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We used two methods to attempt to measure the lifetime and energy level of the ^{229m}Th low-lying nuclear state. Both methods used a ^{233}U source to implant ^{229}Th into a VUV transparent substrate. The uranium source had a low concentration of ^{232}U (0.08 ppm) and was chemically purified one week prior to the start of the experiment. In the first method, the time decay method, sensitive to ^{229m}Th lifetimes from 10 minutes to 15 hours, the VUV substrate was irradiated for 4 to 36 hours to ensure a sufficient transfer of ^{229m}Th ions into the substrate. Then the signal from the substrate was observed using a PMT for 15 hours. A bandpass filter was placed between the substrate and PMT to provide a means of measuring the ^{229m}Th photon energy. We used nine filters with center transmissions ranging from 140 nm to 244 nm. In the second method, direct detection, sensitive to lifetimes from zero to 10 minutes, the substrate was observed directly with the PMT and filter during the deposition process. For both methods our results were limited by the strong background fluorescence and phosphorescence due to the alphas and betas intrinsic to the decay of the ^{233}U and its daughters. We did not see unambiguous signs of photons from ^{229m}Th. This may be due to one or more of the following: lifetime of ^{229m}Th too short or too long, branching ratio to produce ^{229m}Th from ^{233}U is too small, energy of ^{229m}Th transition is outside of our observed range of 5.1 to 8.9 eV, and/or most likely, ^{229m}Th ions drop to a +1 or 0 charge state, and thus lose their nuclear excitation through non-radiative processes.

Since 1975 when Kroger and Reich [1] first reported the $I^{\pi}=3/2^{+}$ band head in ^{229m}Th was located approximately 0.1 keV above the $5/2^{+}$ ground state, a search has been underway to both confirm the existence of the low lying ^{229m}Th transition and to isolate the energy and measure the lifetime of the transition. To aid the search for the energy of the transition Beck, et al. [2] conducted a high precision measurement of the ^{229m}Th gamma ray emission and indirectly determined the energy of the transition to be 7.6(5) eV. Subsequently, Beck, et al. [3] further refined this indirect measurement and reported the energy to be 7.8(5) eV. Recently, the existence of the ^{229m}Th transition was confirmed by Wense, et al. [4] who reported the transition energy to be in the range of 6.3 to 18.3 eV. They also reported the lifetime of the transition to be longer than 60 seconds for the $^{229m}Th^{2+}$ ion. Zhao, et al. [5] reported a direct detection of the ^{229m}Th nuclear VUV photon by fitting their data to a single exponential. This result was subsequently disputed by Peik and Zimmermann [6], due to the presence of ^{212}Pb decays. While Zhao has probably been discredited for the detection of the ^{229m}Th nuclear VUV photon, Zhao's experimental procedure appeared to be sound. In our preliminary analysis for this experiment, we used the data from Fig. 3 of Zhao [5] and fit them to three exponentials. One of the exponential lifetimes was fixed to the ^{212}Pb 1/e lifetime of 921 minutes which showed clearly in Zhao's data, and the other two exponentials were allowed to float. [Throughout this paper we use 1/e lifetimes = (half-life)/ln(2).] The result of the three exponential fit to Zhao's data revealed a rapid decay phosphorescence signal with a lifetime of about 20 minutes and an additional signal with a count rate approximately 1 count/s and a lifetime of 102 minutes. This 1 count/s rate was consistent with Zhao's expected count rate, and 102 minute lifetime was consistent with the wide range of expected lifetime of ^{229m}Th, thus indicating the potential for a positive detection of the ^{229m}Th nuclear VUV photon. The failure of Zhao appeared to be driven by the high concentration 8 ppm of ^{232}U contained in their ^{233}U sample, which led to an exceedingly large ^{212}Pb signal.

We have used an apparatus similar to Zhao, et al. [5] with some improvements. A major improvement is to use a ^{233}U sample with a ^{232}U concentration of less than 0.1 ppm. Another improvement was to use bandwidth filters to allow determination of energy of the VUV decay photon. We were not able to detect VUV decay photons from ^{229m}Th between 140 - 244 nm.

