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229Th the Bridge Between Nuclear and Atomic Interactions

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^{229}Th the Bridge Between Nuclear and Atomic Interactions

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Abstract

The precise measurement of time has been a goal of physicists for centuries. With every new increase in our ability to measure time we have discovered new phenomena. The most advanced clocks available to us currently are atomic clocks that use electronic transitions to track the passage of time. In this proposal, I put forward the framework for the first nuclear clock estimated to be 1000 to 10000 times more precise than the current atomic clocks. This research will explore in detail the atomic nuclear interactions and help perfect and refine current atomic-nuclear interaction models. The realization of a ^{229}Th nuclear clock will allow tests of cosmology by measuring the change of the fine structure constant as a function of time. The results of these experiments could dramatically alter our view of the universe, its past and future evolution.

Precision clocks - with fundamental physics applications - require a long-lived quantum transition (two-level system) that is immune to external perturbations. Nuclear transitions would be better suited than atomic transitions for these applications except that nuclear transitions are typically much higher in energy and therefore cannot be accessed with table-top lasers. There is, however, one promising nuclear transition: the doublet between the ground and first excited states of the ^{229}Th nucleus discovered by Helmer and Reich [1]. This doublet has an energy splitting of 7.6±0.5 eV [2], a spin difference of 1 h-bar, and an excited state half-life that could be as long as hours. A precision clock based on the ^{229}Th nuclear doublet has been proposed by Peik et al. [3]. Their design is similar to the ion clock research being conducted at NIST in Boulder, CO. However, the NIST researchers use atomic transitions for their frequency standards. In [3] the ^{229}Th nuclear doublet transition is the frequency standard while atomic transitions are used to cool the ions and for probing the state of the ^{229}Th nucleus. Recently, Campbell *et al.* [4] have trapped and cooled $^{232}Th^{3+}$ at Georgia Institute of Technology. This is a large step forward in the realization of a nuclear clock. The Georgia Tech group is already a collaborator on this project and we are in discussions with the NIST Boulder group about collaboration.

In order to determine the suitability of the ^{229}Th nuclear doublet for a precision clock, the half-life of the excited-state needs to be measured. Current estimates of the half-life vary from 10 μs to 1000 hours. The longer the half-life, the narrower the natural linewidth of the state and the more desirable the transition is for potential applications.

In this proposal, I outline the necessary research to be conducted to determine the half-life and exact wavelength of the nuclear doublet transition in ^{229}Th. This research will lead to a deeper understanding of atomic-nuclear interactions important for our knowledge of high energy density

science. It will provide a spectroscopy measurement of the lowest known nuclear transition ever and open the doorway for the development of a nuclear clock with unprecedented precision.

Introduction

The energy of the first excited-state of the ^{229}Th nucleus is the lowest of all known isotopes at a mere 7.6±0.5 eV [2] above the ground state; this transition energy corresponds to a wavelength of approximately 160 nm. The spin difference is 1 h-bar and the excited-state is meta-stable with a half-life as long as hours. This makes ^{229}Th the premier candidate for bridging the atomic-nuclear coupling using a tabletop laser to transition between the two states of this doublet. The ability to apply the arsenal of precision laser spectroscopic techniques (where frequencies/energies can be measured to a fractional precision of 10^{-15}) to the nuclear domain would be a breakthrough on par with the Nobel prize winning work of Mössbauer. Laser manipulation of the ^{229}Th nucleus could lead to unprecedented studies of the interplay between atomic and nuclear systems [3], provide a new frequency standard [4] and improve the search for time-variation of fundamental constants by as many as six orders of magnitude [5-6].

