Observation of the Deexcitation of the ^{229m}Th Nuclear Isomer

Xinxin Zhao, Yenny Natali Martinez de Escobar, Robert Rundberg, Evelyn M. Bond, Allen Moody, and David J. Vieira

Phys. Rev. Lett. **109**, 160801 — Published 18 October 2012

DOI: 10.1103/PhysRevLett.109.160801

Observation of the de-excitation of ^{229m}Th nuclear isomer

Xinxin Zhao, Yenny Natali Martinez de Escobar, Robert Rundberg, Evelyn M. Bond, Allen Moody, and David J. Vieira
Los Alamos National Laboratory, Los Alamos, NM 87545, USA

The ^{229}Th nucleus possesses the lowest-energy nuclear isomeric state. Two widely accepted indirect measurements of the transition energy place it within reach of existing laser capabilities. Direct searches for the isomer de-excitation have proven elusive despite extensive effort over the past couple of decades. There is now a growing interest in finding this unique transition because of its potential applications in nuclear, atomic, condensed matter and optical physics, quantum information, metrology and cosmology, including the development of a new type of clock based on this nuclear transition. In this work we report the first direct observation of the de-excitation of the lowest-lying isomeric state in ^{229}Th. By collecting ^{229}Th recoils following the alpha decay of ^{233}U into MgF_2 plates and measuring the subsequent light emission, we have isolated the isomer de-excitation and measured the transition's half-life to be 6±1 hours. Through comparison measurements with ^{235m}U isomer, we found that the observed ^{229m}Th de-excitation signal originates from photon emission rather than internal conversion (IC) electron emission. This discovery lays the ground work for optical and laser spectroscopy of ^{229m}Th nuclear isomer and the development of a ^{229}Th nuclear clock.

PACS numbers: 06.30.Ft, 21.10.Tg, 23.20.Lv, 27.90.+b

The existence of an extremely low-lying nuclear state in ^{229}Th has been deduced from nuclear spectroscopy since the 1970's [1]. Early studies [2] indirectly determined the energy of the nuclear isomeric state in ^{229}Th to be 3.5±1.0 eV, and suggested that such a nuclear state could be excited with lasers. Since then several direct searches have failed to find the isomer de-excitation [3-6]. Recently, researchers performed an improved indirect measurement [7, 8] and placed the isomer energy at 7.8±0.5 eV with an estimated half-life $t_{1/2}$ of 5 hours for the γ de-excitation channel based on energy scaling of a known M1 transition involving the same Nilsson levels in ^{233}U.

A direct observation of the ^{229m}Th γ de-excitation would open up many exciting possibilities in fundamental and applied research, including the development of a "nuclear" clock [9-13]. The inferred transition wavelength (160±10 nm) from the most recent measurement [8] is within the range of high harmonic lasers [14-16], making laser spectroscopy of the nuclear isomer possible. Moreover, a ^{229m}Th nuclear clock is estimated to have a quality factor of $Q = f/\Delta f \sim 10^{20}$, and be inherently more robust than atomic transitions with respect to variations in the environment. In addition, similar to testing the temporal variation of the fine structure constant

using atomic transitions [17], the ^{229m}Th isomeric transition could probe the relative temporal variation of the electromagnetic and strong interactions [18] which unification theories predicted to exist in an expanding Universe [19]. This letter describes the direct observation of the ^{229m}Th nuclear isomer de-excitation.

