Clock transition excitation efficiency determination using an additional short clock $\pi$-pulse

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Abstract. We demonstrate a simple and efficient readout procedure of the 1.14 $\mu$m clock transition excitation efficiency in neutral thulium atoms. Our method utilizes additional but short clock $\pi$-pulse to measure number of atoms excited by spectroscopic clock pulse, that allows to determine excitation probability insensitive to fluctuation of number of atoms in the trap. It is important that this method can be used for both hyperfine components of the clock transition and does not require any additional lasers in the setup.

1. Introduction
In the past decades optical clocks based on neutral atoms and single ions reached the level of total systematic uncertainty at or lower than $1 \times 10^{-18}$ [1, 2, 3]. Such devices are the most precise tools for fundamental experiments like searching for dark matter and fundamental constants drifts [4]. At the same time, some applications like precision gravimetry, geodesy and navigation require transportable devices of comparable precision [5, 6, 7]. Creating a compact optical clock with low uncertainty is a challenging task. There are several choices of the atomic species and clock transitions with their own advantages and disadvantages. While optical clocks based on neutral atoms have better stability, single ion optical clocks are typically less sensitive to the environment and more compact. Nowadays there are a number of projects of transportable optical clocks based on single ions [7, 8] and neutral atoms [5, 9]. Optical lattice clock based on neutral thulium atoms may combine the advantages of both types of these platforms.

In previous works we demonstrated Fourier-limited spectra of the clock transition $|4f^{13}(2F'')6s^2, J = 7/2, F = 4, m_F = 0\rangle \rightarrow |4f^{13}(2F'')6s^2, J = 5/2, F = 3, m_F = 0\rangle$ at 1140 nm in thulium and experimentally studied different sources of its frequency shifts and uncertainties [10]. Here $F$ is the total angular momentum, and $m_F$ is the projection of the total angular momentum on the quantization axis. Further in the article we refer to the ground state as $|g\rangle$ and to the upper state of the clock transition as $|c\rangle$. Unlike many other clock transitions in neutral atoms, the clock transition in thulium has low sensitivity to static electric fields and blackbody radiation (BBR), similar to that of clock transitions in ions. The highest frequency shift and uncertainty in thulium clock transition comes from a second-order Zeeman shift. To address this problem we plan to use second clock transition $|g, F = 3, m_F = 0\rangle \rightarrow |c, F = 2, m_F = 0\rangle$ between other hyperfine levels. This clock transition has exactly the same absolute value of the
second-order Zeeman shift but with the opposite sign. Therefore, the sum of frequencies of the two clock transitions, which we call the synthetic frequency, should be free from a quadratic Zeeman shift [10].

In our experiments we can easily measure the number of atoms $n_{g,4}$ in the ground state using fluorescence on a strong transition $|g, F = 4\rangle \rightarrow |b, F = 5\rangle$, where $|b\rangle = |4f_{12}^{12}(3H_5)5d_{3/2}6s^2, J = 9/2\rangle$. In order to accurately deduce the excitation probability we have to measure the number of atoms $n_{c,3}$ in $|c, F = 3\rangle$ state as well. Then we can calculate the excitation probability:

$$\eta_{4-3} = \frac{n_{c,3}}{n_{g,4} + n_{c,3}}$$

In previous work [11] we used an auxiliary 402 nm laser to transfer the atomic population from the clock level to the ground state via an intermediate level, which complicates the experimental setup. Moreover, to eliminate the second-order Zeeman, similar technique has to be applied for the other clock transition. This implies realization of 650 MHz frequency shift of the 402 nm laser radiation, that complicates the setup even more.

In this work we elaborate a method to measure population of the upper clock levels, and hence the excitation probability, without the use of 402 nm or another additional laser. We use additional clock pulses to transfer population from the upper level to the ground one. A similar technique was used, for example, in work [12]. We show that this method can be easily applied to both clock transitions mentioned above.