However, before the transition can be excited with laser light, it is imperative that the natural linewidth of the state and the wavelength of the transition be determined to better guide any laser scanning search for the transition. We propose an experiment to determine the half-life (and therefore natural linewidth) of the first excited-state by measuring the exponential decay of photon and electron emission from specially prepared ^{229}Th samples. In studying the mechanisms by which ^{229}Th decays we will also discover the best approach to use in measuring the 7.6 eV gamma ray wavelength directly using EUV spectrometry. Once these two key pieces of knowledge are obtained, ^{229}Th can then be turned into a nuclear clock through collaborations with colleagues at the Georgia Institute of Technology and National Institute of Standards and Technology at Boulder. Please refer to figure 1 for a level scheme of the ^{229}Th nucleus.

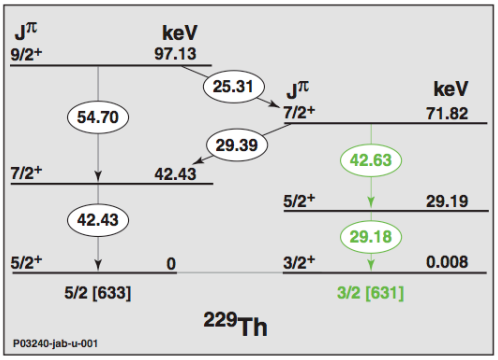

Figure 1: is the ^{229}Th nuclear level diagram from Beck [2]. The ground state has a spin of 5/2+ while the first excited state has a spin and parity of 3/2+.

Background

The low-lying nuclear level in ^{229}Th has attracted the attention of scientists all over the world and has been the subject of much experimental and theoretical interest in recent years. Several members of my current research group have already performed ground-breaking research to understand the properties of this state. E. Browne, E.B. Norman and collaborators [7] have placed the only limits on the half-life (<6 hours or >20 days) by studying its feeding from the alpha decay of ^{233}U. Only recently B.R. Beck *et al*. [2], discovered that the energy of the first excited state is 7.6±0.5 eV. This result revealed that the excited-state energy is a factor of two larger than previously believed. All of the previous direct searches for the decay of this state were insensitive at these energies.

The experimental method to determine the half-life and the decay mode involves creating a very thin source of ^{229m}Th on a catcher plate and moving this plate in front of an electron detector and an UV-sensitive photon detector. This technique has been previously used to measure the half-life of the 76 eV state of ^{235m}U. Our technique has several advantages: (1) it is simple, inexpensive, and can be executed quickly, (2) it has been previously demonstrated and optimized for measurements of the half-life of the 76 eV excited-state of ^{235}U (with a statistical precision of 0.1%), and (3) it allows both low-energy electrons and photons to emerge from the target for efficient detection. Other research groups around the world are planning challenging experiments to study the properties of this isomeric state, including performing collinear laser spectroscopy on ^{229}Th ions to study the hyperfine interaction [8], photon counting of ^{229}Th atoms guided to a target using a radiofrequency ion guide and buffer gas technique [9], and bombarding the ^{229}Th atoms with intense x-ray beams from the Advanced Photon Source at Argonne National Laboratory [10].

Proposed research and methods

In the following sections I will discuss the research approach that will be applied to determining the half-life of ^{229}Th, the role of atomic-nuclear interaction theory, the benefits of a nuclear clock compared to an atomic clock, recent research of the constancy of the fine structure constant and finally the capabilities and expertise that will be brought to bear while performing research at LLNL.

1) Experimental technique for determining the half-life

The ^{229m}Th half-life has never been measured and calculations are unreliable, ranging from 10 μs [11] to 5 hours [2]. The half-life and decay mode can be measured by directly detecting conversion electrons or photons following the internal transition decay of the ^{229m}Th state. Measurements of this

type were performed 35 years ago using a ^{239}Pu source to study the energy and half-life of the 76 eV state in ^{235m}U state [12] which decays by conversion electron emission with a half-life of 26 minutes. The same basic techniques can be applied to the ^{229m}Th measurement. In fact, studies of the ^{235m}U decay provide a convenient way to optimize the apparatus.