^{233}U ($t_{1/2}$ = 1.592 x 10^5 yr) undergoes alpha (α) decay, resulting in ^{229}Th ($t_{1/2}$ = 7400 yr) recoils with a maximum energy of 83 keV and ~2% branching to the ^{229m}Th isomeric state. Fig. (1a) illustrates the relevant energy levels and decay mechanisms for the isomer transition. We collect ^{229}Th α-recoils from eight ^{233}U source plates on a set of four MgF_2 plates (2 cm x 2cm x 1 mm from Almaz Optics) as depicted in Fig. (1b). The $^{233}UO_2$ samples were electroplated [20] onto gold plated stainless steel plates. Each sample is 2 cm in diameter and nearly transparent. SRIM calculations show that 83 keV ^{229}Th recoils travel about 15 nm (or an areal thickness of 17μg/cm^2) in uranium oxide (UO_2) which sets an optimal total ^{233}U activity at ~4.5 μCi for our experiment. After a suitable collection period, we move the MgF_2 plates close to a photomultiplier tube (~ 3 mm away from the PMT window) and measure the light emission as a function of time. We used two PMTs (Hamamatsu R8487 and R8486) to search for the isomer emission and to determine its spectral range. The PMT are cooled to 10.0±0.2 °C and their quantum efficiencies are shown in Fig. (1c). For a 25 cm^2 total surface area of 4.5 μCi UO_2, we estimated a ^{229m}Th implantation rate of ~1000 isomers/s and PMT signal on the order of 1 Hz if the isomer de-excites through a photon emission. We assumed 25% recoil implantation efficiency, 1% optical coupling to the PMT sensor (~1.7 cm away from the PMT window), and 10% PMT QE. The system is enclosed in a vacuum chamber (~2x10^{-7} Torr) to eliminate air ionization and optimize vacuum ultra-violet (VUV) light detection. Our ^{233}U sources also contain 8 ppm of ^{232}U ($t_{1/2}$ = 69.8 yr) which decays to ^{228}Th ($t_{1/2}$ = 1.9 yr).

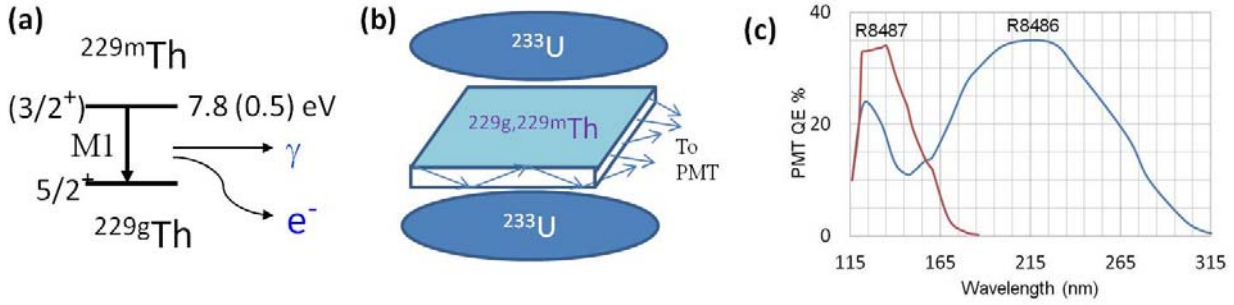

FIG. 1. ^{229}Th nuclear energy levels and experimental details. **(a)** Ground state and lowest isomeric state of ^{229}Th showing the two possible decay channels for the isomer: γ-decay and

internal conversion (IC) electron emission. (**b)** Schematic of experimental setup used to measure the γ–decay of the ^{229m}Th isomer: ^{229}Th recoils from ^{233}U α-decay are implanted into MgF_2 plates for several hours. The ^{233}U sources are then removed and the light emission is measured with a PMT. (**c)** Quantum efficiencies (as provided by the manufacturer) for Hamamatsu PMTs R8487 and R8486.

Several processes emit light after recoil implantation into the MgF_2 plates from the ^{233}U source: (1) MgF_2 phosphorescence after alpha and beta radiation exposure [21]; (2) MgF_2 scintillation from the radioactive decay of implanted $^{228,\ 229}$Th and their daughters; (3) fast decaying atomic fluorescence associated with the recoil implantation; and (4) ^{229m}Th γ-ray emission, if it exists. Earlier work on MgF_2 phosphorescence after beta (β) radiation exposure [21] demonstrated that it consists of a broad emission spectrum, with the shortest wavelength band peaking near 225 nm. We therefore used PMT R8487 to search for the isomer emission while minimizing the phosphorescence background. MgF_2 light emission was first measured after collecting recoils from a ^{232}U source in order to study non-isomeric processes. The ^{232}U source has an activity of 3.6 μCi and 99% of its decay daughters were chemically removed beforehand. Fig. 2 shows the MgF_2 emission (red dots) measured after implanting recoils from the ^{232}U source for 80 minutes. The emission rises initially and subsequently evolves into a double exponential decay with the half-lives of ^{212}Pb ($t_{1/2}$ = 10.64 hr) and ^{224}Ra ($t_{1/2}$ = 3.63 day). The initial rise indicates that the light emission results from Cherenkov radiation produced by high-energy β particles (2.25 MeV) from ^{212}Bi ($t_{1/2}$ = 1.009 hr) decay. ^{212}Bi does not implant efficiently but grows-in from implanted ^{212}Pb and eventually reaches secular equilibrium [22] with the ^{212}Pb and ^{224}Ra present in the MgF_2.

