Nuclear Science and Techniques (2025) 36:20
https://doi.org/10.1007/s41365-024-01632-w

RESEARCH HIGHLIGHT

Triggering highly nonlinear responses in ^{229}Th nuclei with an intense laser

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Received: 28 November 2024 / Revised: 28 November 2024 / Accepted: 28 November 2024 / Published online: 9 January 2025

In a recent paper published in Phys. Rev. Lett. 133, 152503 (2024), H. Zhang, T. Li, and X. Wang predicted that modern intense lasers can induce highly nonlinear responses in the ^{229}Th nucleus for the first time, which is an astonishing effect of light-nucleus interactions. This phenomenon is underpinned by two key factors: (1) the presence of a very low-lying nuclear excited state and (2) a nuclear hyperfine mixing effect that significantly enhances light-nucleus coupling. The resulting highly nonlinear responses facilitate efficient nuclear excitation and enable coherent light emission from the nucleus, resulting in high harmonic generation. ^{229}Th presents a promising platform for advancements in both laser-nuclear physics and nuclear clock development. The pioneering work by Zhang et al. marks a new frontier in light-matter interactions.

Lasers with intensities exceeding 10^{13} W/cm^2 can induce highly nonlinear responses in atoms and molecules, resulting in novel phenomena, such as multiphoton and above-threshold ionization [1, 2], nonsequential double ionization [3–5], high harmonic generation [6, 7], and laser-induced electron diffraction [8–10]. A particularly significant outcome of intense laser-atom interactions is the generation of attosecond (1 as = 10^{-18} s) pulses [11, 12], which is a breakthrough that achieved the 2023 Nobel Prize.

The ability to induce highly nonlinear responses in atoms arises when the laser-atom interaction energy E_{I} becomes a non-negligible fraction of the atomic transition energy ΔE. The former is the product of the atomic transition moment D and laser field amplitude $\mathcal{E}_0$. Thus, the following ratio serves as an indicator of whether highly nonlinear responses can be initiated:

$$\eta = \frac{E_{\mathrm{I}}}{\Delta E} \approx \frac{D\mathcal{E}_0}{\Delta E}. \tag{1}$$

If $\eta \ll 1$, the response remains linear, while if $\eta \sim 1$, highly nonlinear responses can occur. For atoms with $D \sim 1$ a.u. (atomic units) and $\Delta E \sim 1$ a.u., a laser intensity of 10^{13} W/cm^2 (equivalent to $\mathcal{E}_0 \sim 0.01$ a.u.) results in $\eta \sim 0.01$, which is typically sufficient to trigger highly nonlinear responses.

The induction of highly nonlinear responses in atomic nuclei presents a significant challenge. First, a nucleus is approximately five orders of magnitude smaller than an atom, resulting in correspondingly smaller transition moment values D. Second, the nuclear transition energy ΔE is typically five orders of magnitude greater than that of atoms. Consequently, to achieve a similar η value for nuclei, the laser field amplitude $\mathcal{E}_0$ must be ten orders of magnitude higher. This translates to a required laser intensity of 10^{33} W/cm^2, which is prohibitively high and far beyond current technological capabilities (with the latest intensity record being 10^{23} W/cm^2 [13]).

However, this estimation applies to "typical" nuclei, whereas nature provides certain exceptional cases. One such nucleus is thorium-229 (^{229}Th), which features an extremely low-lying excited state just 8.4 eV above the nuclear ground state. The minimized transition energy of the ^{229}Th nucleus is $\Delta E \approx 0.3$ a.u., which significantly increases η. However, the transition moment remains very small at $D \sim 10^{-7}$ a.u. Even at the highest currently achievable laser intensity of 10^{23} W/cm^2 (equivalent to $\mathcal{E}_0 \sim 10^3$ a.u.), the resulting $\eta \sim 10^{-4}$ is still insufficient to trigger highly nonlinear responses.

The second key element is the nuclear hyperfine mixing (NHM) effect. An inner-orbital electron generates a strong electromagnetic field at the nucleus site, causing mixing between nuclear states. This effect has significant

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implications for the ^{229}Th nucleus. The lifetime of the isomeric state for the bare nucleus is of the order of 10^3 s. In contrast, this lifetime significantly decreases by five orders of magnitude to 10^{-2} s for the hydrogen-like ionic state, where the 1s electron contributes to the NHM effect. This reduction occurs because the NHM effect enhances the coupling between the nucleus and light field, effectively modifying the transition moment from D to $D' = D + b\mu_e$. Here, $b \approx 0.03$ is the mixing coefficient, and μ_e is the magnetic moment of the 1s electron. The additional term $b\mu_e \sim 10^{-4}$ a.u. is approximately three orders of magnitude larger than D, highlighting the substantial influence of NHM on the nuclear dynamics.

As the transition moment increases to $D' \sim 10^{-4}$ a.u., it can be seen that η becomes ~ 0.1 for a laser intensity of 10^{23} W/cm^2, which is sufficient to trigger highly nonlinear responses in ^{229}Th. This was recently confirmed by numerical results obtained by Zhang et al. [14].

Figure 1a illustrates the nuclear excitation probability as a function of laser intensity for both the bare nucleus (Th^{90+}) and hydrogen-like ion (Th^{89+}). In the case of Th^{89+}, the nuclear excited state splits into two levels due to hyperfine splitting, and the excitation probabilities for both levels (with total angular momentum $F = 1$ and 2) are displayed. For Th^{89+}, the excitation probabilities deviate from linearity at intensities above 10^{17} W/cm^2, exhibiting highly nonlinear characteristics and rapidly climbing to the 10% level at an intensity of 10^{21} W/cm^2. In contrast, for the bare nucleus Th^{90+}, the excitation probability remains linearly dependent on laser intensity all the way up to 10^{23} W/cm^2, indicating that the response is still linear and fails to evoke nonlinear effects. Moreover, the absolute value of the excitation probability remains relatively small.

