Precise modulation of laser radiation by an acousto-optic modulator for stabilisation of the Nd: YAG laser on optical resonances in molecular iodine

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Abstract. A system of precise frequency modulation of laser radiation by an acousto-optic modulator, which makes it possible to stabilise the radiation power and simultaneously suppress the residual amplitude modulation to a level of 10^{-8} of the total laser power at the third harmonic of modulation frequency (~500 Hz), is presented. The use of this system for the Nd : YAG/I₂ optical frequency standard and application of digital signal synthesis and processing methods provided a level of frequency standard instability as small as ~ 10^{-15} for ~ 6×10^4 s.

Keywords: saturated-absorption spectroscopy, lasers, optical frequency standards, luminescence, acousto-optic modulator, molecular iodine.

1. Introduction

Currently, frequency-doubled Nd:YAG lasers, with frequency stabilised on optical nonlinear resonances in molecular iodine, are compact and most widespread optical frequency standards. There exist many methods for detecting saturated-absorption resonances. Basov and Letokhov [1] proposed to observe resonances in luminescence intensity when absorption is saturated in a standing wave. In some cases this method has a number of advantages, which provide long-term stability and good reproducibility of laser frequency [2, 3]. According to our data, the best result on the long-term frequency stability (4×10^{-15} for a measurement time of 10^4 s) was obtained in [4] using a four-pass I₂ cell, which provided interaction between Nd:YAG laser beams and iodine vapour on a length of 180 cm.

The luminescence method for detecting nonlinear resonances implies the use of the stabilisation technique based on third-harmonic zero in the response to probe frequency modulation of a laser beam. Here, one of the largest frequency shifts, determining the reproducibility and long-term stability of the frequency of the Nd:YAG/I₂ optical standard is the shift related to the magnitude and quality of the probe signal of laser frequency modulation. Therefore, the development of the methods of digital signal processing makes it possible to solve the problems related to precise laser frequency modulation and lock-in signal detection based on radically new prin-

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2. Experimental setup

The experimental setup contained two identical optical standards based on single-frequency Nd: YAG lasers with intracavity frequency doubling (model ILP 1064/532-30/50-2A, developed at the Institute of Laser Physics, Siberian Branch, Russian Academy of Sciences). The laser was described in detail in [5, 6]. Preliminary stabilisation with a highly stable optical cavity was used to obtain high short-term stability of the Nd:YAG laser frequency. The Pound-Drever-Hall method with probe phase modulation of laser radiation by an electro-optic modulator (EOM) was applied. Stabilisation on the second harmonic of laser radiation (532 nm) on saturatedabsorption resonances in an iodine absorbing cell was performed to obtain high long-term frequency stability. Stabilisation on the third-harmonic zero of probe modulation frequency was applied. An external acousto-optic modulator (AOM) was applied to prevent the direct frequency modulation of the laser.

To improve the technique of recording molecular iodine luminescence in the absorbing cell, photomultipliers were replaced with silicon photodiodes (see [2, 3]), which have a higher quantum yield. The sensitivities of a silicon photodetector and a multi-alkali photocathode (S20) are compared in Fig. 1. To date, silicon photodiodes with a rather large receiving area ($\sim 1 \text{ cm}^2$), low dark current ($\sim 200 \text{ pA}$), and high detecting ability ($\sim 1.5 \times 10^{-14} \text{ W Hz}^{-0.5}$) have been developed; examples are FDS10×10 photodetectors (Thorlabs). A necessary condition for operations with weak signals is a high load resistance (~1 M Ω); i.e., one should use a transimpedance amplifier with a low current noise (~ $2.5 \times 10^{-15} \text{ A}^{-15} \text{ Hz}^{-0.5}$), based, for example, on the ADA4817-1 integrated circuit. The signal-to-noise ratio of a detector composed of ten such photodiodes exceeds that of a previously used PTM-125 photomultiplier.

A block diagram of the Nd:YAG/I₂ optical frequency standard is shown in Fig. 2. Each standard was located on an individual platform. The laser frequency was automatically tuned to the frequency of the nonlinear optical resonance in molecular iodine using third-harmonic zero in the response signal to probe modulation.

To implement probe modulation, the second harmonic of Nd:YAG laser radiation was frequency-modulated using an AFM-402A20 AOM (working frequency \sim 40 MHz). The modulation frequency was chosen to be \sim 500 Hz (the choice of a sufficiently low modulation frequency was substantiated in [3]).The optimal frequency deviation for the stabilisation



Figure 1. Wavelength dependences of (1, 2) the current sensitivity of (1) silicon photodetector and (2) multi-alkali photocathode S20, (3) the theoretical limit for the current sensitivity at unit quantum yield, and (4) the luminescence signal of molecular iodine upon excitation of the R56(32-0) absorption line.