To eliminate the scintillation fluorescence associated with $^{232,\ 233}$U decay daughters when searching for the ^{229}Th isomer, we used a 4.5 μCi ^{233}U source (also containing 8 ppm of atomic ^{232}U) that was chemically purified to remove more than 99.99% of the U daughters. The uranium source was alpha counted right after the chemical purification to determine the activities of the daughters. The residue activities of the daughters (<0.2 nCi) come from the ^{232}U decay chain with negligible amount (non-detectable) from the ^{233}U decay daughters due to the long half-life of ^{229}Th. We found no measurable emission signal after a recoil implantation time of 80 minutes (as shown in green dots in Fig. 2). An emission signal of ~50 mHz with a decay time constant consistent with the 60 hr phosphorescence component [21] was observed only until the

collection period was increased to 31 hr. We therefore conclude that there is no isomer emission between 115-160 nm (see Fig. 1c).

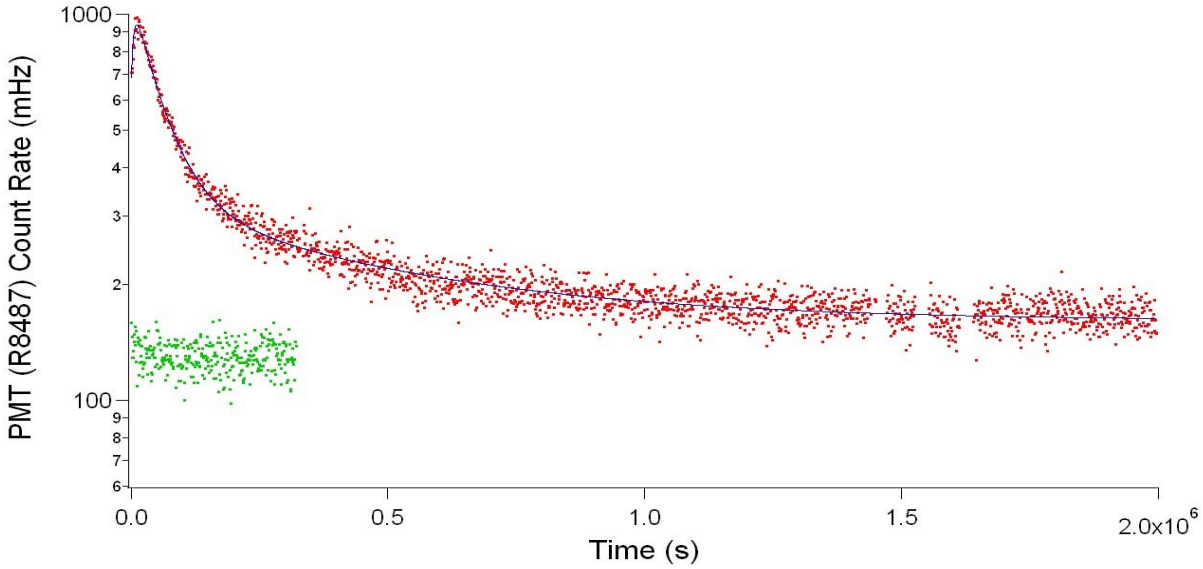

FIG. 2. Results with R8487 PMT. Emission decay curve measured after an 80 minute collection period (red dots) and the fit (blue line) using a ^{232}U source. The data fits well to the function: $f(t) = A_0 + A_{Pb}\,(e^{-t/\tau_{Pb}} - 0.684\, e^{-t/\tau_{Bi}}) + A_{Ra}\, e^{-t/\tau_{Ra}}$, where the lifetimes ($\tau = t_{1/2}/\ln 2$) of all radio nuclides are held fixed, and the amplitude ratio of ^{212}Bi to ^{212}Pb was calculated from the implantation time t_i using a derived formula [22]: $-\frac{\tau_{Bi}}{\tau_{Pb}}\left(\frac{1-e^{-t_i/\tau_{Bi}}}{1-e^{-t_i/\tau_{Pb}}}\right)$. Emission measured after 80 minute (green dots) collection period using a chemically purified ^{233}U source. The data shows no ^{229}Th isomer emission detected with PMT R8487.

