JEDRSKA URA

JAN JURKOVIČ

Fakulteta za matematiko in fiziko
Univerza v Ljubljani

Natančnost časa postaja vse bolj uporabna in pomembna, zato se rojevajo novi načini merjenja časa. Do danes najbolj natančnih meritev časa in frekvence pridemo z atomskimi urami. Te so že tako natančne, da če bi delovale več deset milijard let, bi imele dano napako manjšo od ene sekunde. Kljub tako veliki natančnosti bi jih v prihodnosti lahko prekosile nove ure, imenovane jedrske ure. Medtem ko atomske ure delujejo na podlagi vzbujanja elektronov atomov v višja stanja, bodo jedrske ure delovale na podlagi vzbujanja atomskega jedra. To je zelo obetavno, saj na atomsko jedro okolica ne vpliva tako močno kot na elektrone v atomski ovojnici. Za atomsko uro lahko uporabimo atome različnih elementov, jedrska ura pa bi lahko delovala le z enim izmed vseh 176.000 znanih jedrskih stanj. To stanje je prvo vzbujeno jedrsko stanje ^{229}Th. Kljub temu je do prve delujoče jedrske ure potrebno raziskati še veliko nejasnosti.

NUCLEAR CLOCK

As time precision becomes more and more useful and important, new ways of time measurement have been born. Today's most precise time and frequency measurements are performed with atomic clocks. The most precise ones work so well, that if they worked for tens of billions of years, they would be less than a second off. However, in the future they could be even outperformed by the so-called nuclear clocks. While the atomic clock is based on the excitation of the electrons in an atom, the nuclear clock would work with excitation of an atomic nucleus, which is promising, because the atomic nucleus is less affected by the effects of the surroundings like the electrons in the atomic shell. For an atomic clock, atoms of different elements can be used, however the nuclear clock could be based only on one of the known 176.000 nuclear levels on the first excited nuclear state of ^{229}Th. But to actually build a working nuclear clock there is still so much left to discover.

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1. Introduction

Accuracy is a very respected virtue. To be at the exact place in the exact time, is now a lot easier with the evolution of the technology. And the basis of all this is a precise definition for the unit of time - the second.
The latest definition of a second is the duration of 9192631770 periods of the radiation, corresponding to the transition between two energy levels of the ground state of the caesium 133 atom. Why is it defined this way?

2. Atomic clock

Well, this definition is the result of the most precise clock known today - the atomic clock.
Here, the before mentioned caesium comes into play. Liquid caesium is heated in an oven and vaporised. The created gas is then collimated into a beam that consists of caesium atoms with two different energy levels. There is a very small difference between the energies of these states (3.78×10^{-5} eV).

In atomic physics, fine structure (fine energy levels) refers to small shifts in energy levels of atoms. It is the result of energy changes produced by electron spin - orbit coupling (interaction forces from orbital and spin motion of electrons). Because the electron spin can be oriented along or opposite to magnetic field, two energy states occur. This effect is called Zeeman effect. So the hyperfine structure refers to even smaller shifts. These occur due to the energy of the nuclear magnetic dipole moment in the magnetic field generated by the electrons.

The reason for this small energy gap can be explained with laws of quantum mechanics. Caesium has 55 electrons and 54 of them are in very stable orbitals. Then there is the 55th electron that is in the 6s orbital. It does not interact much with other electrons, but it does interact with the atomic nucleus, with the nuclear spin to be more exact. The nuclear spin is responsible for the magnetic field, which is responsible for the interaction. Because of two different options for spin of the valence electron, there are also two different energy states - the two hyperfine ground states of caesium. The collimated beam of these two states then exits the oven and enters the magnetic field of two magnets. Because the atoms are in two different energy states, they are separated into two beams. The quantum numbers n, l, m_l and m_s for both of these these states are $n = 6, l = 0, m_l = 0, m_s = 1/2$, the difference between them is the sum of the nuclear spin ($I = 7/2$) and the total angular momentum of the valence electron ($J = 1/2$). So the difference is in the quantum number m_F, where $F = I + J$. So there are two options for m_F, the first is $m_F = 4$, when spin of the electron is parallel with the nuclear spin and the second option is $m_F = 3$, when electron spin is antiparallel with the nuclear spin. You can see the schematic of caesium clock in Figure 1.

For the atomic clock to work we need the beam with lower energy, which enters the core of the atomic clock - a tuneable microwave cavity, while the other one, consisting of higher energy caesium atoms, is guided away from the cavity by the magnetic field and detected. Like in any other clock, there is a frequency reference at the heart of it. This is an object that oscillates at a constant rate. In case of the atomic clock this is a resonator that confines electromagnetic fields in the microwave region of the spectrum, while in the olden days, a pendulum was the frequency reference - one swing of the pendulum one meter long equals one second. The electronic oscillator that is connected to the cavity is adjusted to the wavelength of approximately 3.3 cm, so the low energy caesium atoms jump to the higher energy state. These then flow through another set of magnets and are detected. If the frequency of the radio waves is exact, the higher energy caesium atoms continue to flow into the detector at the maximum rate. If the frequency is not quite right, because of the electronic's noise, then not all of the atoms jump into the higher state when passing the cavity and thus fewer detections are made. So just the right feed-back signal is sent to the oscillator, altering the frequency in the cavity, inducing that all caesium atoms start resonating again. We are feeding the output information back in. This is called a feedback loop. The feedback signal keeps the oscillator tuned in resonance with the frequency of the electronic transition of the caesium. This means that the atomic clock is self adjusting [3].

