Ab initio calculations of the highest-multipole electromagnetic transition ever observed in nuclei

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Abstract

High multipole electromagnetic transitions are rare in nature. The highest multipole transition observed in atomic nuclei is the electric hexacontatetrapole E6 transition from the T1/2 = 2.54(2)-min J = 19/2− isomer to the 7/2− ground state in 53Fe with an angular momentum change of six units. In the present work, we performed ab initio calculations for this unique case by employing chiral effective field theory (EFT) forces. The in-medium similarity renormalization group is used to derive the valence-space effective Hamiltonian and multipolar transition operators. Bare nucleon charges were used in all the multipolar transition rate calculations, providing good agreement with the experimental data. The valence space takes the full fp shell. In 53Fe, the low-lying states were dominated by the 0f7/2 component. Two different versions of the chiral EFT two- plus three-nucleon interaction were used to test the dependence on the interaction used. We also tested the convergence of the transition rate calculations against the harmonic oscillator parameter Ω and basis truncations emax and E3max for two- and three-nucleon forces, respectively.

Full Text

Preamble

We present ab initio calculations of the highest-multipole electromagnetic transition ever observed in nuclei. High multipole electromagnetic transitions are rare in nature, and the highest multipole transition observed in atomic nuclei is the electric hexacontatetrapole (E6) transition from the $T_{1/2} = 2.54(2)$-min $J^\pi = 19/2^-$ isomer to the $7/2^-$ ground state in ^{53}Fe, involving an angular momentum change of six units. In this work, we perform ab initio calculations for this unique case using chiral effective field theory (EFT) forces. The in-medium similarity renormalization group is employed to derive the valence-space effective Hamiltonian and multipolar transition operators, with bare nucleon charges used in all multipolar transition rate calculations, yielding good agreement with

experimental data. The valence space encompasses the full fp shell, where low-lying states in ^{53}Fe are dominated by the $0f_{7/2}$ component. We test the dependence on the interaction using two different versions of the chiral EFT two- plus three-nucleon interaction, and we examine the convergence of the transition rate calculations with respect to the harmonic oscillator parameter $\hbar\Omega$ and basis truncations $e_{\max}$ and $E_{3\max}$ for two- and three-nucleon forces, respectively.

Keywords: Isomerism, Highest-multipole electromagnetic transitions, Ab initio calculations, Chiral two- plus three-nucleon forces, Valence-space in-medium similarity renormalization group

Introduction

The $T_{1/2} = 2.54$-min $J^\pi = 19/2^-$ isomer at 3.0 MeV in ^{53}Fe is unique in atomic nuclei in that it has a direct decay branch to the $J^\pi = 7/2^-$ ground state, requiring a hexacontatetrapole electric transition (E6) [**?**, **?**, **?**]. This transition was recently confirmed experimentally with improved precision [**?**], providing a sensitive probe for nuclear structure [**?**, **?**]. More broadly, this scenario offers unique insights into the highest-order shapes and correlations in nuclei and tests the applicability of nuclear models under extreme conditions. In the present work, we focus on ab initio calculations with bare nucleon charges that can simultaneously explain both ^{53}Fe excited-state energies and electromagnetic decay transition rates.

In nature, a free photon carries angular momentum of $1\hbar$, whereas in a high multipole transition, a single photon carries away large angular momentum. Naively, the probability of emitting a high-spin photon should be very small. In ^{53}Fe, a single E6 photon mediates a spin change as large as $6\hbar$. No other isolated physical system is known to exhibit such high multipole or single-photon emission. By comparison, infrared spectra have revealed $6\hbar$ absorption in solid hydrogen [**?**], and magneto-optical studies of atomic ^{85}Rb have reported $6\hbar$ spontaneous emissions [**?**]. Thus, a small group of physical environments exists where high multipole radiation provides access to novel and exceptional physics.