We then proceeded with measurements using PMT R8486. To effectively eliminate scintillation fluorescence and study phosphorescence more carefully, we block the low-energy recoil ions (~50 nm path length) by covering the ^{233}U source with aluminized Mylar foil (2.2 μm thick) during an implantation period. Comparing phosphorescence measurements performed with single and double layers of Mylar foil established that a scaling factor of 1.15±0.01 compensated for single layer Mylar foil attenuation. By measuring the phosphorescence before and after each successive recoil ion implantation, we found that the phosphorescence efficiency of MgF_2 increased with thorium doping but the enhancement rate is reduced after ~10 hours of ^{229}Th exposure. We therefore pre-exposed the MgF_2 plates for ~10 hours to a ^{233}U source before undertaking isomer search measurements as discussed below. Small temperature modulations of the phosphorescence were controlled by stabilizing the vacuum chamber temperature.

We then searched for the ^{229m}Th isomer emission with a series of 3.5 hr implantation

measurements using a chemically-purified 4.2 μCi ^{233}U source. The phosphorescence background with Mylar-covered ^{233}U source was measured before and after each ion implantation period with judicious care taken to match the experimental conditions, i.e. position and temperature of the uranium and MgF_2 plates. A set of three measurements are shown in Fig. (3a), where emission after ^{229}Th implantation is shown in red crosses and the phosphorescence measured given as blue and green dots. Since the increase in phosphorescence after each thorium implantation was small, we averaged the two background measurements after attenuation scaling to establish the phosphorescence background. After subtracting off this background, we obtained the net signal associated with the implanted recoil ions as shown in black dots in the Fig. (3a). We fit the data to an exponential function to obtain its amplitude and half-life. Repeating these measurements revealed that the net signal increased with the age of the ^{233}U source because of the in-growth of ^{232}U decays daughters, as shown in Fig. 3b. The data shows that the net amplitude increases at a rate of 1.2±0.4 mHz per day and has an initial amplitude of 228±22 mHz when extrapolated to zero in-growth time. The half-life of the net signal, also increases slowly with the age of ^{233}U source because of the increasing contributions from ^{212}Pb and ^{224}Ra, has an extrapolated value of 6±1 hr at zero in-growth time (shown in Fig. 3c). Measurements using several different ^{233}U sources show that the net signal amplitude at zero in-growth time depends on the thickness and uniformity of the ^{233}U sources. The purity of every sample was checked after chemical purification using α-spectroscopy analysis to rule out the contribution of daughters. It was critical to prepare a thin and uniform source for good recoil collection efficiency

The net signal at zero in-growth time has three possible origins: emission of the isomer γ-ray, MgF_2 scintillation from emitted isomer IC electrons, and MgF_2 phosphorescence associated with recoil implantation. To investigate the contributions of the latter two processes, we made MgF_2 emission measurements using a 4.9 μCi ^{239}Pu source in the same setup. ^{239}Pu α-decays (5.1 MeV in energy, $t_{1/2}$=2.41x10^4 yr) with a ~100% branching ratio to the ^{235m}U isomer (E^*=65 eV), which de-excites entirely through IC electron emission with a half-life of ~26 minutes [23]. Phosphorescence emission of similar decay profile to that present with the ^{233}U source was observed, but no net optical signal was detected after a 3.5 hr implantation period, as shown in Fig. (3d). We therefore conclude that the observed ^{229}Th implantation signal is from ^{229m}Th γ-ray emission. Combined with the null PMT R8487 measurement, we conclude that the emission

wavelength is at least longer than 160 nm. For this ultra-violet γ-ray emission, it is possible that the lifetime and the signal amplitude could be changed by the "super-radiance" effect [24].

