Results of a Direct Search Using Synchrotron Radiation for the Low-Energy ^{229}Th Nuclear Isomeric Transition

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We report the results of a direct search for the ^{229}Th ($I^\pi = 3/2^+ \leftarrow 5/2^+$) nuclear isomeric transition, performed by exposing ^{229}Th-doped $LiSrAlF_6$ crystals to tunable vacuum-ultraviolet synchrotron radiation and observing any resulting fluorescence. We also use existing nuclear physics data to establish a range of possible transition strengths for the isomeric transition. We find no evidence for the thorium nuclear transition between 7.3 eV and 8.8 eV with transition lifetime (1–2) s $\lesssim \tau \lesssim$ (2000–5600) s. This measurement excludes roughly half of the favored transition search area and can be used to direct future searches.

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Almost four decades ago, the existence of a low-lying nuclear excited state in ^{229}Th was indirectly established through the spectroscopy of γ rays emitted following the α decay of ^{233}U [1]. Subsequent indirect measurements placed this excited, isomeric state ($I^\pi = 3/2^+$) to be (3.5 ± 1.0) eV above the ground state ($I^\pi = 5/2^+$) [2]. The prospects of a laser-accessible nuclear transition touched off a flurry of proposals to utilize this apparently unique nuclear transition as a sensitive probe of both nuclear structure and chemical environment [3], to constrain the variability of the fundamental constants [4–6], to check the exponentiality of the decay law of an isolated metastable state [7], to demonstrate a γ-ray laser [8], and to construct a clock with unprecedented performance [9–11].

For these applications to proceed, it was necessary to first more precisely determine the transition energy. Therefore, several efforts were undertaken to spectroscopically resolve the expected ultraviolet (UV) emission from this magnetic dipole ($M1$) transition, where the excited state was typically expected to be populated in the α decay of ^{233}U [12]. Despite initial claims of observation [13,14], these searches were unsuccessful [15–17].

In 2007, using a significantly improved γ-ray spectrometer, the ^{229}Th isomeric transition was remeasured and found to actually be in the vacuum ultraviolet (VUV) portion of the electromagnetic spectrum [18], with an energy of (7.8 ± 0.5) eV [19], thus explaining why previous VUV insensitive searches failed, and reenergizing the community in the search for this nuclear transition. Recently, the result of a search for VUV emission from the nuclear excited state, again populated in α decay of ^{233}U, suggests that the transition energy is <7.75 eV [20], though it is the subject of controversy [21].

In experiments using the decay of ^{233}U it is difficult to differentiate whether the observed signal is indeed from the ^{229}Th isomeric transition or from known systematic effects [15,16,21]. Therefore, it is desirable to perform a direct measurement, where the nuclear transition is excited by an external source of electromagnetic radiation and the resulting fluorescence monitored.

In 2008, we proposed an experiment to utilize a VUV transparent crystal [22] doped with ^{229}Th to provide a high density sample suitable for a search using a broadband synchrotron light source [10,23]. Here, we report the first results of this direct search. In the remainder of this manuscript, we outline the experimental apparatus and protocol, present sample data, and interpret the data to place constraints on the transition lifetime as a function of transition energy. We also establish a favored range for the possible transition energy and lifetime, and conclude with a discussion and estimation of possible systematic effects. This result excludes roughly half of the favored region and can be used to direct future searches.

Conceptually, the experimental apparatus, shown in Fig. 1, and protocol are simple. A VUV transparent ^{229}Th-doped crystal is illuminated for time T_e with VUV photons. If the VUV photons are resonant with the isomeric transition, a fraction of the ^{229}Th nuclei are excited to the ($I^\pi = 3/2^+$) isomeric state. Then, the VUV photon source is shuttered and any subsequent fluorescence, which results from spontaneous decay back to the ($I^\pi = 5/2^+$) ground state, is recorded by opening two low-profile shutters to expose two photomultiplier tubes (PMTs) to the crystal for a time T_d. This simple approach is complicated by the generation of tunable VUV light, availability of high-purity ^{229}Th, and construction of a ^{229}Th-doped, VUV transparent crystal.