The ^{229m}Th samples will be prepared using a simple "hot atom" technique [12-13] in which ^{229m}Th atoms recoiling from the surface of a thin ^{233}U source following α decay will be collected on a catcher plate, see figure 2 and figure 3. Atoms collected in this manner sit on or near the surface where the eV-energy electrons and photons from ^{229m}Th decay can escape. A ^{233}U source with thickness of 20 μg/cm^2 and area 20 cm^2 would have an activity of 4 μCi producing a source of ^{229}Th atoms at a rate of 1.5×10^5 atoms per second. A source with similar dimensions containing ^{239}Pu (instead of ^{233}U) would have an activity of 40 μCi (the half-life is 10 times shorter) and would yield 1.5×10^6 ^{235m}U ions per second. The decay of the ^{235m}U isomer is used to optimize the experimental apparatus. Systematic studies of the efficiency of this technique under different operating conditions have been extensively summarized in several references [12-13]. This technique effectively eliminates the background noise from the alpha decay of the ^{233}U source. The "noise" arises from the 5 MeV alphas emitted by the ^{233}U source as it decays which can liberate electrons from any material they strike and also cause fluorescence of most materials. By collecting only the recoil ^{229}Th atoms, the technique reduces the source noise by 10^5 times. A third stage uses a silicon detector to measure the gradual build up of ^{229}Th on the catcher plate. Another background to consider is the ground state alpha decay of ^{229}Th whose half-life is 7340 years. After an accumulation time of 1 day (assuming 0.4 of 4Pi for solid angle collection) there would be the potential for a 0.9 Hz background from ^{229}Th alpha decay. The "hot atom" technique is effectively a very efficient microscopic isotope recoil separator.

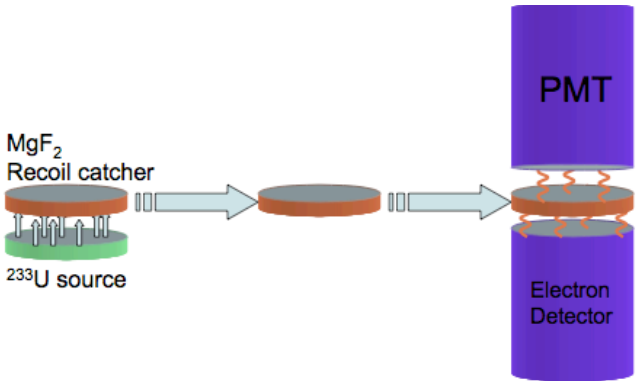

Figure 2: The "hot-atom" technique eliminates the background from ^{233}U alpha decay. The background from ^{229}Th alpha decay (with a 7340 year half-life) is negligible.