The frequency that is needed for the wanted electronic transition is 9129631770 Hz. So when we record 9129631770 periods of these radio frequency waves, 1 second has passed. This way the second is defined. Today the National Institute of Standards and Technology or NIST in the USA has an atomic clock that is so precise that if one had been running since the Big Bang, it would now be less than a second off.

But why caesium?

- it has only one stable isotope ($A = 133$),
- it needs a high frequency for the electronic transition, therefore we can make a more accurate measurement in a shorter period of time,
- caesium can be vaporised and has an odd number of nucleons.

If we combine all the properties listed above, we are not left with many options.

Atomic clocks are widely used in our daily life. Their most common use is within the global positioning system or the GPS. For a GPS satellite to work out your precise location, the timings of the signals it sends and receives have to be just right. The signals travel at the speed of light, which means that an error of even a single microsecond translates to an error of 300 m on the ground. Consequently, the timing has to be well-controlled, so that even tiny effects that we see in quantum mechanics have to be tracked. Until today the best known way to do that is the atomic clock described above.

But could it get better? The latest researches show that in the future it may be possible to create an even more precise clock. A nuclear clock.

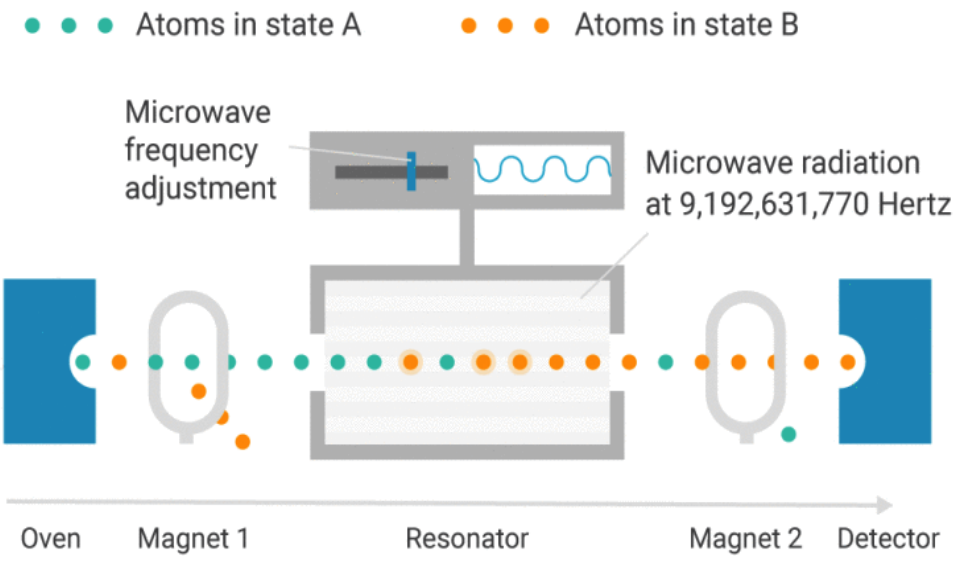

Figure 1. Schematic of caesium atomic clock [8].

3. Nuclear clock

While the atomic clock is based on the electronic transition of the atoms, the nuclear clock would be based on nuclear transitions. With nuclei, the measurements could be even more precise, because they are less sensitive to external conditions (thermal, electrical,...) than the atom. But physicists cannot build a nuclear clock with any nuclei. They must have a transition energy appropriate for optical excitation and they also have to have a short half-life. There is only one nuclear state that has these characteristics, which is an isomeric excited state of thorium (Th).

3.1 Isomeric excited state.

An isomeric excited state is a metastable state of an atomic nucleus where one or more nucleons are excited. Nuclei are found to have many distinguishable states. Of these, the ground state is indefinitely stable - has the lowest possible energy. Each of the other states has a higher energy and of all these other states, the metastable states have a lifetime from 100 to 1000 times longer than the most short-lived states of the same nucleus [5].

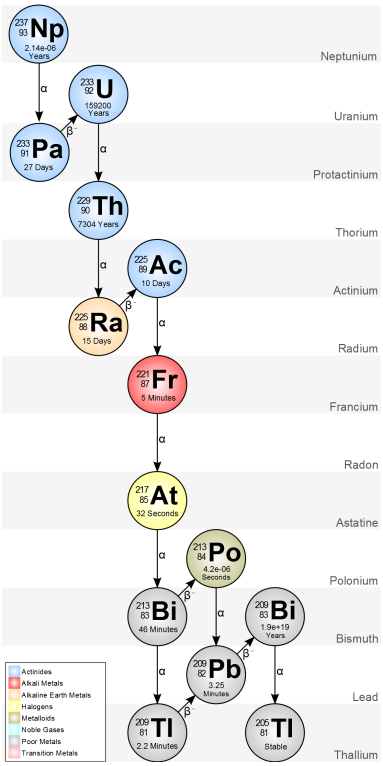

Figure 2. Neptunium decay chain [7].