Our ^{233}U source consists of two flat stainless steel one inch planchets electrodeposited with uranium oxide. The ^{233}U was purchased from the National Isotope Development Center at Oak Ridge National Lab (ORNL), from the UTHX001 material, which has a ^{232}U impurity of less than 0.1 ppm. Prior to deposition, the uranium was chemically purified at Pacific Northwest National Lab (PNNL) to remove any daughter products. Optima grade hydrochloric and nitric acids were used during the preparation of the ^{233}U sources. The ^{233}U sample obtained from ORNL was converted to 3M HNO_3 and then loaded onto two 2 mL Eichrom® Technologies, Inc. UTEVA resin cartridges (50-100 μm) in series using a vacuum box setup. The UTEVA cartridges were conditioned with 3M HNO_3 prior to sample loading. Rinses of 3M HNO_3 were used to quantitatively transfer the sample to the UTEVA cartridges and then additional 3M HNO_3 was used to wash the cartridges. After converting the resin to the chloride form using 9M HCl, a solution of 9M HCl-0.05M oxalic acid was used to elute thorium and the other decay products. The uranium was eluted using 0.1M HCl. An aliquot of the purified ^{233}U was analyzed by gamma spectroscopy to confirm that decay products had been removed from the solution. The electrodeposited planchets were prepared by the methods described in Glover, et al. [7]. Each planchet had 1.5 μCi of ^{233}U deposited in a very flat cylinder of 1.15 cm radius,

from which we calculated an approximately 40 nm thick layer of uranium oxide. We believe only the top 16 nm of the UO_2 layer contributes to any possible Th ion emission, since 85 keV heavy ions such as Th^+ will stop in about 16 nm of solid UO_2, according to an SRIM calculation [8]. Alpha spectrometry was used to confirm the activity of each source.

A simplified schematic of the time decay apparatus is given in Fig. 1 where the essential components are displayed (the top and bottom detectors are essentially duplicate systems and allow two wavelengths to be measured at once). The ^{233}U planchets were placed into a Teflon mount that separated the two planchets by 7 mm allowing unhindered insertion of a 1 to 5 mm thick VUV transparent substrate made of either CaF_2, MgF_2, or Al_2O_3. A substrate was placed between the ^{233}U planchets under 10^{-6} mbar vacuum, (or immersed in 1 atm of Ar), for 4 or more hours to allow $^{229m}Th^{2+}$ (or higher charge state) ions to be deposited and implanted into the substrate. (The deposition in/through Ar would stop Th ions, but still allow background alpha and beta rays to reach the substrate.) The substrates were selected because of their ability to transmit VUV photons for wavelengths greater than 130 nm.

