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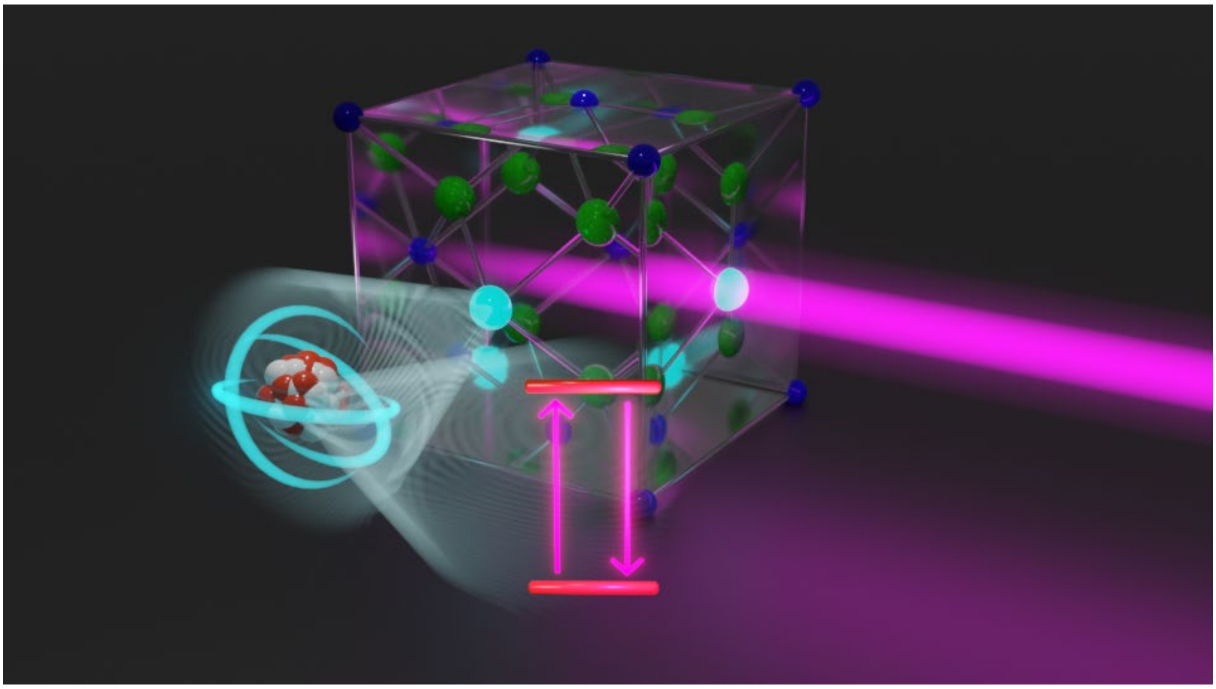
Atomic Nucleus Excited with Laser: A Breakthrough after Decades

The "thorium transition", which has been sought after for decades, has now been excited for the first time with lasers. This paves the way for revolutionary high precision technologies, including nuclear clocks.



A laser beam changes the state of a thorium nucleus, embedded in a crystal. That way, the long-sought thorium transition could finally be found and measured precisely. (Copyright: TU Wien)

Physicists have been hoping for this moment for a long time: for many years, scientists all around the world have been searching for a very specific state of thorium atomic nuclei that promises revolutionary technological applications. It could be used, for example, to build an nuclear clock that could measure time more precisely than the best atomic clocks available today. It could also be used to answer completely new fundamental questions in physics - for example, the question of whether the constants of nature are actually constant or whether they change in space and time.

Now this hope has come true: the long-sought thorium transition has been found, its energy is now known exactly. For the first time, it has been possible to use a laser to transfer an atomic nucleus into a state of higher energy and then precisely track its return to its original state. This makes it possible to combine two areas of physics that previously had little to do with each other: classical quantum physics and nuclear physics. A crucial prerequisite for this success was the development of special thorium-containing crystals. A research team led by Prof. Thorsten Schumm from TU Wien (Vienna) has now published this success together with a team from the National Metrology Institute Braunschweig (PTB) in the journal "Physical Review Letters".

