Exciting the Isomeric ^{229}Th Nuclear State via Laser-Driven Electron Recollision

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We propose a new approach to excite the isomeric ^{229}Th nuclear state, which has attracted much attention recently as a potential "nuclear clock." Our approach is based on a laser-driven electron recollision process, the core process of strong-field atomic physics. Bringing together knowledge of recollision physics and of the related nuclear physics, we calculate the isomeric excitation probability. This new approach does not require precise knowledge of the energy of the isomeric state. The excitation is well timed which may be exploited to control the coherence of the isomeric state. Experimental realization is within reach using tabletop laser systems.

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Among all known nuclei the ^{229}Th nucleus has a unique low-lying isomeric state with an energy of about 8 eV above the ground state [1–4]. The existence of this isomeric state has fascinated the scientific community with the possibility of a nuclear clock that complements or even outperforms current electronic-shell-based atomic clocks in precision and in robustness against environmental perturbations [5,6]. Substantial progress has been made during the past few years in characterizing this isomeric state [4,7–11].

One of the major problems yet to be solved is how to prepare the isomeric state in a controllable and efficient way. Naturally, the isomeric state can be obtained via α decay of the ^{233}U nucleus (half-life 1.592×10^5 years) with 2% of the resultant ^{229}Th nuclei in the isomeric state. However, this decay is largely uncontrollable: the ^{229}Th nuclei are left with a recoil energy of 84 keV into random directions and various ionic states. Direct optical excitation from the nuclear ground state is conceptually the most straightforward way, yet experimental attempts have not been successful [12–15], possibly due to inaccurate knowledge of the isomeric state energy [16]. An alternative excitation approach that has been successfully demonstrated experimentally is by Masuda *et al.*, who use 29-keV photons from synchrotron radiation to pump the population from the ground state to the second excited state, which then decays into the isomeric state [17]. Proposals exploiting electronic bridge processes have been made for various ionic states [18–21]. A proposal exploiting photon excitation by the fifth harmonic of Ti:sapphire lasers has also been made [22].

The goal of the current Letter is to propose a new, controllable, and efficient approach for the isomeric excitation. Our approach is based on a laser-driven electron recollision process. In an intense laser pulse, one (or several) of the outer electrons of the ^{229}Th atom can be pulled out by the laser electric field. The emitted electron, which has not yet escaped the control of the laser field albeit in the continuum, may be driven back to collide with and excite the nucleus. A schematic illustration of the idea is shown in Fig. 1.

Recollision [23–26] is the core process of strong-field atomic physics. It has been well understood that the recolliding electron may (i) recombine radiatively to the ion core leading to high harmonic generation [27–29]; or (ii) kick out another electron (or electrons) leading to nonsequential double (multiple) ionization [30–32]; or (iii) elastically scattered by the ion core leading to a diffraction pattern that encodes the instantaneous ion-core structural information [33–35]. The recolliding electron usually has an energy of several tens of electronvolts with typical laser parameters, therefore it is not energetic enough to have an effect on a typical nucleus. With the ^{229}Th nucleus, however, the new channel of recollision-induced nuclear excitation (RINE) is energetically opened.

This RINE process is a combination of two processes: one is electronic excitation of the ^{229}Th nucleus, and the other is laser-driven electron recollision. Both processes are in fact well understood and the required work here is to build a bridge between them.

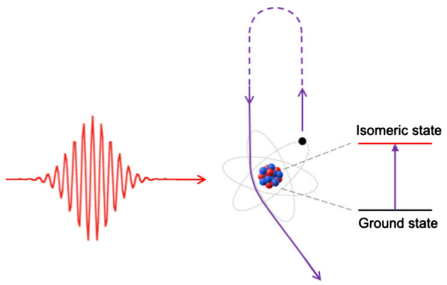

FIG. 1. Schematic illustration of our approach. When exposed to an intense laser pulse, a ^{229}Th atom (or ion) loses an electron due to strong-field ionization. The emitted electron may later be driven back to recollide with and excite the nucleus from the ground state to the isomeric state.