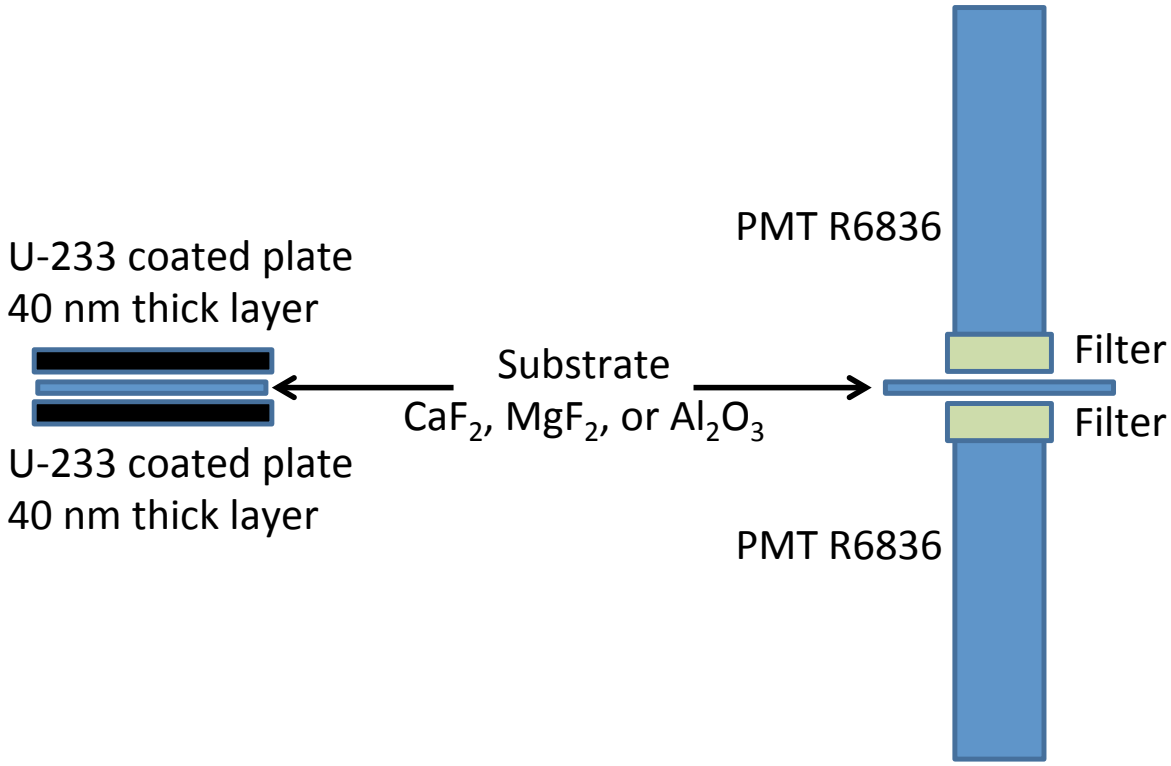

Fig. 1 Simplified diagram of the experimental apparatus. The substrates are implanted with ^{229m}Th through alpha decay of ^{233}U. After some time the substrate is transferred to the detection region, where counts are accumulated as a function of time.

After the deposition into the substrate was completed, the substrate was removed from the U and placed between two Hamamatsu R6836 end-on PMTs. During the 5 to 10 minute transfer from deposition chamber to PMT chamber, the substrates were exposed to room air. The R6836 PMTs have a detection range between 115 and 320 nm, and a quantum efficiency of between 9% and 13%, typically 11%, in our wavelength range of 140 to 244 nm, as stated in the Hamamatsu

documentation. End-on PMTs were selected because of their large acceptance angle which increased signal strength 10 times relative to a side-on PMT. Placed between the PMTs and substrate were two Acton VUV optical interference filters (one for each PMT). The center of the bandpass of the filters ranged from 140 to 244 nm with about a 20 nm full width at half maximum. The filters' peak transmission was about 20%. By acquiring data with these filters, the wavelength and thus energy of the ^{229m}Th transition could be located to within about 10 nm. The two signals from the two PMTs were separately amplified using Stanford Research SR445 amplifiers, then fed into two Agilent 53220A frequency counter/timers, which recorded the PMT counts in 10 s bins for up to 15 hours. The PMTs and uranium source deposition were inside a vacuum chamber that typically achieved pressure on the order of 10^{-6} mbar. The entire vacuum chamber and contents, in particular the PMTs inside, were maintained at a temperature of 17 C to prevent thermal fluctuations from affecting the measurements.

The direct detection method is similar to the above, but the bottom PMT and filter assembly is replaced by the ^{233}U planchet (source). In this case the top PMT views the ^{233}U planchet through a substrate and a filter. Also in this method a 3 μm Al foil can be inserted on the ^{233}U source, to block heavy ions, but allow alphas and betas through.

We calculated the expected ^{229m}Th VUV photon signal count rates, using our experimental geometries, the various isotope decay rates, the transmission of the filters, and the detectivity (quantum efficiency as a function of wavelength) of the PMTs. Our maximum possible expected signal rate is calculated as follows: In the 16 nm layer of $^{233}UO_2$, there are about 1.5×10^{17} ^{233}U atoms. Given the decay rate, we expect 2.1×10^4 of the uranium atoms to decay in 1 s to ^{229}Th. Of those decays, we expect 2% of them to be to ^{229m}Th. In the simplest approximate geometry of our experiment, we expect 25% of the ^{229m}Th to get out of the U source and go across to implant in the substrate. Then 50% of those ^{229m}Th will emit a nuclear VUV photon up into the PMT (the other 50% will emit photons away from the PMT). The VUV interference bandpass filters we use have a maximum transmission of 20%. The quantum efficiency of the PMT is 11%. Thus our maximum expected signal is $2.1 \times 10^4 \times 0.02 \times 0.25 \times 0.5 \times 0.2 \times 0.11 = 1.2$ VUV photon counts/s. We expected up to about 1 count/s of ^{229m}Th photons in our apparatus at the start of the time decay (decaying exponentially thereafter), assuming most of the ^{229m}Th would not deexcite in production or deposition. Given these optimistic assumptions, our apparatus would need about 350 ^{229m}Th nuclei to produce 1 measured VUV photon.

