

Laser-induced quenching of the Th-229 nuclear clock isomer in calcium fluoride

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The 10-min radiative lifetime of the first excited $^{229}\text{Th}^{4+}$ nuclear state in ionic crystals provides narrow spectroscopic linewidths, enabling the realization of a solid-state nuclear clock. Due to the $4+$ noble gas configuration, electronic readout or state initialization schemes known from atomic clocks are inaccessible. This elongates the interrogation cycle, which will deteriorate the clock performance. To address this limitation we demonstrate laser-induced quenching (LIQ) as a method of depumping the ^{229}Th isomer population in CaF_2 . We provide experimental evidence for LIQ at different wavelengths (148–420 nm) and temperatures (100–350 K), achieving a threefold reduction in the isomer lifetime with 20 mW of laser power.

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Introduction. The nuclear isomer state of ^{229}Th is intensively studied due to its unique low excitation energy at approximately 8.4 eV [1]. The first laser excitation of this nucleus was recently demonstrated [2] and quickly confirmed [3]. Additionally, the lifetime of its excited state ^{229m}Th has been determined in both vacuum and various solid-state environments [2–6] and the hyperfine splitting due to the crystal electric field gradient has been resolved in precision laser spectroscopy [7]. This unique isomeric state has inspired numerous proposals for applications [8–11], particularly focusing on a nuclear optical clock that could potentially outperform atomic clocks based on electronic excitations [12,13]. The ^{229}Th nuclear clock would be highly sensitive to new physics because its transition frequency is strongly influenced by both the nuclear and electromagnetic forces [14–18].

$^{229}\text{Th}^{4+}$ doped in a large band gap solid is a strong candidate for a compact nuclear clock: a far greater number of nuclei can be interrogated in an ionic crystal compared to an ion trap and the ions in the lattice are deeply in the Lamb-Dicke regime [11]. Optical clocks operate in cycles consisting of state preparation, excitation of the clock transition, and state detection. Ideally, the time used for excitation should dominate the duration of the cycle, because it will determine the Fourier-limited frequency resolution. However, in the $\text{Th}:\text{CaF}_2$ optical clock, the nuclear coherence time

of ≈ 10 ms, which determines the longest efficient excitation time, is much shorter than the ≈ 600 s lifetime of the isomer. This presents an obstacle to the effective operation of a solid-state nuclear clock, thereby deteriorating the expected clock's overall performance [19,20]. Previous studies have demonstrated that quenching, i.e., shortening the lifetime of the isomeric state, is possible using x rays [21]. This is however impractical for the tabletop operation desired in solid-state nuclear clock applications. Furthermore, $^{229}\text{Th}^{4+}$ ions have a noble gas electronic configuration, which lacks optically accessible electronic transitions that could be used to rapidly quench the isomer nuclear state via an electronic bridge process [22].

A laser-induced quenching (LIQ), as opposed to x-ray-induced quenching, of the isomer state could enable tabletop controlled initialization of the Thorium-229 nucleus into its ground state, which merits further investigation. Aside from radiative decay, the isomeric state can decay via internal conversion where it transfers its excitation energy to an electron in the surrounding environment, exciting it to the conduction band, potentially with a much shorter time constant [1,20,23]. Recently, it has been suggested that fluoride vacancies that are created due to irradiating CaF_2 can cause electronic levels to emerge, which are resonant with the isomer energy, that can be used for LIQ [24].