In nuclear isomeric excited states, nucleons occupy a nuclear orbital with a higher energy that is available for lower lying nuclear orbital. While most of the metastable isomers decay through gamma ray emission, they can also decay by **internal conversion**. This is a process in which the released energy of the nuclear excitation is used to accelerate one of the inner shell electrons of the atom and is not released as a gamma ray. This happens when an inner atomic electron penetrates the nucleus and finds itself in intense electric fields.

A metastable isomeric excited state can be produced by the nuclear reactions. They are designated with a letter 'm' next to the mass number of the atom, for example ^{229m}Th. We can also find atoms with more than one metastable state and we distinguish them with 'm_1', 'm_2', where the increasing indices refer to the increasing excitation energy of the state.

Thorium (^{229}Th) is one of the daughter nuclides in the decay chain of isotope ^{237}Np (Figure 2). A very small amount of neptunium can be found in the nature, but the majority of it is created as a byproduct in nuclear power plants. It can be created via neutron capture in ^{235}U. Created isotope ^{237}Np has a half-life long enough to be easily extracted. [6]

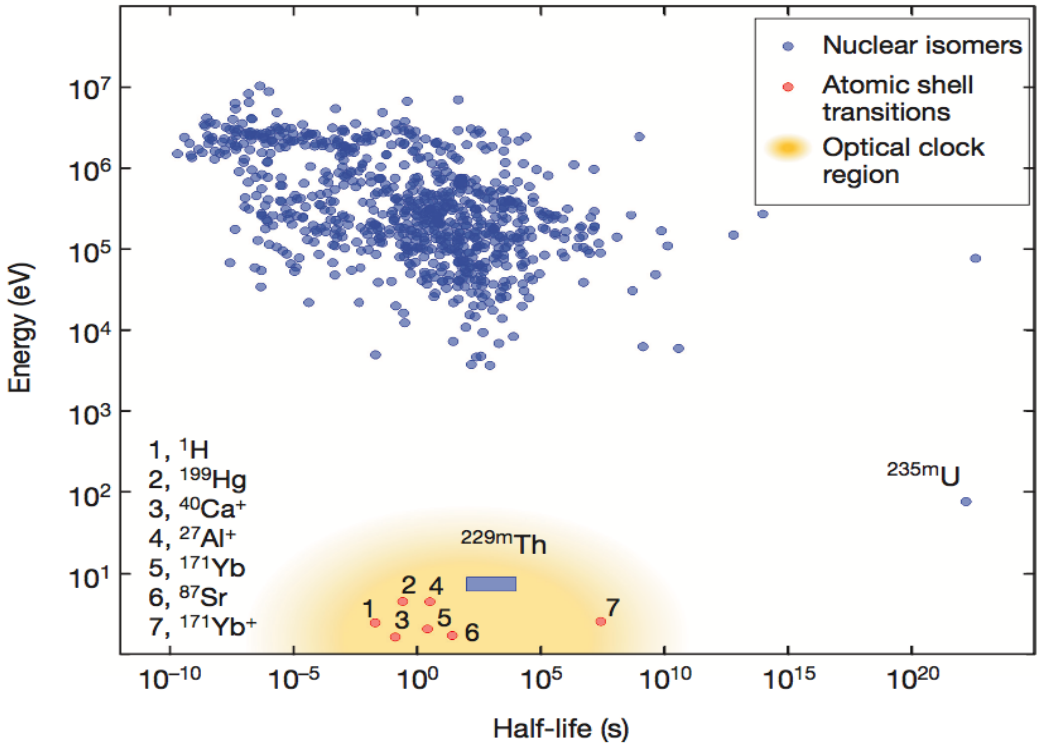

Figure 3. Energy - half-life distribution of isomeric nuclei. Each blue circle represents one of the known isomeric states. The red circles are for some of the electronic transitions and they are all in the orange region, which represents a space of parameters accessible to optical clocks. On the graph we can also see one blue box instead of a circle. It represents ^{229m}Th. The shape of it in this distribution is a box, because the excitation parameters are not yet exactly known. As we can see it is the only isomeric state in the orange region, making it the only promising isomer for the development of a nuclear clock. *Reproduced from: Direct detection of the* ^{229}Th *nuclear clock transtition, Nature* ***533*** *(2016), 47–51.*

The excited (isomeric) state of ^{229m}Th is the most exotic state of all nuclear states. It possesses the lowest excitation energy of all known states. The nucleus of ^{235m}U shows only nuclear states with excitation energies lower than 100 eV. But looking at Figure 3, we can see that ^{229m}Th also has a much shorter half-life than ^{235m}U.