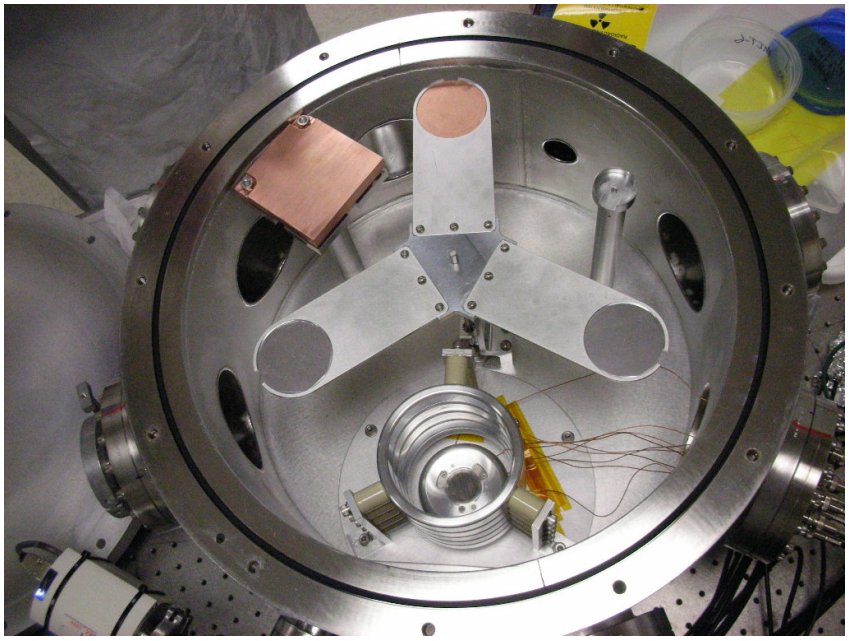

Figure 3. shows the actual vacuum chamber with 3 catcher plates mounted on a rotating stage. The MCP detector is shown at the bottom of the figure with its electrostatic guide. The source is contained under the copper shield in the top left portion of the figure. This shield reduces the background due to alpha particles being emitted from the sample.

2) Expected Rates

Uranium-233 alpha decay populates the ^{229m}Th state ~2% of the time, while ^{239}Pu decay populates the ^{235m}U state ~100% of the time. Using a 10 μCi ^{233}U source, and assuming 25% of the recoils are collected on the catcher plate, the initial ^{229m}Th activity will be 1850 decays/sec. The catcher plate is then moved in front of an electron and/or photon detector. Conversion electrons from this transition, with energies of ~2 eV will emerge only from decays on the surface of the catcher plate but can be accelerated and focused for efficient (~10%) detection using a Multi-Channel Plate detector. Optical and UV-sensitive photomultiplier tubes have quantum efficiencies of 10% and 20% for photons and with a 30% solid angle coverage they would detect photons with ~3% efficiency. Therefore, an initial signal rate of 10-40 decays/sec should be observed.

3) Recent progress

We have already demonstrated that we can efficiently collect and count the conversion electrons from ^{235m}U decay. We have performed several measurements this year to validate this technique using various catcher plate materials. At this point in the research, the most efficient catcher

plate is a thin coating of CsI powder evaporated onto a copper substrate. We have observed initial count rates for ^{235m}U internal conversion electrons in the 3000 Hz regime as shown in Figure 4 below. This is the highest initial count rate ever observed for ^{235m}U decay to our knowledge.

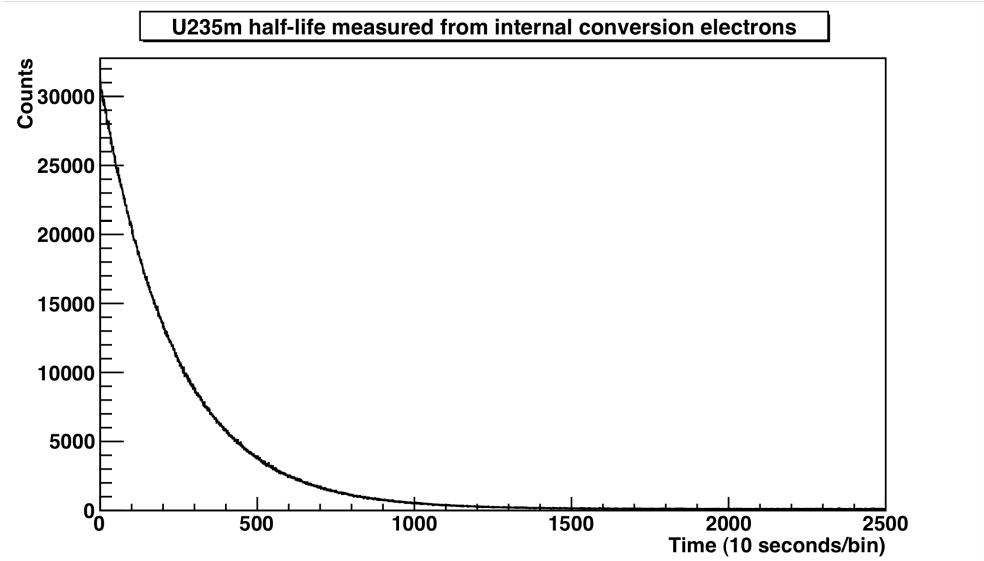

Figure 4 shows the rate of internal conversion electrons following the decay of ^{235m}U populated from the alpha decay of ^{239}Pu using the hot atom technique. Note each time bin is a 10 second integral count of the rate.