Here we report the demonstration and extensive investigation of LIQ of the ^{229}Th isomer population in calcium fluoride. We show that the isomer state decays unperturbed over a temperature range between 100 K and 350 K when no quenching light is present. Furthermore, using LIQ, the isomer can be forced to decay up to three times faster at 297 K using 30 mW of average laser power, independent of wavelength between 148 nm and 420 nm. At longer wavelengths, tested for 729 nm and above, LIQ is not present. Finally, LIQ can be suppressed by cooling. LIQ offers an effective and easy-to-implement method for reducing the total duration of

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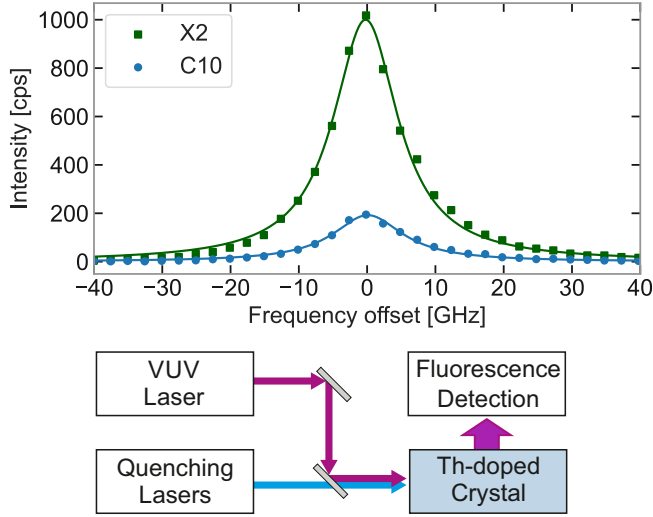


FIG. 1. Laser spectroscopy setup and excitation spectra of the processed nuclear resonance fluorescence signals of the two $^{229}\text{Th}:\text{CaF}_2$ crystals X2 and C10 with different doping concentrations $5 \times 10^{18} \text{ cm}^{-3}$ and $3 \times 10^{17} \text{ cm}^{-3}$ [26].

the clock interrogation cycle and can improve the stability of a solid-state nuclear clock.

Experimental details: VUV spectroscopy setup. The experimental setup consists of a crystal spectroscopy vacuum chamber with a ^{229}Th -doped crystal and a photomultiplier tube (PMT) fluorescence detection scheme [2], a vacuum ultraviolet (VUV) laser system [25], and a set of lasers emitting at different wavelengths to study laser-induced effects in a Th-doped crystal (see Fig. 1). Improving on the setup used in [2], we implement a frequency stabilization of the VUV laser at each scanning point. The processed nuclear resonance fluorescence signals of two $^{229}\text{Th}:\text{CaF}_2$ crystals (referred to as C10, X2) with different doping concentration [26] are shown in Fig. 1. Signal records are obtained with 100 s irradiation and 250 s detection times. Postprocessing compensates the asymmetric shape of the recorded spectra when the scanning speed exceeds the decay rate (see [2]). The C10 crystal was freshly polished and fluorinated which improved its transmission to $>60\%$ at 150 nm [27]. The nuclear transition frequency in CaF_2 is measured to be 2020407.8(15) GHz, which corresponds to an energy of 8.355734(6) eV, in agreement with the previous results reported in [2,7]. These results also agree with those for ^{229}Th doped into LiSAF, showing the influence of the host crystal on the nuclear transition frequency to be less than 1 GHz [3].

For the investigation of the laser-induced effects in the crystal, we use two laser sources. One is a 10 ns pulsed tunable Ti:Sa laser with a repetition rate of 1 kHz and a frequency conversion of its fundamental radiation to the second and third harmonics. This laser is used to irradiate the crystals at 280 nm, 420 nm, 729 nm, and 820 nm wavelength. The second source is a continuous wave (cw) diode laser at 402 nm. The 402 nm laser is used to compare between radiation-induced effects caused by a pulsed and a cw laser. The radiation of these lasers is superimposed with the VUV radiation and steered to the spectroscopy vacuum chamber. The VUV and quenching

