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Ultrastable laser system for spectroscopy of the $^1S_0-{}^3P_0$ clock transition in Sr atoms

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Abstract. **A laser system with a spectral linewidth less than 1 Hz for spectroscopy of the $^1S_0-{}^3P_0$ clock transition in strontium atoms has been demonstrated. A semiconductor laser emitting at a wavelength of 698 nm was stabilised to an external high-finesse Fabry–Perot cavity with vibration and temperature compensation near the zero expansion point. After laser cooling to a temperature below 3 μK, ^{88}Sr atoms were loaded into an optical lattice at a magic wavelength of 813 nm. The laser system was used to characterise the ^{88}Sr clock transition by magnetically induced spectroscopy. The resonance spectral width was determined to be 130±17 Hz, which corresponds to a quality factor of 3×10^{12}.**

Keywords: *Sr atom, laser cooling, optical lattice, magnetically induced spectroscopy, clock transition, stable cavity.*

1. Introduction

Frequency standards play an important role in modern basic and applied physics. In the last decade, optical atomic clocks have reached an extremely high level of stability and accuracy, having surpassed caesium fountains – primary microwave frequency standards – by two orders of magnitude. The relative instability of the best optical clocks today reaches a few parts in 10^{18} [1, 2], whereas the best fountain-type references are limited to 2×10^{-16} [3]. This offers the possibility of utilising optical clocks as extremely accurate tools, allowing fundamental theories to be tested at a new level of accuracy and opening up new possibilities of using such clocks in practical applications.

Atoms or ions trapped and cooled to temperatures of a few microkelvins are at the heart of optical clocks. Having an extremely narrow clock transition, with a natural linewidth typically under 1 Hz, they are used as precision frequency references. One important problem is to shield the atoms and ions from the effect of external fields or minimise their effect on the clock transition. For the excitation and spectroscopy of such narrow transitions, an ultrastable cw coherent light source comparable in spectral linewidth to the transition being interrogated is needed. It is desirable that the spectral width of laser light be smaller than the expected measurement uncertainty, because laser emission spectrum asymmetry can make a significant contribution to the systematic error of the optical clock. Otherwise, the symmetry of the laser emission spectrum should be investigated [3]. Moreover, the long-term stability of an optical clock can be influenced by the Dick effect, which can also be suppressed by narrowing the laser emission linewidth [4].

In the framework of the federal targeted programme GLONASS 2012–2020, researchers at the All-Russia Research Institute of Physical and Radio Engineering Measurements aim to create an optical frequency standard using cold ^{87}Sr atoms trapped in an optical lattice. In the case of the odd isotope ^{87}Sr, which has a nuclear spin $I = 9/2$, the $^1S_0-{}^3P_0$ transition is weakly allowed owing to the hyperfine interaction – which mixes the $^3P_0$ state with the $^3P_1$, $^3P_2$ and $^1P_1$ states – and has a natural linewidth $\gamma = 1$ mHz. The ^{87}Sr isotope is, however, less abundant than ^{88}Sr and requires additional laser fields for deep cooling [5]. Because of this, the even isotope ^{88}Sr was initially chosen for adjusting a magneto-optical trap, secondary cooling [5, 6] and retrapping in an optical lattice. It is also reasonable to employ the even isotope for assessing the capabilities of the laser system produced in this study.

The $^1S_0-{}^3P_0$ clock transition, at a wavelength of 698 nm, is completely forbidden for the even isotope ^{88}Sr. According to previous work [7], however, the forbidden transition can be excited in an external magnetic field, which mixes levels. This approach was successfully used to implement spectroscopy of even-isotope clock transitions in a number of laboratories in the world [8–11].

