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Spectroscopy of the quadrupole clock transition of ytterbium-171 ions for optical frequency standard development

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Abstract. **We outline the results of the spectroscopic investigation of the quadrupole transition of a single ytterbium-171 ion as applied to the development of an optical frequency standard with a relative uncertainty and long-term frequency instability of less than 10^{-17}. The spectral width of the resonance recorded at the centre transition frequency amounts to about 30 Hz.**

Keywords: *laser cooling, spectroscopy, frequency stabilisation optical frequency standard.*

1. Introduction

Optical frequency standards based on spatially localised single laser-cooled ions are presently among the most stable ones [1–3]. The key advantage of these systems is that the ion, which has a forbidden transition (and the corresponding ultranarrow line) in its energy level structure employed as an optical frequency reference, resides in a well-controlled medium isolated from external actions.

An ytterbium-171 ion is the optimal candidate for the development of an optical frequency standard with the prospect of an on-board employment. This is due to the coincidence of several factors: technically, the possibility of using compact diode lasers for ion cooling and detecting the clock transition along with the use of fibre lines for delivering laser radiation; physically, the special features of the energy level structure of ytterbium ions, which permit employing different transitions for the development of the optical frequency standard. We chose the quadrupole $^2S_{1/2} \rightarrow {}^2D_{3/2}$ transition with a wavelength of 436 nm (Fig. 1) and a natural linewidth of 3.1 Hz. Furthermore, ytterbium ions have an octupole transition ($^2S_{1/2}$, $F = 0$) → ($^2F_{7/2}$, $F = 3$) at a wavelength of 467 nm with a natural linewidth of several nanohertz, which is also employed for developing optical frequency standards [1].

In this work we present the results of spectroscopic investigations of the quadrupole transition of a single ytterbium-171 ion, which were performed in the framework of the project aimed at developing an optical frequency standard with a relative uncertainty and long-term frequency instability under 10^{-17}.

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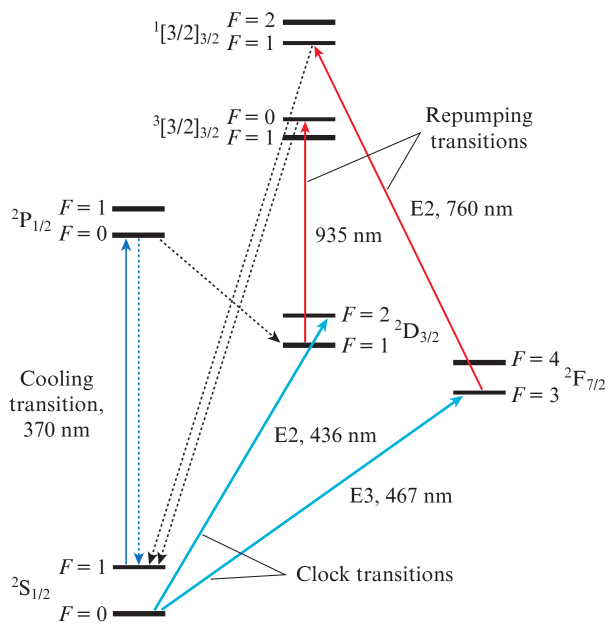

Figure 1. Energy level diagram of Yb.

2. Optical frequency standard based on a single ytterbium-171 ion

Figure 2 shows the block diagram of the optical frequency standard harnessing a single ytterbium-171 ion developed in the ILP, SB of the RAS.

A radio-frequency Paul trap with endcap electrodes [4] is employed to capture and confine the ion. Laser cooling of the ion is performed on the quasi-cyclic dipole transition $^2S_{1/2}$ ($F = 1$) → $^2P_{1/2}$ ($F = 0$) with a natural linewidth of 23 MHz and a wavelength of 370 nm [5,6]. To excite the quadrupole transition $^2S_{1/2}$ ($F = 0$) → $^2D_{3/2}$ ($F = 2$) at a wavelength of 436 nm, use is made of a narrow probe laser (or a so-called clock laser). To depopulate the $^2D_{3/2}$ and $^2F_{7/2}$ levels, advantage is taken of diode repump lasers with output wavelengths of 935 and 760 nm, respectively.

