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Laser frequency stabilisation on narrow resonances of cold magnesium atoms at the $^1S_0-^3P_1$ transition

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Abstract. **Experimental studies aimed at developing an optical frequency standard based on ultracold magnesium atoms with a relative uncertainty of $\Delta\nu/\nu < 10^{-16}$ are performed. The frequency of the clock laser system at a wavelength of 457 nm is stabilised to narrow optical resonances (Ramsey fringes) in time-separated laser fields interacting with cold magnesium atoms localised in a magneto-optical trap (MOT). The frequency stability of the laser system is investigated using a femtosecond comb based on a Ti:sapphire laser. Long-term frequency stability (determined by the Allan function) of $\sim 5\times10^{-15}$ at averaging time $\tau = 1000$ s is obtained.**

Keywords: *laser cooling, magnesium, frequency standards.*

1. Introduction

Optical frequency standards based on cold atoms are of great interest both for various applications in navigation and metrology and for fundamental physics [1]. Experimental studies aimed at developing optical frequency standards using cold magnesium, calcium, ytterbium, strontium, mercury, and thulium atoms are in progress [2–9]. To date, the best results have been obtained for strontium and ytterbium atoms under their cooling and localising in 'optical lattices'. The instability and frequency uncertainty of modern optical frequency standards based on cold strontium and ytterbium atoms are one to two orders of magnitude lower than the corresponding parameters of the primary frequency standard based on caesium atom fountain. Magnesium atoms have a number of properties favourable for their application in optical frequency standards. These properties are as follows: small frequency shift due to thermal radiation; simplicity of electronic configuration; the presence of a strong closed transition, which allows for fast and efficient cooling to $\sim$3–5 mK and localising atoms in a magneto-optical trap (MOT); and the presence of narrow optical transitions between the ground state $^1S_0$ and triplet states $^3P_{0,1,2}$. The intercombination transition $^1S_0-^3P_1$, having a natural width of $\sim$30 Hz, is the narrowest one among such transitions in alkaline-earth atoms, which are interesting for optical frequency standards. This transition is considered as promising for obtaining high, long-term stability of optical frequency standards. The strongly forbidden $^1S_0-^3P_0$ transition is of particular interest for developing a frequency standard with a relative uncertainty less than 10^{-16}, based on magnesium atoms localised in an optical lattice [10]. Despite some complexities in the use of magnesium atoms for an optical frequency standard, related to the necessity of their deep cooling to temperatures of $\sim$10 μK [11–13] and a relatively large magnitude of the quadratic Zeeman effect [2], encouraging results have been obtained recently, which demonstrate good prospects of magnesium atoms as a base of an optical standard with a relative uncertainty of $10^{-17}-10^{-18}$ [14]. In this paper, we report the results of the experimental studies aimed at developing a magnesium frequency standard at the Institute of Laser Physics of the Siberian Branch of the Russian Academy of Sciences.

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2. Experimental setup

Previously we developed an experimental setup, including laser systems for cooling and localising magnesium atoms in a MOT and carrying out ultrahigh-resolution spectroscopy of the narrow intercombination transition $^1S_0-^3P_1$ at a wavelength of 457 nm [15]. Cooling and localisation of atoms in a MOT was performed using the strong resonance transition $^1S_0-^1P_1$ at a wavelength of 285 nm (Fig. 1). Magnesium atoms were loaded into a MOT from a thermal beam and then cooled to a temperature of $\sim$3–5 mK.

We used a cloud consisting of 10^6 atoms, cooled to $\sim$3–5mK, to carry out ultrahigh-resolution spectroscopy of ^{24}Mg on the intercombination transition $^1S_0-^3P_1$ in time-separated laser fields [16]. In contrast to the strongly forbidden $^1S_0-^3P_0$ transition (whose use for a frequency standard requires sub-Doppler cooling on the $^3P_2-^3D_3$ transition [17], localising atoms in an optical lattice, and using magnetically induced spectroscopy [18]), the intercombination transition in the magnesium atom allows for direct excitation and has a small natural width ($\Gamma = 36$ Hz), due to which one can design a relatively simple optical frequency standard with a high long-term stability. When carrying out clock transition spectroscopy, the MOT is 'switched off', and one detects the inter-

