

Photoinduced quenching of the ^{229}Th isomer in a solid-state hostJ. E. S. Terhune¹, R. Elwell¹, H. B. Tran Tan^{2,3}, U. C. Perera², H. W. T. Morgan^{4,5}, A. N. Alexandrova⁴,
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The population dynamics of the ^{229}Th isomeric state is studied in a solid-state host under laser illumination. A photoquenching process is observed, where off-resonant vacuum-ultraviolet (VUV) radiation leads to relaxation of the isomeric state. The cross-section for this photoquenching process is measured, and a model for the decay process, where photoexcitation of electronic states within the material band gap opens an internal conversion decay channel, is presented and appears to reproduce the measured cross-section. By engineering defects into ^{229}Th -doped solid-state hosts, this previously unrecognized photoquenching process may be used to reduce the clock transition readout time and thereby increase the stability of the nuclear clock.

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The ^{229}Th nuclear isomeric state provides a laser-accessible nuclear transition that can be driven even when doped into a high-band-gap solid [1,2]. The unique properties of the system are expected to allow access to a number of novel applications, including the construction of a solid-state nuclear clock [2]. With the recent laser excitation and measurement of the isomer energy and lifetime in these systems [3–6], work can now begin towards constructing and optimizing timekeeping devices based on thorium-doped crystals.

The stability of an optical clock operating by collecting fluorescence from the clock transition is limited to [7] $\sigma = \sqrt{(T_e + T_c)/T} / (2\pi QS)$, where $Q = f_0/\Delta f$ is the transition quality factor, f_0 is the transition frequency, Δf is the transition linewidth, S is the signal-to-noise ratio (SNR), T_e is the excitation time, T_c is the fluorescence collection time, and T is the averaging time. Thus, optimization of clock performance requires maximizing S , while minimizing $T_e + T_c$ without degrading Q . The transition linewidth in ^{229}Th doped fluoride crystals is expected to be ~ 1 kHz [2], which is $10^6 \times$ larger than the intrinsic linewidth of the transition. As a result, simply put, optimum clock performance is realized by exciting all ^{229}Th nuclei in a time of order 1 ms and having them decay as quickly as possible.

Given that laser excitation of a nucleus in a solid-state environment has only just been demonstrated [3,4], it is not

yet known what values of S , T_e , and T_c are attainable. In particular, Ref. [4] observed that not all ^{229}Th in the crystal were laser excitable and speculated that impurities and/or crystal defects could quench the transition or alter its decay lifetime [8,9]. This observation, combined with observations of photoinduced quenching by x-ray pumping [10], suggests that the lifetime of the nuclear transition could depend on the quantum state of nearby electrons, potentially allowing optimization of clock performance by improving S and reducing T_e and T_c .

To determine if such a process is possible, here we present a study of the excitation dynamics of ^{229}Th nuclei doped into a LiSrAlF_6 crystal. We find that the vacuum-ultraviolet (VUV) photons used to excite the ^{229}Th nuclei also lead to a process that quenches nuclear excitation. We study this process to determine the cross-section of the photoquenching process and provide a description of mechanisms that could be the origin of the phenomenon. We conclude with a discussion of the use of photoquenching for optimizing solid-state nuclear clock performance and suggest directions of future work to better understand the process.

The experiments reported here were conducted via monitoring the resulting fluorescence from a $^{229}\text{Th}:\text{LiSrAlF}_6$ crystal (crystal 3.1 from Ref. [4]) following illumination with a VUV laser (see inset Fig. 1). Using the system described in Ref. [4,11], VUV radiation was produced via resonance-enhanced four-wave mixing of two pulsed dye lasers (PDL) in Xe gas. The frequency of the first PDL (referred to as the UV PDL), ω_u , was locked to the $5p^6(^1S_0) \rightarrow 5p^5(^2P_{3/2})6p^2[1/2]_0$ two-photon transition of Xe at ~ 249.63 nm. The frequency of the second PDL, ω_v , was scanned to produce VUV radiation in the Xe cell given by the difference mixing relation $\omega = 2\omega_u - \omega_v$. All three laser beams then impinge off-axis