The preparation and examination of the ion energy states is performed with a specially selected sequence of laser pulses. The probability of $^2D_{3/2}$ ($F = 2$, $m_F = 0$) energy level excitation (the so-called quantum jumps [7,8]) is recorded as a function of the clock laser frequency. The detected spectrum consists of several resonances, which contain information about the

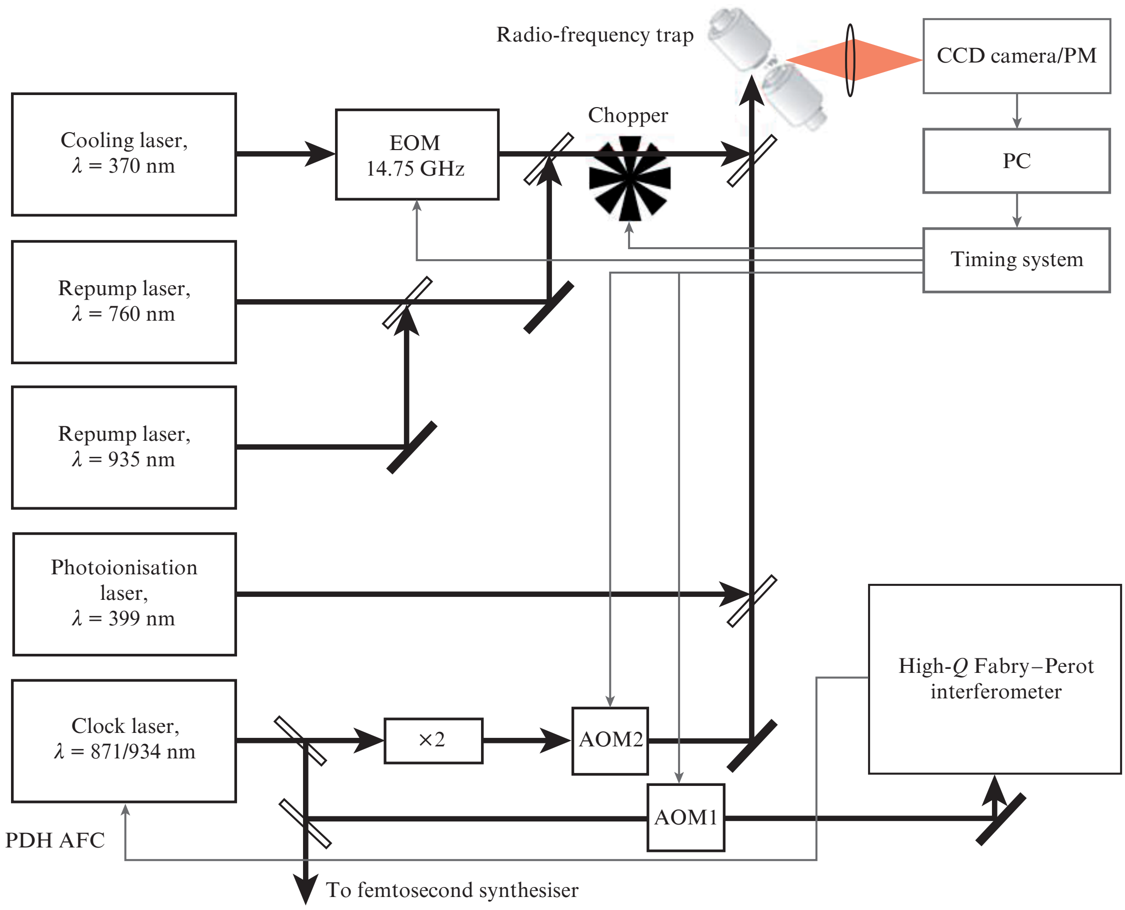

Figure 2. Block diagram of the optical frequency standard with a single ytterbium ion.

state of the ion, its motion in the trap, and its interaction with the environment.