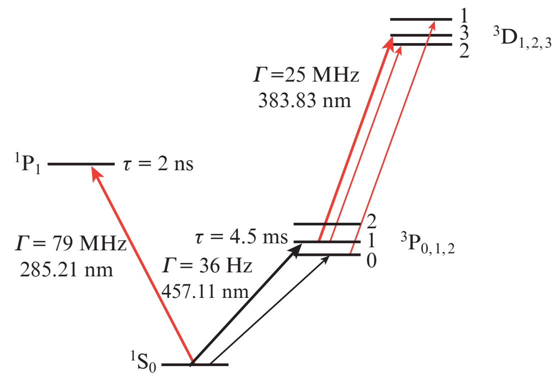

Figure 1. Energy level diagram of the Mg atom (the splitting of the ^3P and ^3D levels is shown on a highly enlarged scale).

action of radiation with atoms that freely move away with velocities of ~1 m s^{-1}. In this case, the Doppler and recoil effects limit the development of a high accuracy standard [19]; however, due to the large number of atoms, one can significantly increase the signal-to-noise ratio. This circumstance, in turn, facilitates improvement of long-term frequency stability.

We cooled and localised magnesium atoms in a MOT using a laser system (λ = 285 nm) based on a dye laser and frequency doubling in a BBO nonlinear crystal, which was placed in an external cavity to increase the UV radiation power [20]. The 285-nm beam, whose power reached 100 mW, was directed to the MOT; the parameters and design of the latter were described in detail in [15].

The main parameters of the MOT and cloud of localised atoms are as follows: power of each of six laser beams (5 mW), laser beam diameter ($2w_0 \approx 4$ mm), red frequency detuning from the absorption line centre (40–50 MHz), magnetic field gradient at the trap centre along the direction perpendicular to the magnetic coil plane (150 G cm^{-1}), the number of MOT atoms (10^6-10^7), the atomic temperature (3–5 mK), and the atomic cloud diameter (0.5–1 mm). The lifetime of the MOT atoms is limited by different factors, in particular, the collisions with the residual gas at a pressure of $(3-4)\times10^{-7}$ Pa (the lifetime is ~15 s) and the collisions with the thermal magnesium beam (1–2 s); in addition, the two-step ionisation through the $^1P_1$ level at a maximum power of cooling radiation of 45 mW (in all six beams) limits the atomic lifetime to 0.3 s. The MOT loading time was 0.1–2 s, depending on the UV power and cooling beam diameter.

The spectroscopy of a cloud of cold magnesium atoms was performed using a laser system based on a Ti:sapphire laser with subsequent frequency doubling in a cavity containing a nonlinear $KNbO_3$ crystal [21]. A schematic of the experimental setup is presented in Fig. 2.

The power of the 457-nm second harmonic was ~100 mW for the 600-mW fundamental beam entering the doubler cavity. The laser system frequency was stabilised by referencing it to the transmission bands of two Fabry–Perot interferometers using the Pound–Drever–Hall technique [22]. The first interferometer (made of Invar) with a finesse F = 600 and a free spectral range FSR = 300 MHz provided preliminary narrowing of the Ti:sapphire laser line; the second (Zerodur) interferometer ($F = 10^5$, FSR = 375 MHz) was used to correct the low-frequency perturbations of the first interferometer by adjusting automatically the length of a piezoelectric transducer onto which one of the interferometer mirrors was installed. The beam was coupled into the second interferometer through a double-pass AOM, which served to tune the clock laser frequency. Both interferometers were placed in