In this paper, we describe stabilised laser systems for spectroscopy of the clock transition in strontium atoms with a spectral linewidth less than 1 Hz. By comparing two identical laser systems, we analyse in detail spectral characteristics of the emission. In addition, we present the first results on the magnetically induced spectroscopy of the $^1S_0-{}^3P_0$ transition in ^{88}Sr atoms with the use of so-called clock lasers, which confirms that the laser systems can be applied in spectroscopy of narrow clock transitions.

2. Deep laser cooling of strontium atoms

One advantage of optical frequency standards based on neutral atoms is the possibility of accumulating a large number of atoms, up to 10^4-10^5, in an optical lattice, which leads to an

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increase in signal-to-noise ratio and a decrease in frequency instability in comparison with ion standards. For loading into an optical lattice that ensures Doppler effect suppression (Lamb–Dicke regime), eliminates the collisional shift and increases the time of interaction with interrogating light, the temperature of the atoms should be lowered to a few microkelvins.

The most widely used approach is laser cooling, which involves two stages in the case of ^{88}Sr atoms. First stage cooling is effected on the cyclic, essentially closed transition $^1S_0-{}^1P_1$, at a wavelength $\lambda = 461$ nm (Fig. 1), which allows the atoms to be cooled to mK temperatures and collected in a magneto-optical trap (MOT). The temperature achievable in this stage is determined by the Doppler limit $T_D = h\gamma_1/(2k_B) =$ 770 μK, where $\gamma_1 = 30$ MHz is the natural linewidth of the $^1S_0-{}^1P_1$ transition. Second stage cooling is effected on the narrow intercombination transition $^1S_0-{}^3P_1$ (wavelength $\lambda =$ 689 nm, natural linewidth $\gamma_1 = 7.4$ kHz) to the temperature corresponding to the Doppler limit $T_D = h\gamma_2/(2k_B) = 200$ nK.

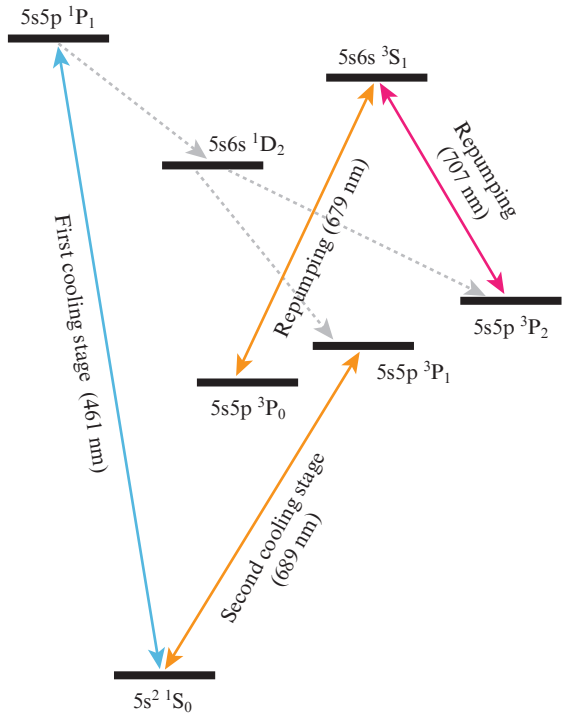

Figure 1. Simplified diagram of the ^{88}Sr levels involved in the experiment and functions of the laser sources used.

Even in the second cooling stage, the spectral linewidth of the cooling laser should meet heightened requirements, because the natural linewidth in the second cooling stage is just 7.4 kHz. The main sources of coherent light at the second stage cooling wavelength $\lambda = 689$ nm in current use in laboratories are diode lasers. The spectral linewidth of diode lasers having a diffraction grating as an external cavity is 0.1 to 1 MHz. For second stage cooling, we made a second stage laser cooling system stabilised to an external high-finesse Fabry–Perot cavity and having a spectral linewidth of 80 Hz [12].

The use of the second stage laser cooling system allowed us to reload 40% of the precooled atoms into a second-stage MOT at a wavelength of 689 nm and cool 10^5 atoms to a temperature of 2.5 μK [5].

