Methods for determining the polarisability of the fine structure levels in the ground state of the thulium atom

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Abstract. We have calculated the scalar and tensor parts of the polarisabilities of the fine-structure sublevels J = 7/2 and 5/2 of the thulium atom ground state. The static polarisabilities are in a good agreement with the values known from the literature. We also present experimental techniques for measuring the dynamic scalar polarisabilities of these levels at a wavelength of 532 nm. The measured values agree with our calculations within the errors. The results of this work can be used for designing an optical frequency standard based on laser-cooled thulium atoms.

Keywords: optical frequency standard, laser cooling, dynamic polarisability, thulium.

1. Introduction

The relative accuracy of modern optical frequency standards based on individual ions and ensembles of neutral atoms has achieved a low 10^{-18} level [1–4]. A significant and sometimes dominant contribution to the error of these standards is introduced by the frequency shift of the metrological transition due to the interaction of the atom or ion with the thermal radiation of surrounding bodies. For example, at room temperature the shift of the clock transition in the strontium atom at a wavelength of 698 nm caused by the black body radiation amounts to 2 Hz or 5×10^{-15} rel. units [3]. To achieve the frequency error at a level of 10^{-18} , two approaches are used: i) the high-accuracy determination of the differential static polarisability of the clock levels and the temperature of

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Received 17 February 2017 *Kvantovaya Elektronika* **47** (5) 479–483 (2017) Translated by V.L. Derbov the environment, which allows high-accuracy calculation of the shift [5, 6]; and ii) placing the atom ensemble in a cryogenic environment that allows the shift reduction [7]. Both approaches are difficult to implement.

The search for atomic and ion systems in which the metrological transition possesses a minimal sensitivity to the thermal radiation remains an important problem [8, 9]. Promising approaches still not implemented experimentally are the spectroscopy of optical transitions in multiply charged ions [10] and the excitation of optical isomeric nuclear transitions in thorium [11]. The polarisability of such systems is extremely small, which provides a weak sensitivity to the black body radiation.

The progress in the laser cooling of lanthanides [12–14] opened new interesting possibilities for the development of optical frequency standards. In lanthanides with an open inner 4f electron shell the fine structure sublevels of the ground state (Fig. 1a) are coupled by the magnetic dipole/electric quadrupole transition, which is shielded from the external electric fields by the closed outer electronic shells $5s^2$, $5p^6$ and $6s^2$. Such intrashell transitions are studied well in the Er³⁺ and Tm³⁺ ions embedded in solid-state matrices and are widely used in IR lasers [15, 16]. Apart from the close values of static polarisabilities of the ground state fine structure sublevels, these levels also possess large lifetimes. Thus, in thulium the lifetime of the metastable state $|4f^{13}6s^2(J=5/2)\rangle$ is nearly 100 ms [17, 18], while in the erbium atom the level $|4f^{12}6s^2(J=4)\rangle$ decays during 6 hours [19]. The combination of the shielding effect and small natural spectral width of the intrashell transitions provides the possibility of using them in optical frequency standards.

The intrashell magnetic dipole transition $|4f^{13}6s^2(J=7/2)\rangle \rightarrow |4f^{13}6s^2(J=5/2)\rangle$ in the thulium atom was previously proposed as a candidate for optical clocks [18] and thoroughly analysed in Ref. [17]. It is known that one of the central problems in the construction of a frequency standard is the study of static and dynamic polarisabilities of the clock levels, which allows the calculation of the frequency shift of the clock transition induced by the black body radiation and the determination of the magic wavelength [20]. Note that for the element with an open inner electron shell the calculations of the wave functions and oscillator strengths are much more difficult than in alkali and alkali-earth atoms. The accuracy check of the calculations is possible either directly by comparison with the experiment, or indirectly by comparison with the results, calculated using other methods [21–23].

In the present work we carried out the analysis of the data existing in the literature on the static polarisability of the clock levels in the thulium atom and compared them with our results, obtained previously [17]. We also implemented the



Figure 1. (a) Energy levels of the thulium atom used in the laser cooling cycle and in the clock transition spectroscopy. The numbers indicate the following transitions: (1) $\lambda = 410.6$ nm, $\gamma = 10$ MHz (primary cooling), (2) $\lambda = 530.7$ nm, $\gamma = 350$ kHz (secondary cooling), (3) $\lambda = 1.14$ µm, $\gamma \sim 1$ Hz (the clock transition). (b) Schematic of a 1D dipole trap for thulium atoms at a wavelength of 532 nm (beam 4) with the recording of atomic luminescence in the probe laser beam at 410.6 nm (beam 1). The acoustooptical modulator (AOM) is driven by a 40 MHz oscillator with the clock transition the radiation at a wavelength of 1.14 µm (beam 3) is used; CCD is a photo camera.

experimental methods that allow the estimation of the dynamic polarisability of the lower (J = 7/2) and upper (J = 5/2) clock levels and the comparison with the calculated data. This increases the reliability of prognosing the magic wavelength values to be used in further experiments, and confirms the estimate of the clock transition shift by the black body radiation.