Switching quantum states

Manipulating atoms or molecules with lasers is commonplace today: if the wavelength of the laser is chosen exactly right, atoms or molecules can be switched from one state to another. In this way, the energies of atoms or molecules can be measured very precisely. Many precision measurement techniques are based on this, such as today's atomic clocks, but also chemical analysis methods. Lasers are also often used in quantum computers to store information in atoms or molecules.

For a long time, however, it seemed impossible to apply these techniques to atomic nuclei. "Atomic nuclei can also switch between different quantum states. However, it usually takes much more energy to change an atomic nucleus from one state to another – at least a thousand times the energy of electrons in an atom or a molecule," says Thorsten Schumm. "This is why normally atomic nuclei cannot be manipulated with lasers. The energy of the photons is simply not enough."

This is unfortunate, because atomic nuclei are actually the perfect quantum objects for precision measurements: They are much smaller than atoms and molecules and are therefore much less susceptible to external disturbances, such as electromagnetic fields. In principle, they would therefore allow measurements with unprecedented accuracy.

The needle in the haystack

Since the 1970s, there has been speculation that there might be a special atomic nucleus which, unlike other nuclei, could perhaps be manipulated with a laser, namely thorium-229. This nucleus has two very closely adjacent energy states – so closely adjacent that a laser should in principle be sufficient to change the state of the atomic nucleus.

For a long time, however, there was only indirect evidence of the existence of this transition. "The problem is that you have to know the energy of the transition extremely precisely in order to be able to induce the transition with a laser beam," says Thorsten Schumm. "Knowing the energy of this transition to within one electron volt is of little use, if you have to hit the right energy with a precision of one millionth of an electron volt in order to detect the transition." It is like looking for a needle in a haystack – or trying to find a small treasure chest buried on a kilometer-long island.

The thorium crystal trick

Some research groups have tried to study thorium nuclei by holding them individually in place in electromagnetic traps. However, Thorsten Schumm and his team chose a completely different technique. "We developed crystals in which large numbers of thorium atoms are incorporated," explains Fabian Schaden, who developed the crystals in Vienna and measured them together with the PTB team. "Although this is technically quite complex, it has the advantage that we can not only study individual thorium nuclei in this way but can hit approximately ten to the power of seventeen thorium nuclei simultaneously with the laser – about a million times more than there are stars in our galaxy." The large number of thorium nuclei amplifies the effect, shortens the required measurement time and increases the probability of actually finding the energy transition.

On November 21, 2023, the team was finally successful: the correct energy of the thorium transition was hit exactly, the thorium nuclei delivered a clear signal for the first time. The laser beam had actually switched their state. After careful examination and evaluation of the data, the result has now been published.

"For us, this is a dream coming true," says Thorsten Schumm. Since 2009, Schumm had focused his research entirely on the search for the thorium transition. His group as well as competing teams from all over the world have repeatedly achieved important partial successes in recent years. "Of course we are delighted that we are now the ones who can present the crucial breakthrough: The first targeted laser excitation of an atomic nucleus," says Schumm.

The dream of the atomic nucleus clock

This marks the start of a new exciting era of research: now that the team knows how to excite the thorium state, this technology can be used for precision measurements. "From the very beginning, building an atomic clock was an important long-term goal," says Thorsten Schumm. "Similar to how a pendulum clock uses the swinging of the pendulum as a timer, the oscillation of the light that excites the thorium transition could be used as a timer for a new type of clock that would be significantly more accurate than the best atomic clocks available today."