The reason CsI coatings have been used as a catcher plate is a result of previous studies have identified it as having a high band gap energy of 6.2 eV and its very low electron affinity of 0.1 eV [14-15]. For these reasons we expect that an appropriately negatively (-100V to 300 V) biased copper substrate coated with a thin layer of CsI should allow efficient release of the low energy internal conversion electrons following the decay of ^{229m}Th. The current uncertainty in the exact energy of the isomer state leads us to consider other methods of observing the half-life that do not depend on the electrons escaping from the catcher plate surface.

4) Optical photons following internal conversion and bound internal conversion decay

If the energy of the ^{229}Th isomer happened to be just below the ionization energy of the ^{229}Th nucleus then internal conversion would be shut off. This still leaves bound internal conversion as a decay channel for the nucleus. In this process electrons from lower orbitals in the valence structure are excited to higher bound states. The subsequent decay of these levels back to the ground atomic state would

emit optical and UV photons for observation. This process is similar yet distinct from x-ray emission following the internal conversion decay of more energetic nuclear states (>3 keV).

We have recently observed the half-life of ^{235m}U using photomultiplier tubes (PMTs) that have optical response sensitivities. To our knowledge this is the first observation of low energy photons emitted following internal conversion decay of a nucleus. The samples were prepared the using the same method as described for the detection of internal conversion electrons. Instead of rotating the sample in front of the MCP detector, the sample was placed in front of an optically sensitive PMT. The background in the PMT (Hamamatsu R980) was approximately 500 Hz prior to the rotation of the sample. Once the ^{235m}U catcher plate was rotated in front of the PMT, the count rate increased to 30000 Hz immediately and follows two distinct decay half-lives, see Figure 5. There are no known contaminants in the sample that can produce this rate. We are vigorously investigating and confirming the origin of this signal.

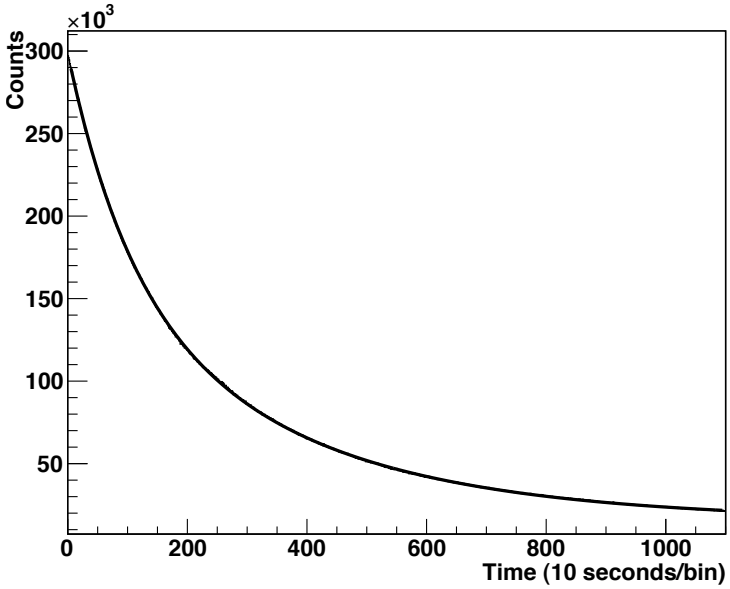

Figure 5 shows the half-life curve observed using an optical photon Hamamatsu R980 PMT with a wavelength response from 300nm to 650nm. The decay curve has been fit to two exponentials plus a background and yields a half-life of 130 minutes and 437 minutes respectively.