The high short-term frequency stability of the standard is achieved by stabilising the frequency of the diffracted radiation of the probe laser at the acoustic modulator (AOM1) output. The frequency is stabilised to the reflection resonances of a high-Q Fabry–Perot interferometer by the Pound–Drever–Hall method with the use of an automatic frequency control system (PDH AFC). The long-term stability of the system is achieved by tuning the AOM1 frequency in such a way that the second harmonic of the probing laser frequency is coincident with the peak of $^2D_{3/2}$ ($F = 2$) level excitation probability for the single ion localised in the radio-frequency trap. A digital computer-based AFC corrects the AOM1 frequency over a characteristic time which permits providing the signal-to-noise ratio required to achieve the desired stability (in the case of a single ion, the characteristic time is typically no shorter than 100 s). The error signal for determining the position of the second harmonic of the probe laser relative to the probed transition centre is the difference of the excitation probabilities in the detuning to either side of the resonance centre by a half of its width, which is realised by probe radiation frequency tuning with the help of AOM2. Therefore, the short-term radiation frequency stability of the standard over a short time (less than 100 s) is defined by the properties of the Fabry–Perot interferometer, while the characteristics of long-term stability and precision of the standard are defined by the parameters of the resonance on the ultra-narrow optical transition forbidden in the dipole approximation.

3. Trapping and laser cooling of a single ytterbium ion

Ion motion is the main factor that limits the attainment of a high frequency stability of the standard: the time-of-flight broadening of the resonances limits the quality $Q = \nu/\Delta\nu$ of the frequency reference, the residual linear Doppler effect entails the frequency shifts due to the wavefront curvature of the probe radiation, the quadratic Doppler effect and the recoil effect are responsible for an asymmetric profile distortion of the reference spectral line and, as a consequence, for systematic errors in determining its centre. Major progress in the improvement of the metrological characteristics of frequency standards was achieved with the use of ion localisation and cooling. Single-ion optical standards differ advantageously by precisely the absence of the Doppler effect. This is so because the ions in a trap are confined to dimensions much smaller than the wavelength of detecting field (the so-called Lamb–Dicke regime) and the vibrational level structure is spectrally well resolved.

The most convenient technique of confining ions in a limited space involves the use of an alternating electric field to form a three-dimensional potential (the Paul trap) [9]. In the present paper we used an ion trap with an endcap configuration of the electrodes (Fig. 3). Unlike the classical trap with a central ring electrode, this trap type is structurally more convenient, which facilitates ion loading and the introduction of laser radiation. The trap electrodes, an oven for vaporising ytterbium ions, an electron gun, and two additional electrodes

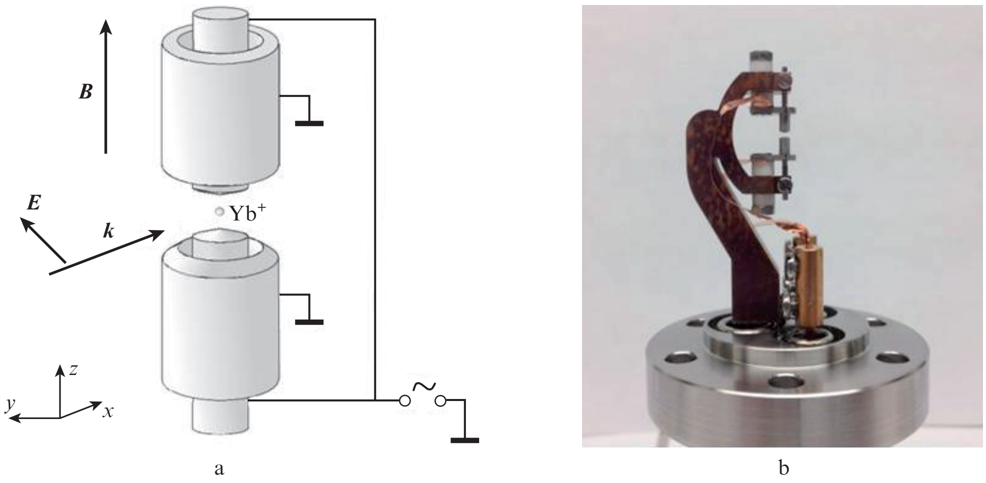

Figure 3. (a) Configuration of the electrodes and voltage application circuit in the endcap ion trap and (b) trap electrodes assembled on the vacuum flange 50 mm in diameter.