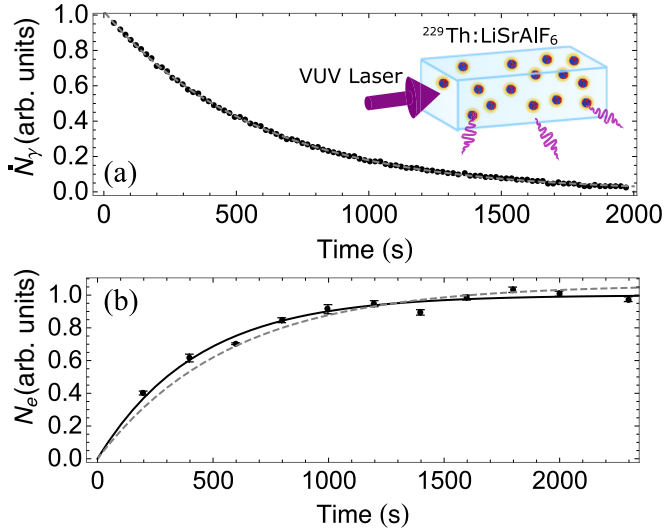


FIG. 1. (a) Photon emission rate of $^{229\text{m}}\text{Th}$ following laser excitation determining the radiative decay rate in LiSrAlF_6 to be $\tau = 573.4(29)\text{ s}$. (b) The total number of excited $^{229\text{m}}\text{Th}$ as a function of laser excitation time. The excited state population grows with a lifetime of $\tau_{\text{eff}} = 431(70)\text{ s}$. The dashed gray line is a fit to the data with τ_{eff} fixed to τ .

with respect to a MgF_2 lens, whose chromatic dispersion is used with downstream pinholes to spatially filter the VUV beam and pass it towards the $^{229}\text{Th}:\text{LiSrAlF}_6$ crystal. The laser system delivers 30 pulses per second to the crystal with a typical VUV pulse energy of $\sim 5\text{ }\mu\text{J/pulse}$.

The crystal under study is held in a vacuum chamber consisting of a crystal mount, two VUV-sensitive photomultiplier tubes (PMTs; Hamamatsu R6835), and a pneumatic shutter system to protect the PMTs from direct exposure to the VUV laser. The PMTs are operated in a cathode-grounded configuration, and their output waveforms are recorded by a 1 Gs/s waveform digitizer (CAEN DT5751) for subsequent post-processing. The VUV laser beam terminates on a custom VUV energy detector. The crystal chamber is maintained with an Ar atmosphere at a pressure of $\sim 10^{-2}$ mbar to provide high VUV transmission while minimizing browning of optics due to hydrocarbon deposition [4,11].

We model the laser excitation and quenching of the ^{229}Th nuclei with a simple rate equation. Assuming the quenching process is linear in photon number, the number of excited thorium nuclei is

$$\dot{N}_e = -\frac{N_e}{\tau} + \sigma_e \dot{\phi} \left(p N_g - \frac{g_g}{g_e} N_e \right) - \sigma_q \dot{\phi} N_e, \quad (1)$$

where N_g is the number of ground state nuclei, τ is the isomeric state radiative lifetime in the crystal, g_g (g_e) is the degeneracy of the ground (excited) state, σ_e is the wavelength-dependent absorption cross section, $\dot{\phi}$ is the number of photons per second per unit area passing through the crystal, and σ_q is the cross-section describing the strength of the photoquenching. The coefficient p , which we dub the participation factor, is introduced to account for the observation in Ref. [4] that not all ^{229}Th nuclei present in the crystal participate in the nuclear transition.