The process of Coulomb excitation of nuclei has been treated in detail in the literature, e.g., in the classic article of Alder *et al.* [36]. In Fig. 2(a) we plot the isomeric excitation cross section of ^{229}Th by electrons. The electric dipole ($E1$) channel is forbidden due to a parity consideration, and the excitation is mainly through the electric quadrupole ($E2$) and the magnetic dipole ($M1$) channels. By calculating the cross section we have used the reduced transition probabilities $B(E2) = 27$ W.u. (Weisskopf units) and $B(M1) = 0.0076$ W.u., as suggested by Minkov and Pálffy [37]. Because of page limit, we shall not list the detailed cross section formulas here, and the reader can refer to Eqs. (II E.40–46) of [36]. One sees that for electron excitation, the $E2$ channel is more important than the $M1$ channel. This is different from the case of radiative decay or direct light excitation, in which the $M1$ channel dominates. The domination of the $E2$ channel has also been found by Bilous *et al.* when the internal conversion or the electronic bridge processes are considered, both involving coupling with electrons [38].

For the reader who is not familiar with recollision, we give a brief explanation to it. Let us first neglect the ion-core potential and assume that an electron is emitted with zero velocity. (These are the assumptions of the intuitive "simple-man model" [39].) The electron is mostly emitted around field peaks. For recollision to happen later, however, the electron needs to be emitted *after* a field peak. Recollision happens about 3/4 cycles later, around field zeros. The relation between the recollision time t_r and the emission time t_i is given in Fig. 2(b). (Here, we only consider first-time recollisions. Higher-time recollisions are neglected due to more severe wave-packet spreading.) Different t_i will lead to different kinetic energies at recollision, as shown in Fig. 2(c). The electron emitted at $t_i = 17°$ after a field peak will have the highest recollision energy of $3.17U_p$, where $U_p = E_0^2/4\omega^2$ is the ponderomotive energy. E_0 is the laser electric field and ω is the angular frequency.

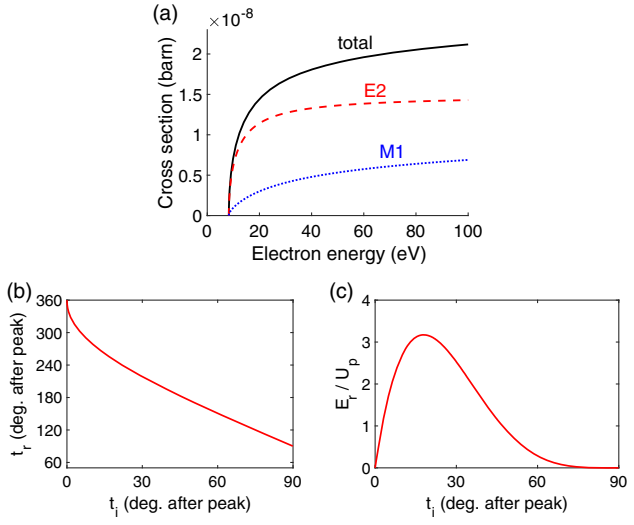

FIG. 2. (a) Isomeric excitation cross section of ^{229}Th by electrons. $E2$ denotes the cross section from the electric quadrupole channel and $M1$ denotes that from the magnetic dipole channel. (b) Relation between the recollision time t_r and the ionization time t_i within a laser cycle, given in degrees starting from a field peak. (c) Kinetic energy of the electron at the time of recollision as a function of the ionization time t_i. E_r is in units of the ponderomotive energy U_p.

In order to obtain an effective recolliding-electron flux density, we perform a numerical simulation going beyond the simple-man model. Now let us consider a single electron (which could be the first-emitted, second-emitted,…, electron). The electron is assumed to be emitted via quantum tunneling at each time with an Ammosov-Delone-Krainov (ADK) tunneling rate [40]

$$w(t) = \frac{C_l^2}{2^{|m|}|m|!}\frac{(2l+1)(l+|m|)!}{2(l-|m|)!}\frac{1}{\kappa^{2Z_c/\kappa-1}} \times \left(\frac{2\kappa^3}{|E(t)|}\right)^{2Z_c/\kappa-|m|-1} e^{-2\kappa^3/3|E(t)|}, \quad (1)$$

where l and m are the angular momentum quantum numbers of the atomic state from which the electron is emitted, Z_c is the charge of the ion core seen by the electron, $\kappa = \sqrt{2I_p}$ with I_p the ionization potential, and $E(t)$ is the laser electric field. C_l is a constant on the order of unity. There has been no study reporting the values of C_l particularly for the ^{229}Th atom, and here we put $C_l = 1$ a.u. Reported C_l values are between 1 and 3 a.u. mostly for rare gas atoms [40]. The ionization probability is

$$P_{\rm ion}(t) = 1 - \exp\left[-\int_{t_0}^{t} w(t')dt'\right], \quad (2)$$

where t_0 is the time when the laser pulse starts.