are used to apply a dc voltage, which compensates for the field asymmetry in the trap arising from the structure imperfection or the presence of parasitic fields.

An alternating voltage with an amplitude of 600 V and a frequency of 14 MHz is used to form the confining potential. In this case, the potential well depth is equal to 18 eV, the secular trap frequencies $\nu_z = 2\nu_r = 1.2$ MHz, and the stability parameters $q_z = 2q_r = 0.26$, $a_{z,r} = 0$ (the trap is stable when $|q|,|a| < 1$). To lengthen the ion lifetime in the trap, it is accommodated in a vacuum chamber with a residual gas pressure of less than 5×10^{-10} Torr, which minimises the ion loss due to collisions with gas molecules.

Frequency-modulated radiation at a wavelength $\lambda = 370$ nm is used for Doppler cooling and detecting the ion state (Fig. 4). The diode laser radiation is modulated with an electro-optical modulator (EOM) at a frequency of 14.75 MHz to produce the spectral component which excites the superfine component $^2S_{1/2}$ $(F = 0) \to$ $^2P_{1/2}$ $(F = 1)$ of the cooling transition. This component is not excited by the resonance cooling radiation. The modulation component intensities amount to several percent of the carrier intensity. This is sufficient for shortening the ion residence time in the $^2S_{1/2}$ $(F = 0)$ state below 10^{-4} s and improving the efficiency of cooling, which is performed with the radiation up to 10 μW in power focused to a waist of 50 μm.

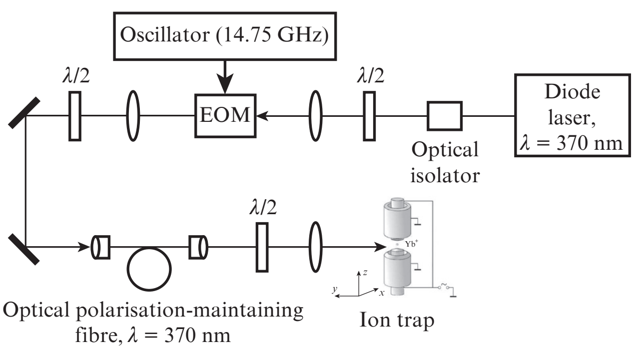

Figure 4. Laser system for Doppler ion cooling.

The ion fluorescence induced by the cooling radiation is projected onto a photomultiplier (PM) and a CCD camera with the use of a multi-lens objective (Fig. 2). The image on the CCD array is used for determining the number of particles captured in the trap as well as for controlling the ion posi-