In order to quantify the quenching it is necessary to first measure the other parameters of the model. By exciting the nuclear transition for $T_e = 1200\text{ s}$ and then collecting the resulting fluorescence for $T_c = 2000\text{ s}$ [see Fig. 1(a)], τ is found from a nonlinear least-squares fit to be $\tau = 573.4(29)\text{ s}$ —here and throughout $()$ denote a 68% confidence interval. It is difficult to independently measure p as the number of excited nuclei is also affected by the quenching process. Nonetheless, as seen below, the quenching process is not detectable at lower laser power. Therefore, using data taken at low laser power, the radiative lifetime extracted from Fig. 1(a), the system detection efficiency, and the solution of Eq. (1), it is estimated that $p = 0.14(1)$. This yields an effective density of $n_p = 7.0(5) \times 10^{14}\text{ cm}^{-3}$ of participating ^{229}Th nuclei.

To ascertain the relative importance of the quenching dynamics, we first monitor the number of excited thorium nuclei as a function of excitation time. Given the narrow linewidth of the nuclear transition ($\sim 1\text{ kHz}$) relative to the excitation laser linewidth ($\sim 10\text{ GHz}$), $\sigma_e \dot{\phi}$ is on the order of 10^{-9} . Thus the number of excited ^{229}Th nuclei as a function of laser excitation time can be approximated as $N_e(t) \approx \sigma_e p \dot{\phi} \tau (1 - \exp(-t_e(1/\tau + \sigma_q \dot{\phi}))) = \sigma_e p \dot{\phi} \tau (1 - \exp(-t_e/\tau_{\text{eff}}))$. Figure 1(b) shows the total number of detected photons $N_{\gamma}^{(q)}$ following excitation, a quantity proportional to N_e , with a VUV laser power of $P \approx 90\text{ }\mu\text{W}$ versus laser exposure time t_e . The solid black line is a variable lifetime fit of $N_e(t)$ to the data. This fit finds an effective lifetime of $\tau_{\text{eff}} = 431(70)\text{ s}$, while the dashed gray line is a fit of $N_e(t)$ with τ_{eff} fixed to τ . Both the reduced χ^2 and Akaike information criteria model test prefer the model with variable lifetime, though neither is conclusive. Further, the fitted effective lifetime of $\tau_{\text{eff}} = 431(70)\text{ s}$ differs from τ by approximately two standard errors and suggests quenching by an amount of $\sigma_q \approx 1\text{ Mb}$.

To confirm the quenching process, we perform an additional experiment where after laser excitation for $T_e = 1200\text{ s}$, the excitation laser is detuned by approximately 100 GHz so that it does not efficiently excite the nuclei, but may still quench excited nuclei. We then record the emitted photons as a function of this variable “quench time” t_q , as shown in Fig. 2(a) for $t_q = 0, 100\text{ s}$, and 500 s . As can be seen, the total number of emitted photons decreases with an increase in quench time. This process is repeated for several quench times, and σ_q is extracted from the data with a model that accounts for the small off-resonant excitation rate that can occur during the quench time; larger laser detunings are not possible in our system. The results are shown in Fig. 2(b) and lead to an average value of $\sigma_q = 0.29(3)_{\text{stat}}[12]_{\text{sys}}\text{ Mb}$, where the systematic error is dominated by uncertainty in the VUV beam waist. Here and throughout, we use the convention $()_{\text{stat}}[12]_{\text{sys}}$ to emphasize that the systematic error is a scaling factor that affects both the reported mean and statistical standard error. Thus these data confirm the quenching process. Similar photoquenching, at several wavelengths and temperatures, has been observed in $^{229}\text{Th}:\text{CaF}_2$ [12].

To further investigate the photoquenching process, the dependence of σ_q on the photon flux $\dot{\phi}$ was studied. The data in Fig. 3 are taken in the same manner as that of Fig. 2, but here the quenching laser is applied for a fixed time of $t_q = 500\text{ s}$ and its power is varied. The laser power is controlled