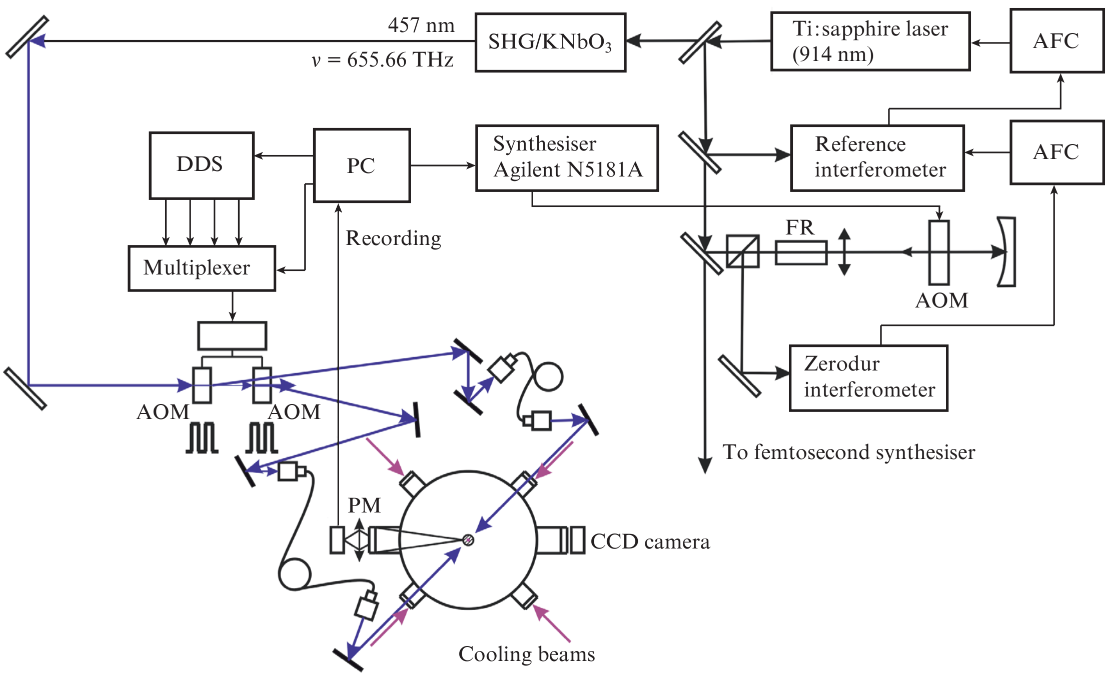

Figure 2. Schematic of the setup for spectroscopy of magnesium atoms in a MOT and frequency stabilisation for a clock system based on a Ti:sapphire laser using narrow Ramsey resonances in time-separated laser fields: (AFC) automatic frequency control; (SHG/$KNbO_3$) frequency doubler based on a nonlinear $KNbO_3$ crystal in an external cavity; (DDS) four-channel direct digital synthesiser; (AOM) acousto-optic modulator; (FR) Faraday rotator; (PM) photomultiplier.

vacuum chambers and thermally stabilised. Their vibration isolation was provided by a Minus K 100 BM-4 vibration-isolation table (the first interferometer) and a spring suspension (the second interferometer). The bandwidth of the frequency-stabilisation feedback loop of the Ti:sapphire laser was 200 kHz; this value made it possible to reduce the 457-nm linewidth to less than 100 Hz. The laser frequency drift, stabilised with respect to the Zerodur interferometer, ranged approximately from 1 to 10 Hz s^{-1} (depending on the ambient temperature stability in the laboratory room); it was caused by the aging of the material of the Zerodur interferometer base and the imperfection of its thermal stabilisation system.

To record narrow resonances in time-separated optical fields, the laser system frequency was tuned using a frequency synthesiser (Agilent N5181A), whose signal controlled the AOM in the frequency stabilisation system with respect to the second interferometer, and a fluorescence signal from a cloud of cold magnesium atoms was recorded (using a PM) on the resonance transition at a wavelength of 285 nm. Light field pulses were formed by two AOMs, and the AOM frequency and pulse parameters (duration, delay between pulses, and duration of cooling/testing cycle) were controlled by a multichannel timer, a multiplexer, and a computer-controlled four-channel direct digital synthesiser (DDS). The pulse duration τ was 4 μs, while the time delay T for a pair of unidirectional pulses could be varied, depending on the desired spectral resolution $\Delta\nu$ (half-width at half maximum of Ramsey fringe): $\Delta\nu = 1/(8T_{\rm eff})$, where $T_{\rm eff} = (4/\pi)\tau + T$. Cooling atoms in MOT and testing the clock transition were separated in time; the time diagram was described in detail in our previous study [15]. The repetition frequency of the cooling/testing cycle was 500 Hz. The best (to date) spectral resolution, $\Delta\nu$ = 390 Hz, was obtained at T = 310 μs. The Ramsey resonances recorded at T = 310 μs are presented in Fig. 3.