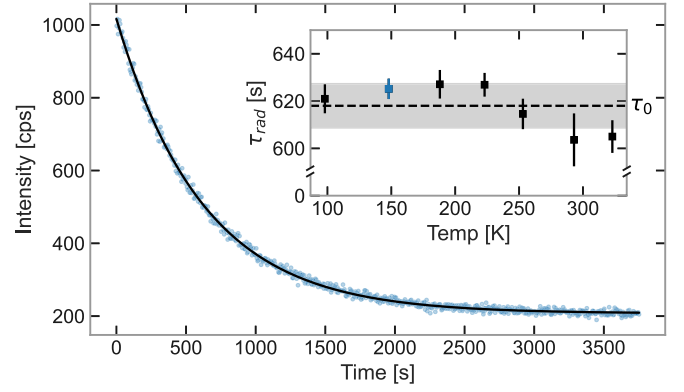


FIG. 2. Nuclear fluorescence decay curve of ^{229}Th in CaF_2 after resonant excitation, recorded at 150 K (C10 crystal). Inset: radiative isomer lifetime measured at various temperatures. The dashed line corresponds to the mean value and the gray area is the standard deviation.

light are focused to ≈ 0.5 mm diameter at the Th-doped crystal and the overlap of both beams is monitored by a CCD camera. Further details on setup can be found in the Supplemental Material (SM) [28].

Crystal production. Two $^{229}\text{Th}:\text{CaF}_2$ crystals were grown at TU Wien, referred to as X2 and C10 [2,26]: different pieces of these crystals were used in VUV spectroscopy experiments at SPring-8 [21], PTB [2], and JILA [7,29] and demonstrated suitability for precision nuclear laser spectroscopy. The two crystals differ in their ^{229}Th doping concentration, thus changing the number of nuclei participating in the spectroscopy, as well as the crystal properties. The concentrations are (cm^{-3}): 5×10^{18} and 3×10^{17} with column densities (mm^{-2}): 8×10^{15} and 1×10^{15} , for X2 and C10, respectively [2].

Results and discussions: ^{229}Th isomer radiative lifetime over temperature. We start by investigating the temperature dependence of the radiative isomer lifetime in CaF_2 . The C10 crystal, mounted on a cryogenic liquid nitrogen cold finger with a resistive heater, was temperature stabilized at different values between 100 K and 350 K. For each temperature, the crystal was irradiated for 500 s with the on-resonance VUV laser followed by a recording of the isomer fluorescence decay using a PMT. Such a decay curve recorded at 150 K, as well as an exponential fit, is shown in Fig. 2. No statistically significant changes in the decay constant are observed over the investigated temperature range, as presented in the inset. The average of these lifetime measurements yields $\tau_0 = 618(9)$ s, which is in agreement with the previously reported lifetime measurements in CaF_2 [2,7,21]. We observe identical excitation energy and radiative lifetime for the C10 and X2 crystals (further details in the SM [28]).

^{229}Th isomer population dynamics. While we have ascertained that the isomer excitation energy and radiative lifetime do not change with crystal temperature (within experimental uncertainty), we observed a substantial reduction of the isomer signal strength, decreasing by a factor of 3 as the temperature is increased from 100 K to 350 K, and laser power and irradiation duration remained constant. This decrease indicates the presence of a competing decay process

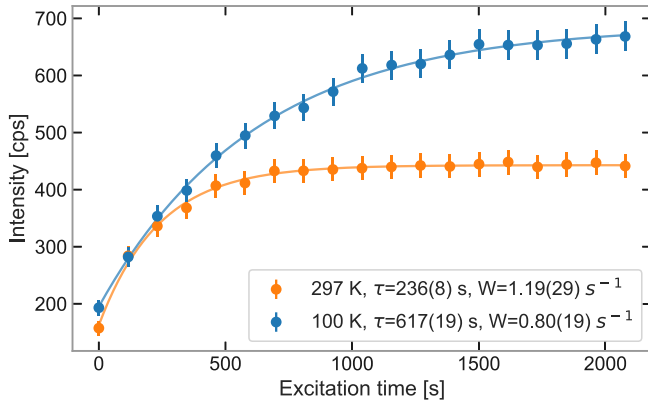


FIG. 3. Excitation dynamics of the ^{229}Th isomer in the C10 crystal at two different temperatures. Blue: measured at 100 K with an extracted $\tau = 617(19) \text{ s} \approx \tau_0$ and no LIQ observed. Orange: measured at 297 K with an extracted $\tau = 236(8) \text{ s}$ and clear indication of LIQ.