But it is not just time that could be measured much more precisely in this way than before. For example, the Earth's gravitational field could be analyzed so precisely that it could provide indications of mineral resources or earthquakes. The measurement method could also be used to get to the bottom of fundamental mysteries of physics: Are the constants of nature really constant? Or can tiny changes perhaps be measured over time? "Our measuring method is just the beginning," says Thorsten Schumm. "We cannot yet predict what results we will achieve with it. It will certainly be very exciting."

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Full paper (preprint): https://www.tuwien.at/fileadmin/Assets/tu-wien/News/2024/Thorium_Preprint.pdf

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Historical Background:

The Long Search for the Thorium Transition

The story of a decades-long mystery: How much energy is needed to switch a thorium nucleus from the lowest energy state to the next higher energy state?

At first glance, one might think that the situation is quite simple: a thorium nucleus can be in different states - in the ground state or in a metastable state, both of which have almost the same energy. If you hit the thorium with a laser beam whose wavelength exactly matches the energy difference between these two states, you can make the thorium nucleus switch from one state to the other.

The problem is that for this to work, you have to know exactly how much energy is required. You will only see an effect if you hit the bullseye exactly. You have to work with a precision in the range of millionths of an electron volt, otherwise the laser beam simply has no effect.

This becomes even more difficult due to the fact that it is not possible to tell immediately whether the right energy has been hit or not. "If you excite the atomic nucleus, switching it from the lower state to the higher state, it initially remains in this higher state for a while," says Thorsten Schumm. "The excited state has a lifetime of over ten minutes. Only then does the atomic nucleus return to the ground state and emit light, which can then be measured."

Reliably testing a specific wavelength can take hours. "For a long time, it was therefore considered impossible to simply try out all conceivable energy values one after the other," says Thorsten Schumm. "If you don't know where to look, the search is hopeless." So the question is: how can you narrow down the energy range to be searched?

Skyscrapers versus the kerb

As early as the 1970s, uranium nuclei, which transform into thorium nuclei through radioactive decay, were investigated. The radiation from this decay revealed that thorium-229 should have two energy states that are extremely close together – with an energy difference of less than 100 electron volts. It was not possible to be more precise at the time.

In the 1990s, efforts were made to measure this energy difference more precisely. A special trick was used for this: the thorium nuclei were put into a different state with much higher energy than the two states that were actually to be investigated (in the range of many thousands of electron volts). After a while, the thorium nucleus will then switch to one of the two closely neighboring low-energy states – the ground state or the closely neighboring metastable state. The distance between the two low states can be indirectly calculated from the radiation that is produced.

However, the accuracy of this method is limited. "It's a bit like measuring the height of a kerb by first going much higher and dropping a ball onto the street from the roof of a skyscraper," says Thorsten Schumm. "The ball can fall onto the street, or onto the kerb. The difference between these two possibilities is tiny, compared to the height of the skyscraper. But if you measure both distances very precisely, you can in principle learn something about the kerb."

A race for the highest precision

Initial results using this method yielded extremely low values for the energy of the sought-after thorium transition: 3.5 electron volts was an early assumption, then in 2005 a value of 5.5 electron volts was published – with an estimated accuracy of one electron volt.

Research teams from Europe, the USA and Japan competed for the best results, with the estimated value shifting slightly upwards: in 2009, a value of 7.8 ± 0.5 electron volts was measured at the

Lawrence Livermore Lab (USA); in 2019, a team from LMU Munich with the participation of Thorsten Schumm finally arrived at 8.28 ± 0.17 electron volts; in 2020, Schumm and colleagues from Heidelberg published data indicating 8.10 ± 0.17 electron volts.

The accuracy had increased over the years, but the results were still far too imprecise to hope to actually hit the energy transition exactly with a laser.

An essential step in improving these measurements was the production of thorium-containing crystals – a difficult task that Thorsten Schumm and his team at TU Wien solved. In this way, a large number of thorium nuclei can be examined simultaneously.