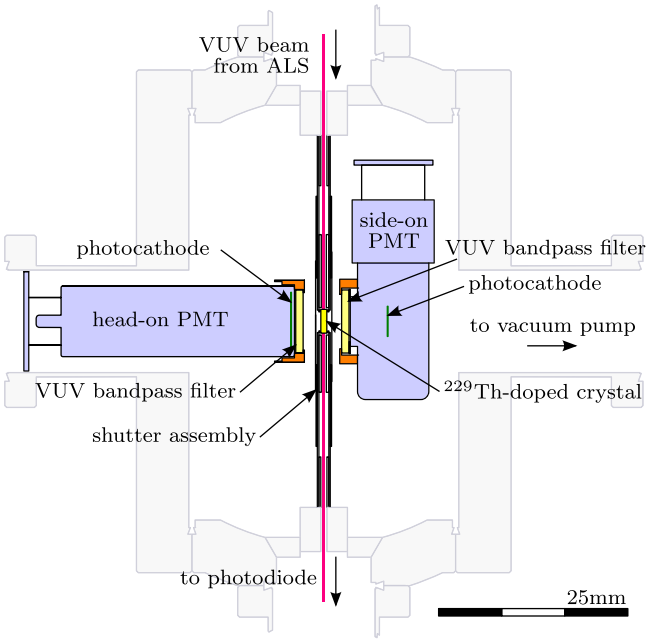

FIG. 1 (color online). Experimental schematic. A ^{229}Th-doped crystal is illuminated with synchrotron radiation along its long axis. Any resulting fluorescence is collected perpendicular to this axis by a head-on type and side-on type PMT. Low-profile mechanical shutters (UniBlitz) shield the PMTs from scattered light during illumination. The assembly is held under a pressure $<10^{-5}$ mbar.

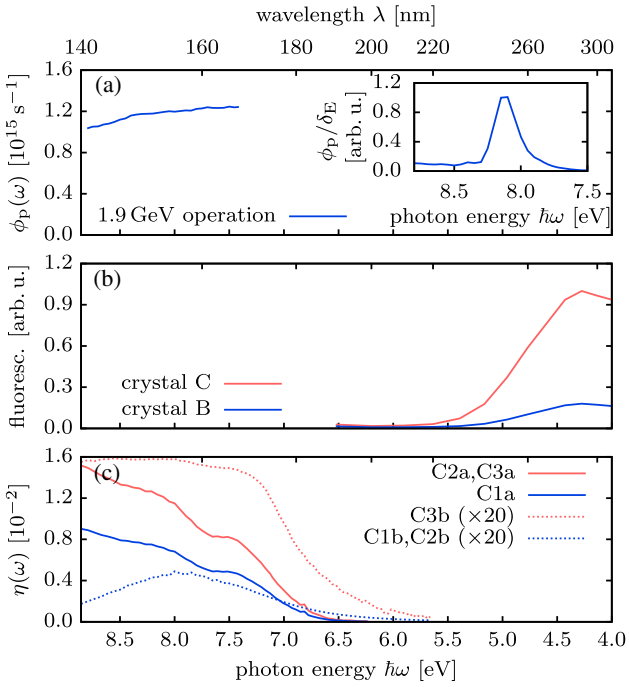

FIG. 2 (color online). Photon energy (wavelength) dependence of relevant experimental parameters: (a) VUV photon flux ϕ_p of the ALS beam line 9.0.2.1 for normal (1.9 GeV) operation; inset shows sample ALS spectral line shape taken with monochromator resolution $\delta_E \approx 0.05$ eV (courtesy of Oleg Kostko). (b) UV fluorescence spectrum of the crystals due to radioactive decay, recorded with a McPherson 234/302 monochromator and a Hamamatsu R1527P PMT. (c) Total detection efficiencies (see Table I).