The ^{229}Th atom has 90 electrons, so the first- (second-, third-) emitted electron sees the nucleus plus the remaining 89 (88, 87) electrons. To take into account effects of the remaining electrons, we use an effective potential given by Green, Sellin, and Zachor (the so-called GSZ potential) [41]

$$V(r) = \frac{1}{r}\left[-(Z-N) - \frac{N}{(e^{r/d}-1)\xi+1}\right], \quad (3)$$

where $Z = 90$ is the nuclear charge number, N is the number of the remaining electrons. The parameters d and ξ take values of 0.927 and 5.58 a.u., respectively [41].

From the tunneling picture the electron appears in the continuum at the tunneling-exit point, the position of which can be determined by equating the total potential of the electron to the value $-I_p$. The velocity of the electron right after tunneling ionization is usually assumed to be zero along the longitudinal direction and a Gaussian distribution along the transverse direction [42–47]. The Gaussian

distribution has a form of $P(v_\perp) \propto \exp(-v_\perp^2/\eta^2)$ with $\eta^2 = |E(t)|/\sqrt{2I_p}$, as derived by Ivanov *et al.* [48].

In our calculation, a large number of trajectories are launched to simulate a single electron. The laser pulse is divided into small time steps with $\Delta t = 0.1$ a.u. At each time step, 10^5 trajectories are initiated at the tunneling exit with randomly assigned transverse velocities. Each trajectory is assigned a weight according to the ADK rate and the Gaussian transverse-velocity distribution. The total weight of the 10^5 trajectories at an emission time t_i is set to be $w(t_i)\Delta t[1 - P_{\rm ion}(t_i)]$, where the value in the square bracket is the survival probability at the time.

After being launched, the trajectories will be propagated according to classical mechanics under the influence of the laser field and the ion core GSZ potential (neglecting the energy loss due to nuclear excitation). The Hamiltonian equations of motion are integrated

$$\frac{dr_i}{dt} = \frac{\partial H}{\partial p_i}; \qquad \frac{dp_i}{dt} = -\frac{\partial H}{\partial r_i}, \tag{4}$$

where $i = x,\ y,\ z$ and $H = (p_x^2 + p_y^2 + p_z^2)/2 + V(r) + zE(t)$. The laser field is linearly polarized along the z axis.

We follow each trajectory and record the position (x_0, y_0) when the electron crosses the plane $z = 0$, if it does so (i.e., recollision). Only trajectories with relatively small recolliding radius $R_0 \equiv \sqrt{x_0^2 + y_0^2}$ contribute effectively to the nuclear excitation. The effective recolliding radius can be estimated using the following procedure [36]: (i) We follow each trajectory and obtain the distance $r(t) = \sqrt{x(t)^2 + y(t)^2 + z(t)^2}$. (ii) We calculate the potential $V(t) = V[r(t)]$ using Eq. (3). (iii) We Fourier transform $V(t)$ and look particularly for the component $\tilde{V}(\omega_0) = \int_{-\infty}^{\infty} V(t)e^{-i\omega_0 t}dt$ with $\omega_0 = 8.3$ eV the energy of the isomeric state. It is known that the width of the potential $V(t)$, noted δt, and the energy range of possible excitation, noted $\delta\omega$, satisfy an uncertainty relation $\delta t\delta\omega \approx 1$. The above procedure can be understood from this uncertainty relation. The dependency of $|\tilde{V}(\omega_0)|^2$ on R_0 is shown in Fig. 3. One sees that trajectories with $R_0 > 5$ a.u. do not contribute to the excitation. Trajectories with $R_0 < 1$ a.u. are most effective to the excitation. Although Fig. 3 is obtained for a single time step, other time steps yield similar results.

The effective flux density most relevant to the nuclear excitation can be obtained by summing over the weights of the recolliding trajectories with $R_0 < 1$ a.u., then dividing by the area π a.u. and the time interval Δt. The nuclear excitation rate at a given recollision time t_r is the product of the excitation cross section and the effective recolliding-electron flux density:

$$\Gamma(t_r) = \sigma(E_r)j(t_r). \tag{5}$$

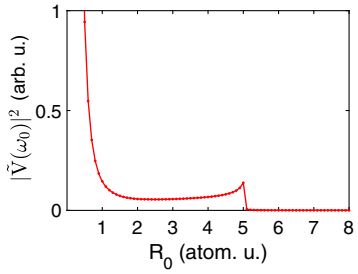

FIG. 3. Fourier component $|\tilde{V}(\omega_0)|^2$ as a function of R_0, with ω_0 the energy of the isomeric state.