Most energetic laser we have today works with a wavelength of 0.15 nm (around 8 keV), which is enough for excitation of the nucleus of ^{229}Th to ^{229m}Th. Detection of these transitions can be used for a new clock, a nuclear clock. For this we need to know the exact excitation energy of the isomeric state. The first step towards an even more accurate clock is the measurement of the exact excitation energy and the half-life of ^{229m}Th.

3.2 Measurements

The first measurements of the excitation energy were indirect. This means that they have not detected the ^{229m}Th directly, but they concluded about its existence from the gamma-ray spectrum from the decay of ^{233}U. Today the most widely accepted value for the transition energy is (7.8 ± 0.5) eV from 2009 [2]. However, this value is still not and cannot be considered as exact, until a direct measurement of energy is performed. This has motivated physicists to carry out an experiment that would directly detect the isomer ^{229m}Th. They succeeded a year ago. The only drawback of the experiment was that they optimised it for detection rather than for precision.

Now let's take a closer look at the experiment from 2016 [1], with the first direct detection of isomer ^{229m}Th. This was a huge step towards the determination of all decay parameters, which would help with the optical excitation.

3.2.1 Direct detection of the isomeric state

As said before, the isomeric state of thorium atoms decays predominantly by internal conversion (IC). The main goal of this experiment was to detect the emitted electron from the IC decay. But first we need to obtain thoriums isomeric states. These are populated in alpha decay of ^{233}U, and only 2 percent of the daughter nuclides are the ones we are looking for.

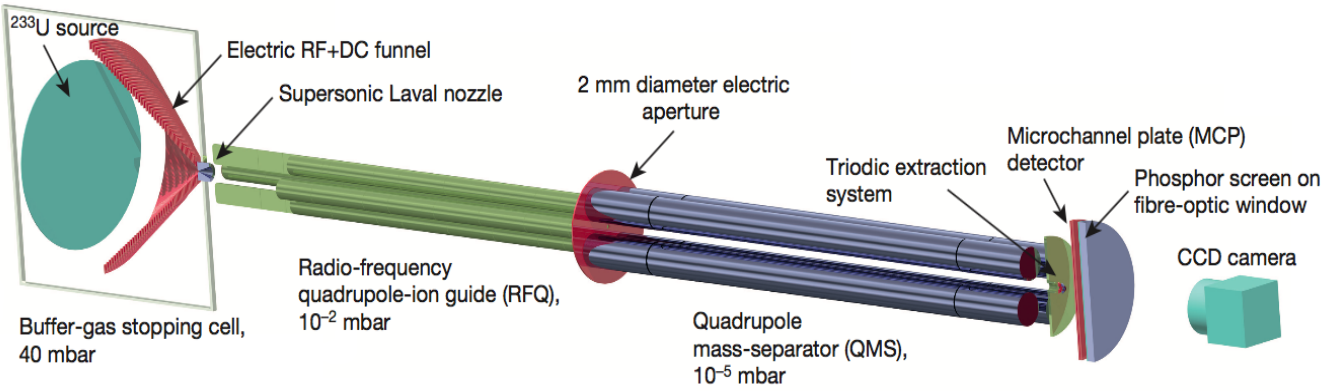

Figure 4. Schematic of the experimental setup 1. The source is placed in a buffer-gas stopping cell. The alpha decay ions are guided through the RF + DC funnel system into the RFQ (radio-frequency quadrupole system), where an ion beam is created. It is guided through the QMS (quadrupole mass-separator) for mass purification. Only the ions with the right mass-charge ratio are attracted onto the MCP (micro channel plate detector). With the help of a phosphor screen and CCD camera behind it, the signals of the ^{229m}Th isomeric decay are detected. *Reproduced from: Direct detection of the* ^{229}Th *nuclear clock transtition, Nature* ***533*** *(2016), 47–51.*

First we put the ^{233}U source, which consists of an $^{233}UF_4$ evaporated onto a stainless steel plate, in a buffer gas-stopping cell. This is a cell filled with helium at 40 mbar and its function is to stop all the daughter products of ^{233}U alpha decay, through collision with helium atoms. This is very important for the next stages of the experiment and the reader can follow them in Figure 4 and Figure 6. While the products are stopping, they loose energy by ionising the ambient gas. In Figure 6, can be seen all the daughter nuclides and the ions that are produced during the stopping process. Thorium is then mostly in 2+ and 3+ charge states.