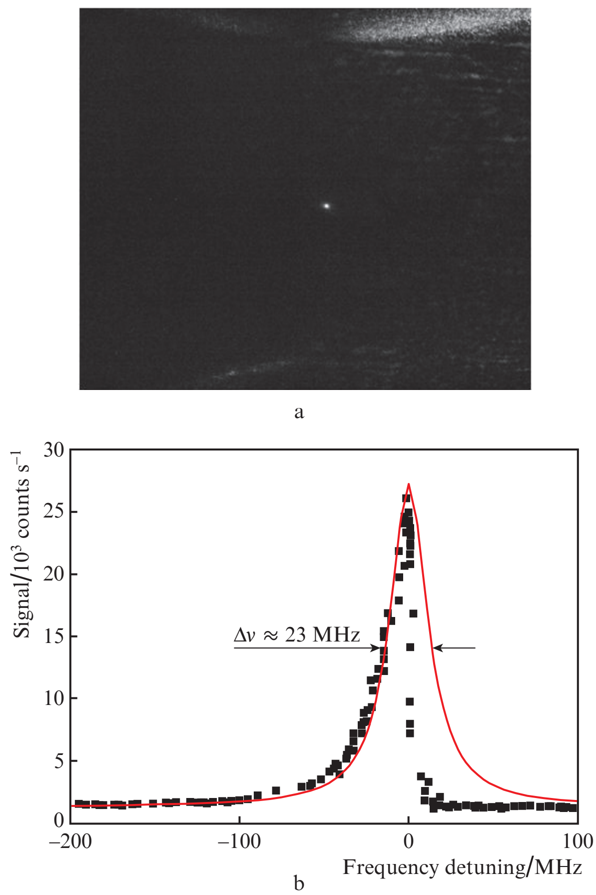

Figure 5. (a) Single trapped ^{171}Yb ion, and (b) resonance fluorescence signal from a single cooled ion obtained in the cooling laser frequency scanning and spectral line approximation with a Lorentzian profile.

tion in the trap. The PM signal serves to determine the total fluorescence intensity with a high temporal resolution.

Figure 5 shows the image of a single ^{171}Yb ion confined in the radio-frequency trap as well as the resonance fluorescence signal at the cooling transition, whose half-width (FWHM) is equal to ~23 MHz. This testifies to the absence of Doppler broadening and, consequently, to the efficiency of ion cooling.

4. Detection of the quadrupole transition

To excite the $^2S_{1/2}$ $(F = 0) \rightarrow$ $^2D_{3/2}$ $(F = 2)$ quadrupole clock transition, use is made of the second harmonic of the fundamental $\lambda = 871$ nm radiation of an external cavity diode laser (Fig. 6). To decrease the spectral linewidth to ~1 Hz, the output laser frequency is stabilised to a reference Fabry–Perot cavity made of glass with a low thermal expansion coefficient (ULE glass, Corning). Mechanical perturbations are among the main noise sources that affect the stability of the reference cavity. To suppress low-frequency vibrations, the chamber housing of the cavity is accommodated on a passive vibration-isolating plate. Furthermore, to suppress the residual perturbations we developed a system of suspensions similar to that described in Ref. [10], which lowered the cavity sensitivity to vibrations. A significant suppression of the effect of ambient temperature on the position of the resonances of the high-quality interferometer was reached by stabilising the temperature of the optical cavity about the 'zero' point of the cavity, which is characterised by a nearly zero temperature expansion coefficient of the cavity base material. Apart from the action of temperature and mechanical perturbations, the reference interferometer length is also liable to changes due to the aging of the material which the interferometer base is made of.

To determine the 'zero' reference point and its drift rate, the clock laser frequency was stabilised to the reference transmission peak and mixed with the frequency of one of the spectral components of the femtosecond supercontinuum stabilised to an optical standard. Measurements were made of the temperature dependence of the beat signal frequency, which characterises the reference drift. Long-term (over several years) observations of the variation of the stabilised probe laser frequency made it possible to confidently determine in 2018 the magnitude of the linear drift of the resonance frequencies of the high-Q reference employed to narrow the laser spectrum. The linear drift rate was shown to decrease with time and presently amounts to 0.04 Hz s^{-1} (Fig. 7). Taking this effect into account in the system of laser frequency stabilisation with the use of the high-Q Fabry–Perot reference allowed us to significantly lengthen the resonance detection time without distortion of its shape caused by the probe laser frequency drift.

