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Detection of the clock transition (1.14 μm) in ultra-cold thulium atoms

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Abstract. **The magnetic dipole transition between fine structure sublevels of the ground state of thulium atom $4f^{13}(^2F^o)6s^2$ has been directly excited in a cloud of ultra-cold atoms by a frequency-stabilised laser. This transition at the wavelength $\lambda = 1.14$ μm is planned to be used as the clock one in an optical frequency reference on laser-cooled thulium atoms.**

Keywords: *clock transition, frequency reference, laser cooling, magneto-optical trap, ultra-cold atoms, thulium.*

1. Introduction

Precise measurements of frequency and time are important both in fundamental science and in such applications as navigation systems and telecommunications. Creation of femtosecond optical frequency synthesisers [1] and development of new methods for cooling, capturing and exciting atomic ensembles have stimulated rapid development of optical frequency standards. Main development activity is associated with the standards on single ions and on ensembles of neutral atoms trapped in optical lattices [2]. For example, the optical frequency reference on a single aluminium ion has the systematic inaccuracy of $\sim 8.6 \times 10^{-18}$ [3], whereas for the optical frequency standard on strontium atoms in JILA [4] the total relative error is 6.4×10^{-18}. In addition, due to a large number of interrogated atoms, systems on neutral atoms possess a substantially lower level of quantum projection noise, which provides the relative frequency instability of $\sim 10^{-17}$ for the measuring duration of 100 s [4]. Note that under the International System of Units (SI), the second has been defined in terms of the frequency of the microwave transition in caesium atoms with the relative error of 10^{-16} [5], which is by more than an order of magnitude worse than errors in modern optical standards.

We suggest using the transition $4f^{13}(^2F^o)6s^2(J = 7/2) \rightarrow 4f^{13}(^2F^o)6s^2(J = 5/2)$ with the wavelength $\lambda = 1.14$ μm as a clock transition in the optical frequency reference [6] on the ensemble of thulium atoms trapped in an optical lattice. In the LS coupling approximation, the probability of magnetic dipole emission on this transition can be estimated by the formula (see, for example, [7]):

$$W = \frac{4\omega^3}{3\hbar c^3}\frac{1}{2J+1}\left(\frac{e\hbar}{2mc}\right)^2 \times \tag{1}$$

$$\frac{(L+S+J+2)(L+S-J)(S+J-L+1)(J+L-S+1)}{4(J+1)},$$

where L, S and J are the orbital, spin and total moments of electrons in the upper state, respectively. The estimated value of W yields the natural spectral line width of 1.6 Hz. This result is confirmed by the numerical modelling performed with the COWAN software [8]. The contribution into the natural spectral line width determined by the electric quadrupole transition between the levels mentioned above is negligible and is equal to 0.02 Hz.

The magnetic dipole transition seems promising as a clock transition because it occurs inside the electron 4f shell, which is shielded by the external closed shells $5s^2$ and $6s^2$. This substantially reduces the sensitivity of the transition frequency to an external permanent electric field and the differential polarisability of combination levels with $J = 7/2$ and 5/2. Correspondingly, the frequency shift due to the interaction with blackbody radiation, which presently introduces the greatest error into optical clock on strontium or ytterbium atoms [4], also reduces.

For the first time the value of fine splitting for the ground state of thulium atom was found in 1942 by the difference between frequencies of strong dipole transitions [9]. Then in 1983 the transition was directly observed in a cell with thulium vapours [10]. In the present work we present first results on laser excitation of the clock transition in thulium atoms captured in a magneto-optical trap (MOT).

2. Simulation of excitation probability

Laser excitation of a clock transition is difficult due to its small natural spectral width. The most popular method for detecting weak transitions in a cloud of laser-cooled atoms consists in observing the fall in the luminescence intensity of atoms on a strong cyclic transition (in our case it was the cool-

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ing transition with λ = 410.6 nm) when the radiation frequency of the clock laser matches with the frequency of the transition $|3\rangle \to |1\rangle$ (Fig. 1). The depths of 'dips' in luminescence signals were determined by solving optical Bloch equations [11] for the density matrix ρ of the three-level V-scheme comprising the ground state of thulium atom $|1\rangle$, upper level of the cooling transition of the MOT $|2\rangle$ and upper level of the clock transition $|3\rangle$, which interacts with two laser fields that excite the transitions $|2\rangle \leftrightarrow |1\rangle$ (cooling transition 1) and $|3\rangle \leftrightarrow |1\rangle$ (clock transition 2). In this case, the intensity of luminescence at λ = 410.6 nm is proportional to the parameter ρ_{22} that determines the population of level $|2\rangle$, and the normalised signal of luminescence is ρ_{22}/ρ_{22}^0, where ρ_{22}^0 is the population of level $|2\rangle$ in the absence of radiation of a clock laser.

