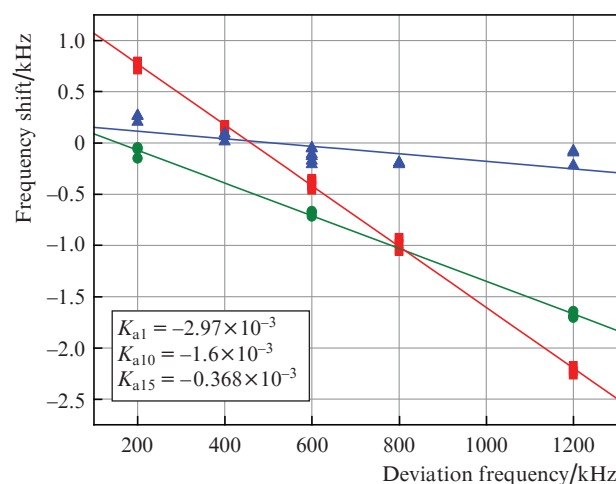

Figure 3. Measurement results of the frequency shift vs. deviation amplitude of probe modulation for hyperfine structure components (■) a1, (●) a10, and (▲) a15 of the molecular iodine absorption line R(56) 32−0.

In addition, the nonlinear saturated absorption resonance is revealed in the refractive index dispersion that possesses odd symmetry. The Gaussian cross-section profile of a laser

radiation field under frequency tuning together with the feature of nonlinear resonance mentioned above results in the self-focusing and defocusing (depending on the frequency detuning sign) of the laser radiation. This, in turn, makes the resonance asymmetric regardless of the presence of the total Doppler profile. For low-frequency hyperfine structure components, the nonlinear resonance asymmetry increases due to both the reasons, whereas for high-frequency components it is partially compensated for, because the sign of the total Doppler profile changes. The increase in the frequency shift at a greater deviation of the probe modulation at high pressures is explained by the growing width of nonlinear resonances at a higher pressure of molecular iodine. At the same asymmetry but greater width of the resonance, the shift should be greater. Coefficients K_{an} in Fig. 3 characterise the frequency shifts at the deviation of probe modulation by 1 Hz for the nth component of the hyperfine structure.

The frequency of the second harmonic of laser radiation is $\nu \approx 5.64 \times 10^{14}$ Hz. In order to obtain a relative frequency instability $\Delta\nu/\nu$ less than $\sim 10^{-15}$ the frequency error $\Delta\nu$ should be less than 0.56 Hz. The error $\Delta\nu/K_{a1}$ of the probe modulation deviation for component a1 should be less than 188 Hz. The relative error of the deviation will be 3.76×10^{-4} for the deviation amplitude of probe modulation 500 kHz.

The frequency shifts measured for the hyperfine structure component a1 of the molecular iodine absorption line R(56) 32–0 are shown in Fig. 4 as functions of the molecular iodine vapour.

The frequency shift dependence on the molecular iodine vapour pressure in Fig. 4 at zero amplitude of frequency deviation is obtained by extrapolating data from Fig. 2 towards a zero value of frequency deviation. The factor β, which characterises the slope of the frequency shift dependence on pressure, is approximately -266 Hz mTorr^{-1}. This frequency shift is related to iodine molecule collisions and can be more correctly described by the expression:

$$\Delta\nu_{\text{coll}}(t_{\text{fing}}, t_{\text{cell}}) = -n(t_{\text{fing}}, t_{\text{cell}})\sigma_{\text{coll}} V_{\text{rel}}(t_{\text{cell}} + 273.15), \quad (1)$$

where $\Delta\nu_{\text{coll}}(t_{\text{fing}}, t_{\text{cell}})$ is the frequency shift due to collisions between iodine molecules;
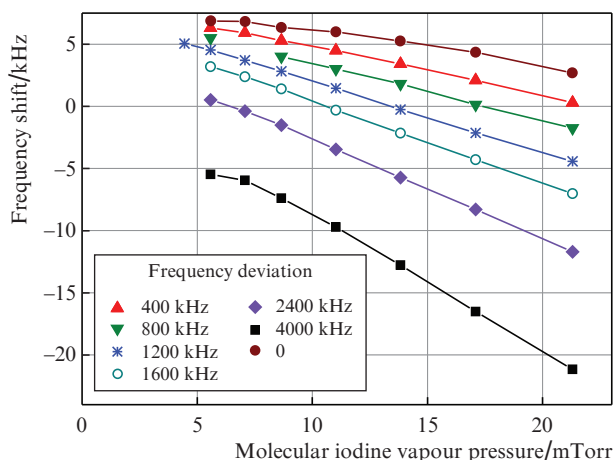

Figure 4. Frequency shifts of the optical standard vs. pressure of molecular iodine vapour at various deviation frequencies of probe modulation for the hyperfine structure component a1 of the molecular iodine absorption line R(56) 32–0.