Figure 2. Bock diagram of Nd: YAG/I₂ optical frequency standard: (ACS1) Pound–Drever–Hall automatic control system; (PD, PD1) photodetectors; (EC) evacuated cavity; (PBS) polarisation beam splitter; (PEC) piezoelectric ceramics; (GP) Glan polaroid; (EOM) electro-optical modulator; (AOM) acousto-optic modulator; (ACS2) automatic control system stabilising laser power; (LPD) luminescence photodetector; (T1) thermal stabilisation of the optical scheme with AOM; (T2) thermal stabilisation of the iodine cell extension; (T3) thermal stabilisation of the iodine cell; (DSS) digital signal synthesiser; (DACS) digital automatic control system; (OCXO 10 MHz) quartz generator.

using third-harmonic zero in the response signal is ~1.64 Γ , where Γ is the optical resonance half-width at half maximum. To stabilise the Nd: YAG/I₂ optical frequency standard, we chose component *a*1 of the R56(32-0) transition with a contrast of the observed nonlinear optical resonance $k \approx 3 \times 10^{-2}$ and characteristic resonance half-width $\Gamma \approx 4 \times 10^5$ Hz. In the case under consideration (optical resonances in molecular iodine), the optimal deviation for probe modulation turned out to be ~0.66 MHz.

The use of an external AOM has a number of advantages: the frequency of output laser radiation is not modulated, and only part of radiation that is introduced into the iodine absorbing cell is subjected to modulation. In addition, using an AOM, one can stabilise the optical radiation power by controlling the power of the high-frequency signal applied to the modulator due to an automatic control system (ACS).

To observe optical resonances, we used radiation transmitted through the AOM in the first diffraction order on a travelling sound wave, shifted by the frequency of the highfrequency signal applied to the modulator. Due to difference in the frequency shifts in two different optical standards, one can obtain a signal at the beat difference frequency, which is convenient for measuring the Allan variance.

A laser beam was introduced into the iodine cell through a single-mode polarisation-maintaining fibre, with a Glan polaroid mounted at its output to perform additional polarisation control (see Fig. 2). This design excludes angular deviation of the laser beam in the cell with a change in the sound wave frequency. Part of radiation transmitted through the fibre and Glan polaroid arrived at photodetector PD1, and the signal from it served to suppress the laser beam amplitude noise using an active stabilisation system ACS2 (a component of the AOM control unit).

When the modulation frequency is tuned in the abovedescribed scheme, the angular deviation of the laser beam polarisation appears and residual amplitude modulation arises when introducing the beam into the fibre. Some versions of double-pass laser beam transmission through an AOM have been developed to compensate for the angular deviation of radiation with a change in the modulation frequency [7]. However, along with the mentioned factor, leading to risidual amplitude modulation, there is another factor, related to the nonideality of the travelling acoustic wave propagating through the modulator. The characteristic sizes of the laser beam cross section are 0.1-1 mm, while the sound wavelength is the product of dividing the speed of sound (4170 m s⁻¹) by the sound frequency (~40 MHz), which is ~ 0.1 mm. If the AOM working length contains an integer number of sound half-waves, a resonance of sound vibrations is observed, which is related to the interference of forward and backward sound waves; this circumstance affects the modulator efficiency and changes the fraction of the beam power diffracted into the first order.

Figure 3 shows the changes in the laser power observed on a photodetector, depending on tuning the working frequency of the modulator. One can see that, at a deviation from the centre working frequency by less than 400 kHz, the second mechanism of risidual amplitude modulation (the change in the signal caused by partially standing sound wave) becomes the key one. The double-pass transmission of a laser beam through an AOM compensates for the angular deviation; therefore, the second mechanism makes a larger contribution. However, the double-pass scheme leads to a large loss of optical power. For this reason, we chose the block diagram of the frequency standard presented in Fig. 2.

The amplitude-frequency and phase characteristics of the suppression of laser beam noise and risidual amplitude modulation, performed by the active power stabilisation system (a component of the AOM control unit), are shown in Fig. 4.

The working band of the laser power stabilisation system has a fundamental limitation: the operating speed of the AOM controlling the laser amplitude. The operating speed is determined by the sound propagation time from the boundary of the modulator working medium, where it is excited, to the laser beam and through the beam. The AOM amplitude–frequency and phase characteristics are also shown in Fig. 4. The use of the modulator made it possible to implement a working band of the laser power stabilisation system



Figure 3. Frequency dependences of the signal power after the passage through the AOM and optical fibre: (1) a change in the signal upon tuning the modulator frequency with respect to the centre working frequency; (2) the same in the absence of frequency tuning; (3) a polynomial approximation of the change in the signal due to the risidual modulation, caused by the change in the input angle into the optical fibre; (4) the signal distortion (with modulation subtracted) due to the change in the input angle into the fibre, caused by the presence of a partially standing sound wave; and (5) a sinusoidal approximation of the signal distortion caused by the presence of partially standing sound wave.



Figure 4. (a) Amplitude-frequency and (b) phase characteristics of the (1) AOM and (2) system of laser power stabilisation in the AOM control unit.

with a unit coefficient at a frequency of ~ 300 kHz; under these conditions, the suppression coefficient increased with a change in the astatism from the first to the third order with a decrease in frequency. The suppression coefficient on the third harmonic of the probe modulation frequency (~1.5 kHz) was ~ 10^5 .

