Highly Nonlinear Light-Nucleus Interaction: Supplemental Material

Hanxu Zhang,[1,*] Tao Li,[1,*] and Xu Wang[1,2,†]

[1] Graduate School, China Academy of Engineering Physics, Beijing 100193, China
[2] Southern Center for Nuclear-Science Theory, Institute of Modern Physics, Chinese Academy of Sciences, Huizhou, Guangdong 516000, China

(Dated: July 20, 2024)

1. The NHM effect

The hyperfine interaction between the electron and the nucleus can be written as a summation over the scalar product of two irreducible tensor operators [S1]

$$V_{\mathrm{HF}} = \sum_{\tau=E,M} \sum_{lm} \frac{4\pi}{2l+1} (-1)^m \mathcal{M}_{\mathrm{n}}^{(\tau l-m)} \cdot \mathcal{N}_{\mathrm{e}}^{(\tau lm)}, \tag{S1}$$

where $\mathcal{M}_{\mathrm{n}}^{(\tau l)}$ is the nuclear multipole moment operator of type τ (E for electric, M for magnetic) and rank l, and $\mathcal{N}_{\mathrm{e}}^{(\tau l)}$ is the multipole transition operator for the electron. They can be written as

$$\mathcal{M}_{\mathrm{n}}^{(Elm)} = \int \rho_{\mathrm{n}} r^l Y_{lm}(\theta,\phi)\, d\tau, \tag{S2}$$

$$\mathcal{M}_{\mathrm{n}}^{(Mlm)} = -\frac{i}{c(l+1)} \int \boldsymbol{j}_{\mathrm{n}} \cdot \boldsymbol{L} \left[r^l Y_{lm}(\theta,\phi) \right] d\tau, \tag{S3}$$

$$\mathcal{N}_{\mathrm{e}}^{(Elm)} = \int \frac{\rho_{\mathrm{e}}}{r^{l+1}} Y_{lm}(\theta,\phi)\, d\tau, \tag{S4}$$

$$\mathcal{N}_{\mathrm{e}}^{(Mlm)} = -\frac{i}{cl} \int \frac{\boldsymbol{j}_{\mathrm{e}} \cdot \boldsymbol{L} \left[Y_{lm}(\theta,\phi) \right]}{r^{l+1}}\, d\tau. \tag{S5}$$

In the above formulas $\rho, \boldsymbol{j}, \boldsymbol{L}$ represent the charge density operator, current density operator, and angular momentum operator, respectively.

It is convenient to use the basis of total angular momentum states, coupled by the $1s$ electron with angular momentum $J = 1/2$ and the nucleus with angular momentum I:

$$|F, m_F; J, I\rangle = \sum_{m_I, m_J} C^{F,m_F}_{J,m_J,I,m_I} |J, m_J\rangle \otimes |I, m_I\rangle, \tag{S6}$$

where C^{F,m_F}_{J,m_J,I,m_I} is a Clebsch-Gordan coefficient, F is the total angular momentum quantum number, m_I and m_F are the magnetic quantum numbers of I and F. In $^{229}\mathrm{Th}^{89+}$, the hyperfine interaction splits the nuclear ground state ($I_{\mathrm{gs}} = 5/2$) into two levels with $F = 2$ and $F = 3$, and the isomeric state ($I_{\mathrm{is}} = 3/2$) into two levels with $F = 2$ and $F = 1$.

With Eqs. (S1-S6) and the Wigner-Eckart theorem [S2], the matrix elements of V_{HF} can be written as

$$\begin{aligned} &\langle F, m_F; J, I | V_{\mathrm{HF}} | F', m'_F; J, I' \rangle \\ =&\delta_{Fm_F, F'm'_F} (-1)^{J+I+F} \sum_{\tau l} \frac{4\pi}{2l+1} \begin{Bmatrix} J & I & F \\ I' & J & l \end{Bmatrix} \langle I || \mathcal{M}_{\mathrm{n}}^{(\tau l)} || I' \rangle \langle J || \mathcal{N}_{\mathrm{e}}^{(\tau l)} || J \rangle. \end{aligned} \tag{S7}$$