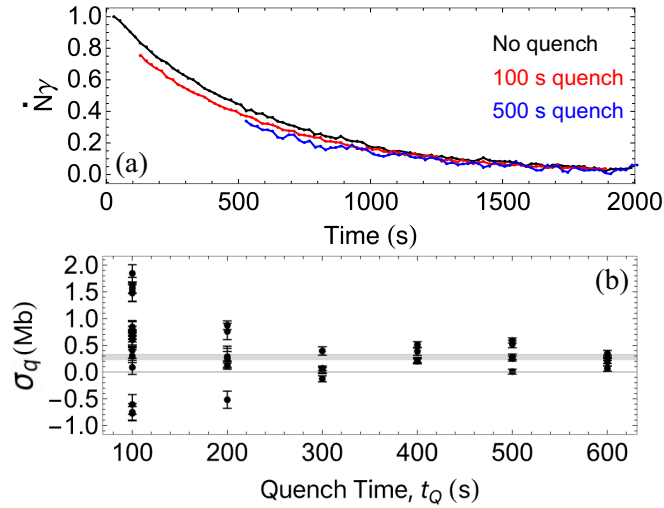


FIG. 2. (a) Photon emission rate of ^{229m}Th following laser excitation and a variable amount of quenching by an off-resonant laser. Each trace represents the average of several trials and clearly shows the reduced ^{229m}Th population with quenching. (b) The extracted quenching cross-section σ_q from each trial is shown as a function of quenching time t_q . At short quenching times, variability in laser parameters leads to a spread in excitation probability and therefore the extracted σ_q .

during the quench process by detuning the UV PDL a variable amount away from the Xe resonance. In this way, the change in Xe excitation controls the realized VUV power. Shown alongside the data as a black line is a prediction of the quenching based on the fit to the data in Fig. 2, which was taken at $90\ \mu\text{W}$, and an assumption of linear dependence of the

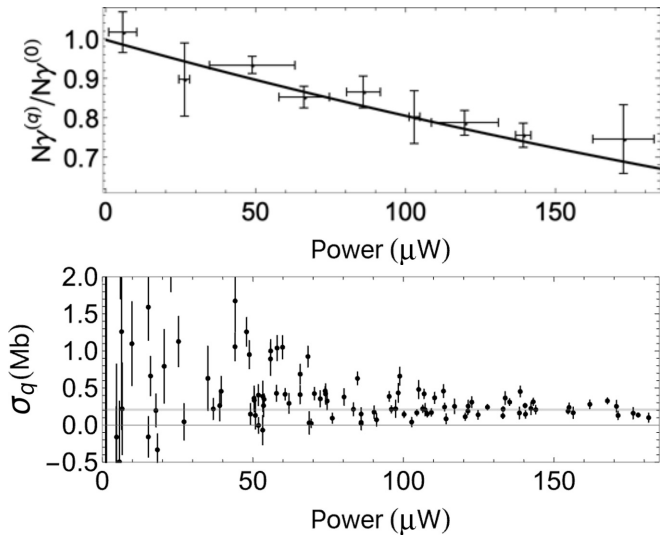


FIG. 3. (a) Ratio of quenched to unquenched photon emission, $N_{\gamma}^{(q)}/N_{\gamma}^{(0)}$, following laser excitation at variable quenching laser power. Each data point is an average of at least 5 trials. (b) The extracted quenching cross-section from each trial as a function of quenching laser power. The weighted average of these points is $\sigma_q = 0.23(1)_{\text{stat}}[9]_{\text{sys}}$ Mb.

photoquenching on ϕ . While this prediction agrees with the data, it is possible that the quenching process saturates at around $100\ \mu\text{W}$. Confirmation of this saturation is not possible in the present apparatus due to laser instabilities above $150\ \mu\text{W}$. Saturation of the photoquenching could arise, for example, if not all of the excited ^{229}Th undergo the photoquenching process, which as described later could arise if the quenching process depends on the nature of localized electronic states within the band gap. A fit to the data that allows for a variable quenching fraction indicates that roughly 50% of the ^{229}Th can be photoquenched and experience a considerably higher photoquenching cross section of $\sigma_q \sim 5$ Mb. The possibility of quenching saturation will be the study of future work when reliable higher power laser operation is available.