As applied to our problem, the Bloch equations take the form:

$$\dot{\rho} = -\mathrm{i}\hbar[H,\rho] + \Lambda(\rho) + D(\rho) + L(\rho), \tag{2}$$

$$\rho = \begin{pmatrix} \rho_{11} & \rho_{12} & \rho_{13} \\ \rho_{21} & \rho_{22} & \rho_{23} \\ \rho_{31} & \rho_{32} & \rho_{33} \end{pmatrix}, \quad H = \begin{pmatrix} 0 & \Omega_1/2 & \Omega_2/2 \\ \Omega_1/2 & \delta_1 & 0 \\ \Omega_2/2 & 0 & \delta_2 \end{pmatrix}, \tag{3}$$

where H is the interaction Hamiltonian for the atom in two laser fields in the rotating wave approximation; $\Omega_i = \Gamma_i\sqrt{s_i/2}$ ($i = 1, 2$) are the Rabi frequencies; s_i are the saturation parameters for the corresponding transitions; δ_i are the angular frequency detunings of laser fields with respect to the frequencies of transitions $|2\rangle \to |1\rangle$ and $|3\rangle \to |1\rangle$, respectively; $\Gamma_1 = 2\pi\cdot10^7$ s^{-1} is the probability of spontaneous emission from level $|2\rangle$; and $\Gamma_2 \sim 2\pi\cdot1.6$ s^{-1} is the probability of spontaneous emission from level $|3\rangle$. The first term in the right-hand side of Eqn (2) describes the coherent part of the interaction of radiation with the atom. The second term makes allowance for processes of spontaneous emission (Lindblad superoperator) [11, 12]:

$$\Lambda(\rho) = \frac{1}{2}\sum_{i=1}^{2}([V_i\rho, V_i^+][V_i, \rho V_i^+]),$$
$$V_1 = \begin{pmatrix} 0 & \sqrt{\Gamma_1} & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \quad V_2 = \begin{pmatrix} 0 & 0 & \sqrt{\Gamma_2} \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \tag{4}$$

where V_i^+ is the Hermitian conjugate operator. The third term is responsible for the de-coherence processes related to the finite spectral width of the clock laser radiation line Δ_0 [13]. Elements of matrix D are determined by the expressions

$$D_{ij}(\rho) = \rho_{ij}\Delta_{ij}, \quad \Delta = \begin{pmatrix} 0 & 0 & \Delta_0 \\ 0 & 0 & \Delta_0 \\ \Delta_0 & \Delta_0 & 0 \end{pmatrix}. \tag{5}$$

The last term in (2) determines variation in the number of atoms in the MOT. Elements of matrix L are given by the expressions

$$L_{ij}(\rho) = \rho_{ij}O_{ij} + R_{ij},$$

$$O = \begin{pmatrix} -1/\tau_{\rm vac} & 0 & 0 \\ 0 & -\gamma_{\rm leak} - 1/\tau_{\rm vac} & 0 \\ 0 & 0 & -1/\tau_{|3\rangle} - 1/\tau_{\rm vac} \end{pmatrix}, \tag{6}$$

$$R = \begin{pmatrix} \kappa & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix},$$

where $1/\tau_{\rm vac}$ is the loss rate of atoms in the MOT due to collisions with residual gases in a vacuum chamber; $\gamma_{\rm leak}$ is the loss rate of atoms [14] due to the transitions from level $|2\rangle$ to levels neglected in the present consideration; κ is the loading rate of atoms into a trap from the Zeeman slower beam; and $1/\tau_{|3\rangle}$ is the loss rate of atoms from level $|3\rangle$ determined by the fact that being excited to level $|3\rangle$ the atom no longer experiences a confining force of the MOT and leaves the trap domain in a characteristic time $\tau_{|3\rangle}$. The characteristic temperature of atoms in the thulium MOT is 100 μK which corresponds to the root-mean-square atom velocity of ~7 cm s^{-1}. At the laser beam radius of 3 mm, the atom in level $|3\rangle$ will escape from the MOT in a time interval $\tau_{|3\rangle} \sim 50$ ms.