This expected signal rate can be compared to the 1 count/s of Zhao et al. [5] Our PMT data collection system had a dark count rate of about 1 count/s, thus the expected 1 count/s of ^{229m}Th photons should have been easily visible, even with some extra background.

The lifetime of the isomeric state ^{229m}Th would affect the expected count rate, however we deposed the substrates (accumulated ^{229m}Th into the substrates from the ^{233}U source) for at least 4 hours, and in some cases for 1.5 days. We assumed or guessed that the ^{229m}Th has a 1/e lifetime of about 6000 s = 1.67 hours in the substrate, thus the number of ^{229m}Th in the substrate would reach a steady state equilibrium by the end of the deposition, (where the number added by the U equal the number decaying, each second). The loading rate of ^{229m}Th into the substrate is given as part of the previous signal calculation: $2.1 \times 10^4 \times 0.02 \times 0.25 = 105$ ^{229m}Th per second. From this, the presumed number of ^{229m}Th in the substrate at steady state is $105/\lambda = 6.3 \times 10^5$ atoms,

where λ = 1/6000 s = assumed decay constant of ^{229m}Th. In case we were underestimating the ^{229m}Th lifetime, we did try several 36 hour long depositions, but did not see any sign of ^{229m}Th for those runs either.

The decay chain from ^{233}U (half-life 1.59×10^5 years = 1/e lifetime 7.25×10^{12} s) produces ^{229}Th (half-life 7917 years = 1/e lifetime 3.60×10^{11} s), then subsequent decays produce daughters down to ^{209}Bi. We calculated the populations and activity of each of these daughters in our system. They all produce alphas or betas, which can produce VUV photons, either prompt or delayed, fluorescence or phosphorescence, by interaction with substrates. We also modeled the decay series for ^{232}U in a similar way, using the expected U isotopic ratio in our highly purified source. In general we found that none of the daughters of either U isotope would be numerous enough to mask our expected ^{229m}Th signal, as long as the ^{233}U source is less than six months after chemical purification. Our data were taken less than one month after purification. In particular, and for example, ^{213}Bi is a daughter of the ^{229}Th decay, will travel as an ion along with the Th ions from the source into the substrates, and has a lifetime (4000 s) similar to that of the expected optical photon decay lifetime of ^{229m}Th. Thus it would mimic the desired decay of ^{229m}Th. We found the ^{213}Bi contamination count rate was negligible relative to the expected ^{229m}Th count rate. ^{212}Pb and ^{212}Bi are daughters of ^{232}U, and also have similar lifetimes to that expected for ^{229m}Th. These also had a negligible effect in our system, due to the high isotopic purity of our ^{233}U source, which has only 0.08 ppm of ^{232}U. However apparently the ^{212}Pb was not negligible in Zhao, et al.'s [5] measurements, and constituted a large fraction of their time decay signal, since their U source had 8 ppm of ^{232}U. (^{232}U has a half-life approximately 2300 times shorter than ^{233}U, thus much larger activity relative to its concentration.)

A typical time dependence of the count rate using a MgF_2 substrate is shown in Fig. 2. The signals for two data collections are presented. In one case, the data were collected after a deposition occurred in a vacuum. Under these conditions, the ^{229}Th ion should be deposited into the substrate and some of those should be in the excited ^{229m}Th state and result in a ^{229m}Th VUV photon signal. For the other case, the deposition occurred in one atmosphere of argon. With the argon, the thorium ions should be blocked and only alpha and beta particles should make it to the substrate. Therefore, by differencing the resulting signals, a clearly visible ^{229m}Th signal should be seen. Unfortunately, it is clear from the difference shown in Fig. 2 that no ^{229m}Th VUV signal is present in the data. We measured many of these decay curves for various substrates, deposition times, and with Th ions not blocked/blocked by vacuum/Ar gas deposition.