For tunable VUV light, we utilize beam line 9.0.2.1 at the Advanced Light Source (ALS) synchrotron [24]. In normal operation (1.9 GeV electron energy), this beam line can tune within $\hbar\omega \approx$ (7.4–30) eV. We characterize the photon flux ϕ_p with an Opto Diode Corporation (ODC) SXUV-100 VUV damage resistant photodiode, calibrated against a NIST-calibrated ODC AXUV-100G photodiode, and find $\phi_p = (1.0\text{–}1.25) \times 10^{15}$ s^{-1} [see Fig. 2(a)]. The VUV spectrum is nearly Lorentzian with linewidth $\hbar\Gamma \approx 0.19$ eV and exhibits a constant "tail" up to ≈10 eV [see inset Fig. 2(a)], which reduces the useful photon flux to $\xi \approx 0.7$ of its measured value.

The ^{229}Th dopant used in this work was purchased from U.S. DOE and was extracted from ORNL mass-separated ^{233}U, batch UTHX001, which contains ppb levels of ^{232}U. The mass distribution of thorium isotopes in this sample was: ^{229}Th (75.6%), ^{228}Th ($\lesssim$0.1%), ^{230}Th (0.48%), and ^{232}Th (23.8%).

The development of a suitable ^{229}Th-doped crystal has been one of the most significant challenges of this work [25,26]. Briefly, we have grown and tested a variety of VUV transparent crystals doped with the common ^{232}Th isotope to find a material that satisfies all of the requirements of this experiment, which include high VUV transparency, absence of long-lived fluorescence, resulting from, e.g., color center formation, resistance to the effects of nuclear radiation, and low radiation-induced scintillation. From this work, we chose $LiSrAlF_6$ and have produced three ^{229}Th-doped $LiSrAlF_6$ crystals with dimensions ≈ 3 mm $\times$ 3 mm $\times$ 10 mm. The amount of ^{229}Th in each crystal was assayed by γ-ray spectroscopy (Ortec GMX-50220-P) [27] and found to be (115 ± 5) nCi, (290 ± 40) nCi, and (2.2 ± 0.3) μCi. In this work, we use only the latter two crystals, referred to as crystals B and C, with thorium atomic densities of $n_{Th} \approx 5.8 \times 10^{16}$ cm^{-3} and $n_{Th} \approx 4.1 \times 10^{17}$cm^{-3}, respectively.

Because of scintillation following the radioactive decay of ^{229}Th (4.8 MeV α decay with a half-life of (7917 ± 48) y [28]) and its daughter isotopes, the crystals emit photons in the UV region [see Fig. 2(b)]. This UV fluorescence together with scintillation induced directly in the PMTs by the radioactivity is the dominant background.

At a given ALS beam energy, $\hbar\omega$, in the limit of weak excitation, the number of detected photons from the nuclear fluorescence, N_d, in a time interval $[t_1, t_2]$ is given as

$$N_d = N_0(1 - e^{-T_e/\tau'})(e^{-t_1/\tau'} - e^{-(t_1+t_2)/\tau'})$$
$$\text{with} \quad N_0 = \frac{2}{3}\eta(\omega)\frac{\lambda_0^2}{2\pi}\frac{\xi\phi_p'(\omega)n_{Th}l}{\Gamma}\frac{1}{1+4(\frac{\omega-\omega_0}{\Gamma})^2}, \qquad (1)$$

where τ' is the lifetime of the ^{229}Th transition inside the crystal, ϕ_p' is the photon flux transmitted through the crystal, $\eta(\omega)$ is the total efficiency of the detection system,

ω_0 (λ_0) is the unknown isomeric transition energy (wavelength), and l is the length of the crystal.

For each configuration, $\eta(\omega)$ depends on the PMT quantum efficiency η_{PMT}, its solid-angle fraction η_{SAF}, and the transmission of an optional VUV bandpass filter η_{filter}. Both η_{PMT} and η_{filter} are characterized using a McPherson 234/302 monochromator, deuterium lamp, and the above-mentioned photodiodes, and agree with the calibrations provided by the respective manufacturers. The different η_{SAF} are determined using both commercial (Zemax) and homemade ray-tracing software.

To better discriminate any ^{229}Th fluorescence from spurious signals, we simultaneously employ two PMTs of differing technology. Specifically, three overall configurations are used (see Table I) and their resulting total detection efficiencies are shown in Fig. 2(c). Finally, the PMT signals are (optionally) amplified (Stanford Research SR445A) and recorded using a digitzer (CAEN DT5751).