during excitation that reduces the overall population of excited ^{229}Th nuclei within the crystal. A similar effect, termed “x-ray-induced quenching,” was observed in [21] in which the irradiation of the $^{229}\text{Th}:\text{CaF}_2$ crystal with a 29 keV x-ray beam reduced the isomer population.

With a spectral power density of $\approx 0.1 \text{ pW/Hz}$, the nuclear laser excitation is performed significantly below the saturation of the isomeric transition. Therefore, while increasing the excitation time t and keeping the VUV laser power constant, the isomer population approaches a steady state level where the excitation rate equals the isomer decay rate. The excitation dynamics can be described by [30]

$$n_{\text{isomer}}(t) \approx W\tau(1 - e^{-t/\tau}), \quad (1)$$

where n_{isomer} is the normalized population of the excited state, W is the excitation rate, and τ is the lifetime resulting from the combination of quenching and radiative decay (see the SM [28]). In the case of pure radiative decay without additional decay channels, $\tau = \tau_0$. Finding an equilibrium timescale $\tau < \tau_0$ with an associated reduction of the fluorescence yield indicates the presence of additional decay channels during the excitation phase, which we refer to as LIQ.

We examine the reduction of isomer lifetime of the C10 crystal during excitation at two different temperatures, using Eq. (1). The results can be seen in Fig. 3. At 100 K the time constant τ matches the radiative decay constant τ_0 , indicating no LIQ. At room temperature, the time constant τ was reduced by a factor of 2.6, indicating an additional decay process present during laser irradiation.

Active off-resonant isomer quenching. We conjectured that the LIQ observed in the excitation dynamics is related to the intense off-resonant VUV laser radiation. To study LIQ independently from the excitation dynamics, we implemented isomer quenching after nuclear excitation, primarily using the higher-doped X2 crystal. C10 behaves qualitatively the same as X2, however, there is a significant quantitative difference with stronger LIQ observed for the lower-doped crystal.

For these measurements we excited the isomer on resonance for up to 500 s, followed by fluorescence detection

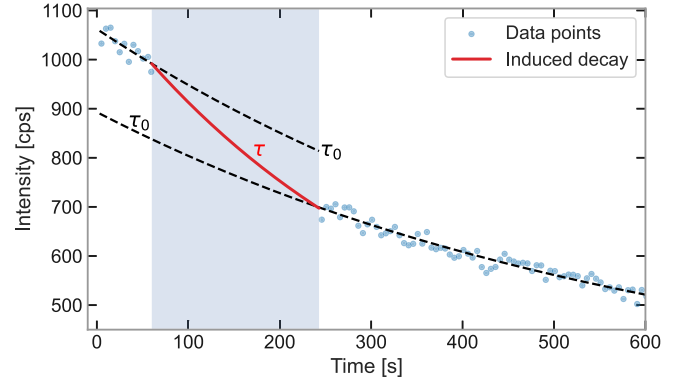


FIG. 4. Quenching of the ^{229}Th isomer with off-resonant 148 nm pulsed laser radiation: Exponential isomer decay signal (X2 crystal) with the radiative lifetime τ_0 recorded before and after off-resonant laser irradiation. Blue shaded area: time window in which the crystal was irradiated off resonantly. Red curve: laser-induced exponential decay with a lifetime $\tau < \tau_0$.

over a period of 60 s. Thereafter, the isomer was exposed to off-resonant laser light for up to 200 s where the VUV center frequency was shifted by approximately 100 GHz. Additionally, we studied LIQ using wavelengths between 280 and 840 nm. The off-resonance light does not induce further excitation of the isomer, but would cause LIQ.