All the ions are then guided towards the cell exit with an electric field through a DC funnel system. There is also a radio-frequency electric field in the funnel system, which only permits the ions to

exchange charge with the electrodes. At the exit there is a Laval nozzle and through it, the ions are injected into a radio frequency quadrupole structure (RFQ). It consists of four rods, divided into 12 segments. These are the source of an electric field gradient (1, 8 V to 0 V) and are constructed in a way that forms the extracted ions into a beam. On account of electric field gradient the beam is guided towards the exit and to the next stage of the experiment. At this stage, most of the daughter nuclides from the alpha decay are still present, but this will change after the ion beam enters the quadrupole mass-separator (QMS).
The QMS is used to extract the desired ions from the whole ion beam. It consists of four rods, similar to the RFQ. Two of the rods of the QMS are set at positive voltages. There is a DC offset between the two of them and therefore an electric field is created. The second pair of rods are set at negative voltages, again with a DC offset between them, creating an electric field. All four rods are connected to RF voltages, so the electric field in QMS is alternating. All the ions in the ion beam, which enter the QMS, have different mass-charge ratios and are affected by the alternating electric field differently. We can set the DC offsets between the rods, the frequency as well as the amplitude of the RF voltages manually. With the right setting we choose which ions will complete the whole path through the whole QMS, without hitting one of the rods or anything else.

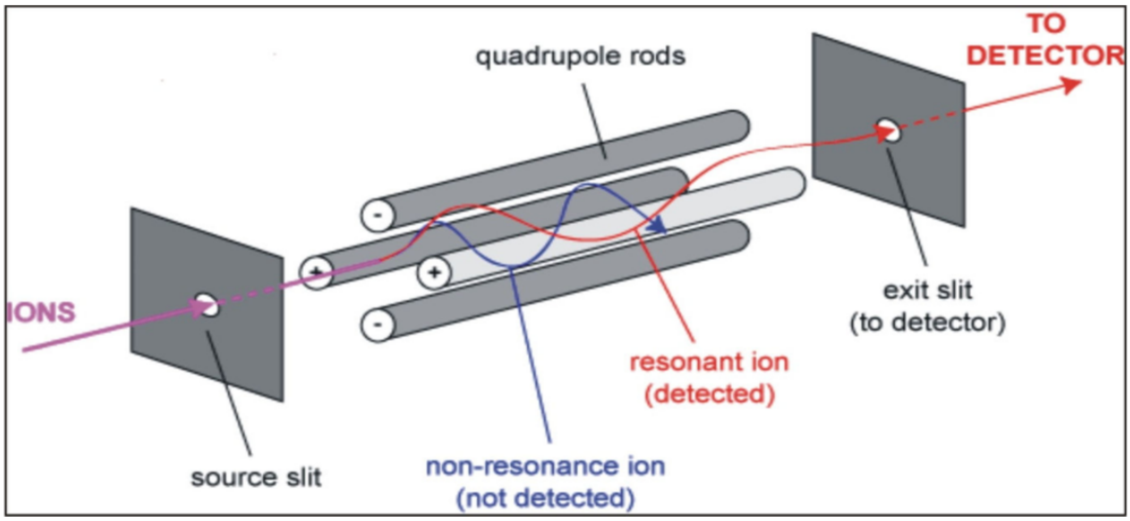

Figure 5. Schematic of a quadrupole mass separator (QMS), with represented trajectories of two ions with different mass-charge ratio [9].

Ions with small mass-charge ratio are affected mostly by the frequency of the RF voltages. If the frequency is too low, then they are attracted to one of the rods before the direction of the electric field changes. If the frequency is too high, the ion cannot follow the changes of the electric field direction and behaves the same as it would in an constant electric field. But if the frequency would be set right (for that ion), then the ions would alternate between the two rods towards the end of the QMS. But the ions with higher mass to charge ratio are not affected by the alternating field this much, because they are too heavy. So if the heavy ion has a positive charge it is attracted to a negative rod and if it has a negative charge, then to a positive rod. In this experiment the physicists observed the decay of isomeric excited state of thorium. It enters the QMS in 2+ and 3+ charged states. The positive ions with low mass-charge ratio pass between the negative rods and the positive ions with high mass-charge ratio pass between the positive rods. With the right RF voltages and right DC offset, both criteria are met and we guide through only the ions with the desired mass-charge ratio towards the exit.

For the extraction of Th^{3+}, a frequency of 925 kHz, an RF amplitude of $600,5$ V_{pp} (peak to peak voltage) and a DC voltage of 50.15 V is required, while for Th^{2+}, the respective values are 925 kHz, 901.5 V_{pp} and 75.23 V.

The ions are then guided towards a micro channel plate detector (MCP), presented in Figure 7. It operates at -25 V voltage, so the ions are collected at low kinetic energy. The MCP plate is the detector for low-energy electrons from the IC decay. But how does it work? First the ^{229m}Th ions are collected on its surface. Because of the positive charge the ions exchange charge with the surface and capture electrons, so they become neutral. With this process of charge exchange phonons are emitted for each electron exchange. When thorium is no longer an ion, the IC decay occurs. An IC electron is released, which triggers emission of secondary electrons.