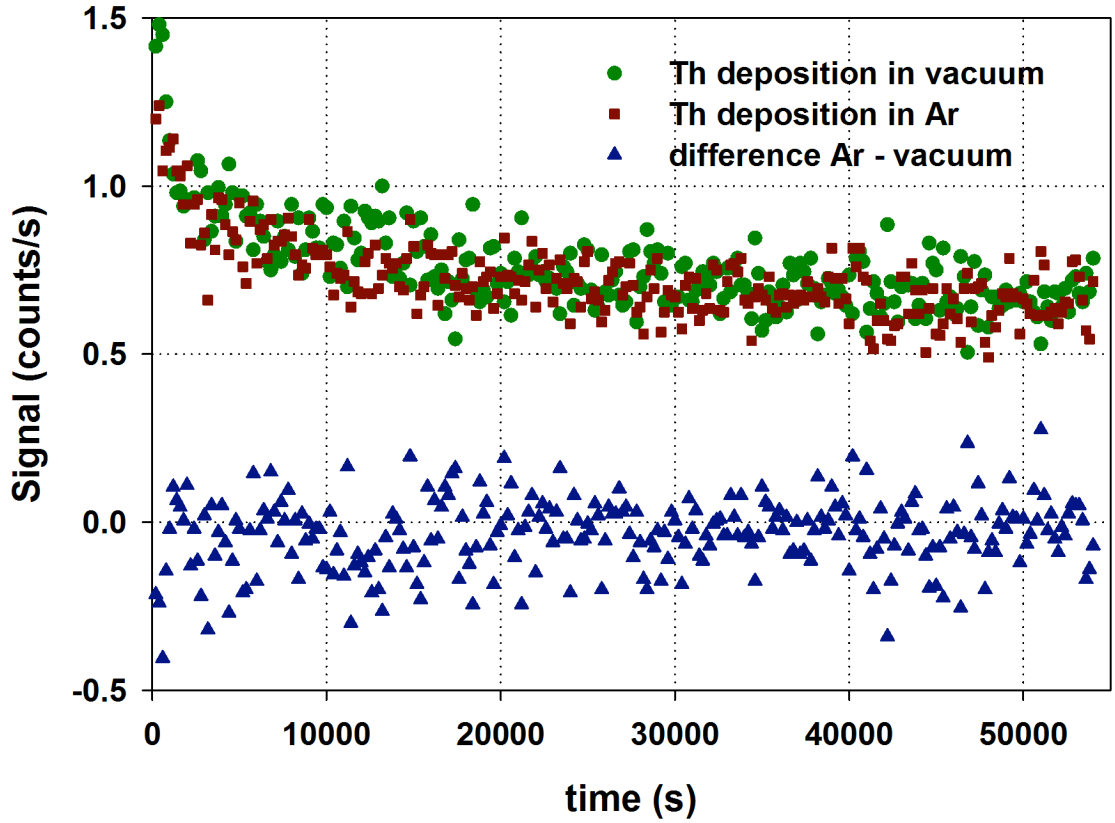

Fig. 2 Typical time decay signal from MgF_2 substrate for the cases where the ^{229}Th deposition occurred in either vacuum (green dots) and argon (red squares). If there were a ^{229m}Th signal it should be visible in the difference argon minus vacuum of the two signals (blue triangles). [These data were acquired in 10 s bins, but were put into 200 s bins for the purposes of this figure.]