2. Static polarisability data analysis of the clock levels J = 7/2 and 5/2 in thulium atom

To calculate the magic wavelength of an optical dipole trap and the frequency shift of the clock transition due to the interaction with thermal radiation, it is necessary to determine the static and dynamic polarisabilities of the clock levels. To solve this problem, modern computational methods are usually applied (see [21, 23] and references therein), together with the high-precision measurements of the static polarisability (see, e.g., the experiments with strontium and ytterbium atoms [24, 25]).

To calculate the static and dynamic polarisability of thulium atoms, we computed the contribution from the discrete and continuous spectrum. The contribution of the discrete spectrum α_{discr} to the polarisability of the clock levels J =7/2, 5/2 was calculated in the second order of the perturbation theory using the software package COWAN [26]. This algorithm is based on the Hartree-Fock calculation of single-electron wave functions in the central field approximation, followed by the variation of free parameters to fit the calculated energy levels to the experimental data. The obtained configurations were used to calculate the transition matrix elements. The contribution of the continuous spectrum α_{cont} was determined using the photoionisation cross-section spectrum of the thulium atom clock levels, calculated by means of the software package FAC [27]. Since the contribution of the continuous spectrum to the scalar polarisability appeared to be much smaller than that of the discrete spectrum (Table 1), it was not taken into account in the calculations of the tensor polarisability. The obtained values of the static polarisability for the lower, J = 7/2, and upper, J = 5/2, clock levels of the thulium atom are presented in Table 1.

The methods used in the calculations by other groups differed from ours. In Ref. [21] the calculation was based on the separation of the contributions to the polarisability from the closed inner shells (the core), the electrons of 4f shell, calculated in the random-phase approximation, and the valence 6s² electrons, and was implemented using the CI + MBPT method ('interaction + many-body perturbation theory' configuration). The authors of [21] demonstrated the legitimacy of including the 4f electrons into the core, which, in fact, was implemented in our calculations. In Ref. [28] they used the approach based on the theory of linear response of the atomic polarisability in the self-consistentfield approximation with the construction of Green's functions. The calculations in Ref. [29] were based on the timedependent density functional method. In Ref. [30] the method of configuration interaction in combination with a few different methods of taking the atomic orbitals into account was used.

In all theoretical papers, except ours, the fine interaction was not taken into account. However, the value of the scalar polarisability of the electronic term α_L^s in the LS-coupling approximation for the considered level coincides with α_J^s and does not depend on the total electronic angular momentum J [31]. As seen from the values of the polarisability of the levels J = 7/2, 5/2 obtained by us (and the arguments in Ref. [19]), such approximation appears to be quite adequate for the considered sublevels of the ground state fine structure. This is also confirmed by the coincidence of the rate of the magnetic dipole transition between these levels, calculated in the LS-coupling approximation [32], with more precise calculations and measurements [17]. Note also that in Ref. [21] only the contribution

Table 1. Contributions of the discrete (α_{discr}) and continuous (α_{cont}) spectrum to the static scalar (α^{s}) and tensor (α^{t}) polarisabilities of the clock levels J = 5/2, 7/2, calculated by our group and adopted from the literature. The values are presented in atomic units, 1 a.u. = $4\pi\varepsilon_0\alpha_0^3 = 1.65 \times 10^{-41}$ J (V m⁻²)⁻².

Polarisability	Clock levels, J	$\alpha_{ m discr}$	$\alpha_{\rm cont}$	$\alpha_{\rm discr} + \alpha_{\rm cont}$	Theory	Experiment
α^{s}	7/2	138.06	8	146	144.3 [21], 147 [28]	_
	5/2	138.003	10	148	161 [29], 152 [30]	-
α^{t}	7/2	-2.7	0	-2.7	-2.27 [29]	-2.7(2) [33]
	5/2	-2.3	0	-2.3		-

of discrete spectrum was considered, while in Refs [28-30] the contributions of both the discrete and continuous spectrum were taken into account.

The calculated values are in a good agreement within the inaccuracy less than 10%, which is a good result, keeping in mind the difficulty of calculations for the atoms having open inner shells. One can conclude that the method used by us in Ref. [17] allows the calculation of the static polarisability of the clock levels in thulium atom with the inaccuracy not exceeding 10%. Since the calculation of the dynamic polarisability is based on the same initial parameters as those, used in the calculation of the static polarisability, one can expect a similar inaccuracy in the calculation of the dynamic polarisability.