The electronic reduced matrix elements $\langle J || \mathcal{N}_{\mathrm{e}}^{(\tau l)} || J \rangle$ are calculated using the electron wave function, whereas the nuclear reduced matrix elements $\langle I || \mathcal{M}_{\mathrm{n}}^{(\tau l)} || I' \rangle$ are obtained by relating them to established nuclear multipole moments and reduced transition probabilities. Taking magnetic dipole as an example, the nuclear magnetic moments

$$\mu_I = \sqrt{\frac{4\pi}{3}} C^{I,I}_{I,I,1,0} \frac{\langle I || \mathcal{M}_{\mathrm{n}}^{(M1)} || I \rangle}{\sqrt{2I+1}}, \tag{S8}$$

and reduced transition probabilities

$$B(M1; I' \to I) = \frac{\left|\langle I||\mathcal{M}_{\mathrm{n}}^{(M1)}||I'\rangle\right|^2}{2I'+1} \quad (I \neq I'). \tag{S9}$$

The nuclear magnetic moments have experimental values $\mu_{\mathrm{gs}} = 0.360\ \mu_N$ [S3], $\mu_{\mathrm{is}} = -0.376\ \mu_N$ [S4], with μ_N being the nuclear magneton. The reduced transition probabilities are adopted from numerical calculation results of Minkov and Pálffy [S5] with $B(M1; \mathrm{is} \to \mathrm{gs}) = 0.008$ and $B(E2; \mathrm{is} \to \mathrm{gs}) = 42.9$ in Weisskopf units.

According to Eq. (S7), states with the same quantum numbers F and m_F have non-zero non-diagonal elements, i.e., these states are mixed. The mixing effect becomes clearer by diagonalizing the matrix of V_{HF} to obtain the eigenstates of the system, which has been shown in Eq. (1) in the main text. The mixing coefficient b is calculated to be -0.03066.

2. Light-nucleus interaction

Here we present more details on the interaction between the intense laser pulse and the $^{229}\mathrm{Th}^{89+}$ ion. The Hamiltonian of this laser-nucleus-electron system can be written as

$$H = H_0 + H_{\mathrm{I}}(t) = H_{\mathrm{e}} + H_{\mathrm{n}} + V_{\mathrm{HF}} + H_{\mathrm{I}}(t), \tag{S10}$$

where $H_{\mathrm{I}}(t)$ is the interaction Hamiltonian with the laser:

$$H_{\mathrm{I}}(t) = -\frac{1}{c}\int \left[\boldsymbol{j}_{\mathrm{e}}(\boldsymbol{r}) + \boldsymbol{j}_{\mathrm{n}}(\boldsymbol{r})\right] \cdot \boldsymbol{A}(\boldsymbol{r}, t)\ d\tau. \tag{S11}$$

The vector potential of the laser field is written as

$$\boldsymbol{A}(\boldsymbol{r}, t) = \frac{\boldsymbol{\epsilon}}{2} A_0 f_A(t) e^{i(\boldsymbol{k}\cdot\boldsymbol{r} - \omega t)} + \mathrm{c.c.}, \tag{S12}$$

where $\boldsymbol{\epsilon}$ is the polarization vector, A_0 and $f_A(t)$ are the amplitude and envelope function of the vector potential. $\boldsymbol{k}$ and ω are the wave vector and frequency. The vector potential can be expanded in multipole series [S6]

$$\begin{aligned}\boldsymbol{A}_\nu(\boldsymbol{k}; \boldsymbol{r}) =& \hat{e}_\nu e^{i\boldsymbol{k}\cdot\boldsymbol{r}} \\ =& -\nu\sqrt{2\pi}\sum_{lm}\sqrt{2l+1}\, i^l D^l_{m\nu}(\phi, \theta, 0)\left[A_{lm}(\boldsymbol{k}; \boldsymbol{r}, M) + i\nu A_{lm}(\boldsymbol{k}; \boldsymbol{r}, E)\right],\end{aligned} \tag{S13}$$

where $\hat{e}_\nu$ is a spherical vector, $\hat{e}_0 = \hat{z}$, $\hat{e}_{\pm1} = \mp\frac{1}{\sqrt{2}}(\hat{x} \pm i\hat{y})$. $D^l_{m\nu}(\phi, \theta, 0)$ denotes the Wigner-D function and (θ, ϕ) gives the direction of wave vector $\boldsymbol{k}$. $A_{lm}(\boldsymbol{k}; \boldsymbol{r}, \tau)$ are transverse vector spherical harmonics