2. Method description

As previously, we perform excitation of a clock transition by 80 ms $\pi$-pulse. Then we directly measure the number of atoms $n_0 = n_{g,4}$ left in the ground state using a 0.1 ms pulse of a radiation resonant with $|g, F = 4\rangle \rightarrow |b, F = 5\rangle$ (solid blue arrow on fig. 1). This produces signal on a CMOS-camera proportional to $n_{g,4}$ and removes almost all the atoms in this state from the trap due to re-scattering of many photons. We confirmed experimentally that the pulse length of 0.1 ms is enough to remove atoms from the trap since we do not observe any atoms left in the ground state after this (using subsequent pulses and CMOS-camera shots). Longer pulses result in larger signal but could off-resonantly excite (and subsequently remove) atoms in $|g, F = 3\rangle$ state, that is undesirable for population readout of this state. After that we apply short (1 - 4 ms) $\pi$-pulse of the clock laser radiation to transfer atoms from the upper clock state to the ground state. This pulse length is a compromise between robustness of the population transfer against clock laser frequency fluctuation and simplicity of the experimental realization. Finally, we measure the number of atoms $n_1$ in the ground state again which is now proportional to the number of atoms $n_{c,3} = \xi_1 n_1$ excited by the spectroscopic clock pulse. We expect that $\xi_1 > 1$, since some of the initially excited atoms could spontaneously decay to the ground states before or during the transferring pulse which results in lower signal $n_1$. For the same reason, the value of $\xi_1$ depends on the timing used in the detection scheme.

The same procedure with minor adjustments can be used to measure excitation probability for $|g, F = 3, m_F = 0\rangle \rightarrow |c, F = 2, m_F = 0\rangle$ transition. Namely, directly after spectroscopic clock pulse we remove atoms from the $|g, F = 4\rangle$ state by 0.1 ms probe pulse. After this we simultaneously repump atoms from the $|g, F = 3\rangle$ to $|g, F = 4\rangle$ state using radiation resonant with $|g, F = 3\rangle \rightarrow |b, F = 4\rangle$ transition (two light blue lines on fig. 1) and detect them using 0.7 ms probe pulse. This gives us the number of atoms $n_2$ in the $|g, F = 3\rangle$ state. The long readout pulse allows us to increase signal since initial population of the $|g, F = 3\rangle$ state is 10 times smaller than that of the $|g, F = 4\rangle$ state. The rest steps are the same: population transfer using short $\pi$-pulse of the corresponding clock laser radiation and another readout with the
repump pulse that gives us the number of atoms $n_3$ that is proportional to $n_{c,2} = \xi_3 n_3$. As for $\xi_1$, we expect $\xi_3 > 1$ due to spontaneous decay before or during transferring pulse.

Figure 1. Transitions involved in the clock transition excitation measurements. Levels notations are introduce in the text.

3. Experiment
Schematic of the optical setup of the clock laser system is shown on fig. 3. As a source of laser radiation for the clock transition excitation we use Toptica DL Pro laser system stabilized to an ULE cavity [10]. Pulses resonant with $|g, F = 4, m_F = 0\rangle \rightarrow |c, F = 3, m_F = 0\rangle$ are formed by a double-passed AOM

Figure 2. Pulse sequence plots for the excitation probability measurements of (a) $|g, F = 4, m_F = 0\rangle \rightarrow |c, F = 3, m_F = 0\rangle$ and (b) $|g, F = 3, m_F = 0\rangle \rightarrow |c, F = 2, m_F = 0\rangle$ transitions.

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Figure 3. Schematic of the clock laser system which generates pulses for the spectroscopy experiments. The clock laser radiation splits into two paths and their frequencies are shifted with AOM that are driven by either SRS or DDS generators.

For spectroscopy, we change frequency of the SRS generator, while the DDS generator remains in resonance with the clock transition. The power of each generator is determined experimentally to maximize the efficiency of population transfer at the exact resonance, thus forming a $\pi$-pulse. For a typical 80 ms SRS-generated $\pi$-pulse the observed
linewidth is about 10 Hz. For the DDS$_{43(32)}$-generated $1 - 4$ ms $\pi$-pulse the linewidth is 200-1000 Hz, which is more than one order of magnitude greater than possible fluctuations of laser source frequency. This ensures that the transfer efficiency is constant during the experiment.