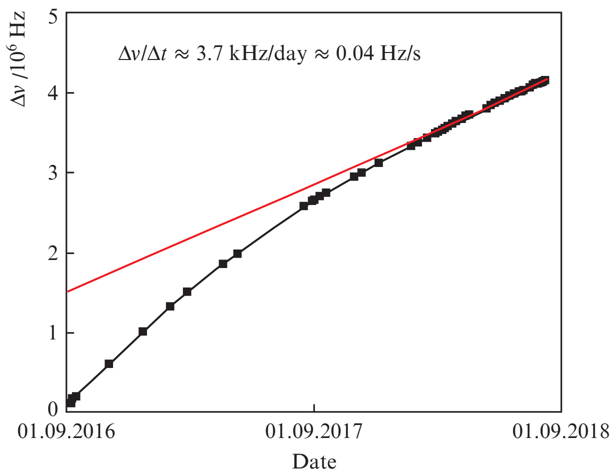

Figure 7. Resonance frequency drift $\Delta\nu$ of the reference Fabry–Perot cavity.

The clock laser linewidth (Fig. 8) stabilised to the reference transmission peak was estimated by the mathematical processing of the error signal at the output of the AFC system with the inclusion of the measured frequency discriminator slope with the use of the method presented in Ref. [11]. The half-height width of the line is equal to ~0.8 Hz. This estimate characterises the degree of laser frequency noise suppression by the AFC system and does not take into account the broadening caused by interferometer parameter variations due to vibration, thermal noise, etc.

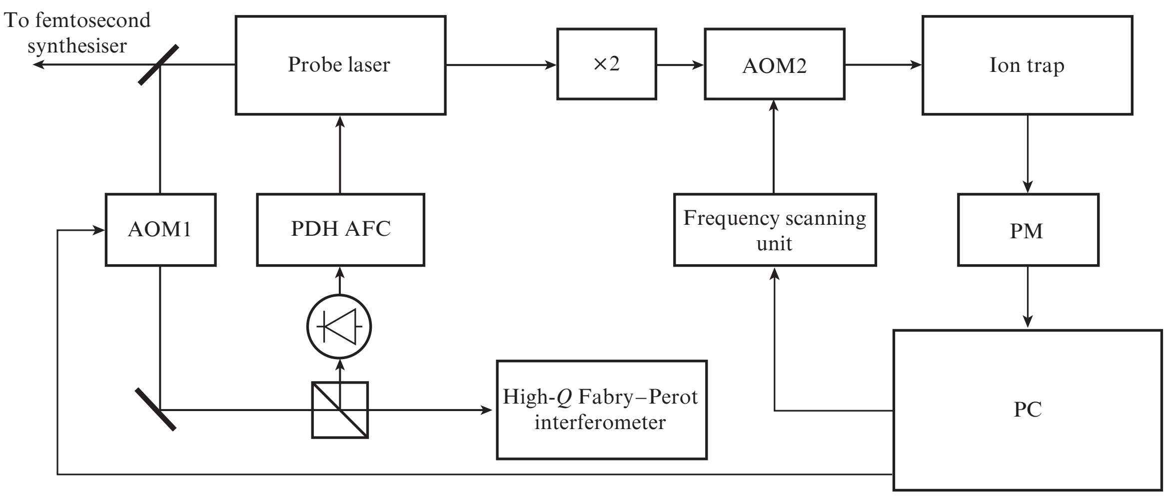

Figure 6. Laser system intended for the excitation of the quadrupole transition of ytterbium ion.

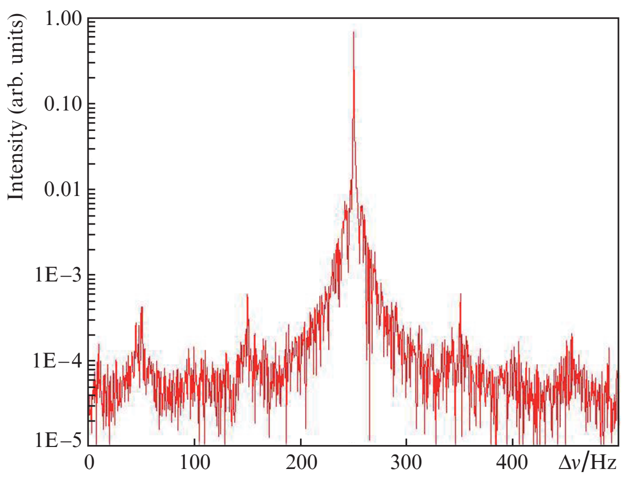

Figure 8. Clock laser line reconstructed neglecting the contribution of the reference noise.