We fit these decay curves to single and double exponentials to extract amplitude and decay time values, which would presumably be the count rate and lifetime of the ^{229m}Th VUV photon decay transition and/or any other signal either from daughter products or phosphorescence. The uranium sample used was exceptionally pure eliminating daughter product signals and by using vacuum and argon deposition, the phosphorescence signal could be identified and removed. The wavelength of the ^{229m}Th photon, thus the energy of the ^{229m}Th state, would be obtained by which VUV bandpass filter transmitted the photons (and the rest of the filters would not). The time decay constants (lifetime) of the time decays we measured varied substantially from run to run, and varied for each of the three substrates,, but typically we found a "fast" or "short" lifetime of approximately 1200(600) s and a "slow" or "long" lifetime of approximately 30000(15000) s. We did not see a repeatable significant difference in the pattern of lifetimes for the depositions with/without Ar to block/not block the Th ions (but not block the alphas and betas). The count rates (and lifetimes) were similar for the various bandpass filters we used, and we did not see a repeatable spectral feature due to heavy ions in the range 140 nm $< \lambda <$ 244 nm. In addition to using argon to shield the ions, we also heated the substrate to 150 C during the deposition. For the cases where the substrate was heated all signal was eliminated regardless of

the substrate. Since it has been shown [9] that heating the substrate can quickly eliminate phosphorescence signal, it is very likely that the signal detected was due to phosphorescence due to damage in the substrate caused by alpha and beta bombardment, and not from the ^{229m}Th nuclear VUV photon.

The second method, called the direct detection method, to observe the ^{229m}Th VUV nuclear photons was to have the PMT directly observe the ^{233}U source through a substrate, with and without a 3 μm thick Al sheet on the U source to block Th ions, and with a range of VUV bandpass filters to provide spectroscopy. Fig. 3 shows typical signals as a function of filter wavelength, for the Al_2O_3 substrate.