$$A_{lm}(\boldsymbol{k}; \boldsymbol{r}, M) = \frac{1}{\hbar\sqrt{l(l+1)}}\boldsymbol{L}\left[j_l(kr) Y_{lm}(\hat{r})\right], \tag{S14}$$

$$A_{lm}(\boldsymbol{k}; \boldsymbol{r}, E) = \frac{-i}{\hbar k\sqrt{l(l+1)}}\nabla \times \boldsymbol{L}\left[j_l(kr) Y_{lm}(\hat{r})\right]. \tag{S15}$$

Using Eqs. (S11-S15) and the long wavelength limit $j_l(kr) \approx (kr)^l/(2l+1)!!$ characterized by $kr \ll 1$, $H_{\mathrm{I}}(t)$ can be written as a linear combination of $\mathcal{M}_{\mathrm{n}}^{(Elm)}$ and $\mathcal{M}_{\mathrm{e}}^{(Elm)}$, where the form of $\mathcal{M}_{\mathrm{e}}^{(Elm)}$ involves replacing the nuclear charge density and current density in Eqs. (S2-S3) with those of the electron. Similar to Eq. (S8), the electron multipole moment is proportional to the reduced matrix element (e.g. $\mu_{\mathrm{e}} \propto \langle J||\mathcal{M}_{\mathrm{e}}^{(M1)}||J\rangle$). The magnetic dipole moment operator of the system is $\hat{\mathbf{m}} = \mathcal{M}_{\mathrm{n}}^{(M1)} + \mathcal{M}_{\mathrm{e}}^{(M1)}$ [used in Eq. (5) of the main text].

The interaction Hamiltonian can also be written in the following form: $H_{\mathrm{I}}(t) = E_{\mathrm{I}}\mathcal{F}(t)$, where E_{I} is a time-independent interaction energy, and $\mathcal{F}(t) = f_A(t)e^{i\Omega t}\left[e^{-i\omega t} - (-1)^l e^{i\omega t}\right]/2$ is a time-dependent envelope. Here $\Omega = \Delta E/\hbar$ is the frequency gap between the two levels under consideration. $0 \le f_A(t) \le 1$ is the envelope function of the vector potential. For $l = 1$, $\mathcal{F}(t) = f_A(t)e^{i\Omega t}\cos\omega t$, so $0 \le |\mathcal{F}(t)| \le 1$. The laser field is assumed to be polarized along $\hat{x}$ and propagating along $\hat{z}$.

Taking the transition between $|F=2;\mathrm{down}\rangle$ and $|F=2;\mathrm{up}\rangle$ for example. Consider the magnetic quantum numbers of the two states to be $\{m_{\mathrm{down}}, m_{\mathrm{up}}\}$, then the interaction energy can be written as

$$\begin{aligned} E_{\mathrm{I}} =&\mathcal{E}_0 \frac{5\sqrt{6\pi}}{3} \sum_{\nu=\pm 1} \begin{pmatrix} F & 1 & F \\ m_{\mathrm{up}} & \nu & -m_{\mathrm{down}} \end{pmatrix} \left[(1-2b^2)\langle I_{\mathrm{is}}||\mathcal{M}_{\mathrm{n}}^{(M1)}||I_{\mathrm{gs}}\rangle \begin{Bmatrix} F & 1 & F \\ I_{\mathrm{gs}} & J & I_{\mathrm{is}} \end{Bmatrix} \right. \\ &\left. + b\sqrt{1-b^2} \sum_{I=I_{\mathrm{gs}},I_{\mathrm{is}}} \langle I||\mathcal{M}_{\mathrm{n}}^{(M1)}||I\rangle \begin{Bmatrix} F & 1 & F \\ I & J & I \end{Bmatrix} + \langle J||\mathcal{M}_{\mathrm{e}}^{(M1)}||J\rangle \begin{Bmatrix} F & 1 & F \\ J & I & J \end{Bmatrix} \right]. \end{aligned} \tag{S16}$$