Given that the next nuclear excited state is nearly 30 keV above the isomeric state [13], the quenching process is presumably driven by photoexcitation of crystal electronic states. Such spatially localized, electronic defect states with energies inside the band gap can result from impurities, crystalline defects, and doping ^{229}Th into LiSrAlF_6 . Refs. [8,9] have considered the electron bridge process as a means for nuclear quenching in the crystalline environment, however, the process occurs at second order in perturbation theory with a likely reduced quenching rate compared to our model. Reference [14] has demonstrated that if the energy, ε_d , of the ^{229}Th defect state $|d\rangle$ with respect to the valence band (VB) maximum (set at zero energy) is less than the nuclear excitation energy, $\varepsilon_d \leq \varepsilon_n$, the nuclear excitation can typically relax by resonant excitation (i.e. first order in perturbation theory) of a defect-hole pair. This process is estimated [14] to occur in $\sim 10^{-3}$ s and is presumably responsible for the deviation of the participation fraction p from unity. It also suggests that the ^{229}Th doping sites responsible for the nuclear decay signal observed here have electronic defect states with energies that are greater than the isomer energy.

To excite defect states with $\varepsilon_d > \varepsilon_n$, the energy of the VUV laser used here a first-order process requires either (i) a multiphoton or (ii) single photon + multiphonon absorption process. Since the data in Fig. 3(a) do not show a ϕ^2 dependence, the second possibility is more likely. The phonon bath can be populated thermally or via optical absorption by impurities (with energies ε_i) throughout the crystal. (Here, we refer to optically active Th doping sites as “defects” and to nonactive sites and other non-Th defects as “impurities.”) Optical absorption by the impurities generates holes in the valence band (VB) that rapidly relax to the top of the VB, generating a phonon excess [15]. This phonon-bath pumping process stops if the quenching laser frequency ω_q drops below the impurity energy ε_i . This predicts a low-frequency photoquenching threshold at ε_i which could be significantly less than the nuclear excitation energy ε_n . Analysis of the photoluminescence of the $^{229}\text{Th}:\text{LiSrAlF}_6$ crystals when illuminated by the VUV laser reveals a roughly 100 nm wide feature centered around 425 nm (see SI), indicative of impurity states satisfying $\varepsilon_i < 5$ eV.

Once the phonon bath is populated, the excitation of the Th defect state at ε_d can proceed by multi-phonon absorption [16] as the defect state excitation is accompanied by the lattice relaxation [14]. This suggests that the excess phonons can

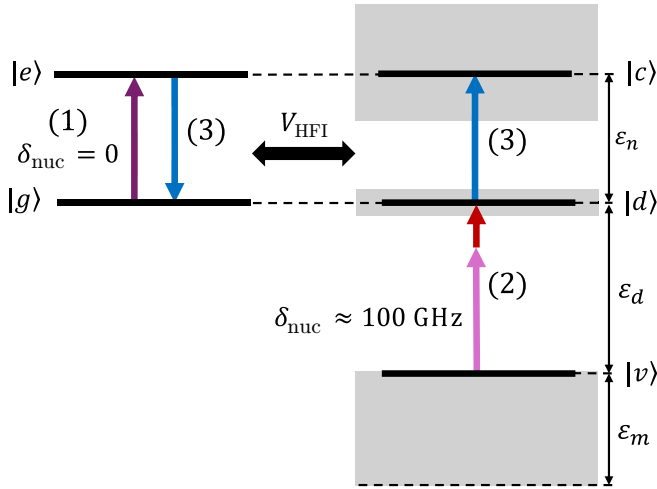


FIG. 4. Example photoquenching mechanism. Valence, defect, and conduction band states are denoted by (v, d, c) , respectively, while the ^{229}Th ground and isomeric states are denoted by (g, e) , respectively. (1) Resonant VUV laser drives the nucleus to the isomeric state. (2) The VUV laser and, in one process, multi-phonon absorption drives an electronic transition from $|v\rangle$ to $|d\rangle$. (3) If $\varepsilon_d + \varepsilon_n > \varepsilon_c$, an internal conversion channel is opened and the isomer can decay by exciting an electron into the conduction band.

be removed by cooling the crystal, leading to reduced photoquenching rates at lower temperatures. The quenching rate may also be sample-dependent as the nature and concentration of impurities are not controlled during the current doping process.