The agreement with the experiment is apparently a more valuable confirmation of validity of the calculation results. As far as we know, to date only the tensor polarisability of the thulium atom ground state (J = 7/2) is measured [33] with the result that agrees with ours calculations within 10%. In the existing configuration of our experimental setup, the measurement of the static polarisability of the thulium atom ground state is impossible, since it requires the installation of a precision capacitor in the vacuum chamber in the region of magnetooptical trap (MOT) [25, 34]. However, it is possible to load the atoms into the dipole trap at the wavelength 532 nm and to excite the clock transition between the levels J = 7/2 and J = 5/2, which allows the measurement of the dynamic polarisability of these levels at 532 nm and the comparison of the obtained results with those of the calculations.

3. Method of measuring the dynamic polarisability in the optical dipole trap

After reloading of thulium atoms from the MOT into the 1D optical dipole trap at 532 nm [35] we can experimentally estimate the dynamic polarisability of both the lower, J = 7/2, and the upper, J = 5/2, clock levels. Since at this stage of the experiment the pumping of atoms to certain magnetic sublevels is not implemented, it is possible to measure only the scalar polarisability α^{s} of the atom.

Below we present the results of measuring the dynamic polarisability of the clock levels J = 7/2, 5/2 at a wavelength of 532 nm, using two different methods. In the first method, the parametric resonances in the dipole trap are detected and their frequencies are measured. In the second methods, the frequency shift of the magnetic dipole transition at a wavelength of 1.14 µm is measured at a different dipole trap radiation power (see Fig. 1b).

In both experiments, the thulium atoms are preliminarily cooled and trapped first in the primary MOT [15] and then in the secondary one [35]. As a result, the cloud of ~10⁶ atoms at the temperature of 20 μ K is formed. During the entire cooling cycle, the dipole trap is switched on, which does not affect the operation of the MOT. The dipole trap is formed by the focused radiation of a single-frequency continuous-wave laser, generating at 532 m and providing up to 8 W of radiation power. The beam waist radius in the MOT region is $w_0 = 60 \pm 10 \,\mu\text{m}$ at the 1/e² intensity level; this error includes the astigmatism from beam injection into the vacuum chamber, as well as the error related to the inaccuracy of matching the beam waist with the cloud of atoms in the MOT. The error of the w_0 measurement is dominant in the measurements described below.

3.1. Measuring the dynamic polarisability of the ground state J = 7/2 at 532 nm

The dynamic polarisability of the ground state J = 7/2 in the thulium atom can be found by measuring the spectrum of vibrational resonances in the dipole trap. The vibration eigenfrequencies in the radial and axial direction in the approximation of harmonic potential [36] are

$$f_{\rm r} = \frac{2}{\pi w_0^2} \sqrt{\frac{a_0^3 \alpha_{J=7/2}^8 P}{cm_0}}, \ f_z = \frac{\lambda}{\sqrt{2} \pi w_0} f_{\rm r}, \tag{1}$$

where *P* is the radiation power; $\alpha_{J=7/2}^{s}$ is the ground state polarisability in atomic units; a_0 is the Bohr radius; *c* is the speed of light; and w_0 is the beam waist radius.

According to Ref. [37], the harmonic modulation of the trap potential depth at the frequency 2f/n, where f is one of the vibration eigenfrequencies and n is an integer, will parametrically excite the oscillations of atoms, giving rise to heating and reducing the number of atoms in the trap. In our dipole trap, we managed to detect only transverse resonances, since for the longitudinal mode the potential is too flat, the corresponding frequencies being about 1 Hz.

After reloading the atoms into the dipole trap, we switch on the harmonic modulation of the trap depth to parametrically excite the oscillations. This is implemented via the amplitude modulation at the frequency $f_{\rm m} \approx 100-1000$ Hz of the radio frequency signal of the oscillator that drives the acoustooptical modulator (Fig. 1b). In 50–100 ms the number of trapped atoms is measured by the signal of atomic luminescence, excited by a beam of probe resonance radiation at the wavelength 410.6 nm with the duration 1 ms.