With Eqs. (S2-S3), Eqs. (S8-S9), and the Wigner-6j symbols evaluated, Eq. (S16) can be derived into the following relatively simpler form

$$\begin{aligned} E_{\mathrm{I}} =&\mathcal{E}_0 \frac{5\sqrt{6\pi}}{3} \sum_{\nu=\pm 1} \begin{pmatrix} 2 & 1 & 2 \\ m_{\mathrm{up}} & \nu & -m_{\mathrm{down}} \end{pmatrix} \\ &\times \left[\frac{(1-2b^2)}{5\sqrt{3}} \sqrt{B(M1)} + b\sqrt{1-b^2} \left(\frac{\sqrt{5}}{2\sqrt{2\pi}}\mu_{\mathrm{e}} - \frac{7}{5\sqrt{10\pi}}\mu_{\mathrm{gs}} + \frac{3}{2\sqrt{10\pi}}\mu_{\mathrm{is}} \right) \right]. \end{aligned} \tag{S17}$$

The transition energies between other levels of $^{229}\mathrm{Th}^{89+}$ (wavy lines in Fig. 1 of the main text) have similar forms and are numerically on the same order of magnitude. The electric quadrupole ($E2$) terms can be neglected due to the dominance of magnetic dipole ($M1$) terms.

For the bare nucleus, the electronic current density in Eq. (S11) is zero. The interaction energy can then be obtained by a similar derivation

$$E_{\mathrm{I}} = \mathcal{E}_0 \frac{4\sqrt{\pi}}{\sqrt{6}} \sum_{\nu=\pm 1} \begin{pmatrix} I_{\mathrm{gs}} & 1 & I_{\mathrm{is}} \\ m_{\mathrm{gs}} & \nu & -m_{\mathrm{is}} \end{pmatrix} \sqrt{B(M1)}. \tag{S18}$$

It can be seen from Eq. (S17) and Eq. (S18) that the main difference between $^{229}\mathrm{Th}^{89+}$ and $^{229}\mathrm{Th}^{90+}$ is the presence of the magnetic dipole moment of the $1s$ electron, which is about three orders of magnitude lager than the nuclear moments μ_{gs}, μ_{is}, or $\sqrt{B(M1)}$. For the purpose of conciseness in presentation, when we explain the interaction energy in the main text, we have neglected some detailed coefficients in these two equations.

The time-dependent Schrödinger equation [Eqs. (2-4) in the main text] can be numerically solved with the detailed terms given in Eqs. (S10-S18). In this study, we employ the Runge-Kutta method with 8th-order and 9th-order pairs for numerical computations [S7]. This widely adopted method offers both high accuracy and computational efficiency.

* H. Z. and T. L. contributed equally to this work.
† xwang@gscaep.ac.cn

[S1] K. Alder, A. Bohr, T. Huus, B. Mottelson, and A. Winther, Study of nuclear structure by electromagnetic excitation with accelerated ions. Rev. Mod. Phys. **28**, 432 (1956).
[S2] R. D. Cowan, *The Theory of Atomic Structure and Spectra* (University of California Press, Berkerley, 1981).
[S3] M. S. Safronova, U. I. Safronova, A. G. Radnaev, C. J. Campbell, and A. Kuzmich, Magnetic dipole and electric quadrupole moments of the ^{229}Th nucleus. Phys. Rev. A **88**, 060501(R) (2013).
[S4] J. Thielking, M. V. Okhapkin, P. Gowacki, D. M. Meier, L. von der Wense, B. Seiferle, C. E. Düllmann, P. G. Thirolf, and E. Peik, Laser spectroscopic characterization of the nuclear-clock isomer $^{229\mathrm{m}}$Th. Nature **556**, 321 (2018).
[S5] N. Minkov, and A. Pálffy, $^{229\mathrm{m}}$Th isomer from a nuclear model perspective. Phys. Rev. C **103**, 014313 (2021).
[S6] J. M. Eisenberg, and W. Greiner, *Excitation mechanisms of the nucleus* (North-Holland, 1987), Vol. 2.
[S7] J. H. Verner, Numerically optimal Runge-Kutta pairs with interpolants. Numer. Algor. **53**, 383-396, (2010).