3. LS1 and LS2 clock laser systems

The ability to reduce the spectral linewidth of a clock laser is critical for successful realisation of an optical clock. To perform spectroscopy of a clock transition without degrading its characteristics, the spectral linewidth of the interrogating laser should be smaller than the spectral width of the transition. A spectral line of atoms experiences broadening due to various mechanisms: the interrogating light power, interaction time or the effect of an external magnetic field, as in the case of ^{88}Sr. In most experiments, it is convenient if the measured spectral linewidth of a clock transition lies in the range 1–10 Hz. This is related to a measurement cycle, which typically takes less than 1 s. Thus, to achieve the required stability of an optical clock, the spectral linewidth of the interrogating laser should be less than 1 Hz. One way of narrowing the emission linewidth of a laser is by stabilising it to an external high-finesse Fabry–Perot cavity.

The emission frequency of a Toptica DL pro semiconductor laser operated at $\lambda = 698$ nm was stabilised by the Pound–Drever–Hall technique [13] to the transmission peak of an external high-finesse Fabry–Perot cavity. We fabricated two, essentially identical cavities from ultralow thermal expansion glass (ULE, Corning [14]): ULE3 and ULE4. The cavity design and stabilisation scheme were similar to those used to stabilise the emission frequency of the second stage cooling laser and were described in detail elsewhere [12]. The finesse of both cavities was evaluated from the attenuation of the power circulating in the cavities and was above 250 000. To study the spectral characteristics of the emission, we compared two, essentially identical laser systems, LS1 and LS2, stabilised to the transmission peak of the ULE3 and ULE4 cavities, respectively. Concurrently, the emission frequency of the lasers was measured using a femtosecond frequency comb (FFC). Figure 2 presents a schematic of the comparison process.

Comparison was carried out using a heterodyne beat signal recorded by a photodetector (PD1) with a 1-GHz bandwidth. After amplification, the signal frequency was

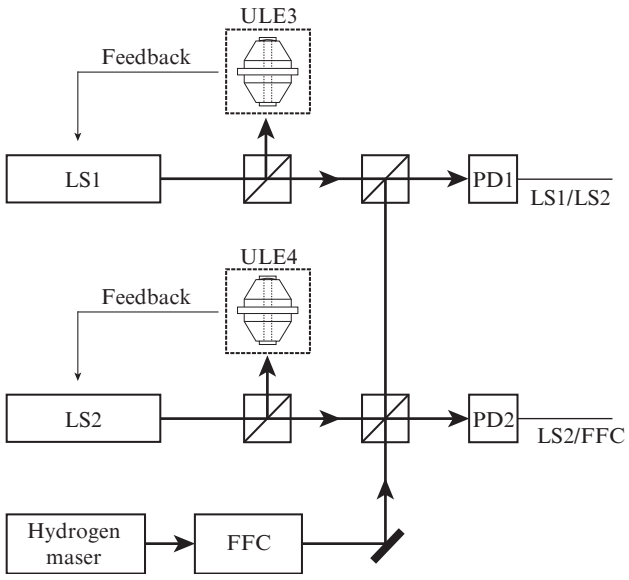

Figure 2. Laser system comparison scheme (the thin lines represent electrical signals and the thick lines represent optical paths).

measured using a dead-time-free FXE counter [15] and the data were stored on a computer. Spectral characteristics of the beat signal were examined using an Agilent N9010 spectrum analyser.

To reduce the effect of temperature fluctuations on the frequency of cavity eigenmodes, the cavities should be maintained at the zero expansion temperature of the glass. Since the critical temperature depends on ULE glass preparation conditions and may range widely (spread from –20 to +40 °C was observed), the zero expansion temperature of the glass should be determined experimentally. The thermal expansion coefficient η of ULE glass can be approximately represented as

$$\eta \approx 10^{-9}(T - T_c)^2. \quad (1)$$

At the critical temperature T_c, the cavity has the smallest linear dimensions, which can be demonstrated by measuring the frequency of the beat signal between the lasers. To this end, the temperature of one cavity was maintained constant and it was used as a frequency reference, whereas the temperature of the other cavity was changed every other day, following which the beat signal frequency was measured. This long time between the measurements was needed because of the long time constant of the cavity temperature stabilisation system.