The process can be described in analogy to the x-rays observed following internal conversion of higher (>10 keV) energy levels in nuclei. Instead of the inner electron shells coupling to the nucleus to allow the decay to proceed, the outer valence electron shells couple to the nucleus. Following the ejection of an outer valence electron from its orbital, the atomic electron shells reorganize themselves very quickly to accommodate the sudden hole created in the valence electron configuration. Upon filling this hole state,

multiple optical photons may be emitted as the higher lying electrons cascade down to fill the hole. The energy spectrum of these photons would yield the information about which orbitals were involved in the internal conversion process. The relative intensities of the photon spectra would tell us the probability of the transitions being involved in the internal conversion process. The same argument applies if the process is governed by bound internal conversion. Uranium having a [Rn] $5f^3$ $6d^1$ $7s^2$ valence structure and the ionization energy of outer valence electrons is 6.05 eV, 14.7 eV, and 19.1 eV respectively would require the use of a EUV spectrometer that we have at LLNL.

5) Estimates of the ^{229}Th half-life

The search for the ^{229m}Th decay is complicated by the fact that the half-life could be anywhere from microseconds to multiple hours long. We envision a typical duty cycle of collecting ^{229m}Th recoils on the catcher plate for three half-lives followed by shuttling it in front of the electron and photon detectors to look for the decay of the signal for five half-lives. This cycle will be repeated until sufficient statistics are collected to observe the decay curve. We plan to conduct these measurements optimized for half-lives ranging from seconds to many hours. Mechanical movement of the catcher plate will limit the accessible half-lives to >500 ms. If we find that the limit of the half-life is below 500 ms then we will use a modified version of the "hot atom" technique.

6) Modified "hot atom" technique

In this method we allow the recoils to fly clear of the source and guide them electrostatically down a flight tube. The ions are similarly caught on a catcher plate as before where the MCP and PMTs can observe their decay. The technique differs in that we can shunt the electric guide field so that the ions are directed away from the catcher plate in microseconds. This will allow us to produce a sample and observe its decay with microsecond timing resolution between creation and decay. There are currently various vacuum flight tubes available for this approach in the current laboratory. Construction of a suitable ion guide and shunt system would need to be undertaken. SIMION, an electrostatics and ion kinematics software program would be used to design an appropriate ion guide system. SIMION has already been used successfully to design the MCP electrostatic guide.

If it is found that ~1% or more of the of the ^{229m}Th decay by photon emission (as opposed to electron emission) then the energy of the doublet can be accurately determined by a spectroscopic measurement of the emitted photons with an energy resolution of about 1 part in 1000. A spectroscopic measurement of the energy would be performed as above except the ejected ^{229m}Th would be collected

onto a line or point. After several half-lives of collecting, the ^{229m}Th recoils would be transported to a UV spectrometer where the energy of the emitted photons would be accurately measured.

Atomic-Nuclear Interaction Physics Theory

The internal conversion rate in ^{229m}Th comes from the coupling of the nucleus and the 7s1/2 valence electrons. Assuming the 7.5 eV energy of the isomer is correct and using the knowledge that the transition is M1 (3/2+ → 5/2+), binding energy of 5.3 eV and the internal conversion electron energy is then 2.2 eV the internal conversion coefficient is 1.95×10^{9}. The bare nuclear photon decay rate is also calculated to be 76 seconds from the Weisskopf model. The corresponding natural line width of this state is 8.7×10^{-18} eV. The internal conversion rate shortens the observed half-life to approximately 12 microseconds.

We are using this value as the lower bound on the ^{229m}Th half-life currently. The half-life can change dramatically depending on the exact energy of the nuclear transition. There is the Beck measurement of 7.6 ± 0.5 eV and that of the revised value that Helmer and Reich in [1] determined by Guimaraes-Filho and Helene at 5.5 eV [16]. The current research apparatus allows us to set an upper limit of approximately 0.5 seconds on the ^{229m}Th half-life. Our recoil separation and catcher plate apparatus would be modified to allow for a time of flight preparation of the sample if we do not observe a signal at longer times.