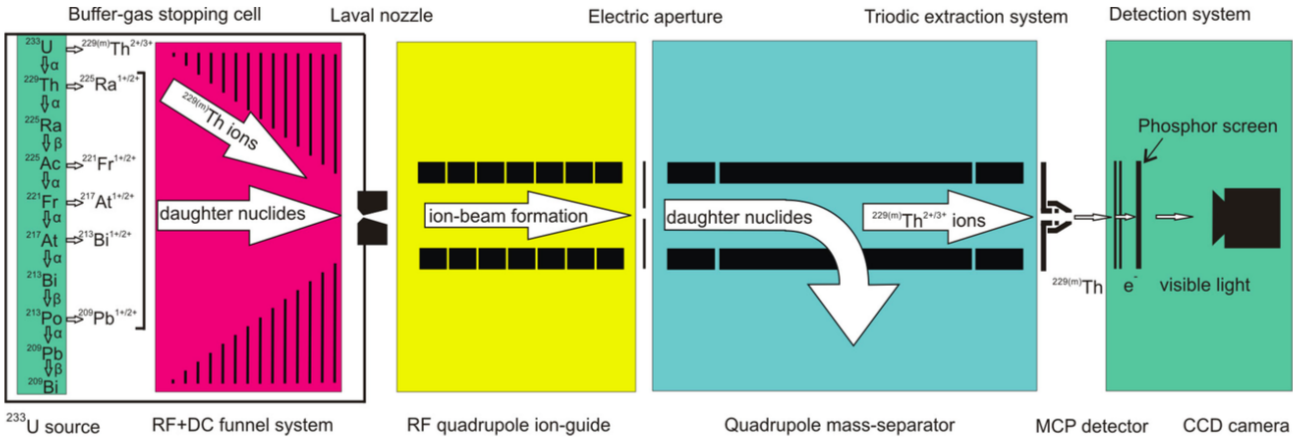

Figure 6. Schematic of the experimental setup 2. *Reproduced from: Direct detection of the* ^{229}Th *nuclear clock transtition, Nature* ***533*** *(2016), 47–51.*

All produced electrons are then accelerated towards the fluorescent screen, placed behind the MCP plate. The ending element of the experiment is a CCD camera. It monitors the fluorescent screen where the electronic-impact signal is converted into visible light.

The signals received from different sources and different ions are presented in Figure 8. One measurement took 2000 s. Such a long measuring time was necessary, as the expected count rate was 0.3 counts per second. The small count rate is due to the low rate of ions in isomeric state (2% only), low MCP detection efficiency for low-energy electrons, and low extraction efficiency of just 0.3% for Th^{+}, 5.5% for Th^{2+} and 10% for Th^{3+}. This is the reason why a clear signal was only seen by extracting Th^{2+} and Th^{3+}. Because the extraction and flight of ions towards the MCP plate takes only a few milliseconds, no significant isomeric decay happens during this process.

3.2.2 The background

The next step was to prove that the detected signal was indeed from the ^{229}Th isomeric decay. For this reason, comparative measurements were performed. The potential background sources are:

- background signal from setup components (stoping gas, extraction, ^{233}U source, QMS, MCP),
- signals from the long-lived (comparable half-life to the one of the isomer) excited states of thorium (excited state of thorium's electrons),
- signal caused by other short-lived nuclides or isomers and not of ^{229m}Th.