Using this system, we were given 96 h of ALS beam time from August 20 to September 5, 2014. We searched the range (7.4–8.8) eV with step size $\leq$0.1 eV and an illumination time $T_e = 2000$ s for each VUV energy.

Figure 3 shows the recorded crystal fluorescence after ALS illumination at $\hbar\omega = 7.7$ eV using configuration C2. These data are representative of all recorded data. From data such as these, we establish an *upper bound* on the transition lifetime using Eq. (1) evaluated for each bin and the Feldman-Cousins prescription [29]. For this analysis, we use a $\Delta\chi^2$ test statistic for the lifetime τ' that is profiled in the background count-rate nuisance parameter [30]. The first 10 s of each data trace are omitted to mitigate the effect of any scattered light and/or short-lived UV/VIS fluorescence. For the *lower* bound on the lifetime, we derive the average background signal from data taken for $t > 200$ s after illumination. Then, we perform a Feldman-Cousins analysis without nuisance parameter using data between [2.2 s, 7.2 s]. For each probed VUV energy, we analyze the data assuming detuning of the ALS beam from the thorium transition by -0.2 eV to $+0.2$ eV in 0.01 eV steps and present the most excluding lifetimes obtained from all configurations.

TABLE I. Configurations of the experimental system and searched VUV photon energy range. All PMTs are manufactured by Hamamatsu, the VUV bandpass filter [(150 ± 27) nm] is an Acton Research Corp. 150-N-MF-1D, and d is the distance from the photocathode to the crystal center.

Config.		Crystal	PMTs	Filter	d [mm]	$\hbar\omega$ [eV]	T_d [s]
C1	a	C	R6835	no	12	7.4–8.8	1000
	b		R8486	yes	25		
C2	a	B	R6835	no	5–6	7.4–8.8	1000
	b		R8486	yes	25		
C3	a	B	R6835	no	6–7	7.4–8.25	1800
	b		R7639	no	25		

Since this data is recorded in a dielectric medium with refractive index n, the $M1$ transition rate is enhanced by a factor of $n^3(\omega)$ relative to the rate in vacuum [31]—for reference, $1.46 \leq n \leq 1.51$ over the scanned photon energy range. Therefore, we convert the bounds to vacuum lifetimes $\tau = n^3\tau'$. The resulting 90% confidence level (CL) excluded region is shown in Fig. 4.

The impact of the exclusion is better understood by considering the theoretically possible isomeric transition energies and lifetimes. While there is general acceptance of the transition energy range established by Ref. [19], there is less consensus on the possible transition lifetime range. Reference [32] employs a quasiparticle plus phonon-model calculation to predict that the transition lifetime is $\tau = 2.23 \times 10^6$ eV^3 s$/(\hbar\omega)^3$. References [2,18], and [20] use the fact that a transition between the same two Nilsson states is observed in ^{233}U at 312 keV, and predict that the transition lifetime is $\tau \approx 8 \times 10^6$ eV^3 s$/(\hbar\omega)^3$. And, Ref. [7] calculates the transition lifetime to be $\tau = 0.66 \times 10^6$ eV^3 s$/(\hbar\omega)^3$ by finding the matrix element of the nuclear transition in terms of another $M1$ transition in the thorium nucleus ($9/2^+5/2[633] \rightarrow 7/2^+3/2[631]$), which was previously measured [33].