^{229}Th as a Precision Nuclear Clock

The doublet between the ground and first excited states of the ^{229}Th nucleus is the only nuclear transition suitable for potential as a frequency standard for precision clocks. A nuclear transition is better suited than any atomic transition for these applications because of its insensitivity to external perturbations. Nuclear transitions are typically too high in energy to be accessed with conventional laser spectroscopy. The doublet between the ground and first excited states of the ^{229}Th nucleus is unique because it is a nuclear transition that could be manipulated using table-top lasers – it has an energy splitting of only 7.6 ± 0.5 eV (163 ± 11 nm) [2], a spin difference of 1 h-bar, and an excited-state half-life that could be as long as hours.

The concept for a clock based on the ^{229}Th nuclear doublet has been proposed by Peik & Tamm [3]. Their design is similar to the clock research being conducted at NIST in Boulder, Colorado [17]. The NIST researchers use atomic transitions for their frequency standard. In [3] the ^{229}Th nuclear doublet transition is the frequency standard while atomic transitions are used to cool the $^{229}Th^{3+}$ ions and for

probing the state of the ^{229}Th nucleus. Recently, Campbell *et al.* [4] have shown they can trap $^{232}Th^{3+}$ ions. This is the first time anyone has been able to trap and laser cool and multiply charged ion. It is a very important and necessary step on the way towards developing the world's first precision nuclear clock.

The primary benefit of the ^{229}Th nuclear clock would be an estimated 1000 time improvement in the precision to which we can measure the passage of time. The main improvements of the precision come from the reduced size of space that the nucleus samples compared to that of an atomic or ion clock. The nuclear dimension being approximately 10^5 times smaller than that of atomic dimensions, for example, reduces the effect non-uniform magnetic fields have on the DC and AC Zeeman effect [17]. This is not to say that new subtle effects may present themselves as this technology and research is developed and we better understand the nuclear-atomic interaction field of physics.

Is the Fine Structure Constant a Constant of Nature?

A fundamental question that one can ask about nature is: "Are the constants of nature constant in time?". In order to address this question the NIST team has recently performed an elegant experiment comparing two ion clock frequencies over the period of approximately one year. The NIST team compared an Al^+ and Hg^+ single ion optical clocks with uncertainties in the frequencies in the 17th decimal place. Using the ratio of the two frequencies they find that the fine structure constant (α) had a linear slope of $\Delta\alpha/\alpha = (-1.6 \pm 2.3) \times 10^{-17}$/year. The result is consistent with zero change in the fundamental constants.

The NIST approach uses leptonic clocks to make the frequency comparison to test the change in the fine structure constant over time. The ^{229}Th clock would instead be a baryonic clock and couple to the strong force of the nucleus. Two separate theorists have predicted that the nuclear clock would enhance the rate of change observable in the fine structure constant by 10^4 and 10^5 times [5,6,18].

A key goal of this research proposal is to determine if ^{229}Th can be turned into a nuclear clock with suitable precision to be able to test whether or not the fine structure constant is truly constant in time. This is Nobel Prize class experiment would alter our view of cosmology and the fundamental forces of Nature.

Summary

The research discussed here entails the development of a new field of physics. Thorium-229 has the lowest known first excited state at 7.6 eV. Understanding its behavior and decay mechanisms will lead to insights in the field of nuclear-atomic coupling or excited state physics. The unknown quantities that need to be measured to understand this nucleus are its half-life and exact wavelength. The research presented here involves a multi-disciplinary approach involving atomic-nuclear theory, nuclear experiment, atomic spectroscopy and a suite of detector measurements including photon counting, electron counting, optical spectroscopy and EUV spectroscopy. The ability to directly control a nuclear state with an external laser will open up a completely new opportunity for the basic science community. We should be able to develop the world's first nuclear clock, which will have unprecedented precision. We will follow on this research by investigating the stability of the fine structure constant. Collaboration between LLNL, NIST Boulder and Georgia Tech will help this final step in the discovery process. The implications of which could change our fundamental understanding of the universe and its evolution.
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