In a stationary case, Eqn (2) is a system of linear algebraic equations with constant coefficients, which has a cumbersome analytical solution. Figure 2a presents the temporal dependence of level $|2\rangle$ population in switching on the clock laser matched with the frequency of the clock transition that was obtained by numerical integration of equations (2). In Fig. 2b one can see the dependence of the luminescence signal on the detuning δ_2 of the clock laser radiation frequency from resonance at the laser radiation intensities corresponding to the saturation parameters $s_2 = 10^7$, 10^8 and 10^9.

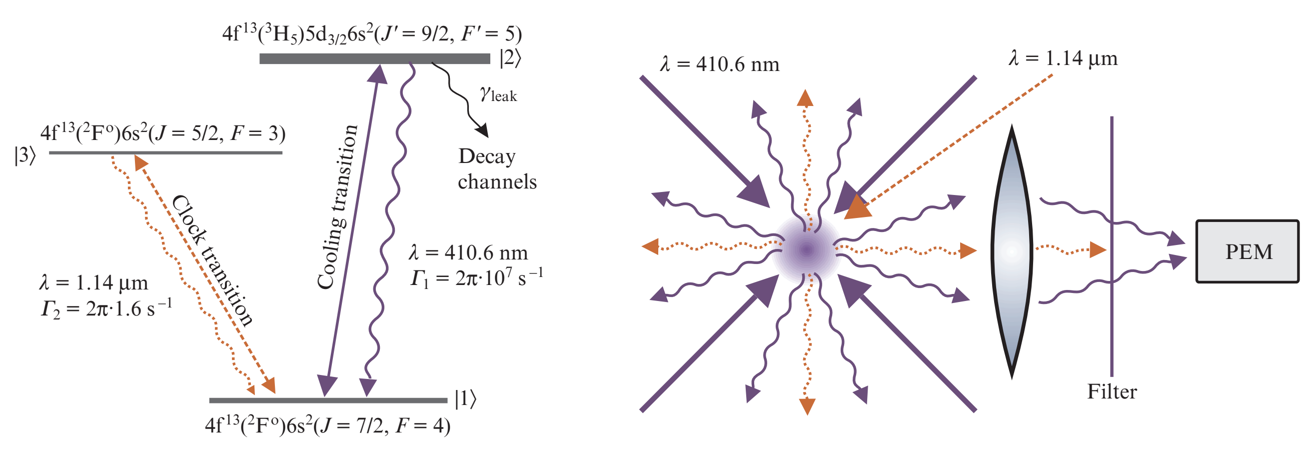

Figure 1. Energy levels of thulium atom involved in the experiment (left) and the scheme of excitation of a cloud of atoms by laser beams and detection of luminescence at λ = 410.6 nm (right).

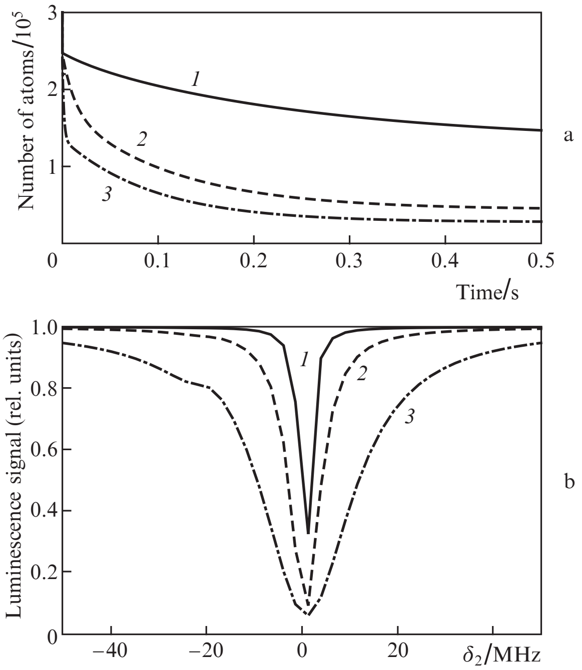

Figure 2. Results of (a) numerical simulation of level $|2\rangle$ population (time starts at the onset of clock laser radiation) and of (b) modelling of the luminescence of atoms in the MOT in the presence of radiation from clock and cooling lasers. The calculation parameters are: $s_1 = 1$, $\delta_1 = -2\Gamma_1$, $\Gamma_1 = 2\pi\cdot10^7$ s^{-1}, $\Gamma_2 = 2\pi\cdot1.6$ s^{-1}, $\tau_{vac} = 2$ s, $\gamma_{leak} = 30$ s^{-1}, $\kappa = 10^6$, $\Delta_0 = 2\pi\cdot10^3$ s^{-1}, $s_2 =$ (*1*) 10^8, (*2*) 10^9 and (*3*) 10^{10}.