$$n(t_{\text{fing}}, t_{\text{cell}}) = \frac{133.3224\, p(t_{\text{fing}})}{k(t_{\text{cell}} + 273.15)} \quad (2)$$

is the concentration of molecular iodine particles (m^{-3});

$$p(t_{\text{fing}}) = A(t_{\text{fing}} + 273.15)\exp\left(\frac{B}{t_{\text{fing}} + 273.15}\right) \quad (3)$$

is the pressure of molecular iodine (Torr); t_{fing} is the temperature of the iodine cell finger (°C); $k = 1.38041 \times 10^{-23}$ J K^{-1} is the Boltzmann constant; $A = 6.978268013771932 \times 10^7$ and $B = 7403.6689551223135$ are constants; t_{cell} is the temperature of cell with iodine (°C); σ_{coll} is the cross section of the molecular iodine collisions resulting in a frequency shift (m^2);

$$V_{\text{rel}}(t_{\text{cell}}) = \sqrt{\frac{16k(t_{\text{cell}} + 273.15)}{\pi M}} \quad (4)$$

is the velocity of molecule heat motion relative each other (m s^{-1}); and $M = 254 \times 1.66056 \times 10^{-27}$ kg is the mass of the iodine molecule.

In addition to the frequency shift due to iodine molecule collisions it is necessary to take into account the shift due to the quadratic Doppler effect, determined by the expression

$$\Delta\nu_D(t_{\text{cell}}) = -\frac{1}{2}\nu\left[\frac{V_0(t_{\text{cell}} + 273.15)}{c}\right]^2, \quad (5)$$

where

$$V_0(t_{\text{cell}}) = \sqrt{\frac{2k(t_{\text{cell}} + 273.15)}{M}} \quad (6)$$

is the most probable rate of molecule heat motion. At room temperature, the shift of the line centre is $\Delta\nu_D(t_{\text{cell}}) \approx 60$ Hz.

The total frequency shift due to iodine molecule collisions and quadratic Doppler effect, depending on temperatures t_{fing} and t_{cell}, will be the sum of the shifts:

$$\Delta\nu_{\text{tot}}(t_{\text{fing}}, t_{\text{cell}}) = \Delta\nu_{\text{coll}}(t_{\text{fing}}, t_{\text{cell}}) + \Delta\nu_D(t_{\text{cell}}). \quad (7)$$

The cross section of collisions in molecular iodine σ_{coll}, which result in a frequency shift, calculated from measured $\beta \approx -266$ Hz mTorr^{-1} is 3.67×10^{-20} m^2.

Frequency shifts of nonlinear resonances in molecular iodine were measured in many papers including our works [3, 5, 6]. In [3, 5], the absorption line R(56) 32–0 of molecular iodine was studied. The frequency shift in [3] was $-555(26)$ Hz mTorr^{-1}, and in [5] it was $-4.2(0.2)$ kHz Pa^{-1}, which corresponds to $-560(27)$ Hz mTorr^{-1}. In the present work we show that a substantial part of the frequency shift is determined by the value of the probe modulation (due to asymmetry of nonlinear resonance, a Doppler pad, imperfect synchronous detection, etc.). By extrapolating the frequency shift to the zero value of probe modulation it was found that the shift due to pressure is $\beta \approx -266$ Hz mTorr^{-1} (the upper curve in Fig. 4).

The frequency shift was numerically estimated under a change of the main bulb cell temperature by one degree at the cell operation temperature of 22 °C and cell finger temperature of –15 °C. As mentioned, for obtaining the relative frequency instability $\Delta\nu/\nu$ less than $\sim 10^{-15}$, the frequency error

should be less than 0.56 Hz. Hence, if the frequency shift is 2.7 Hz under the cell bulb temperature change by one degree, then the admissible error for the cell temperature will be less than $0.21\,°C$.

Also, the frequency shift was numerically estimated under a change of the cell finger temperature by one degree at the finger operation temperature of $-15\,°C$ and main cell bulb temperature of $22\,°C$.

Thus, if at the cell temperature of $22\,°C$ the frequency shift of the standard is 204 Hz under a variation of the finger temperature by one degree, then the admissible error for the cell finger temperature will be less than $0.0027\,°C$.

3. Conclusions

From results of our investigations, one may draw the following conclusion. Obtaining the frequency instability of $\sim 10^{-15}$ for an optical frequency $Nd:YAG/I_2$ standard requires the precision probe modulation with a relative instability of deviation frequency amplitude $\sim 10^{-4}$, stabilisation of the cell finger temperature determining the operation pressure of iodine vapour with the accuracy of $\sim 10^{-3}\,°C$, and stabilisation of the main cell bulb temperature with the accuracy of $\sim 10^{-1}\,°C$.

Modern digital electronics technology is capable of fabricating a voltage generator with precise frequency modulation; however, we need the precise modulation of laser radiation. One of the methods of such precise modulation is realised in the present work: two lasers are used, the radiation of one laser was locked to that of the other laser by phase self-tuning and the reference signal fed a phase detector from a digital synthesiser with precise frequency modulation and programmable amplitude of frequency deviation. The second variant of modulating the laser radiation may be the employment of acousto-optical modulators, which shift the frequency of laser radiation and are controlled by a signal from a digital synthesiser with precise frequency modulation.

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