Of these calculations, the method of Ref. [32] is accurate to within a factor of ~4 for the cases where experimental lifetimes are available, while the method of Refs. [2,18], and [20] provides only a rough estimate since nuclei

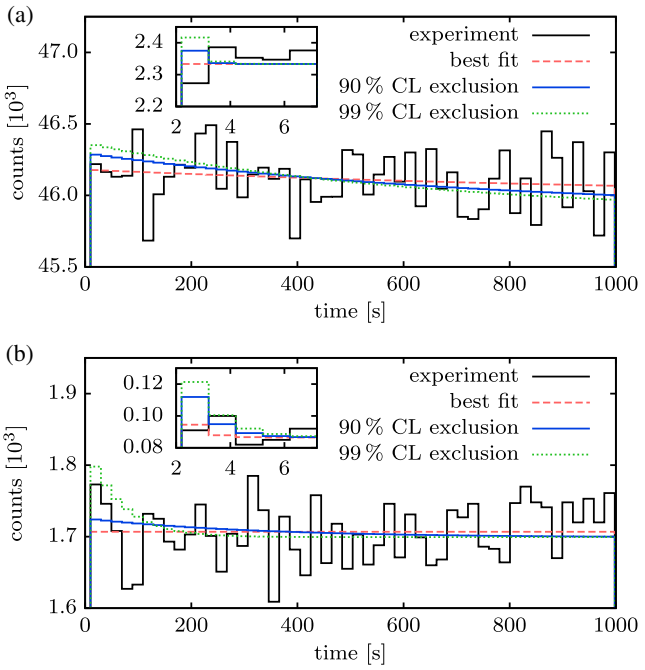

FIG. 3 (color online). Binned photon counts (solid, black) at a VUV energy $\hbar\omega = 7.7$ eV with $T_e = 2000$ s for (a) configuration C2a and (b) C2b, respectively. Also shown are the best-fit result (dashed, red), the curve corresponding to the 90% CL (solid, blue; used for the upper bound on τ'), and 99% CL (dotted, green), according to Eq. (1). Insets show the time interval [2.2 s, 7.2 s] used to obtain the lower bound on τ'.

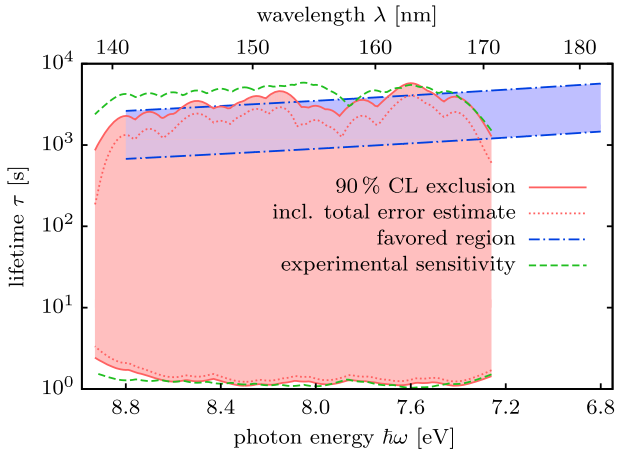

FIG. 4 (color online). The 90% CL exclusion region (red shaded) for the vacuum lifetime τ of the ^{229}Th isomeric transition as a function of the transition energy $\hbar\omega$. Reduction of the sensitivity by the total error budget (Table II) would reduce the excluded region to the area between the red dotted lines. The experimental sensitivity (green, dashed; see text) and the favored region for the transition (blue shaded area between dash-dotted lines; see text) are also given.

specific effects [7] can modify the lifetime. In contrast, the technique of Ref. [7] has been shown to be accurate to within experimental error in the case of other nuclei where data are available. Therefore, we compare our experimental results to this method, with the modification that since the work of Ref. [7] three new experimental measurements of the $9/2^+5/2[633] \rightarrow 7/2^+3/2[631]$ transition lifetime have been made [12,32,34]. Allowing for two standard deviations in all measurements, we have constructed a "favored region" [bounded by 0.46×10^6 eV^3 s$/(\hbar\omega)^3 \leq \tau \leq 1.79 \times 10^6$ eV^3 s$/(\hbar\omega)^3$], where the transition should lie at roughly the 90% CL (Fig. 4, blue shaded region).

Also shown in Fig. 4 (dotted line) is an estimate of the potential impact of systematic errors on the calibration of the experimental sensitivity. This exclusion is created by reducing N_0 by the total error budget given in Table II.

Finally, the experimental sensitivity, defined as in Ref. [29], is determined with the same prescription as used for the bounds of the exclusion region, but by analyzing experimentally recorded background data without prior VUV illumination (Fig. 4, dashed green curve). Our exclusion region is almost at the level of the sensitivity, indicating little excess noise.