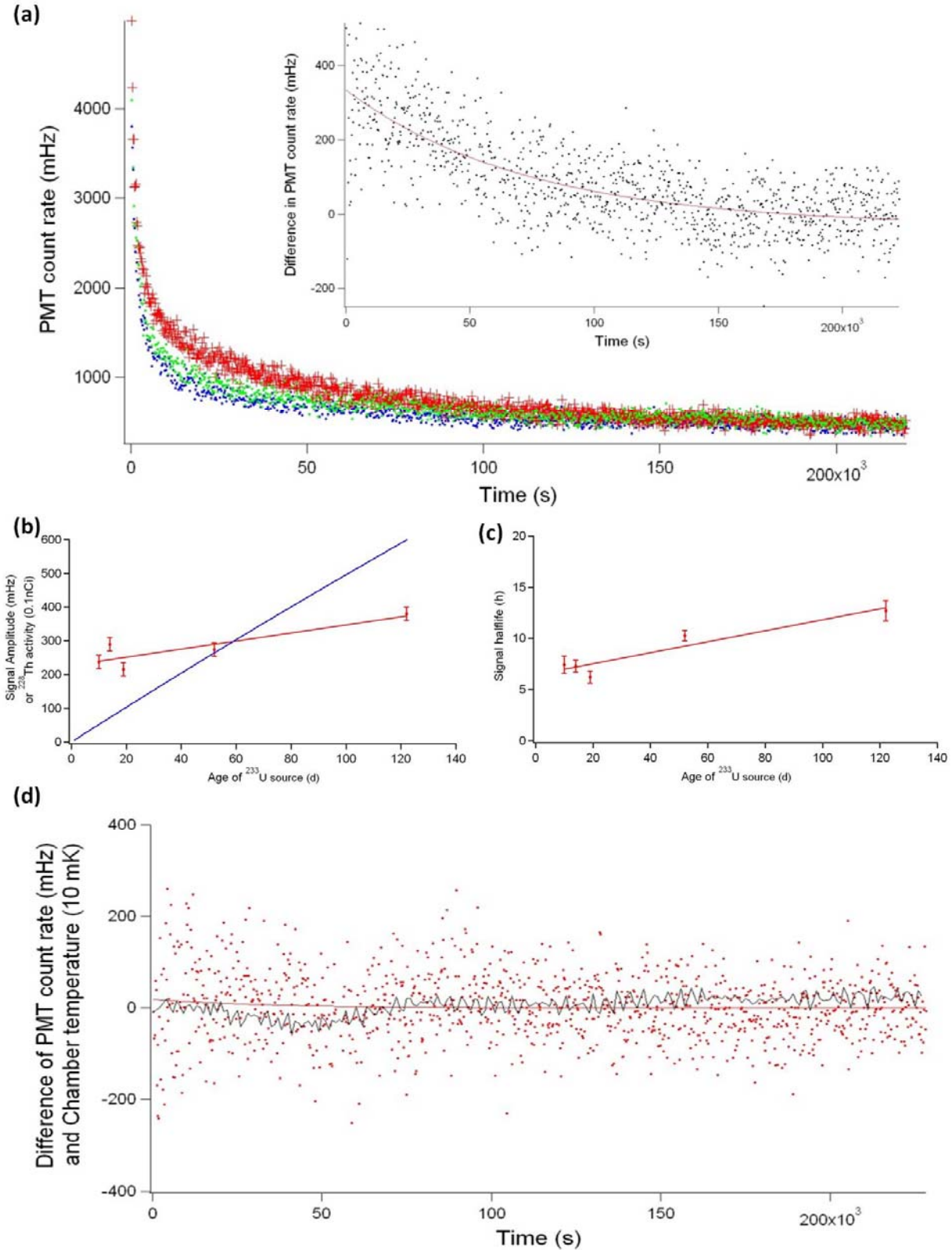

FIG. 3. Evidences of ^{229m}Th γ-ray emission. (a) Emission decay curves measured with PMT R8486 after a 3.5 hour collection period using a 4.2 μCi ^{233}U source (red crosses) and with Mylar foils used as a recoil filter (blue and green dots are phosphorescence data before and after ion implantation, respectively). The PMT signal difference (with the phosphorescence amplitude scaled by 1.15 and then subtracted out) is shown in the inset as black data points. (**b**) Signal amplitude (red) for 3.5 hour implantation period and ^{228}Th activity in the ^{233}U source (blue line) as a function of source age. (**c**) Signal half-life as a function of source age; linear fit to zero source age yields a half-life of 6±1 hours. (**d**) The averaged PMT signal difference for two 3.5 hour recoil ion implantation measurements using a 4.9 μCi ^{239}Pu source (red dots) and the temperature difference for this measurement (black line). The solid red line is an exponential fit with an amplitude of 18±20 mHz using a fixed 6 hour half-life. For a fixed 26 minute half-life, the fit amplitude is -74±50 mHz.