Figure 2 presents the measured dependences of the number of atoms on the modulation frequency $f_{\rm m}$ for the dipole trap radiation power of 2, 3, and 4 W and the modulation depth of 50%. In the inset of Fig. 2, the positions of resonance frequencies $f_{\rm r}$ and $2f_{\rm r}$ are plotted versus \sqrt{P} . As expected from Eqn (1), the dependences are close to linear. Thus, with $w_0 = 60^{+10}_{-10} \,\mu{\rm m}$ the dynamic polarisability is $\alpha^s_{J=7/2} = 420^{+400}_{-200}$ a.u. The contribution of the error of the parametric resonances frequency determination to the error of $\alpha^s_{J=7/2}$ amounts to 60 a.u. Despite the low accuracy, the obtained

Figure 2. Number of atoms in the dipole trap as a function of the modulation frequency at a wavelength of 532 nm and the radiation power P = 2 W (circles), 3 W (triangles) and 4 W (squares). Since the dependence is complex, the positions of the resonances were found by means of fitting the parabolic functions (solid curves) to their centres, which provides a sufficient accuracy. The inset shows the positions of the parametric resonances versus \sqrt{P} .



result agrees well with the result of calculation, $\alpha_{J=7/2}^{s} = 580 \text{ a.u. } [17].$

3.2. Measuring the dynamic polarizability of the upper clock level J = 5/2 at the wavelength 532 nm

If the polarisability of the lower clock level J = 7/2 is known, the scalar polarisability of the upper clock level J = 5/2 can be found from the frequency change Δf_{trans} of the clock transition $|J = 7/2, F = 4\rangle \rightarrow |J = 5/2, F = 3\rangle$ (1.14 µm), depending on the dipole trap power according to the formula

$$h\Delta f_{\rm trans} = U_{J=5/2}^{\rm dip} - U_{J=7/2}^{\rm dip} \approx \frac{4(\alpha_{J=7/2}^{\rm s} - \alpha_{J=5/2}^{\rm s})a_0^3 P}{cw_0^2},$$
(2)

where $\alpha_{J=7/2}^{s}$ and $\alpha_{J=5/2}^{s}$ are the dynamic polarisabilities of the lower and upper clock levels at the wavelength 532 nm.

To implement this method, we used the following experimental scheme. In 20 ms after the dipole trap reloading, the pulse of the probe laser radiation at the wavelength 1.14 μ m was switched on, which excited a part of the atoms to the upper clock level. The pulse duration was 50 ms, the power was 30 μ W. After the excitation, the residual population of the level $|J = 7/2, F = 4\rangle$ was measured.

Figure 3 presents the dependence of the centre position of the clock transition line on the dipole trap power. An example of the detected spectral contour of the magnetic dipole transition and its approximation by the Gaussian function (due to the inhomogeneous broadening of the transition) are shown in the inset. From the linear approximation of the obtained dependence, the slope was found to be $df_{trans}/dP = 0.28(2)$ MHz W⁻¹, which, according to Eqn (2) corresponds to the differential polarisability $\alpha_{J=7/2}^{s} - \alpha_{J=5/2}^{s} = 85^{+30}_{-25}$ a.u. at $w_0 = 60^{+10}_{-10}$ µm. Using the scalar polarisability of the lower clock level $\alpha_{J=7/2}^{s} = 420^{+400}_{-200}$ a.u., we get the scalar polarisability of the upper clock level $\alpha_{J=5/2}^{s} = 335^{+400}_{-200}$ a.u. that agrees with the value $\alpha_{J=5/2}^{s} = 215$ a.u., calculated in [17].

The proposed methods allow the determination of the dynamic polarisabilities of the clock levels in the thulium atom, but their accuracy is limited, mainly by the error in the measurement of the beam waist radius of the optical dipole trap. Nevertheless, this accuracy is enough to confirm the calculations qualitatively, which is important for finding the magic wavelength. For a rough localisation of the magic



Figure 3. Dependence of the $1.14 \,\mu\text{m}$ magnetic dipole transition centre frequency on the optical trap radiation power *P*. The inset shows an example of the measured line shape (circles) and its approximation by the Gaussian function (solid curve).

wavelength and the proof of its existence, it is often sufficient to know which of the levels has a greater polarisability in the given spectral range. The answer to this question unambiguously follows from the dependence presented in Fig. 3. The exact determination of the magic wavelength is usually implemented by the experimental search near the expected value [38].

4. Conclusions

We present the methods for deducing the dynamic polarisability of the clock levels J = 7/2, 5/2 in the thulium atom and the corresponding results. It is shown that the measurements at a wavelength of 532 nm give the values of 420^{+400}_{-200} a.u. and 335^{+400}_{-200} a.u., which agree with our theoretical calculations (580 a.u. and 215 a.u.) [17] within the error. The large error is due to the inaccuracy of determination of the light beam waist radius in the dipole trap.

We have compared the calculations of static polarisabilities of the thulium atom fine structure sublevels, carried out using different methods, including the calculations performed by our group. It is shown that the difference between the results does not exceed 10%, which is satisfactory, keeping in mind the complexity of calculations for the elements with open inner shells.

Acknowledgements. The work was supported by the Russian Foundation for Basic Research (Grant Nos 15-02-05324, 16-29-11723) and the Presidium of the Russian Academy of Sciences (Electrophysics of High-Power Pulsed Systems Programme No.19).

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