Several effects could modify the presented results. First, nonradiative relaxation, so-called internal conversion, could reduce the fluorescence rate predicted by Eq. (1). This process would occur via a photon-mediated nucleon-electron interaction and result in the deexcitation of the nucleus without VUV photon emission [22]. This effect can be estimated from a detailed analysis of the crystal structure (cf. Th:CaF_2 [35]); however, it is difficult to achieve reliable results due to the complexity of $LiSrAlF_6$. Nonetheless, we do not expect nonradiative relaxation to occur here,

TABLE II. Statistical and estimated systematic errors at the 90% CL for the individual contributions to N_0 in Eq. (1). The total is estimated using standard error propagation assuming the parameters (except for ϕ'_p and η_{PMT}) are independent.

Parameter	Symbol	Rel. Error
ALS VUV energy	$\hbar\omega$	0.02
ALS VUV linewidth	Γ	0.10
ALS VUV spectrum	ξ	0.40
Photon flux through crystal	ϕ'_p	0.15
Solid angle fraction	η_{SAF}	0.15
PMT quantum efficiencies	η_{PMT}	0.15
(Eff.) crystal length	l	0.10
^{229}Th density	n_{Th}	0.15
Total		$\approx$0.52

because the $LiSrAlF_6$ crystals exhibit excellent bulk transmission, experimentally confirming that requisite electronic states for nonradiative relaxation are not present, while narrow absorption features around the ^{229}Th resonance or multiphonon processes appear unlikely.

Second, radiation trapping occurs when the on-resonance absorption length L_{res} in the crystal becomes smaller than its length, l:

$$L_{\mathrm{res}} = \left(\frac{2}{3}\frac{\lambda_0^2}{2\pi} n_{\mathrm{Th}} \frac{n^3(\omega_0)/\tau}{\Gamma_B}\sqrt{\pi \ln 2}\right)^{-1}. \qquad (2)$$

For the expected inhomogeneously broadened linewidth of $\Gamma_B \approx 2\pi \times 10$ kHz [10,36], radiation trapping could become important for lifetimes $\tau \leq 50$ s, where $L_{\mathrm{res}} \approx l/2$. However, as verified by Monte Carlo simulations, if radiation trapping occurs it does not affect our ability to detect the transition and in some cases even increases our signal: though excitation starts in a region of size $\sim L_{\mathrm{res}}$ near the entrance of the crystal, subsequent emission and reabsorption events result in excitation of larger portions of the crystal in a random-walk manner. This leads to a slower decaying but considerably larger signal within the detection window as compared to Eq. (1) and would allow us to extend our short-lifetime exclusion significantly beyond the present limit. However, it is possible that the inhomogeneous broadening is >10 kHz; for example, if the thorium atoms substitute in multiple crystal sites, radiation trapping will be negligible [10,36]. Therefore, we report the most conservative limit, which ignores the benefits of any potential radiation trapping.

Finally, coherent effects like superradiance [37] are unlikely, as the thorium number density per wavelength cubed, $n_{\mathrm{Th}}/\lambda_0^3 \approx 250$, is effectively reduced by a factor $n^3(\omega_0)/(\tau\Gamma_B) \approx 5 \times 10^{-5}$ s$/\tau$. Also, coherence-enhanced optical effects in the propagation direction of the exciting light field [38] do not impact a perpendicular detection scheme.

In conclusion, the present result excludes the existence of the ^{229}Th isomeric transition with a vacuum lifetime (1–2) s $\lesssim \tau \lesssim$ (2000–5600) s for the energy range $\hbar\omega = (7.3 - 8.8)$ eV at the 90% CL. This experiment did not

probe energies ≤ 7.4 eV, which cannot be reached in normal operation of the ALS. Future work will concentrate on improving the present limits and probing $\hbar\omega < 7.4$ eV for the first time. To accomplish the former will likely require the use of a VUV laser system, which provides a higher spectral irradiance than the ALS [24,39]; the latter is also possible operating the ALS in a special low-energy mode.

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