The preparation of atoms in two initial ground states of the two clock transitions is described in detail in [13]. Following that we can perform spectroscopy of any of the two clock transitions and calculate the excitation probability. However, first we need to find normalizing coefficients $\xi_1$ and $\xi_3$ introduced in the description above.

Following the procedure of measuring the excitation probability for the clock transition $|g, F = 4, m_F = 0\rangle \rightarrow |c, F = 3, m_F = 0\rangle$ we get signals $n_0$ and $n_1$ (fig. 4.a) from which we calculate:

$$\eta_{4-3} = \frac{n_{c,3}}{n_{c,3} + n_{g,4}} = \frac{\xi_1 n_1}{\xi_1 n_1 + n_0}$$  \hspace{1cm} (2)

The denominator in this equation is the total number of atoms before the clock laser pulse and should not depend on the frequency of SRS$_{43}$ generator. Changing this frequency, however, produces a distinct line shape in the signals $n_1$ and $n_0$. For a correct value of $\xi_1$ this line shape should disappear in the spectrum of the total number of atoms $\xi_1 n_1 + n_0$. Choosing $\xi_1$ with this method we get independence of number of atoms on SRS$_{43}$ generator frequency as seen on fig. 4.b and correct excitation probability spectrum on fig. 4.c. We note that the nonunity excitation at the line center mainly comes from finite clock level lifetime of 112 ms which is comparable with the spectroscopic $\pi$-pulse duration of 80 ms.

Similar calibration method is used for the clock transition $|g, F = 3, m_F = 0\rangle \rightarrow |c, F = 2, m_F = 0\rangle$. We get signals $n_2$ and $n_3$ (fig. 4.d) from which we calculate:

$$\eta_{3-2} = \frac{n_{c,2}}{n_{c,2} + n_{g,3}} = \frac{\xi_3 n_3}{\xi_3 n_3 + n_2}$$  \hspace{1cm} (3)

The results of a similar calibration procedure are presented on fig. 4.e, and correct probability excitation spectrum on fig. 4.f.

Presented method to measure the number of atoms excited by the spectroscopic pulse of clock laser has several advantages over previously used one. First of all, instead of using an auxiliary laser system to repump atoms, we add just one RF generator and an RF-switch. Taking into account necessity of frequency stabilization of additional laser, the experimental setup becomes significantly simpler. Second, short $\pi$-pulse with a higher power can be used to digitally stabilize power of the clock laser radiation that reaches the vacuum chamber. Power of the radiation that forms the spectroscopic pulse is very low and requires more effort to detect. Since we use high power short $\pi$-pulses here anyway, we can measure this power each cycle of the experiment. This gives us the information about long-term fluctuations of clock radiation power, and we can adjust power of SRS and DDS signals to compensate for these fluctuations.

Coefficients $\xi_1$ and $\xi_3$ which we determine by calibration may change over time and should be determined each day before measurement or in case of significant fluctuations in power of the clock laser.

4. Conclusion

We experimentally demonstrated a method of measuring the number of atoms excited by the pulse of the clock laser radiation that does not require any auxiliary laser. To transfer population from the clock level back to the ground state we used a much shorter pulse of the clock laser radiation. This method requires calibration to be performed at least once a day due to fluctuations in power of the clock laser. An example of such calibration procedure was presented together with deduced excitation probability spectra. We showed that this method is applicable to both clock transitions in thulium, that is essential for future realization of the synthetic frequency which is insensitive to the quadratic Zeeman effect.
Figure 4. a) Raw spectra of number of atoms \(n_0\) in the ground state \(|g,F = 4\rangle\) (blue dots) and \(n_1\) in the upper clock state \(|c,F = 3\rangle\) (green dots) after excitation by a 80 ms pulse. Number of atoms is presented in arbitrary units (a.u.) proportional to the signal from CMOS camera. b) spectrum of the total number of atoms \(n_0 + \xi n_1\), which is used for \(\xi\) coefficient determination. c) excitation probability spectrum calculated using equation (2) in the text. d),e),f) - similar results for the transition \(|g,F = 3, m_F = 0\rangle \rightarrow |c,F = 2, m_F = 0\rangle\). Black solid curves are approximations by the profile obtained under excitation of the transition by a rectangular pulse.

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