The main results of modelling with the parameters accessible in experiments are as follows: the signal of luminescence falls by a factor of greater than 2.5 if the frequency of clock laser radiation is in resonance with that of clock transition; the spectral width of the dip in the luminescence signal is greater than 1 MHz; the transient period for establishing equilibrium level populations is ~300 ms; at $s_2 > 10^7$ and the spectral width of the clock laser radiation line $\Delta_0 < 2\pi\cdot10^6$ s^{-1} the latter does not affect the signal of luminescence; and the profile of the dip in luminescence is asymmetric and its centre is shifted relative to the resonance frequency depending on the power and frequency of cooling radiation in the MOT. Thus, one may conclude that successful detection of the clock transition by a variation of the luminescence signal at $\lambda = 410.6$ nm of thulium atoms in the MOT requires that the radiation of the clock laser with a spectral line width of less than 1 MHz and power of 1 mW be focused to a spot of the size of at most 1 mm.

3. Experiment

The clock transition in thulium atom was detected by recording the luminescence signal at $\lambda = 410.6$ nm of cold atoms in the MOT with the resonance probe radiation at $\lambda = 1.14$ μm. A schematic of the experimental setup is shown in Fig. 3. The Zeeman slowing [15] and laser cooling of thulium atoms have been realised using the strong transition $4f^{13}(^2F^o)6s^2(J = 7/2, F = 4) \rightarrow 4f^{12}(^3H_5)5d_{3/2}6s^2(J' = 9/2, F' = 5)$ at $\lambda = 410.6$ nm with the natural width of 10 MHz, which corresponds to the Doppler limit of the atom temperature of 240 μK. The classical MOT scheme was used [16]. A three-dimensional optical molasses was produced by three pairs of mutually orthogonal circularly polarised counterpropagating laser beams with frequencies shifted to the red spectral region from resonance by a frequency of the order of the transition natural width. A quadrupole magnetic field was produced by a pair of coils in the anti-Helmholtz configuration. The cooling radiation at $\lambda = 410.6$ nm was produced by doubling the radiation frequency of a single-mode Ti:sapphire laser (Coherent MBR-110/MBD-200) pumped by a diode laser (Coherent Verdi G-12). The frequency of cooling radiation was stabilised by using a signal of saturated absorption in thulium vapours inside the cell. The MOT for thulium atoms was described in more details in [14]. The characteristic temperature of a cloud of thulium atoms, at which the clock transition was sought and detected, was 100 μK. The characteristic cloud dimension was 200 μm and the number of atoms in the cloud was 10^6. The temperature mentioned is noticeably lower than the Doppler limit due to the sub-Doppler mechanism of cooling [17]. A signal of luminescence from a cloud of cold atoms at $\lambda = 410.6$ nm was detected by a PEM with the corresponding light filter (Fig. 1).

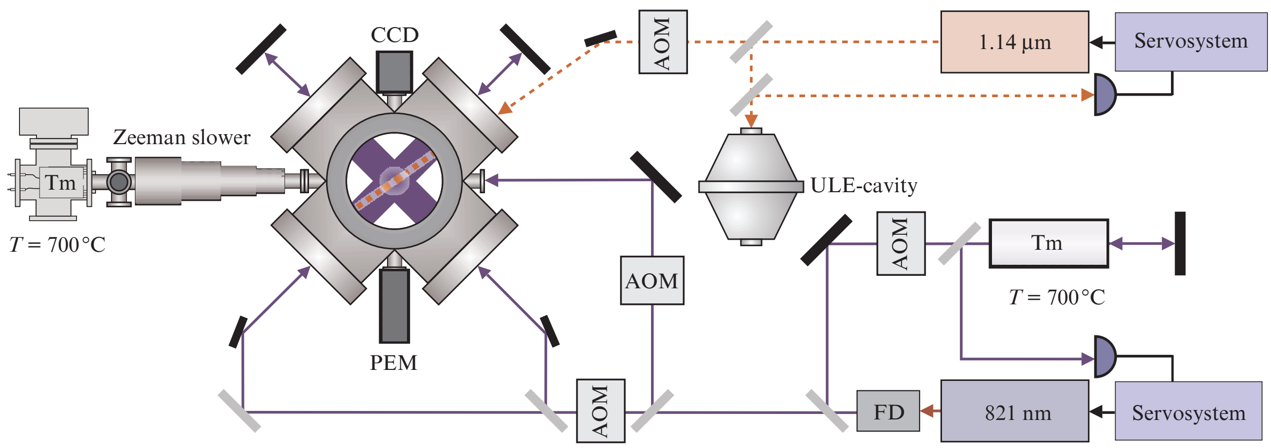

Figure 3. Schematic diagram of the experimental setup (solid and dashed lines correspond to the cooling and clock beams, respectively): (AOM) acousto-optical modulator; (FD) frequency doubler; (CCD) camera; the vertical beam of cooling radiation is not shown (its direction is normal to the plane of the figure).