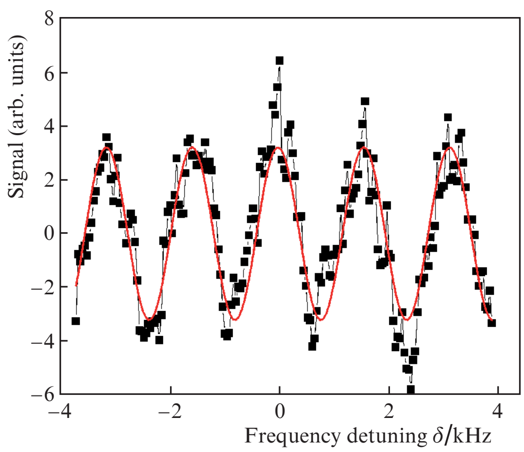

Figure 3. Detected signal of Ramsey resonances in time-separated light fields interacting with a cloud of cold magnesium atoms in a MOT. The time delay between pulses in a pair of unidirectional waves is 310 μs; the total time of coherent interaction of radiation with atoms is 620 μs (the solid line is the approximation by a sinusoidal function $3\sin(2\pi\delta/1.57)$ with a period of 1.57 kHz).

The best results on frequency stabilisation based on narrow Ramsey resonances of cold Mg atoms were obtained at delay times T = 58 and 102 μs, at which the ratio of signal/noise to $\Delta\nu$ is maximum.

3. Frequency stabilisation and measurement of frequency stability

The clock laser system frequency was stabilised with respect to the central Ramsey fringe; the optical frequency was tuned by varying the frequency of the rf signal that was generated by an Agilent N5181A frequency synthesiser and applied to a double-pass AOM. The error signal for the stabilisation system was formed using a digital analogue of stabilisation with respect to the third harmonic of the modulation frequency [4, 23]. To generate an error signal, four rf frequencies were synthesised using a four-channel DDS: $\Delta_1 = f_0 + \Delta/4$, $\Delta_{-1} = f_0 - \Delta/4$, $\Delta_3 = f_0 + 3\Delta/4$, and $\Delta_{-3} = f_0 - 3\Delta/4$, where f_0 is the centre frequency of two AOMs in the system of light field pulse formation, equal to 80 MHz, and $\Delta = 1/(2T_{\rm eff})$ is the Ramsey fringe period. The luminescence signal S_i from the cloud of cold atoms was recorded at four frequency detunings, and the error signal $dS = (S_{-3} - 3S_{-1} + 3S_1 - S_3)$ was calculated. The calculated value was used to tune the AOM frequency in the system of referencing frequency to the Zerodur interferometer. A schematic of detecting the signal from the third derivative is presented in Fig. 4a; a typical third-derivative signal is shown in Fig. 4b.