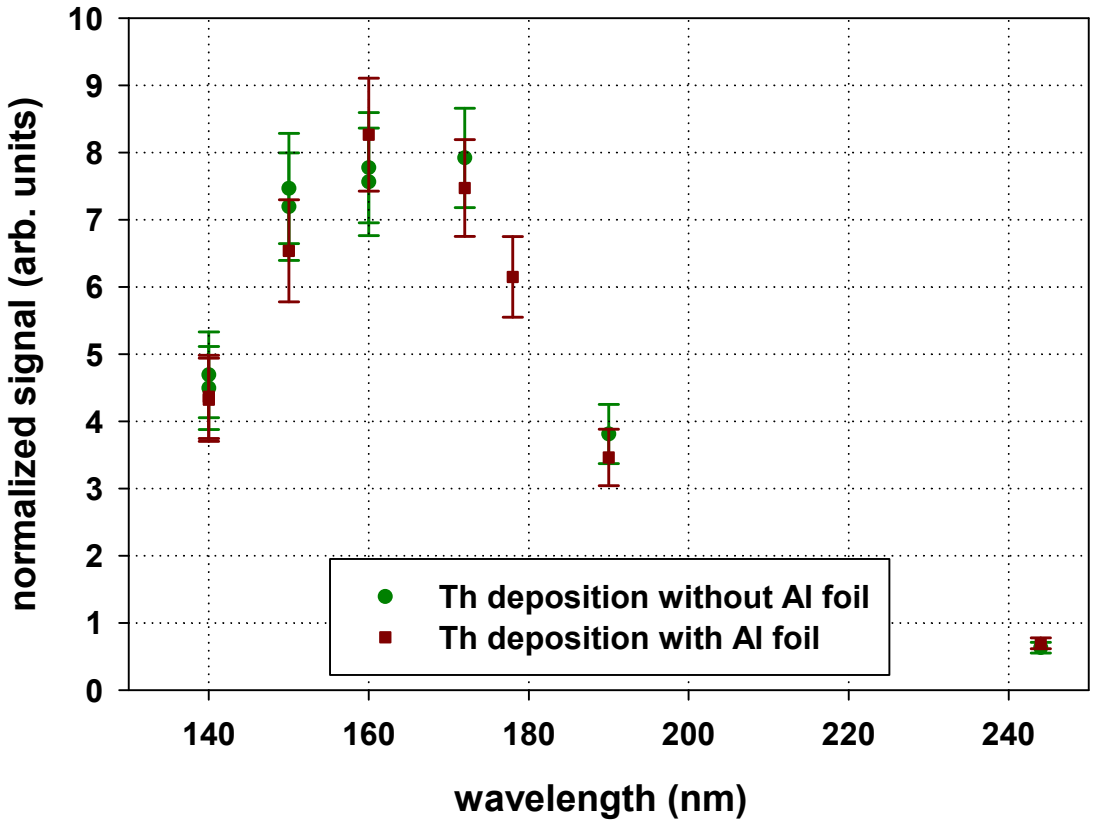

Fig. 3 Count rate of PMT as a function of filter wavelength. Green dots show the signal for the case where the heavy ions are not blocked, and red squares show the signal for the case where the heavy ions are blocked by 3 μm thick Al. Error bars in this figure are set at twice the shot noise of the actual counts, taking into account shot noise, some correlated PMT counts, and other stochastic experimental variations. [Units on the vertical axis of Fig. 3 are proportional to counts/s, but normalized by the area under the curve of the bandpass filter wavelength response, and also normalized by the quantum efficiency of the PMT at the particular wavelengths. The vertical pairs of green dots in the left part of Fig. 3 show repeated measurements at one bandpass filter wavelength.]

Although the feature in Fig. 3 centered around 160 nm is interesting, it appears these data are essentially unchanged with and without the Al foil to block the Th ions, indicating that any

spectral features seen are not due to ^{229m}Th, but instead are due to the VUV fluorescence and phosphorescence of the substrates from the alpha and beta ray bombardment.

This lack of ^{229m}Th signal in both detection methods may be due to four non-exclusive and possibly overlapping explanations:
(1) The 1/e lifetime of the ^{229m}Th state is more than 110000 s (30 hours). (Wense, et al. [4] have found that the ^{229m}Th state has a lifetime greater than 1 minute.)
(2) The branching ratio for production of ^{229m}Th:^{229}Th from the ^{223}U decay is less than the expected 2%. (However the branching ratio must be at least close to 2% otherwise Wense, et al. [4] would not have seen the ^{229m}Th signal they did see.)
(3) The wavelength of the transition is outside our observed range of 140 nm < λ < 244 nm (5.1 eV < E < 8.9 eV). (However the width of our search is approximately centered on Beck, et al.'s [2, 3] predicted energy, and more than five times the full range of its error bars.)
(4) The ^{229m}Th ions quench and the ^{229m}Th nuclei deexcite immediately or in less than 300 s, when produced and/or when deposited in the crystal substrates. A neutral or +1 charge state ^{229m}Th atom would immediately lose its nuclear excitation by internal conversion, likely without emitting a VUV photon.

Different substrates gave drastically different background fluorescence and phosphorescence count rates (but about the same fast and slow lifetimes). Out of the three substrates we used, we found that CaF_2 was by far the worst, giving a background count rate of up to 15 counts/s in our apparatus. The MgF_2 and Al_2O_3 substrates had a much lower background fluorescence and phosphorescence at typically 1 to 2 counts/s. For these above measurements, we used ultra-pure CaF_2 and MgF_2 substrates, at 99.99% chemical purity or better. In the second method, the direct detection method, we found that Ar gas also gave a very large unwanted background fluorescence in the presence of the ^{233}U, so we instead used Al foil to block the Th ions, but let alphas and betas through.

In this experiment, the parent ^{233}U produces the ^{229m}Th with a heavy ion recoil energy of 85 keV in a range of charge states from 0 to +4 or more, then the Th implants into and stops in the substrate. If its charge state is neutral or +1, the nuclear transition can couple to electrons and lead to rapid deexcitation to the nuclear ground state, most likely without emission of a VUV photon. Thus we believe neutralization or partial neutralization (quenching) to the 0 or +1 charge states is essentially synonymous with deexcitation of the ^{229m}Th nuclear state. Only if the Th is in the +2 or higher charge state is it likely to have a reasonably long lifetime in the ^{229m}Th state for our experiment to measure the emitted VUV photon.

There are three possible processes of interest for this quenching/neutralization, for which we have estimated the effect on our measurements. The Th leaves the source, implants into the substrate, and then remains in the substrate. The amount of change in the Th ion charge state for each of these complex solid-state processes is difficult to calculate, or even estimate.