A typical result obtained with the off-resonance scheme is depicted in Fig. 4; the dashed black lines indicate the exponential decay of the isomer, characterized by the radiative lifetime τ_0 , both before and after the off-resonant laser irradiation. In the blue shaded area, during the off-resonance laser irradiation, an isomer lifetime of $\tau < \tau_0$ can be inferred, as shown by the red curve. No fluorescence signal could be taken during the off-resonant laser irradiation period because of stray light blinding the PMT.

Temperature, power, and wavelength dependence. Utilizing the above mentioned off-resonance scheme, we investigated the LIQ in the X2 crystal for different temperatures and with two different lasers: the off-resonant pulsed VUV excitation laser [2,25] operating at an average power of 0.2 mW and a cw laser with a wavelength of 402 nm and a power of 10 mW. For the C10 crystal, the temperature dependence was solely investigated using the VUV excitation laser. We scanned a temperature range from 170 K to 340 K and extracted the reduced lifetime τ .

A clear temperature dependence of τ was observed, as illustrated in Fig. 5. Regardless of laser power, the observed temperature dependence appeared consistent, with stronger LIQ observed at higher temperatures, while LIQ was significantly reduced or completely suppressed at lower temperatures. The quantitative difference in the LIQ between the two types of lasers can be attributed to the large power difference between both lasers, which is investigated further below. The quenching rate difference between X2 and C10 is subject to further investigation. Note that in the case of the off-resonant 148 nm VUV laser, the quenching light is intrinsically aligned to the isomeric nuclei produced inside the crystal while, in the case of the 402 nm laser, the alignment is nontrivial, possibly underestimating the LIQ effect for a given

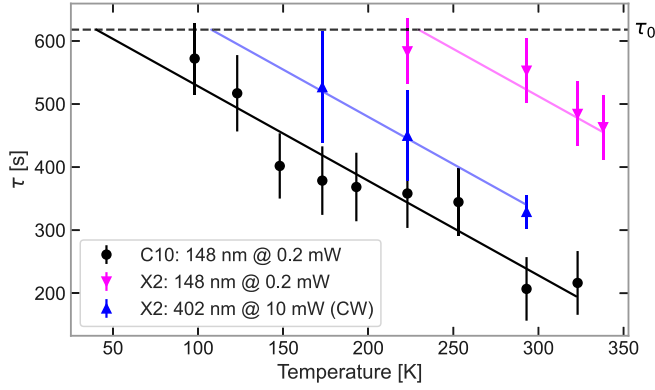


FIG. 5. Isomer lifetime τ affected by laser quenching for different temperatures (X2 and C10 crystal). Dashed black line: unperturbed radiative lifetime τ_0 . Blue: LIQ was induced by a 402 nm cw laser with a power of 10 mW. Purple: LIQ was induced using the pulsed VUV laser shifted by 100 GHz away from the isomer transition frequency. Linear fits to guide the eye have imposed identical slope, yielding -1.5 s/K.

laser power. At the lowest investigated temperature of 100 K, a full suppression of the LIQ ($\tau \simeq \tau_0$ within uncertainty) could be observed for all investigated laser sources and powers.