With the direct observation of the ^{229m}Th de-excitation and evidence that it is a photon emission, we expect exciting advances in optical and laser spectroscopy of ^{229m}Th nuclei and the development of a ^{229}Th nuclear clock.

We thank Anna Hayes, Carol Burns, Todd Bredweg, William Priedhorsky, and Marianne Wilkerson for discussions and support, Alice Slemmons for help on ^{233}U sample preparation, Stosh A. Kozimor and Kevin Boland for preparing ^{232}U solution, Donald Dry for alpha-spectroscopy analysis of U and Pu sources. We thank Justin Torgerson and Richard Greco for their effort to measure MgF_2 phosphorescence in a separate ion beam setup. The work is supported by the Laboratory Directed Research and Development program at Los Alamos National Laboratory, operated by the Los Alamos National Security, LLC for the National Nuclear Security Agency (NNSA) as part of the U.S. Department of Energy under contract No. DE-AC52-06NA25396.

[1] L. A. Kroger, C. W. Reich, Nucl. Phys. A **259**, 29-60 (1976).
[2] R. G. Helmer, C. W. Reich, Phys. Rev. C **49**, 1845-1858 (1994).
[3] G. M. Irwin, K. H. Kim, Phys. Rev. Lett. **79**, 990-993 (1997).
[4] D. S. Richardson *et al*., Phys. Rev. Lett. **80**, 3206-3208 (1998).
[5] S. B. Utter *et al.*, Phys. Rev. Lett. **82**, 505-508 (1999).
[6] R. W. Shaw *et al*., Phys. Rev. Lett. **82**, 1109-1111 (1999).
[7] B. R. Beck *et al*., Phys. Rev. Lett. **98**, 142501 (2007).
[8] B. R. Beck *et al*., *Proc. of the 12th Inter. Conf. on Nucl. Reaction Mechanisms*, Varenna, Italy. Cerutti, F. & Ferrari, A., ed. Vol. **1**, 255-258 (2010).
[9] E. Peik, C. Tamm, Europhys. Lett. **61**, 181-186 (2003).
[10] C. J. Campbell *et al.,* Phys. Rev. Lett. **102**, 233004 (2009).
[11] W. G. Rellergert *et al*., Phys. Rev. Lett. **104**, 200802(2010).
[12] C. J. Campbell, A. G. Radnaev, and A. Kuzmich, Phys. Rev. Lett. **106**, 223001 (2011).
[13] C. J. Campbell *et al*., Phys. Rev. Lett. **108**, 120802 (2012).
[14] T. Kanai *et al.,* J. Opt. Soc. Am. B **21**, 370-375 (2004).

[15] R. J. Jones *et al.*, Phys. Rev. Lett. **94**, 193201 (2005).
[16] C. Gohle *et al.*, Nature **436**, 234-237 (2005).
[17] T. Rosenband *et al*., Science **319**, 1808-1812 (2008).
[18] V. V. Flambaum, R. B. Wiringa, Phys. Rev. C **79**, 034302 (2009).
[19] J. P. Uzan, Rev. Mod. Phys. **75**, 403-455 (2003).
[20] E. M. Bond *et al.,* Nucl. Chem. **276**, 549 (2008).
[21] W. Viehmann *et al.*, Applied Optics **14**, 2104-2115 (1975).
[22] G. Friedlander *et al.*, *Nuclear Radiochemistry*, ISBN 0-471-28021-6, pg. 200 (1981).
[23] J. R. Huizenga, C. L. Rao, C, and D. W. Engelkemeir, Phys. Rev. **107**, 319-320 (1957).
[24] R. H. Dicke, Phys. Rev. **93**, 99-110 (1954).