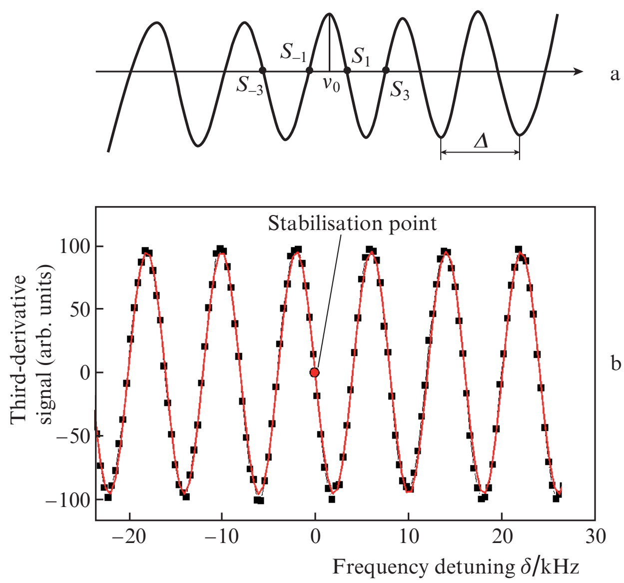

Figure 4. (a) Schematic of the third derivative signal detection and (b) a signal of the third derivative at T = 58 μs (the solid line is an approximation of the experimental curve by the sinusoidal function $f(\delta) = 95\sin(2\pi\delta/8)$ with a period of 8 kHz, where δ is the detuning from the band centre in kHz).

The AOM frequencies were switched by a multiplexer in the following sequence: Δ_3, Δ_{-3}, Δ_1, Δ_{-1} (with a delay of 0.6 s at each frequency). Five signal readings at each AOM frequency were performed over using an analogue-to-digital converter (ADC) with an integration time of 80 ms. The cycle of measuring the clock laser frequency position relative to the reference line lasted ~6 s, including the time necessary for processing and plotting graphical data in order to monitor the stabilisation system operation. The calculated dS value was an error signal for the digital automatic system of frequency tuning, which controlled the Agilent N5181A synthesiser; the synthesiser frequency was corrected with an interval of 1 Hz.

To determine the frequency stability for the radiation stabilised with respect to narrow Ramsey resonances of cold magnesium atoms, some part of clock laser radiation at a wavelength of 914 nm (with power of ~10 mW) was directed through a single-mode 10-m-long polarisation-maintaining optical fibre to a system for measuring optical frequencies using a femtosecond synthesiser based on a Ti:sapphire laser [24]. A schematic of the system for measuring frequency stability is presented in Fig. 5.

The pulse repetition rate of the femtosecond synthesiser was stabilised using the beat signal of one of its modes with a frequency equal to that of optical frequency standard based on an Yb:YAG laser, whose second harmonic was frequency-stabilised using saturated-absorption resonances of molecular iodine I_2 [25]. The frequency stability of the Yb:YAG/I_2 standard determined the accuracy of our measurements. The beat signal of the magnesium frequency standard and the nearest mode of the femtosecond synthesiser was filtered and amplified within a ~100-kHz band using a DDS-based tracking generator, and the frequency of this signal was recorded by a Pendulum CNT-90 frequency counter with a measurement time of 1 s. The accumulated data array was computer-processed.

Figure 6 shows the results of measuring the frequency stability (Allan function); the continuous measurement time was 1 h.

Our studies showed that currently the main limiting factor for frequency stability is the temperature drift of the reference Zerodur interferometer, which is mainly caused by the imperfection of its thermal stabilisation system. The deviation of the measured Allan function from the dependence $\sigma(\tau) = 1.3 \times 10^{-13}/\sqrt{\tau}$ at averaging times of 10–300 s is related to the temperature drift of the reference frequency and the insufficiently high gain of the automatic frequency tuning system. To increase the automatic frequency control (AFC) gain, it is necessary to shorten the signal measurement cycle, which can be done by increasing the signal-to-noise ratio when recording Ramsey resonances and by reducing the graphical data processing time. At high averaging times the measured stability is equal to the reference Yb:YAG laser stability. To measure correctly stability, one must either use a more stable reference generator or determine this parameter from the beat signal of two independent standards. We elaborated and designed the second MOT for magnesium atoms, and these measurements are planned to be performed in the nearest future.

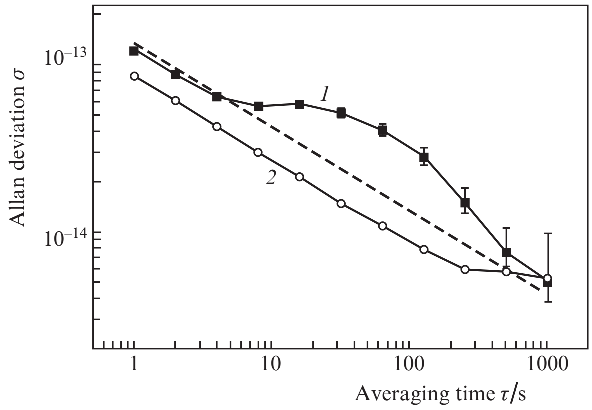

Figure 6. (*1*) Allan function characterising the frequency stability of the developed Mg standard and (*2*) Allan function of the beat signal of two Yb:YAG/I_2 frequency standards, one of which was used to stabilise the parameters of femtosecond optical frequency synthesiser; the dashed line is an approximation of the measured stability by the function $1.3 \times 10^{-13}/\sqrt{\tau}$.