First light from the transition itself

In 2023, a collaboration between LMU Munich and TU Wien succeeded for the first time in directly measuring the sought-after thorium transition using crystals of this kind: It was no longer necessary to indirectly deduce the energy of the thorium transition via another, much higher energy state; it was possible to directly measure the radiation produced during the sought-after transition. This suddenly made a much higher precision possible: 8,338 ± 0.024 electron volts was the new result.

For the first time, there was now hope of being able to excite the state specifically with a laser. Step by step, the now narrowly defined area was scanned until success was finally achieved and a clear signal was obtained – at an energy of 8.355743 ± 0.000003 electron volts.

In five decades, "less than 100 electron volts" thus became a precision in the microelectron volt range. This has finally opened the door to a new field of research with many technical applications.

Potential Applications:

Nuclear Clocks and the Fundamental Constants of Nature

The long-sought thorium transition has been found. What can we do with it now? Very different areas of technology could benefit from this discovery.

For the first time, thorium nuclei have now been successfully transferred from one state to another using a laser – but there has already been speculation for decades about the technical possibilities that would arise if this feat were one day possible. Expectations are high: probably the best-known idea is the nuclear clock, but precision measurements based on the thorium transition could open up new possibilities in completely different areas of research.

Time measurement

The most accurate time measuring devices today are atomic clocks – they have even changed the definition of our units of time. Today, the second is defined as the length of time corresponding to a very specific number of oscillations of the light that must be shone on a caesium atom in order to switch its electrons from one state to another.

Similar to the pendulum of a pendulum clock, the light oscillation of the caesium atoms plays the role of the timer that provides the regular ticking for the most accurate time measurement possible. Today, atomic clocks are used, for example, for the coordination of satellites, they enable the high accuracy of GPS signals and they also play a role in telecommunications.

Instead of cesium atoms, however, thorium atomic nuclei can now also be used for keeping time. Only when the thorium nuclei are irradiated with a laser beam of exactly the right frequency do they change their state. If the laser changes its frequency slightly (e.g. due to external interference), this immediately results in a change of the response of the thorium nuclei. The combination of laser and thorium nuclei therefore makes it possible to keep the laser frequency extremely stable and ensures that the laser frequency does not drift away.

Because atomic nuclei are much smaller than atoms and much less sensitive to electromagnetic perturbations, atomic nuclei can achieve much higher precision than atoms when it comes to measuring time.

Everything gets better with better clocks

With better time measurement, other physical quantities can also be measured more accurately. This is a well-known phenomenon: in nautical navigation, for example, people have struggled for a long time with the problem that although it is easy to determine the latitude based on the position of the sun, an accurate clock is required to calculate the longitude correctly. Better time measurement often makes other measurements possible.

For example, Einstein's theory of relativity states that time does not pass in the same way everywhere: the flow of time depends on the gravitational field. In this way, an extremely precise clock could therefore be used to accurately measure the Earth's gravitational field – with many possible applications ranging from the search for mineral resources to research into plate tectonics and earthquake prediction.

Great hopes are also pinned on the possibility that new, better clocks could clarify previously unsolved fundamental questions of physics: If it is possible to measure the constants of nature much more accurately than before, then it would also be possible to test the theory that these natural constants may not be perfectly constant at all. Perhaps they change over time? Research into dark matter is also hoping to gain new insights through even more precise measurements.

It is not always possible to get closer to the fundamental laws of nature just by looking at the smallest particles in a particle accelerator or by looking at the most distant regions of space. Sometimes you just need greater precision – and this is where the newly discovered thorium transition and all its potential applications should help in the coming years and decades.

The Crucial Crystal Trick:

High-tech Gemstones for Nuclear Science

Special thorium-containing crystals, developed over many years at TU Wien, were crucial in tracking down the long-sought thorium transition.