The measurement results are presented in Fig. 3. The data were fitted with quadratic polynomials. The critical temperature of the ULE3 and ULE4 cavities was determined to be 30 and 31 °C, respectively. For subsequent experiments, the temperature of the cavities was stabilised at their critical points to within 0.1 K. The temperature of one of the inner screens was maintained with a stability of 100 μK.

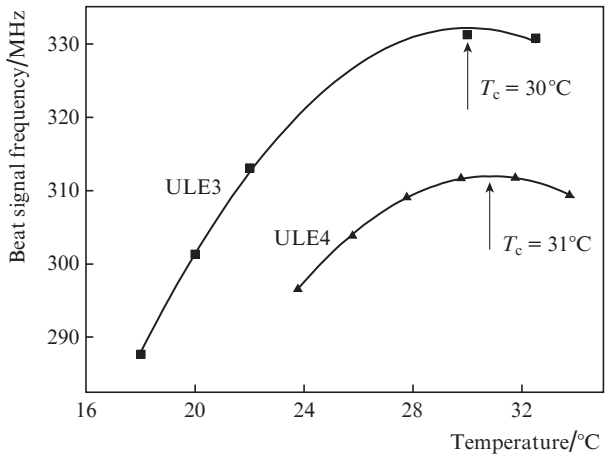

Figure 3. Frequency of the beat signal between the LS1 and LS2 laser systems as a function of the temperature of ULE3 and ULE4.

A spectral analysis of the beat signal between LS1 and LS2 showed that its spectral width at half maximum was 1.5 Hz. Figure 4a shows the average of seven spectra measured with a 1-Hz resolution at a measurement time of 1.8 s. Under the assumption that the two lasers make equal contributions to the spectral width of the beat signal, the emission linewidth of each laser can be estimated at about 1 Hz ($1.5/\sqrt{2}$ Hz). Note that the resolution of the spectrum analyser makes a certain contribution to the width of the measured spectrum, so the above value is the upper estimate.

To measure the Allan deviation, the beat signal was monitored with a counter for several hours. To detect possible cycle slips, the signal was divided into two parts, filtered by different band-pass filters and detected simultaneously in the two channels of the counter. The signal difference was monitored throughout the measurement time. If the signal difference exceeded a certain value, the data point was rejected. The data thus obtained were represented as a normalised Allan deviation.

The frequencies of the laser systems have a linear relative drift at a level of 200 mHz s^{-1}, due to the recrystallization process in the ULE glass. Figure 4b shows the normalised Allan deviation of the LS1/LS2 beat signal, with linear drift removed [curve (*1*)] and not removed [curve (*2*)]. It is seen that, with no allowance for linear drift, the relative laser frequency instability over averaging times from 1 to 100 s reaches $(2-3)\times10^{-15}$, approaching the thermal noise limit [16]. The measured Allan deviation is consistent with the spectral width of the laser spectrum measured using a spectrum analyser.