The radiation frequency of the laser used for exciting the clock transition was stabilised using the Pound–Drever–Hall locking technique [18] by an external high-Q ULE-cavity [19] near the frequency of transition under study that was taken from [20] and was equal to 262.955 THz. The frequency was adjusted by a wavelength metre (Angstrom WS-5) that was preliminarily calibrated with an LGN-109 He–Ne laser having the accuracy of 3 GHz. Then the laser frequency was stabilised by a nearest mode of the ULE-cavity using a fast-response feedback, which provided narrowing of the spectrum to ~100 Hz and the long-term stability of frequency at a level of 1 MHz. The frequency of radiation interacting with a cloud of atoms was scanned by an acousto-optical modulator.

Figure 4 presents a signal of luminescence in the MOT while scanning the radiation frequency of the clock laser near resonance (the rate of scanning is 100 kHz s^{-1}) at the radiation power of 20 μW–1.1 mW (the corresponding saturation parameters are $s_2 = 10^8 – 5 \times 10^9$). As expected in modelling, at a lower power the depth of the luminescence signal dip falls with simultaneous narrowing of the spectral line. The corresponding dependence of the HWHM line contour approximated by a Lorentzian profile is shown in Fig. 4b. Extrapolation to a zero power of the exciting field yields the line width of ~1 MHz, which approximately corresponds to the Doppler line width of laser-cooled atoms in a cloud and to Zeeman splitting of corresponding levels.

The experimental results obtained qualitatively agree with those of numerical simulation. Quantitatively a distinction between measured and theoretical spectral widths may be related to the imperfect model employed (it is one-dimensional and makes no allowance for magnetic sub-levels) and overestimated probability of the clock transition (1).

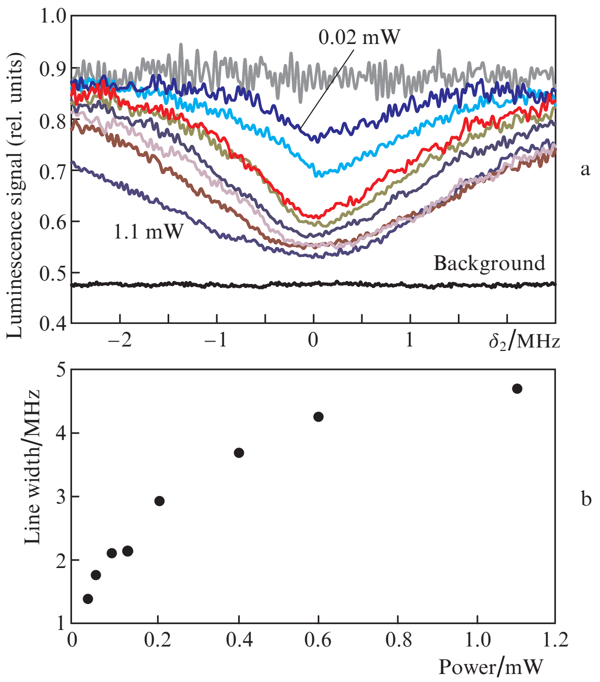

Figure 4. (a) Detected luminescence of atoms in the MOT vs. frequency detuning of the clock laser from resonance at the clock laser power varying from zero (top curve) to 1.1 mW (bottom curve) and (b) dependence of the width of the resonance line detected in the MOT on the power of the clock laser.

4. Conclusions

For the first time the transition between the components of the fine structure of the ground state in cold thulium atoms has been directly excited by a laser at a wavelength of 1.14 μm, which is a promising candidate for creating a frequency reference. At the clock laser power of 20 μW the spectral width of the detected transition line was 1 MHz which is presently explained by the power broadening and broadening due to the Doppler and Zeeman effects. In near future, we plan to study the clock transition in thulium atoms trapped in an optical lattice. Due to the Lamb–Dicke effect [21] the lattice may help escape the Doppler broadening and spectrally resolve Zeeman components. In addition, in an optical lattice we plan to experimentally determine the lifetime of the upper level of the clock transition.

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