The idea of observing resonances in single ions involves the application of quantum jump detection technique – the ion transition from one energy state to another – by the onset of the fluorescence at the cooling transition (by electron shelving [12]). When the fluorescence signal does not appear for some time (for the quadrupole transition it amounts to several milliseconds) after switching off the probe laser and switching on the cooling cycle, the ion is highly likely to be in the $^2D_{3/2}$ ($F = 2$) excited state. As a rule, for the sake of measurement validity the sequence of cooling, excitation, and detection cycles is repeated about twenty times for one frequency of the clock laser.

The total cycle duration amounts to 100 ms (Fig. 9). The resonance is detected for 5 ms prior to a cooling cycle. On completion of the cooling cycle (20 ms), the modulation component of the cooling radiation is turned off and during the next 20 ms the ion is transferred with a high probability to the $^2S_{1/2}$ ($F = 0$) level of the ground state via the nonresonance excitation to the level $^2P_{1/2}$ ($F = 1$), which manifests itself in

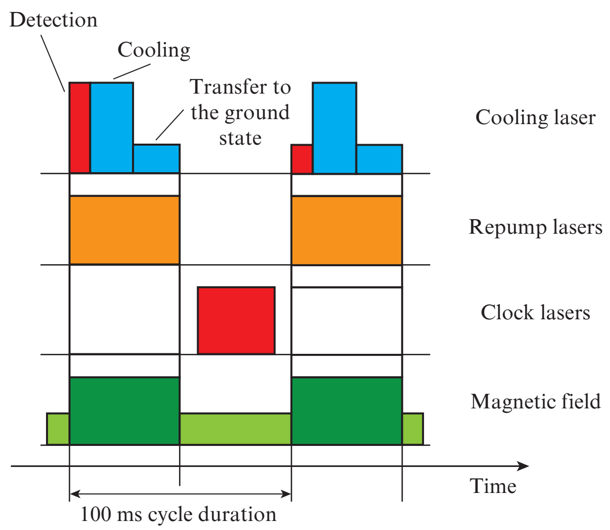

Figure 9. Time sequence of cooling, excitation, and detection pulses for the $^{171}Yb^+$ clock transition.

the disappearance of the fluorescence signal. To prevent the optical pumping of $^2S_{1/2}$ ($F = 1$) nonresonance magnetic sublevels, a permanent magnetic field $B \approx 400$ μT aligned with the trap axis is switched on during the cooling cycle. The polarisation vector of the cooling radiation makes an angle of 45° with vector $\boldsymbol{B}$ to increase the fluorescence probability. To depopulate the $^2D_{3/2}$ and $^2F_{7/2}$ levels, use is made of the diode repump lasers radiating at wavelengths of 935 and 760 nm, respectively. On completion of the cooling cycle, the radiations of the cooling and repump lasers is mechanically blocked and the magnetic intensity is lowered to about 1 μT (to provide the splitting of the clock transition components with different magnetic quantum numbers) for the spectroscopy of the $^2S_{1/2}$ ($F = 0$) → $^2D_{3/2}$ ($F = 2$) transition.

Figure 10 shows the results of detection of the excitation spectrum of the $^2D_{3/2}$ ($F = 2$) ytterbium-171 ion level. The spectrum (Fig. 10a) consists of five components corresponding to $\Delta m_F = 0, \pm 1, \pm 2$. As is clear from Fig. 10, the magnetic splitting of the sublevels amounts to ~ 50 kHz, which corresponds to a residual magnetic field $B \approx 6$ μT in the trap. Figure 10b shows the central component of the spectrum. In this case, a 30-ms long pulse of the clock laser was used to excite the resonance. The spectral width (FWHM) of the resonance recorded at the central transition frequency amounts to about 30 Hz for a level excitation probability of ~ 0.5.