The better performance of the LS1 and LS2 clock laser systems compared to previously made systems for the second

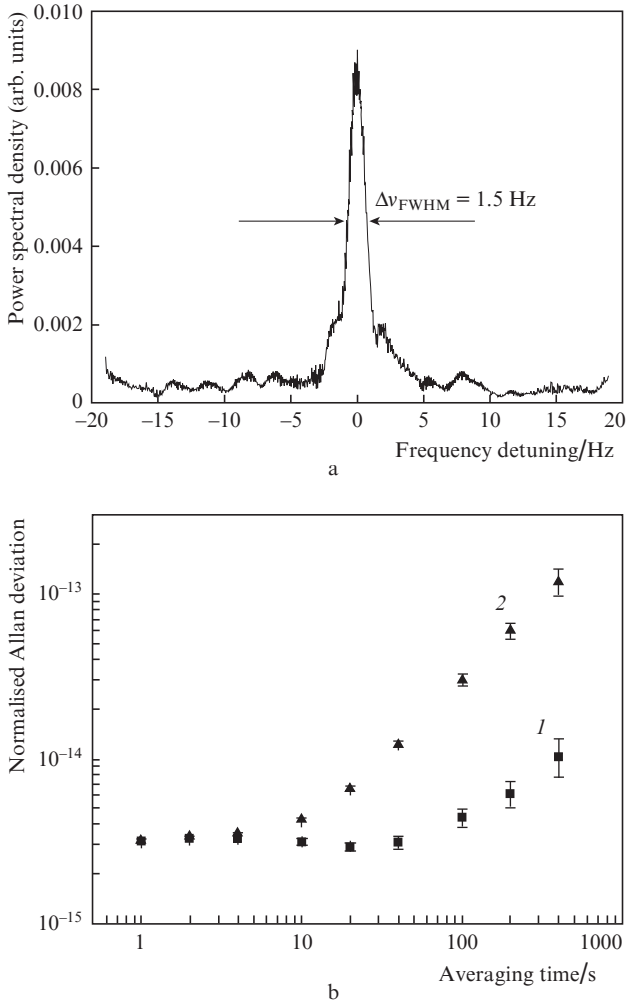

Figure 4. (a) Spectrum of the beat signal between the LS1 and LS2 clock laser systems stabilised to the ULE3 and ULE4 cavities (the average of seven spectra measured with a 1-Hz resolution over a period of 1.8 s) and (b) normalised Allan deviation of the beat signal between the two clock lasers stabilised to the ULE3 and ULE4 cavities, with linear drift (*1*) removed and (*2*) not removed.

stage cooling of strontium atoms [12] is due to the higher cavity finesse and lower input power, as well as to the accommodation of the lasers in a specialised room.

To preadjust to the clock transition, we used a frequency comb stabilised to the hydrogen maser frequency. The clock laser frequency was determined from the LS2/FOFG beat signal with an accuracy of 1 kHz.

4. Spectroscopy of the ^{88}Sr $^1S_0-^3P_0$ transition in a vertical optical lattice

Strontium atoms laser-cooled to a temperature of 3 μK were trapped in a vertical (along gravity) optical lattice at a 'magic' wavelength of 813 nm. As shown by Ye et al. [17], the levels of the clock transition have identical polarisabilities at the magic wavelength, which nullifies the linear term in the dynamic Stark shift. To produce an optical lattice, we used a Toptica DL pro diode laser with a tapered amplifier that ensured 1.2 W of output power. The emission wavelength of the laser was $\lambda_m = 813.42757(62)$ nm and the emission frequency was stabilised with an accuracy of 1 MHz using an Angstrom WS-U wavelength meter. The light was launched into a vacuum chamber through a single-mode optical fibre, which ensured a Gaussian intensity distribution across the beam. The beam power at the fibre output was 600 mW. The beam was focused by an $F = 300$ mm lens to a waist diameter of 30 μm, which corresponded to a calculated lattice trap depth $U_{trap} \approx 100\,E_{rec}$, where E_{rec} is the recoil energy. We were able to load 10^4 atoms into the optical lattice.

We measured the lifetime of the atoms in the optical lattice. The number of atoms was evaluated from the $^1S_0-^1P_1$ fluorescence signal recorded with a 50 μs laser pulse (λ = 461 nm). The measurement results are presented in Fig. 5. The characteristic lifetime of the atoms in the optical lattice was determined to be 400 ms and was limited by both the residual vacuum in the chamber and the constant inflow of hot atoms from the Zeeman slower.