To investigate the power dependence of LIQ, we employed the same off-resonance scheme while maintaining a constant temperature of approximately 297 K and varying the laser power. This investigation utilized five different laser wavelengths: the pulsed VUV laser detuned by 100 GHz from the isomer transition with a 0.2 mW average power, a 280 nm pulsed laser with adjustable power, a 402 nm cw laser with 10 mW, a 420 nm pulsed laser with 32 mW, and an infrared laser operating at 729 nm and 840 nm with 80 mW and 96 mW, respectively. It is important to note that, with the exception of the VUV laser, the alignment for each of these lasers in the crystal varies and the effective laser power can be reduced by up to 50%, caused by imperfect overlap of the quenching laser beam with the excited nuclei. Therefore, it is impossible to give a precise quantitative comparison between different lasers. Figure 6 presents the isomer lifetime τ as a function of laser power for different laser wavelengths. For wavelengths between 148 nm and 420 nm the power dependence of LIQ appears to be independent of wavelength. At near infrared wavelengths of 729 nm and above, even a 100 mW laser fails to induce LIQ. The LIQ process seems to be activated above a (yet to be precisely determined) photon energy threshold in the visible range.

LIQ enhanced clock operation. High-performance frequency standards usually employ some form of Rabi or Ramsey interrogation scheme. In these methods, the obtained error signal is proportional to the difference of the frequency of a laser used for the interrogation and the rotation frequency of the state vector describing the coherent superposition of the two “clock” states, which is equal to the frequency of the clock transition. In addition, these methods require exact knowledge of the quantum state at the beginning of any interrogation cycle. For the ^{229}Th solid-state clock, these methods cannot be applied, because the decoherence time, given by interaction with randomly oriented magnetic momenta of surrounding

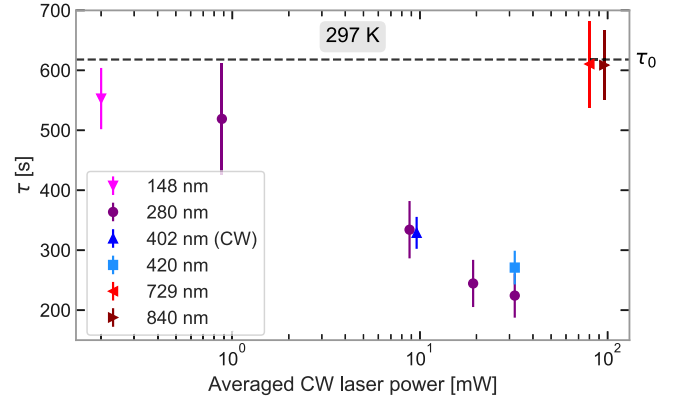


FIG. 6. Off-resonant LIQ in the X2 crystal at different laser powers and wavelengths, at room temperature. The 402 nm laser is cw while all other lasers are pulsed. The dashed black line shows the unperturbed radiative lifetime τ_0 .

fluorine nuclei [19], is of order milliseconds. The time necessary to efficiently transfer the nuclei from the ground to the isomeric state can be much longer and the time τ_0 of spontaneous decay from the isomeric state back to the ground state exceeds the decoherence time by many orders of magnitude.

In [19] we proposed an alternative scheme to stabilize the interrogation laser, based on a comparison of fluorescence signals measured after irradiation of the sample by a laser, detuned from the (assumed) clock transition frequency to the red and blue side, respectively. These fluorescence signals not only depend on the instantaneous parameters of the laser used in the corresponding measurement cycle but also on the number of excited nuclei remaining from previous measurement cycles. While these contributions simply constitute an offset that can be subtracted, the overall signal increases and hence the overall shot noise, increasing the instability in the determination of the frequency offset.

The optimal short-term stability for such a scheme is attained when the time of a single interrogation cycle t_{cycle} is comparable with, or longer than, the radiative lifetime τ_0 , transferring the majority of excited nuclei back to the ground state. In such a scheme, the clock performance is strongly dependent on the intrinsic stability of the laser used for excitation [31].

With the help of controllable quenching, one can reduce or even completely zero the number of excited nuclei remaining from previous interrogation cycles. It allows one to not only reduce the shot noise but also to significantly shorten the interrogation cycle. Figure 7 shows the ratio of the instability in determining the laser frequency offset without (σ) and with LIQ (σ_{LIQ}), assuming instantaneous quenching.