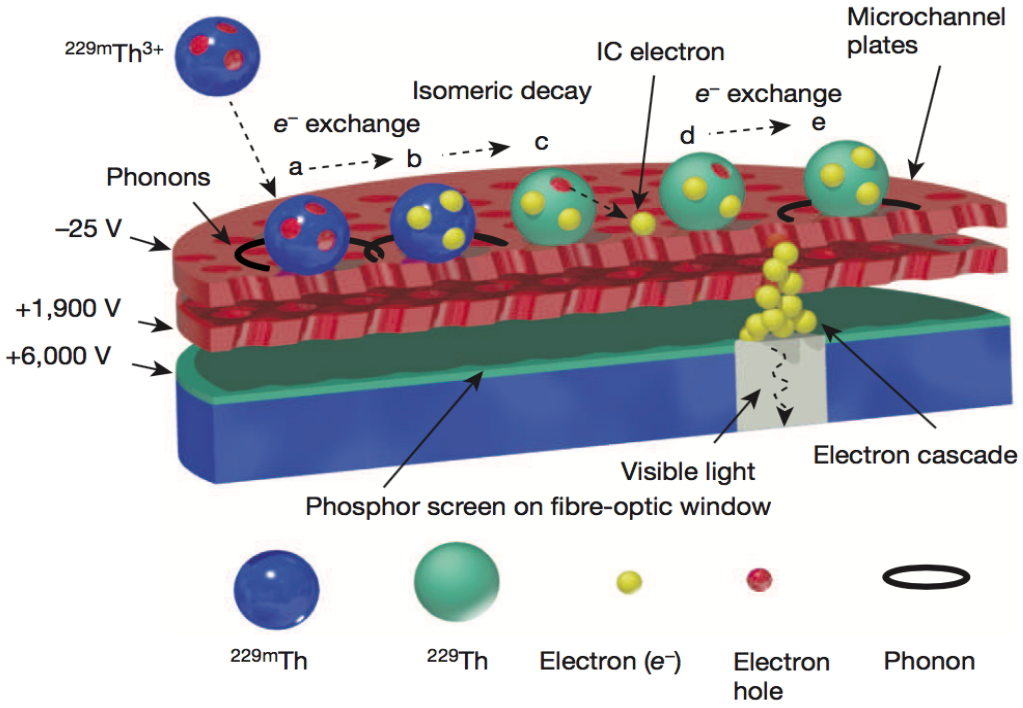

Figure 7. Schematic drawing of the isomer detection on the MCP. Step *a* of the detection shows that an $^{229m}Th^{3+}$ ion (visualised as a blue sphere) flies onto the MCP surface. Step b, the ion captures electrons (visualised as yellow spheres) on the surface. The energy is dissipated in the form of phonons (black circles). Step c, IC decay takes place after it and an IC electron is released. Step d, the IC electron then triggers many secondary electrons by passing through the MCP. The electrons are then accelerated towards the fluorescent screen for detection. Step e, the hole left by the IC electron is afterwards filled by a new electron from the MCP surface. *Reproduced from: Direct detection of the* ^{229}Th *nuclear clock transtition, Nature* ***533*** *(2016), 47–51.*

They measured the signal intensity after extracting $^{233}U^{2+}$ and $^{229m}Th^{2+}$ ions, because of a similar intensity of the mass peak in the decay of ^{233}U (seen in Figure 8). They have not recorded any signal while extracting $^{233}U^{2+}$ but had come to a conclusion that the signal from the impact in the case of uranium is reduced to zero and that only the signal of thorium remains. This experiment again excludes the potential background due to the impact of ions and furthermore also excludes the potential background caused by the setup components, because they would be constant throughout all the measurements.

But there could still be a signal due to the thorium shell effects, i.e. long lived atomic excitations. These effects would be the same in case of ^{230}Th. Therefore, they used a ^{234}U source to produce ^{230}Th ions and collected them on the MCP again for 2000 s. They obtained no signal from it. It was safe to say that the signal does not originate from the atomic shell effects.

What about other radioactive decays of short-lived daughter nuclides? This kind of background was excluded in different ways. One way to do it is to observe the signals of 2+ and 3+ charge states in parallel. The reason behind it is that mostly only thorium is extracted in the 3+ charge state, because of the low third ionisation energy (Table 1).

The results of the experiments show that the magnitude of the extracted short lived daughter nuclides in the 2+ charged state, is four orders of magnitude stronger than in the 3+ charged state. Therefore, if source of the signal were short-lived nuclides, the same drop of magnitude should have

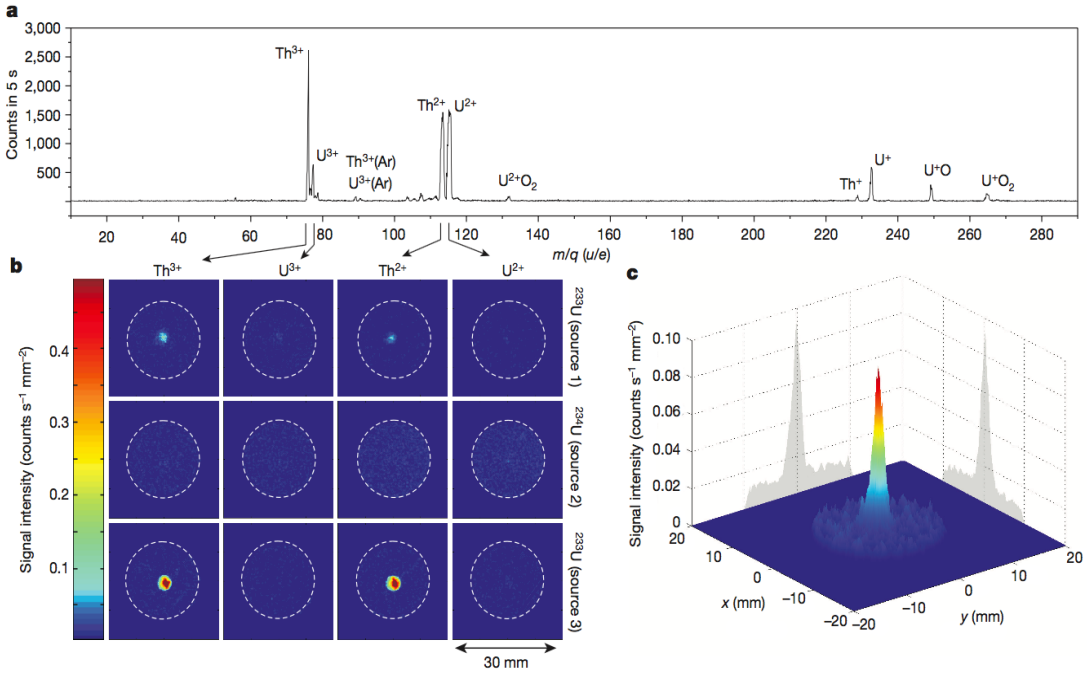

Figure 8. Signal comparison. a. Complete mass scan performed with source 1 (UF_4). The mass-charge ratio (m/q) is presented in the units of atomic mass (u) over electric charge (e). **b.** Comparison of signals obtained with detection of thorium and uranium in 2+ and 3+ charge states, with different sources. Each signal is obtained after a 2000 s measurement, with −25 V surface voltage on the MCP. **c.** Signal of the ^{229m}Th decay, obtained during 3+ charge state extraction, with source 1. The obtained maximum signal intensity is 0.08 counts s^{-1} mm^{-2}. *Reproduced from: Direct detection of the* ^{229}Th *nuclear clock transtition, Nature* ***533*** *(2016), 47–51.*