Since the $^1S_0-^3P_0$ clock transition is completely forbidden for the ^{88}Sr isotope, it is necessary to employ magnetically induced spectroscopy [7]. A uniform magnetic field was generated by the coils of a magneto-optical trap. To this end, before measurements the coils were switched from an anti-Helmholtz to a Helmholtz configuration using electric switches.

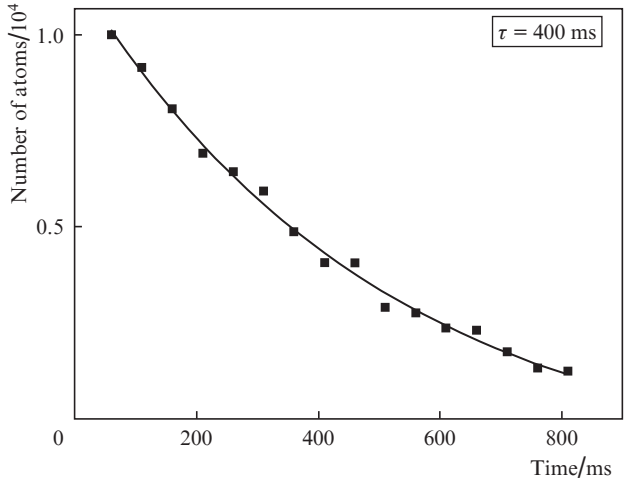

Figure 5. Dynamics of the atomic population loss from the optical lattice. Fitting with an exponential function shows that the characteristic lifetime of the atoms in the lattice is 400 ms.

After pretuning the clock laser frequency to the $^1S_0-^3P_0$ transition using the frequence comb, we carried out experiments aimed at locating the transition. The frequency of the LS2 clock laser was swept using an acousto-optic modulator (AOM). Since spectroscopy of strongly forbidden transitions is a nontrivial issue, it is addressed using an indirect method for assessing the fraction of excited atoms, followed by estimation of the excitation probability. Figure 6 shows a schematic diagram of the experiment and the sequence of light and magnetic field pulses in a measurement cycle.

Atoms trapped in a vertical optical lattice interact with the clock laser light, whose wave vector is collinear with the axis of the optical lattice. A 100-ms clock laser pulse brings some of the ^{88}Sr population from the $^1S_0$ ground state to the $^3S_0$ excited state. The residual ground-state population can be evaluated from the $^1S_0-^1P_1$ fluorescence caused by a 50-μs probe pulse, which heats the atoms and removes them from the trap. The atoms excited by the clock laser pulse to the $^3P_0$ state are brought back to the $^1S_0$ ground state by a repumping laser (679 nm) pulse, and their number is then evaluated from the $^1S_0-^1P_1$ fluorescence signal. In this way, we obtain information about the fraction and total number of excited atoms as functions of clock laser frequency detuning. This allows us to determine the excitation probability in each measurement cycle and reduces the noise related to fluctuations in the number of atoms in the lattice from cycle to cycle.

The measurement cycle is repeated many times, which makes it possible to accumulate data by sweeping the clock laser frequency in the resonance region and obtain the spectrum of the transition line. The optimal clock pulse duration is determined by the Rabi frequency Ω, which depends on the magnetic field B and light intensity I:

$$\Omega = \alpha\sqrt{I}\,|\,B\,|\cos\theta, \tag{2}$$

where $\alpha = 198$ Hz T^{-1} (mW cm^{-2})$^{-1/2}$ and θ is the angle between the polarisation vector of the clock laser light and the magnetic induction vector. In our case, the pulse duration was chosen so as to maximise the clock transition excitation probability. We plan to perform additional studies of the frequency and spectral width of the clock transition as functions of magnetic field and light intensity.