Quenching operation allows us to reach lower fractional frequency instability at shorter cycle times. This significantly reduces the operation time, which will relax the constraints on the interrogation laser stability. This is particularly important in searches for new physics [32] in the high-frequency domain, where a combination of a short probing time and a high sensitivity is crucial.

Conclusion. To conclude, we have demonstrated that a broad range of laser wavelengths can induce accelerated decay

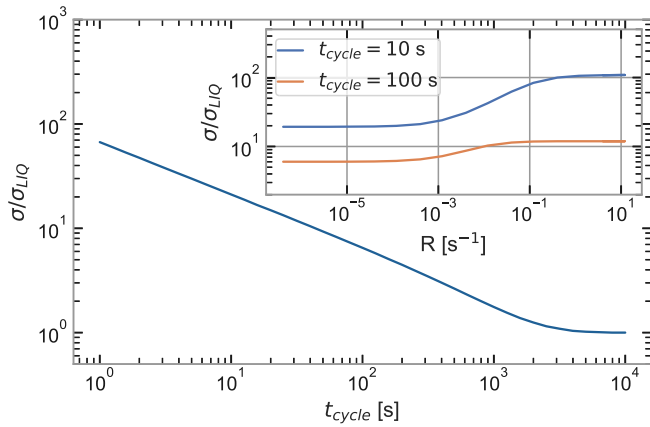


FIG. 7. Ratio of frequency instabilities of a $^{229}\text{Th}:\text{CaF}_2$ clock without (σ) and with LIQ (σ_{LIQ}) based on Ref. [19] as a function of a single interrogation cycle time for an excitation rate $R = \gamma$. The fractions of the interrogation cycle used for excitation, as well as the detunings of the probe field from the assumed clock transition frequency, are optimized for any point. Inset: the same ratio but as a function of excitation rate R for 10 s and 100 s as cycle time (t_{cycle}).

of the ^{229}Th isomer. Within our experimental conditions, LIQ is largely independent of wavelength below 420 nm and is completely absent at wavelengths of 729 nm and above. We observe an approximately linear scaling of LIQ with laser power. Additionally, LIQ is thermally activated with an activation temperature above 100 K.

The underlying physical process of LIQ is currently unknown and will be the focus of further investigations, in particular for different host crystals. LIQ at 148 nm wavelength at room temperature has recently been reported for $\text{Th}:\text{LiSrAlF}_6$ [33]. Our study shows that it requires two factors, possibly proceeding in two separate steps: a (single) photon of approximately 2 eV energy and thermal activation in the range of 0.02 eV. The latter might provide mobility to (laser-induced) defect centers [24,34] within the crystal. These might create intragap electronic states that can interact with the nucleus via electron bridge processes [20,23]. Another possibility is the creation of phonons which mediate the internal conversion process by ensuring conservation of energy and momentum.

Regardless of the specific cause, LIQ of the excited ^{229}Th isomer presents several intriguing applications. First, it demonstrates that the excited nucleus can interact with the crystal environment and potential defect states, offering a different and possibly more efficient excitation pathway. Second, LIQ opens up the possibility of initializing the ^{229}Th nucleus into its ground state in a $^{229}\text{Th}:\text{CaF}_2$ crystal and, with that, an enhanced clock performance [19]. Additionally, LIQ could provide an explanation for previous unsuccessful attempts to excite and detect the isomer using broadband VUV synchrotron sources [35,36]: the significant off-resonance fraction of photons together with the high crystal temperature probably induced significant LIQ. Future solid-state-based ^{229}Th nuclear excitation approaches invoking broadband radiation or multiphoton schemes will require cooling of the crystal and/or suppression of nonresonant photons.

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Data availability. The data that support the findings of this article are openly available [37]; embargo periods may apply.

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Correction: This paper should have published concurrently with J. E. S. Terhune *et al.*, *Phys. Rev. Res.* **7**, L022062 (2025).