As can be seen in Fig. 3, the power of the risidual amplitude modulation signal is less than 10% of the total laser power. The signal has a complex shape, which is periodically repeated with the modulation frequency. Performing expansion in a Fourier series, one can obtain risidual signals at frequencies multiple of the modulation frequency. Estimates show that the risidual signal power on the third harmonic does not exceed 10⁻² of the total radiation power at a desired frequency deviation. In addition, choosing the appropriate values of the AOM working frequency and the probe-signal deviation amplitude and changing the laser beam path at the fibre input, one can reduce the risidual signal to zero. However, the optical scheme becomes misaligned in the course of time because of the change in temperature and other factors. Thermal stabilisation of AOM units and laser beam input into the fibre is used to stabilise the beam path and AOM parameters.

The steps taken to thermally stabilise the units and suppress the risidual amplitude modulation of laser radiation using an active power stabilisation system make it possible to reduce the risidual amplitude modulation to $\Delta P/P \approx 10^{-8}$ of the total radiation power. The slope of the frequency discriminator (for stabilisation with respect to third-harmonic signal zero, with an optimal probe modulation width of $\sim 1.64\Gamma$) in units of relative change in the photodetector signal, with frequency detuned from the optical resonance frequency by 1 Hz, is $K_d = 0.34k/\Gamma = 2.55 \times 10^{-8}$ Hz⁻¹. The error of the laser frequency stabilisation system on a nonlinear resonance, which is caused by the presence of risidual amplitude modulation of the laser beam, is $\Delta v = \Delta P/(PK_d) \approx 0.4$ Hz. Since the second-harmonic laser frequency is $v \approx 5.64 \times 10^{14}$ Hz, the relative frequency instability is $\Delta v/v \approx 7 \times 10^{-16}$.

To perform measurements on the Nd: YAG laser fundamental frequency with a relative error smaller than 10^{-16} , the absolute measurement error for the difference frequency of two optical standards must be less than 7×10^{-2} Hz. The measurement error depends on the working frequency error for each of the AOMs used in optical frequency standards. Since the AOM working frequency is ~40 MHz, the relative error must be smaller than 7.5×10^{-10} .

A reference 10-MHz OCXO RTOS32227 quartz generator with a relative frequency error of 5×10^{-10} per day is used in the digital scheme synthesising a frequency-modulated signal. The difference-frequency signal (~500 kHz) is observed in beats between the first harmonics of the Nd:YAG lasers when both standards are stabilised with respect to resonance *a*1 of the R(56)32-0 line in molecular iodine; to this end, the working AOM frequencies are chosen to be 40 MHz in one of the optical standards and 41 MHz in the other standard. The difference frequency is measured by an electronic counting frequency meter AKIP-5102/1; the reference frequency is applied to this meter from the same quartz generator.

The generally accepted characteristic of frequency instability is the Allan variance. This characteristic is of great practical importance; it can easily be estimated experimentally. For a random sequence x_k (k = 1, ..., N), Allan variance $\sigma_A^2(\tau)$ on averaging interval τ is determined as the mean of the variances of neighbouring data pairs x_k :

$$\sigma_{\rm A}^2(\tau) = \frac{1}{2(N-1)} \sum_{k=1}^{N-1} (x_{k+1} - x_k)^2.$$

When the frequency instability is determined in a measuring device, two signals (measured and reference) are applied at the input of its frequency comparator. As a result, the measured value contains the total frequency instability of both signals; therefore, it is a biased estimate for the signal analysed. The results can be considered reliable only when the reference signal is obviously more stable than the signal under study. A generally accepted method for measuring the frequency instability of optical standards is based on the use of two independent but identical optical standards, the difference frequency of which (biased from zero values by means of an auxiliary heterodyne laser or AOM) is analysed. The expected instability of difference-frequency signal from two identical but independent optical standards should be larger than that for each standard by a factor of $\sqrt{2}$.

The difference frequency was measured once per second, and the measurement result was saved in the computer memory. The measurements lasted approximately three days, after which the results were processed using the Alavar 5.2 programme. Thus, we obtained Allan variance data with minimum and maximum measurement times of 1 and 6×10^4 s, respectively (Fig. 5).



Figure 5. Relative Allan variance σ_A/ν for the Nd:YAG/I₂ optical frequency standard.

3. Conclusions

A system of precise frequency modulation of laser radiation by an acousto-optic modulator with active stabilisation of laser power was developed. Necessary steps were taken to provide thermal stabilisation of the AOM and the system units and to ensure laser power stabilisation. The suppression of residual amplitude modulation of laser radiation by the active system of laser power stabilisation and the steps aimed at thermal stabilisation made it possible to reduce the level of risidual amplitude modulation to $\Delta P/P \approx 10^{-8}$ of the total laser power. The luminescence signals were received using silicon photodiodes, which have a higher quantum yield in comparison with multi-alkali photocathode of photomultiplier.

The above-described upgrade provided a high stability of the Nd: YAG/I₂ optical frequency standard: the Allan variance turned out to be $\sim 10^{-15}$ for a time of 6×10^4 s. Due to the

high stability and compactness of this frequency standard, it can be used as a base for mobile optical clock [8, 9].

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