The cycle in Fig. 6b uses the third probe pulse (461 nm), making it possible to considerably reduce the effect of the background, which is independent of the presence of atoms in the lattice. To raise the signal-to-noise ratio, measurements are typically made in complete darkness. To this end, the vacuum chamber should be isolated from external light. This, however, does not fully resolve the problem of background in the chamber, because there is glare due to the laser light reflected from parts of the chamber and there is always emission from the Zeeman slower, which also operates at a wavelength of 461 nm. Moreover, the background signal is contributed by the dark current of the detection electronics. The third pulse determines the background level for each measurement cycle, and then the background is subtracted, which allows measurements to be made even under typical laboratory lighting conditions.

Figure 7 shows the spectrum of the ^{88}Sr $^1S_0-^3P_0$ clock transition obtained using the above algorithm. The magnetic

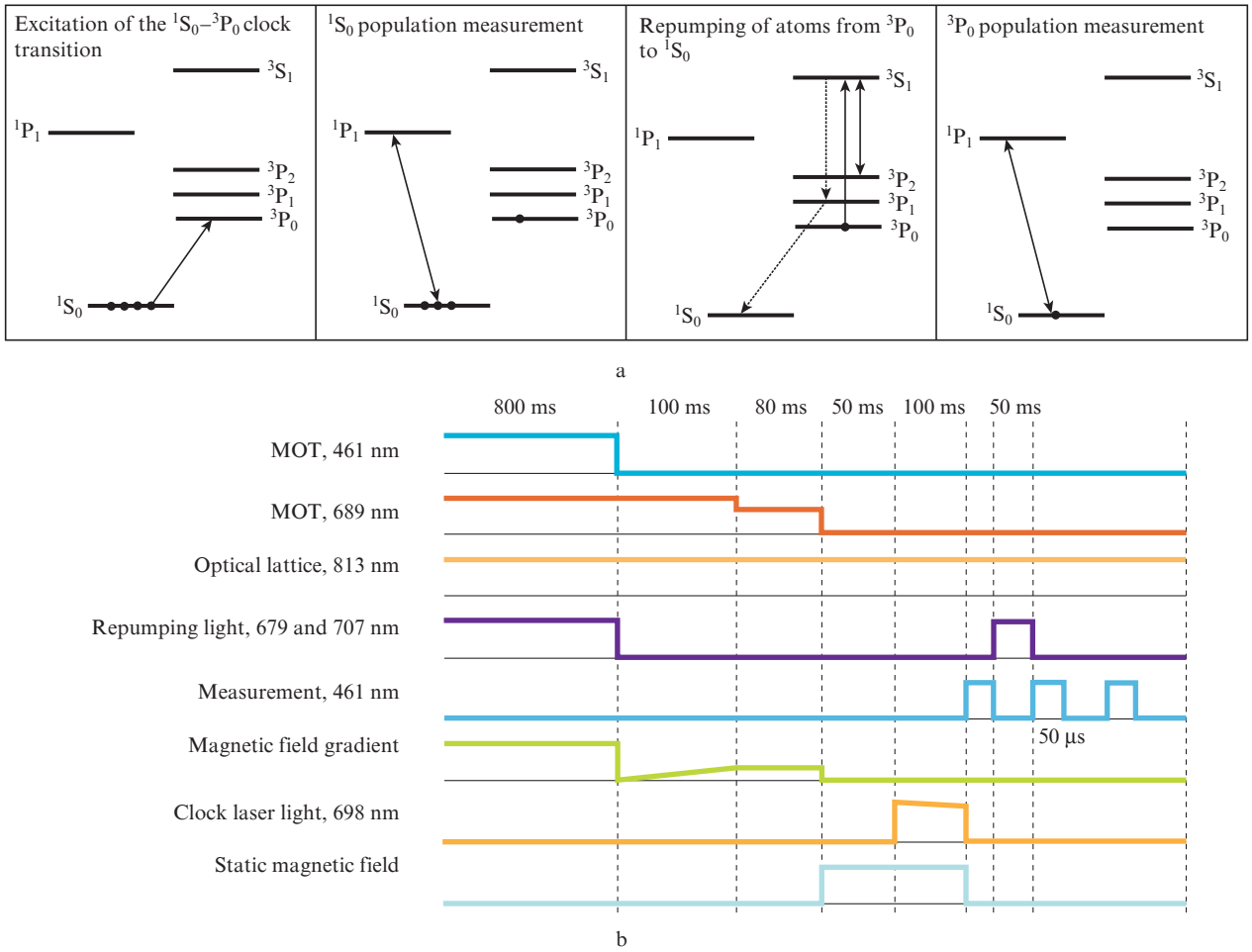

Figure 6. (a) Step-by-step diagram for spectroscopy of the clock transition and (b) timing diagram for the sequence of light and magnetic field pulses used in spectroscopy of the clock transition.

field was 2 mT. The line was well fitted by a Lorentzian with a spectral width at half maximum of 130 ± 17 Hz.

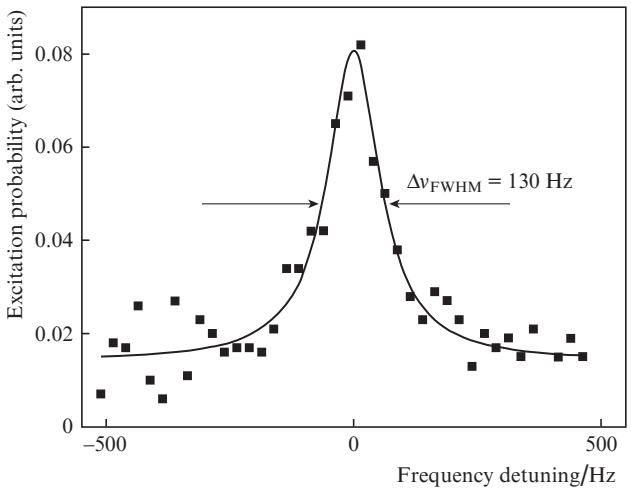

Figure 7. Spectral line of the $^1S_0$–$^3P_0$ clock transition in the ^{88}Sr isotope. The line was fitted by a Lorentzian with a full width at half maximum of 130 ± 17 Hz. The magnetic field was 2 mT. Each data point corresponds to one measurement cycle of 1.5 s duration.