Prof. Thorsten Schumm with a crystal containing thorium. (Copyright: Foto Wilke)

Emerald, ruby, amethyst and many other gemstones have one thing in common: they consist of a perfectly regular crystal structure into which foreign atoms are incorporated in low concentrations. From a physical point of view, these foreign atoms are actually "disturbances", imperfections in the crystal. But they are precisely what give the gemstone its color. Amethyst, for example, has the atomic structure of a simple quartz crystal. However, the addition of a few iron atoms gives it its characteristic violet color. A completely perfect crystal, in which the same arrangement of atoms is repeated exactly over and over again, usually looks quite bland and pale.

The situation is very similar with the crystals that were grown at TU Wien in order to find the long-sought thorium transition: The decisive factor here is also the careful and precise incorporation of foreign atoms - in this case radioactive thorium. Of all the research groups worldwide working on the thorium transition, the team at TU Wien is the only one that can produce such thorium-containing crystals on their own. In the end, this was also the key to their success.

Thorium atoms in the crystal

"If you want to excite thorium atomic nuclei with a laser, you basically have two options," explains Prof. Thorsten Schumm. "Either you use thorium ions, which you trap and hold with electromagnetic fields, or you build the thorium atoms into a solid." Only a very small number of atoms can be trapped in ion traps, so Thorsten Schumm soon realized that he wanted to pursue the solid-state approach. However, there are major technical challenges to overcome.

"The starting material must be completely transparent for the laser. The laser should only have an effect on the built-in thorium atoms," emphasizes Thorsten Schumm. Glass or similar materials that are rather irregular on an atomic level are out of the question, as they are not transparent enough. Only extremely regular crystals, such as calcium fluoride, can be used.

Melting and re-solidifying

But how is it possible to incorporate thorium atoms into an extremely regular calcium fluoride crystal? "It took years to develop this process," says Thorsten Schumm. "We start with a tiny, very regular crystal, to which we add thorium and place it in an ultra-high vacuum. Oxygen would destroy the process immediately." The crystal is then heated in the vacuum chamber and partially melted. This creates a liquid mixture of thorium, calcium and fluorine, while part of the crystal underneath is still solid. The temperature is then lowered again, and the mixture is allowed to solidify – precisely along the geometric pattern defined by the solid crystal underneath.

"There are many technical details that have to be precisely controlled, but if you do everything right, you get a very regular crystal with built-in thorium atoms, a few millimetres in size."

Rare combination of knowledge from different areas

Originally, Thorsten Schumm did not necessarily plan to produce the crystals himself. "There are research institutes and companies that specialize in growing crystals. I had a lot of discussions looking for partners who could produce such crystals, but it was more difficult than I thought," says Schumm.

Most manufacturing processes are optimized for the largest possible crystals. However, small crystals are required to excite the thorium transition: the laser beam used for the experiments only hits a small area of the sample, any material beyond this would only contribute to interference. "When producing small crystals, you have to face completely different difficulties. Surface tension, for example, plays a much more important role on a small scale. It also took us years to go from excellent centimetre-sized crystals to excellent millimetre-sized crystals."

In addition, there are hardly any institutes that have the necessary knowledge and equipment to handle radioactive thorium. "This is of course a great advantage for us at the Institute for Atomic and Subatomic Physics at TU Wien," says Thorsten Schumm. "We are certified for this, we have radiation protection expertise and the necessary equipment to work with thorium." A seemingly simple step such as polishing the crystal becomes a major problem if you have no experience with radioactive material. You can't simply generate thorium dust in the laboratory, which can then perhaps be inhaled.

A completely new field of research

Thorsten Schumm found himself in the strange situation of having to build up world-class expertise in an area that was not really his true field of research: the aim was always to excite atomic nuclei with a laser, i.e. to combine quantum physics and nuclear physics. But as a means to this end, a breakthrough was also needed in materials research – which is actually a completely different area of physics.

"I would never have thought that I would also become a solid-state physicist," says Schumm. "But ultimately that was the key to success: precisely because we produced, characterized and measured the crystals ourselves, we had the decisive edge and, together with our colleagues in Braunschweig, were ultimately the team that achieved the crucial breakthrough."