Note that the energy argument in the σ function should be the asymptotic energy of the electron before being accelerated by the ion-core potential. Therefore E_r as shown in Fig. 2(c) from the simple-man model without taking into account the ion-core potential is appropriate to be used here. The probability of isomeric excitation, which is the time accumulation of $\Gamma(t_r)$, is shown in Fig. 4, together with the ionization probabilities. Two laser intensities are considered, namely, 10^{14} and 10^{15} W/cm^2.

The intensity 10^{14} W/cm^2 is able to pull out the outermost three electrons, as shown in Fig. 4(a). The isomeric excitation probabilities are shown in Fig. 4(b) for each electron. Note that the first-emitted electron does not contribute to the excitation because it is emitted too early during the pulse with a recollision energy below the excitation threshold. Using an over-barrier condition, the laser field at the time of first ionization can be estimated to be $E_1 \approx I_{p1}^2/4 = 0.0134$ a.u. With this field, the maximum recolliding energy is $3.17U_p = 3.17E_1^2/4\omega^2 = 0.044$ a.u. $= 1.09$ eV, which is lower than the excitation threshold. The second and the third electrons do have enough recollision energies. The total excitation probability is on the order of 10^{-18} to 10^{-17}.

The higher intensity 10^{15} W/cm^2 is able to pull out the outermost four electrons, as shown in Fig. 4(c). Similarly, the first electron does not contribute to the excitation. The excitation probabilities from the second, third, and fourth electrons are shown in Fig. 4(d). The total excitation probability is on the order of 10^{-17}, about 6 times higher than the 10^{14} W/cm^2 case. The contribution from the third electron is similar to that from the fourth electron, and both are higher than the contribution from the second electron. This is mostly because the recolliding energies for the third and the fourth electrons are higher than that of the second electron, so the excitation cross sections are higher.

One sees that for RINE, the excitation is well timed and only happens within a fraction of the pulse. The 10^{-17} excitation probability is achieved within about 10 fs. The instantaneous excitation rate can be estimated to be $10^{-17}/10^{-14}$ s $= 10^{-3}$ s^{-1}.

Finally, we consider a minor complication that the recolliding electron may lose some flux when it penetrates

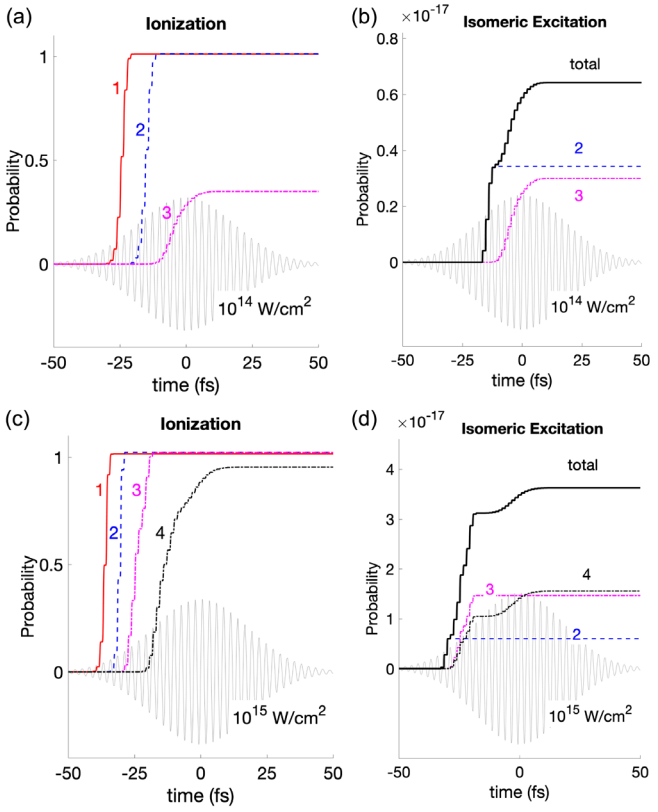

FIG. 4. Probabilities of ionization (a),(c) and of nuclear excitation (b),(d). The laser pulse is Gaussian with duration 30 fs (FWHM), as shown in each figure as background. Two intensities are used, namely, 10^{14} (a),(b) and 10^{15} W/cm^2 (c),(d). The ionization probabilities of individual electrons and their contributions to the nuclear excitation are also separately labeled. For both cases, the first-emitted electron does not have enough recollision energy to excite the nucleus.