4. Conclusions

As a result of the investigations performed, we developed a radiation source with relative long-term frequency instability $\Delta\nu/\nu < 5 \times 10^{-15}$ at an averaging time of 10^3 s. Frequency stabilisation was performed using narrow optical resonances on the intersystem crossing transition $^1S_0-^3P_1$ in magnesium

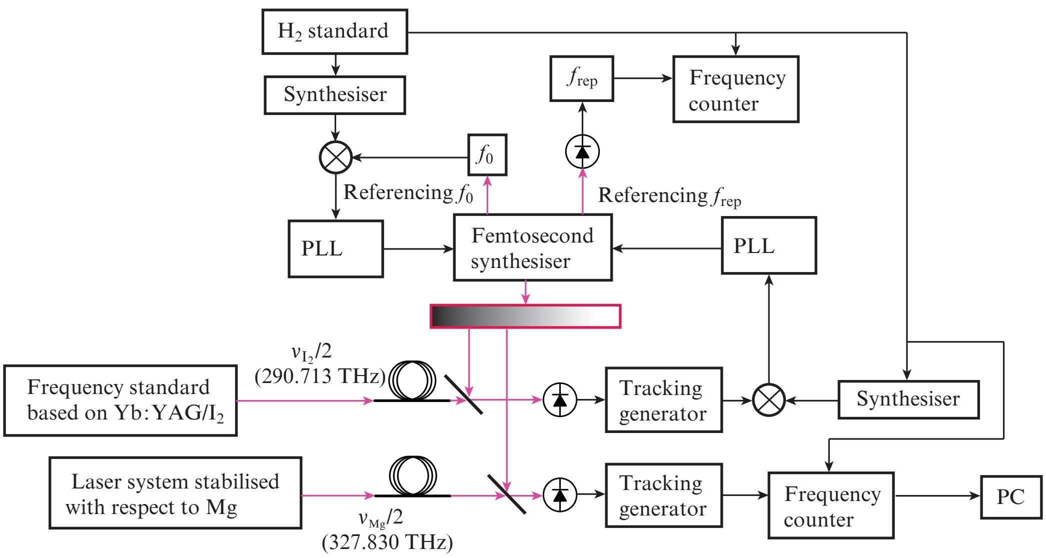

Figure 5. Schematic for measuring the long-term frequency stability of the clock laser system: (H_2 standard) hydrogen frequency standard; (PLL) phase-locked-loop frequency control; (tracking generator) DDS-based tracking generator; f_0 is the comb frequency offset of the of femtosecond optical frequency synthesiser and f_{rep} is the pulse repetition rate of the femtosecond synthesiser.

atoms, which were cooled and localised in a MOT. The frequency stability can be significantly increased by improving the thermal stabilisation system of the reference interferometer and increasing the signal-to-noise ratio when recording Ramsey resonances. To rise the signal-to-noise ratio, we are planning to record the absorption signal on the clock transition using the secondary MOT on the $^3P_2-^3D_3$ triplet transition and reduce the temperature of the cloud of cold magnesium atoms, applying sub-Doppler cooling of atoms in the secondary MOT on the $^3P_2-^3D_3$ triplet transition [11–13]. In future, we are planning to implement sub-Doppler cooling of magnesium atoms to a temperature of 10–50 μK and localise atoms in a one-dimensional optical lattice in order to form an optical frequency standard based on ultracold magnesium atoms with a relative long-term instability and frequency uncertainty at a level of $10^{-17}-10^{-18}$ using the strongly forbidden $^1S_0-^3P_0$ transition.

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