Our search of the literature revealed several papers [10 - 13] that measured the charge state spectrum of various heavy ions recoiling from their parents' alpha decay in a film. In these papers all the decays have an alpha energy of about 5 MeV, and all have a heavy nucleus recoil energy of about 100 keV, similar to this experiment for the case of ^{229}Th recoiling from the ^{233}U

decay. Table 1 summarizes our interpretation of the results of these papers, showing the emitted fraction of ions in each charge state.

Charge state	^{216}Po Ref. [10]	^{222}Ra Ref. [11]	^{208}Pb Ref. [12]	^{208}Tl Ref. [12]	^{206}Pb Ref. [13]
0	0.871	0.20	0.42	0.53	0.60
+1	0.031	0.50	0.40	0.38	0.36
+2	0.021	0.267	0.13	0.06	0.043
+3	0.017	0.083	0.034	0.021	
+4	0.016	0.058	0.013	0.016	

Table 1 Fraction of alpha decay recoiling heavy daughter nuclei emitted in various charge states, from Refs. [10 - 13].

In the emission, there is agreement that the neutrals and singly ionized dominate, which likely do not leave a ^{229m}Th nucleus. For doubly ionized and above, the resulting fraction in these atoms similar to Th is 4% to 41%. In our experiment, if the fraction is 41% or more, the ^{229m}Th signal we expect might just barely rise above the noise, however if the fraction is noticeably less that 41%, we could not see a signal above the noise.

The second and third parts of the problem of maintaining a +2 or more charge state for implanted ^{229m}Th ions are when the ion enters into and stops in the crystal substrate (deposition or implantation). Two papers [14, 15] examine the equilibrium charge states. Equilibrium charge states occur when ionization is balanced by charge capture, which implies free or slightly bound electrons. At zero implantation velocity, almost all the heavy atoms will be neutrals, especially in a metal with free electrons. In an ideal insulator, there will be no free electrons. In our cases, for our substrates, there will always be some very small density of free electrons in the bulk. Also, there can be free electrons on the surface. Our heavy Th ions will be deposited only within about 22 to 30 nm from the surface, according to an SRIM calculation [8]. We estimate that between 0 and 10% of incoming +2 or more charge state ions will stay in their charge state when implanted in the MgF_2, CaF_2, Al_2O_3 substrates, thus we estimate 90 to 100% will quench.

If only 10% of the ^{229m}Th ions are unquenched during emission from the ^{233}U source, and if only 10% of the remaining ^{229m}Th ions are unquenched during deposition (implantation) into the substrate, then our expected signal would be only 0.01 counts/s in this apparatus, which is too small to be observable against the 1 to 2 counts/s background of our system. The most likely reason for the lack of ^{229m}Th signal in this set of measurements is the charge state neutralization/quenching described above, making the fractions of unquenched ^{229m}Th ions, either as produced from the source, or as deposited into the substrates, to be substantially less than 10%. Given the ion quenching and nuclear deexcitation that is apparently occurring, leading to a signal rate of 0.01 counts/s or less, we would need at least 35000 ^{229m}Th nuclei to produce 1 VUV photon count/s out of the PMT.

After many data sets and extensive analysis of the results of both experimental methods, it appears that we only see broadband VUV fluorescence and phosphorescence of the substrates under the alpha and beta bombardment emitted by the radioactive atoms, and we are not able to see VUV photons from the ^{229m}Th. All the signals we see do not go away with Ar gas or Al foil

to block the Th ions, thus the signals we see are apparently due to the approximately 5 MeV alpha and beta bombardment induced broadband VUV fluorescence and phosphorescence of the substrates, which apparently have the same quickly and slowly decaying components in the three substrates. This is likely due to alpha and beta ray induced damage to the crystalline structure, which then emits many broadband VUV photons while relaxing over the 1200 s short and 30000 s long times referenced above. Excited state ^{229m}Th ion deposition does not appear to yield a sufficient concentration on excited ^{229m}Th ions. The rate of charge state 2 or more plus ions ejected from the source appears to be much lower than expected and then coupled with the neutralization of these ions when implanted into the substrate results in a ^{229m}Th nuclear VUV photon signal too low to detect.

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