The detection and spectroscopy of the clock transition constitute an important step towards the creation of an optical atomic clock. The first experiments demonstrate high frequency stability and spectral purity of the emission from the clock laser system made by us. Since short-term optical clock stability is determined by the clock laser stability, we expect that, at the resonance quality factor achieved $f/\Delta f = 3\times10^{12}$, for an ensemble of 10^4 atoms, the relative instability of a ^{88}Sr frequency standard will be no worse than $5\times10^{-15}\tau^{-1/2}$. However, as mentioned above, to reach these parameters one should control systematic effects, such as the collisional shift and Zeeman effect, at a high level. Nevertheless, the obtained estimate suggests that there are good prospects for creating a ^{87}Sr optical frequency standard with a resonance quality factor reaching 5×10^{14} under working conditions.

5. Conclusions

We have demonstrated a laser system with an emission wavelength of 698 nm for spectroscopy of the $^1S_0$–$^3P_0$ clock transition in strontium atoms with a spectral linewidth no greater than 1 Hz. Using this system, we have adjusted to the clock transition and investigated the ^{88}Sr $^1S_0$–$^3P_0$ transition by magnetically induced spectroscopy. In a magnetic field of 2 mT, the spectral linewidth of the clock transition was 130 Hz, which corresponds to a resonance quality factor of 3×10^{12}.

Using spectroscopy of the ^{88}Sr isotope as an example, we have shown that the proposed method ensures the possibility of measuring characteristics of the ^{88}Sr clock transition.

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