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# **Doppler cooling of thorium ions in a multisectional linear Paul trap**

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**Abstract**. Doppler cooling of thorium ions is described. The technique is to be applied for cooling of ions in a multisectional linear Paul trap for precise spectroscopy of ions and eventually detecting an isomeric nuclear transition.

#### 1. Introduction

Today the second is defined according to the transition between the two hyperfine levels of the ground state of the caesium-133, and the best atomic clocks based on <sup>133</sup>Cs reach the uncertainty  $2 \times 10^{-16}$  [1]. However, recent achievements in laser spectroscopy paved the way towards more precise and stable frequency standards [2], which lead to a significant breakthrough in both applied and fundamental physics, e.g. problems of the theory of relativity or constant variance.

One of the ways to increase the uncertainty of a standard is to use higher frequency of oscillations, i.e. move from microwave to optical frequency. It has been done for the neutral atomic ensembles and single ions, with relative uncertainty being as low as  $1.5 \times 10^{-18}$  [3]. Further progress, however, is complicated by systematic frequency shifts due to the sensitivity of electrons to external fields. New approach is therefore necessary.

One of the possible solutions is to use a unique nuclear isomeric transition in thorium-229 with an excitation energy similar to optical frequencies of electron transitions. Since nucleus has low polarizability and is shielded by electron shell, nuclear transitions are far less sensitive to external perturbations, significantly increasing the precision of measurements and making it possible to reach the next level of relative uncertainty  $10^{-19}$  or even lower [4]. The energy of the transition corresponds

to the wavelength  $\approx 170$  nm, which is in the range of such an important tool for frequency standard development as frequency combs.

Thorium-229 is a product of uranium-233 alpha-decay with a half-life 7880 years. Early research [5] of <sup>229</sup>Th nuclear energy levels using gamma-spectroscopy showed that two rotational bands (main and isomer) of the nucleus are close to each other. The difference was estimated to be  $1 \pm 4$  eV at first [6], and then  $3.5 \pm 1.0$  eV later [7]. Further research assisted with understanding the probability of the transition and different decay channels [8]. For example, recent experiment on measuring the lifetime of the upper level of the transition through the electron conversion decay channel (7±1 µs) [9] is in good agreement with theory (10 µs) [10].

Although laser cooling of thorium ions is an essential step towards spectroscopy of the nuclear isomeric level, the excitation of this level is a challenge. The direct excitation has not yet been demonstrated due to a large uncertainty of the energy and long lifetime of the excited state. Therefore, it may be more feasible to use a method of reverse electron bridge [11]. This mechanism, if achievable, will make use of well-known electronic transitions and will have a higher cross-section compared to the direct excitation.

Despite many efforts, precise spectroscopy of the isomeric level has not been done yet. Nevertheless, we are now at the important stage of research towards thorium frequency standard. Our group has recently joined this research as well by developing a multisectional linear Paul trap for thorium ions [12]. Now we have plans to make use of Doppler cooling for spectroscopy of ions and researching ways of excitation of the isomeric nuclear level. In this work our plans of Doppler cooling of  $^{229}$ Th<sup>3+</sup> ions together with  $^{232}$ Th<sup>3+</sup> are presented.

#### 2. Level structure of ions

In order to successfully laser cool ions, precise knowledge of their energy structure is necessary. Spectroscopy of energy levels suitable for laser cooling was performed by the group from the USA [13]. Energy level diagrams for both ions are presented on Fig 1.



Figure 1. Energy levels of both <sup>232</sup>Th<sup>3+</sup> and <sup>229</sup>Th<sup>3+</sup> (hyperfine sublevels omitted) used for cooling

Since nuclear spin of the isotope  ${}^{232}\text{Th}^{3+}$  is zero, it has no hyperfine structure. Transition  $5F_{5/2} \rightarrow 6D_{3/2}$  has a wavelength 1088 nm, and a natural linewidth of the upper level  $\gamma_{3/2} = 145$  kHz, according to calculations [14]. Level  $6D_{3/2}$  has a linewidth  $\gamma_{3/2} = 234$  kHz, and decays to the two fine sublevels of the ground state  $5F_{5/2}$  and  $5F_{7/2}$  with a branching ratio 1:8. Corresponding wavelengths of the transitions are 690 nm and 984 nm.