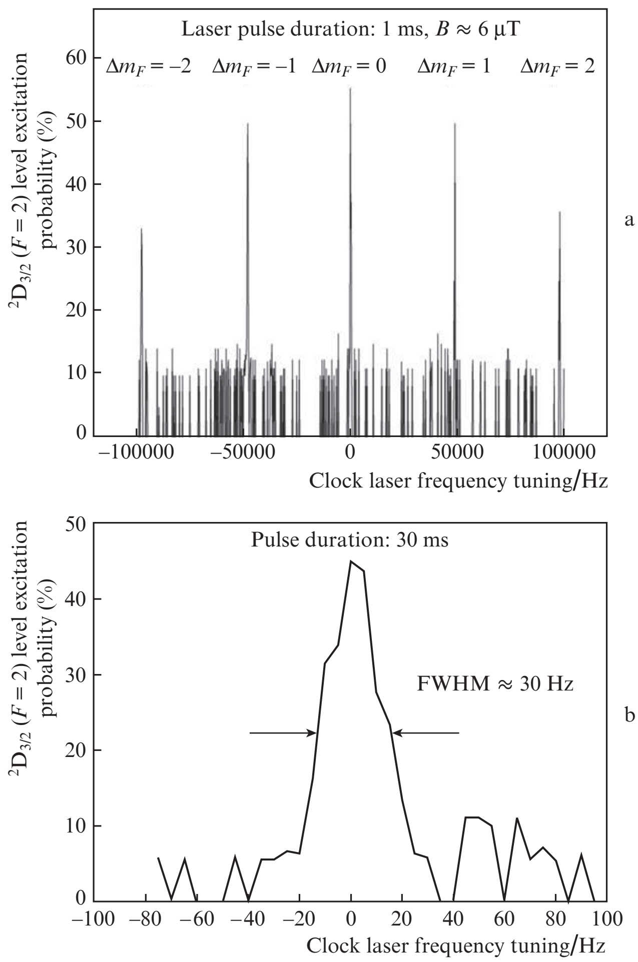

Figure 10. (a) Excitation spectrum of the $^2D_{3/2}$ ($F = 2$) ytterbium-171 ion level and (b) central component of the spectrum. The scan rate of the clock laser frequency is equal to 5 Hz s^{-1}.

In accordance with the method described above, the clock laser frequency was simultaneously stabilised to the transmission peak of the reference Fabry–Perot cavity and the central resonance frequency of the quadrupole transition of ytterbium ions.

5. Conclusions

We have performed spectroscopic investigations of the quadrupole transition of a single ytterbium-171 ion, which were performed in the framework of the project aimed at developing an optical frequency standard with a relative uncertainty and long-term frequency instability under 10^{-17}.

To trap and confine the ion, use is made of a miniature radio-frequency Paul trap with endcap electrodes of original design. The laser cooling of the ion is performed on the quasi-cyclic dipole transition $^2S_{1/2}$ (F = 1) → $^2P_{1/2}$ (F = 0) with a natural width of 23 MHz and a wavelength of 370 nm. A narrow-band clock laser is employed to excite the $^2S_{1/2}$ (F = 0) → $^2D_{3/2}$ (F = 2) quadrupole transition at λ = 436 nm. The laser linewidth was narrowed by stabilising the radiation frequency to a high-Q Fabry–Perot cavity made of glass with an ultra-low thermal expansion coefficient.

The preparation and examination of the ion energy states is performed with a specially selected sequence of laser pulses. The probability of $^2D_{3/2}$ (F = 2, m_F = 0) energy level excitation is recorded as a function of the clock laser frequency. The detected excitation spectrum consists of several resonances, which contain information about the state of the ion, its motion in the trap, and its interaction with the environment. The resonance width recorded at the central transition frequency amounts to about 30 Hz.

The clock laser frequency was simultaneously stabilised to the transmission peak of the reference Fabry–Perot cavity and the centre resonance frequency of the quadrupole ytterbium ion transition.

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