With highly nonlinear responses triggered in Th^{89+}, radiation is emitted at multiple frequencies, which are the high-order harmonics of the laser frequency, as illustrated in Fig. 1b for four different laser intensities. The third and fifth harmonics appear at intensity 10^{18} W/cm^2. As the intensity increases, higher-order harmonics become evident, with the 31st harmonic observable at 10^{21} W/cm^2.

The potential for inducing highly nonlinear responses in nuclei presents several important applications: (1) efficient excitation and manipulation of nuclei—an excitation probability of 10% per nucleus per laser pulse is remarkable, paving the way for further quantum-state manipulation. (2) High harmonic generation as a novel mechanism for nucleus-based coherent light emission, which is distinct from the long-sought nuclear laser concept based on population inversion [15]. (3) High harmonic spectra as a new spectroscopic technique for extracting nuclear information—these spectra encode valuable nuclear data, such as the nuclear transition moment, which can be decoded through detailed analysis.

Beyond serving as a valuable platform for studying laser-nucleus interactions, the ^{229}Th nucleus holds immense promise for the construction of nuclear clocks. A peculiar low-energy nuclear transition in ^{229}Th has been proposed as the basis for nuclear clocks [16], offering distinct advantages over atomic clocks. Substantial progress has been made in recent years [17–24].

Here, I highlight the recent advancements in China: (1) vacuum ultraviolet (VUV) light sources: our group in Wuhan has developed a VUV frequency comb that provides narrowband (< 30 MHz) VUV light centered at 148

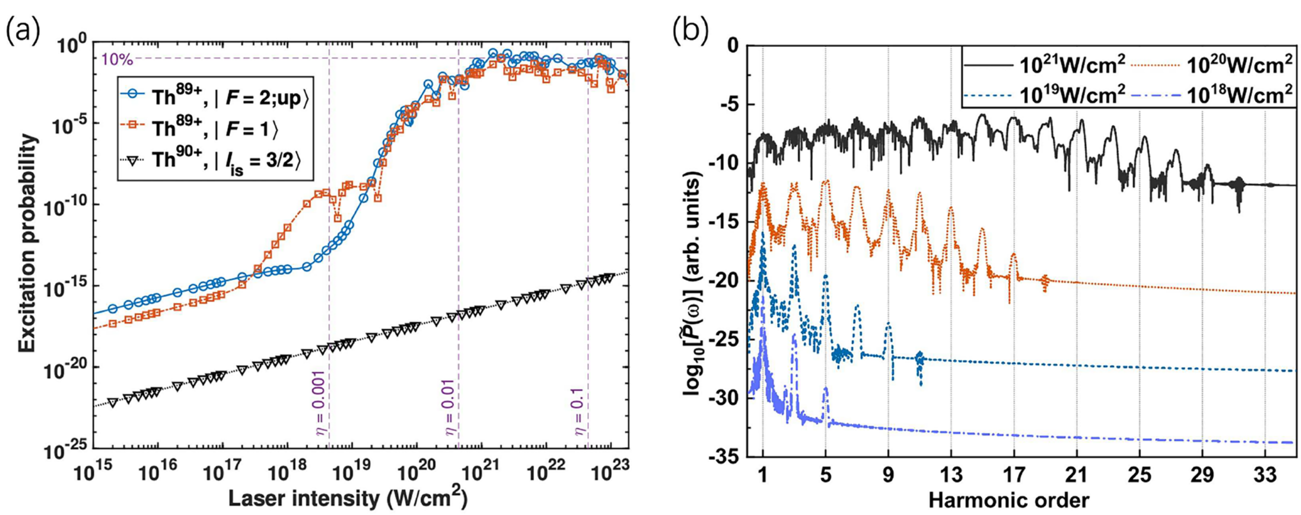

Fig. 1 (Color online) **a** Nuclear isomeric excitation probability as a function of laser intensity for the bare nucleus (Th^{90+}) and hydrogen-like ion (Th^{89+}). In the Th^{89+} case, the nuclear excited state splits into two levels. **b** High harmonic spectra generated from the laser-driven Th^{89+} system under four different laser intensities. Figures adapted from Ref. [14]

nm [25]. Additionally, Xiao et al. at Tsinghua University proposed a VUV laser scheme based on four-wave mixing in cadmium vapor [26]. (2) Th-doped crystals: Gong et al. investigated various Th-doped wide-bandgap crystals as potential materials for building solid-state nuclear clocks [27, 28]. (3) ^{229}Th production: Ma et al. proposed generating ^{229}Th through accelerated ^{232}Th and neutron knocking-out reactions [29], while Wu et al. suggested using γ rays to remove three neutrons from ^{232}Th [30]. (4) Isomeric excitation methods: a variety of theoretical methods for isomeric excitation have been explored, including laser-driven electron recollision [31], electron bridge mechanisms [32, 33], inelastic electron scattering [34], laser-cluster interactions [35], proton Coulomb excitation [36], and electron capture [37].

In conclusion, ^{229}Th stands out as a promising platform for both laser-nuclear physics and nuclear clock development, with substantial advancements already achieved and further progress anticipated.

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