Regardless of the mechanism, if $|d\rangle$ is populated and $\varepsilon_d + \varepsilon_n \geq \varepsilon_c$, where ε_c is the energy of the bottom of the conduction band, the isomer can undergo an internal conversion (IC) decay process by resonantly exciting the defect state electron to the conduction band (see Fig. 4). Following similar arguments to those in Ref. [14], this IC rate is much faster than the radiative nuclear decay rate. This defect to conduction band IC process may be responsible for the observed photoquenching.

To estimate the rate of the quenching process, we perform a more comprehensive rate equation analysis that involves the relevant nuclear and electronic degrees of freedom (see SI). The resulting photoquenching cross-section is found as $\sigma_q \approx \lambda^2 / (2\pi)(\Gamma_{\text{IC}}/\Gamma_L)$, where Γ_{IC} is the internal conversion rate, $\Gamma_L = 2\pi \times 15 \text{ GHz}$ is the bandwidth of the laser system [4], and λ is the wavelength of the nuclear transition. Thus, the observed value of $\sigma_q \approx 0.25 \text{ Mb}$ translates to an IC rate of $\Gamma_{\text{IC}} \approx 2\pi \times 100 \text{ s}^{-1}$. While internal conversion rates from defect states into the conduction have not been calculated, this value of Γ_{IC} is of the same order of magnitude as the partial IC rate for $5f$ states in neutral Th [17] and as shown in Ref. [14] the electronic defects associated with ^{229}Th doping into LiSrAlF_6 have significant $5f$ character. Estimates in solid-state hosts [14] indicate a similar IC rate.

One immediate consequence of this process is that it produces an electronic population in the conduction band. As a result, this process could be confirmed by the observation of

photons whose energy is on the order of the LiSrAlF_6 band gap ($\sim 10 \text{ eV}$) or the electron itself. Such detection schemes may be beneficial in improving S for ^{229}Th clock operation. Further, if the location of the defect states can be determined or engineered, then perhaps resonant excitation of the defects can be used to strongly enhance the photoquenching rate. This raises the possibility of improved performance for the solid-state nuclear clock since the photoquenching provides control of the nuclear excited state lifetime. For example, pulsed excitation to the defect states with a strong laser could be used to quickly quench the excited nuclear state population, which could be detected via conduction band electrons or their $\sim 10 \text{ eV}$ fluorescence. This would obviate the need to wait for the $\sim 1000 \text{ s}$ required for the nucleus to decay during the clock interrogation protocol, translating to improvement in clock instability σ . Deliberately engineering these defects may also introduce the possibility of transferring the excitation to an optically active electronic state, thereby allowing shelving-style detection [18] of the nuclear transition.

Finally, though the uncovered photoquenching mechanism has no direct effect on planned ion clocks [19–21], especially for those operating using quantum logic spectroscopy [22], it suggests a possible means for quickly returning the excited nuclear population back to the nuclear ground state in those systems. Specifically, excitation of the electronic state of, e.g., $^{229m}\text{Th}^{3+}$ will open new electronic bridge decay channels [23,24], potentially dramatically shortening the effective isomer lifetime; of course, this excitation should be kept below $\lesssim 20 \text{ eV}$ so the internal conversion channel [25] is not opened.

In summary, a previously unrecognized phenomenon affecting solid-state nuclear clocks—photoquenching of the ^{229}Th isomer with a laser—was observed and its cross-section is measured. The process appears to result from an internal conversion decay channel that is opened by photoexcitation of electronic defects. A simple model suggests an IC decay rate via these defects of $\sim 2\pi \times 100 \text{ s}^{-1}$, which is similar to recent estimates for IC decay via $5f$ electrons [14,17]. This effect may be useful for optimizing the performance of future solid-state clocks, as the engineering of deliberate electronic defects could reduce the clock interrogation time, enhance the signal-to-noise ratio, and potentially enable alternative readout schemes such as transferring the excitation to an optically active defect [26].

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The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

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Correction: This paper should have published concurrently with F. Schaden *et al.*, *Phys. Rev. Res.* **7**, L022036 (2025).