Unlike  ${}^{232}$ Th ${}^{3+}$  isotope  ${}^{229}$ Th ${}^{3+}$  has nuclear spin I = 5/2, which leads to a rather complex hyperfine structure of energy levels. It can be described as following [15]:

$$\delta E_i = \Delta_i + \frac{K_i}{2} A_i + \frac{3K_i(K_i+1)/2 - 2I(I+1)J_i(J_i+1)}{4I(2I-1)J_i(2J_i-1)} B_i,$$

Where  $K_i = F_i (F_i + 1) - I (I + 1) - J_i (J_i + 1)$ , *I*- nuclear spin,  $J_i$  - total electron momentum, *F*- total atomic momentum,  $\delta E_i$  - difference between level of <sup>229</sup>Th<sup>3+</sup> and the same level of <sup>232</sup>Th<sup>3+</sup>, *i* - index of the corresponding level *e* (excited) or *g* (ground), *A*, *B*,  $\Delta$  - coefficients that are not dependent on *F*. All coefficients *A*, *B* and  $\Delta_e$  -  $\Delta_g$  of all transitions were measured in the work [13].

## 3. Doppler cooling

The choice of isotope <sup>232</sup>Th<sup>3+</sup> was primary dictated by its simple level structure, which facilitates Doppler cooling of these ions compared to <sup>229</sup>Th<sup>3+</sup>, and can later assist with cooling of <sup>229</sup>Th<sup>3+</sup> through the mechanism of sympathetic cooling [12]. Closed transition 1088 nm and three-level lambda system 690 nm – 984 nm can both be used for Doppler cooling. Doppler limit of temperature in this case can be estimated as  $T_D = \hbar \gamma/(2k_B) \approx 1 \mu K$ , which is low enough to form ion Coulomb crystals [16].



Figure 2. The first stage of Doppler cooling of <sup>229</sup>Th<sup>3+</sup> ions using 1088 nm transition.

The hyperfine structure of the <sup>229</sup>Th<sup>3+</sup> isotope complicates such process. In particular, for the transition 1088 ground and excited states have 4 and 6 hyperfine sublevels respectively, with the distance between them being  $100 \div 1000$  MHz. Since an ion can decay from particular upper sublevel *F* to any of the sublevels *F*,  $F \pm 1$  of the ground state, there is no closed two-level system. Addressing many transitions is therefore necessary to avoid any undesirable dark states. It can be done with the use of electro-optical modulators. However, it will no longer be possible to select the desired detuning of laser frequency in this case, which forces us to use more convenient cooling scheme as a second stage. It is worth mentioning that during the first stage described above, the driving transitions optically pump ions to the certain ground hyperfine sublevel  $5F_{5/2}$  (F = 1) (Fig. 2).



Figure 3. The second stage of Doppler cooling of  $^{229}$ Th<sup>3+</sup> ions using 984 nm  $\leftrightarrow$  690 nm lambda system.

This particular state is chosen so that the three-level lambda scheme  $5F_{5/2}(F = 1) \leftrightarrow 5D_{5/2}(F = 0) \leftrightarrow 5F_{7/2}(F = 1)$  can be used for the second stage cooling (Fig. 3). It is a closed system because the transition  $F = 0 \leftrightarrow F = 0$  is forbidden (left branch on Fig. 3). Since only two frequency detunings are to be controlled in this case, efficient Doppler cooling is feasible. Still, non-resonance excitation to another sublevels remains possible, which makes driving repumper transitions necessary.

The two described stages of Doppler cooling of ions can be used simultaneously. At first, when the temperature of ions is high, cooling on the 1088 nm works inefficiently due to high possibility of non-resonance excitation. Still, on average the temperature is decreasing and ions are getting pumped to the  $5F_{5/2}$  (F = 1) sublevel. When the temperature becomes <10 K, the Doppler broadening of all the sublevels becomes <200 MHz, thus allowing to address certain hyperfine sublevels separately with low probability of non-resonance excitation. The second stage cooling becomes relevant at this point, while the 1088 nm transitions remains working as a repumper.

# 4. Conclusion

We have created a multisectional linear Paul trap for thorium ions and are planning to apply laser cooling in order to achieve more precise spectroscopy of ions, the isomeric nuclear transition in particular. The technique of laser cooling is in accordance with that of a group in the USA, and can be used to cool ions to crystallization.

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