through the electron cloud of the remaining ion core before exciting the nucleus. This loss of flux is due to excitation of the ion-core electron cloud. Here, we give an estimation to this process. We use a code ELSEPA [49] to calculate the imaginary absorption potential $iW_{\rm abs}(r)$ for the $^{229}Th^{2+}$ ion, as shown in Fig. 5. Three asymptotic incoming energies are used, namely, 20, 50, and 100 eV, covering the typical energy range of the recolliding electron. The probability of absorption is

$$P_{\rm abs} = 1 - \exp\left[\int_0^\infty 2W_{\rm abs}(r)\frac{dr}{v(r)}\right], \qquad (6)$$

where $v(r) = \sqrt{2(E_i + 2/r)}$ is the electron velocity at distance r. For simplicity we only consider trajectories on the z axis. The result shows that $P_{\rm abs}$ is about 19% for 20 eV, 16% for 50 eV, and 14% for 100 eV. Therefore, the recolliding electron indeed loses some flux, but the majority of the flux can penetrate the electron cloud and contribute to nuclear excitation.

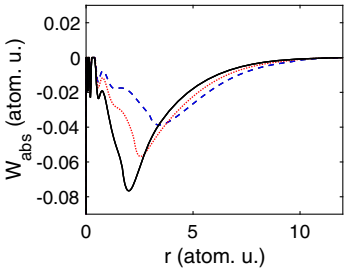

FIG. 5. Absorption potential seen by the colliding electron for the $^{229}Th^{2+}$ ion, calculated using the code ELSEPA, for incoming energies of 20 eV (blue dashed line), 50 eV (red dotted line), and 100 eV (black solid line).

In conclusion, we propose a new approach for the isomeric excitation of ^{229}Th based on laser-driven electron recollision. Compared with existing approaches, our approach has several advantages, some of which are unique. First, it does not require precise knowledge of the isomeric energy because the recolliding electron has a wide energy range. Uncertainties about the isomeric energy on the level of 0.5 eV have very small effects on our results. Second, the excitation is well timed and is achieved within a fraction of the laser pulse. This may be useful for potential coherent operations in which a precise timing of the excitation is important. Third, experimental realization is within reach using tabletop laser systems, and the efficiency of our approach is sufficiently high for practical usage. Current intense laser systems can achieve repetition rates of 100–200 kHz [50–53]. If N_0 is the average number of ^{229}Th atoms that can be effectively radiated by each pulse, then the number of excited nuclei is about $N_0 \times 10^{-17} \times$ [laser repetition rate] $\times$ [time in seconds]. The number density of metallic ^{229}Th is about 3×10^{19} mm^{-3}. Assuming a focal volume of $(30\ \mu\text{m})^3$, N_0 is estimated to be 8×10^{14}, and the number of excited nuclei is about 800 per second, or 48 000 per minute. (For ^{229}Th in the vapor state, the number density can be lower by 3 orders of magnitude. Larger focal volumes, higher repetition rates, and optimized temporal shapes of the laser field, as will be explained below, are key factors that will enhance the excitation yield). Besides, the excited nuclei are free from internal-conversion decay because they are left in well controlled ionic states after recollision.

We notice very recently that newer theoretical values for the reduced transition probabilities are suggested: $B(E2)$ is between 30 and 50 W.u. and $B(M1)$ is between 0.005 and 0.008 W.u. [54]. If these values are used, the cross section for electronic excitation will be higher by as much as 50%. The isomeric excitation probabilities will be increased by a similar ratio.

We envisage that the excitation probability may further be increased by optimizing the temporal shape of the laser field. This works via guiding more recolliding-electron fluxes into the effective excitation radius.

Similar optimizations have been successful in enhancing the yields of high harmonic generation [55–58]. The results will be reported in a subsequent paper.

A possible collaboration between RINE and a mechanism called nuclear excitation by electron capture (NEEC) [59,60] may deserve further studies, although NEEC has mostly been applied to other nuclei. Starting from neutral ^{229}Th atoms, an intense laser pulse arrives and RINE happens first. With the accumulation of free electrons, NEEC may happen and the ^{229}Th ions are (partially) neutralized. When the next laser pulse arrives, the neutralized ^{229}Th atoms (ions) may experience another RINE process. And so on. Detailed calculations are needed to tell more about this RINE-NEEC cycle.

Our scheme of RINE also applies to ^{235}U, which has an isomeric state of energy 76 eV [61], well within the energy range of recolliding electrons. ^{229}Th and ^{235}U are the only two known nuclei with an excited-state energy below 1 keV.

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Correction: The number preceding "eV" in the fourth sentence of the first full paragraph after Eq. (5) was presented with a conversion error and has been fixed.