been observed in the signal from the MCP too. But this was not observed. Another way to exclude the background caused by other short-lived nuclides or isomers was to use a chemically purified ^{233}U source. This is a source which is purified of the short-lived nuclides. If the signals were due to the nuclear background, they should have recorded a reduced signal. But the opposite happened. The signal was increased owing to a larger content of ^{233}U.

So after excluding all possible background signals, they concluded that the obtained signal is from the nuclear isomeric transition in ^{229m}Th. They succeeded in making the first experiment of direct detection of this exotic nuclear state. This was a huge step towards the first nuclear clock, but as discussed before, to actually build one, they needed the exact value of the excitation energy and

Table 1. Ionization energies of elements potentially contained in the ^{233}U source material

Element	Atomic no.	1+ [eV]	2+ [eV]	3+ [eV]
Plutonium	94	6.03	11.5	21.1
Neptunium	93	6.27	11.5	19.7
Uranium	92	6.19	11.6	19.8
Plutonium	91	5.89	11.9	18.6
Thorium	90	6.31	11.9	18.3
Actinium	89	5.38	11.8	17.4
Radium	88	5.28	10.1	31
Francium	87	4.07	22.4	33.5

the half-life of the state. This way they could understand the process of this isomeric decay better and could excite the ground state of thorium with a laser beam and therefore they could develop a nuclear frequency standard, just like the caesium one in atomic clocks. But at the moment there is still no exact experiment known, with which the desired precision can be reached.

3.3 The decay parameters

The experiment was optimised for detection of the isomeric state, therefore no exact value for the transition energy was obtained from it. But some conclusions on the restriction on the transition energy could still be drawn from the measurements. The neutral isomer state of thorium mostly decays by internal conversion. Its half-life is of the order of microseconds. Using this fact, the lower limit for the isomeric transition energy could be specified. Right after the $^{229m}Th^{2+}$ ions have struck the MCP surface (where they exchanged charge), the researchers acquired pictures with the CCD camera. This way they not only confirmed that the isomeric decay is energetically allowed, but also determined that the half-life was less than a second. The main fact that they got from this measurement is, that the isomeric energy must be above the first ionisation potential of thorium, $6,3$ eV. But the half-life of thorium is very charge-state dependent and their prediction for the $^{229m}Th^{1+}$ or higher state's half-life was from minutes to hours, so they predicted also that the IC decay for ions is energetically forbidden. To see if their predictions were right, they used a different gradient of electrical potential in RFQ and stored the $^{229m}Th^{2+}$ ions in it. The maximum storage time was about 60 s. After releasing them from the RFQ and guiding them towards the MCP surface they still obtained a significant signal. Thus the half-life of the $^{229m}Th^{2+}$ ion is longer than 60 s, which can only be explained if the IC decay is energetically forbidden. This way they obtained the second limit for the transition energy and this is equivalent to the third ionisation energy of thorium, 18.3 eV. (The same measurement could not be done with $^{229m}Th^{1+}$ because of the very low extraction rate.)

With predictions of half-life for isomeric atom and ions of thorium they came to a conclusion that the isomeric energy is between the first and third ionisation energy of thorium - between 6.3 eV and 18.3 eV. A very promising fact is that the most precise and accepted value from the indirect measurements is in this range.

4. Conclusion

Precision of time is very important. Not only does it allow us to be on time in a classroom or at work, it also has a much bigger impact on our lives than we think. Exact timings are critical for the internet and GPS which we use every day. Then there are physics researches like those in CERN, NPL, NIST and others where incredible precision is needed. We are currently using the most precise clocks, atomic clocks. With their help the latest definition of the second is precise to ten digits. This could get even better in the future. Direct detection of the isomeric state of thorium was a big step towards creating an even more precise clock, named nuclear clock. With this invention, the second would be redefined to even greater precision, with total inaccuracy approaching 10^{-19}, therefore all devices which depend on precise timings would work even better. The caesium standard known today, would then be replaced by the thorium frequency standard. The isomeric metastable state of thorium ^{229m}Th is the best choice of all known nuclear states to develop a nuclear clock. The main goal for the next research is to determine the exact decay parameters of this state. Last year the first observation of the ^{229m}Th decay was performed, but it was not precise enough. So we are left with an energy range for the excitation energy of this state and it is between 6.2 eV and 18.3 eV. When a more precise excitation energy will be directly measured, we will be able to use a laser

with an exact wavelength to excite thorium atoms into these metastable states. With a possible optical excitation, the first nuclear clock would be made, which promises important improvements in current atomic clock applications.

ACKNOWLEDGEMENTS

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