In a recent experimental study by Palazzo et al. [**?**], the existence of E6 decay in ^{53}Fe was firmly established using empirical shell model calculations. However, Ref. [**?**] does not provide calculations of high multipole electromagnetic transition probabilities because empirical calculations require both proton and neutron effective charges that cannot be determined from the single available set of experimental E6 transition rate data. In empirical models, different effective charges are used for different multipolarities of transitions. To improve upon such ad hoc features, ab initio calculations using realistic interactions and bare nucleon charges are required, offering the prospect of gaining insight into rare high-multipole transitions and testing the ab initio method itself. Ab initio nuclear theory stands at the forefront of current nuclear structure studies, and over the past two decades, significant progress has been made in ab initio many-body methods [**?**, **?**, **?**, **?**, **?**] using nuclear forces based on chiral effective

field theory [**?**].

In this study, we employ the ab initio valence-space in-medium similarity renormalization group (VS-IMSRG) [**?**, **?**, **?**, **?**] to derive the valence-space effective Hamiltonian and effective multipole transition operators for subsequent shell model calculations of the observed excited states and their electromagnetic transitions in ^{53}Fe. Unique bare nucleon charges are used in the calculation of all multipolar transitions. We aim to provide solid calculations that explain the experimental observations and offer deep insight into the structure of the $J^\pi = 19/2^-$ isomer.

II. Theoretical Framework

Our calculations begin from the intrinsic Hamiltonian of the A-nucleon system [**?**, **?**]:

$$H = \sum_i \frac{p_i^2}{2m} + \sum_{i<j} v_{NN} + \sum_{i<j<k} v_{3N}$$

where p_i is the nucleon momentum in the laboratory system, m is the nucleon mass, and v_{NN} and v_{3N} represent the nucleon-nucleon (NN) and three-nucleon (3N) interactions, respectively. Explicit treatment of the three-nucleon force (3NF) in many-body calculations has not yet been accessible for nuclei as heavy as ^{53}Fe. Therefore, we obtain a normal-ordered approximation of the Hamiltonian (1), neglecting the residual 3NF [**?**, **?**, **?**, **?**, **?**].

The IMSRG evolves the normal-ordered Hamiltonian to a block-diagonal form, yielding the valence-space effective Hamiltonian. Using this technique, we can also derive valence-space effective operators for other observables, including multipolar transitions. The Magnus expansion [**?**] was employed in the IMSRG evolution. Operators appearing in the IMSRG equations are truncated at the two-body level, which is referred to as IMSRG(2) [**?**]. The technique of using IMSRG to obtain a valence-space Hamiltonian for shell model calculations is called VS-IMSRG [**?**].

The normal-ordering approximation of the Hamiltonian and the evolution of the VS-IMSRG were implemented with a closed-shell reference state in the Hartree-Fock basis. However, to reduce the effect of the residual 3NF that is neglected, the fractional filling of open-shell orbitals in an open-shell nucleus has been suggested, known as ensemble normal ordering (ENO) [**?**]. Using the ENO approximation of the VS-IMSRG, we can obtain nucleus-dependent valence-space effective Hamiltonians and effective operators for other observables. The resulting effective Hamiltonian is diagonalized using the large-scale shell model code KSHELL [**?**], yielding eigenenergies and one- and two-body transition densities (OBTD and TBTD, respectively) for subsequent calculations of states.

The reduced transition probability is calculated by:

$$B(\sigma\lambda;\psi_i \to \psi_f) = \frac{|\langle\psi_f\|\sigma\lambda\|\psi_i\rangle|^2}{2J_i+1}$$

where σ denotes the electric ($\sigma = E$) or magnetic ($\sigma = M$) transition and λ is the rank of the tensor operator $M_{\sigma\lambda}$ [**?**]. ψ_i (ψ_f) represents the initial (final) state of the transition, with J_i denoting the angular momentum of the initial state.

The free-space bare electric transition tensor operator is defined by $M_{E\lambda} = Q_\lambda$ with tensor components given by [**?**]:

$$Q_{\lambda\mu} = \sum_j e_j r_j^\lambda Y_{\lambda\mu}(\hat{r}_j)$$

where e_j is the natural (bare) charge of the jth nucleon, that is, $e = 1$ (0) for the proton (neutron), and $Y_{\lambda\mu}$ is the spherical harmonic function. The free-space bare magnetic transition tensor operator is defined by $M_{M\lambda} = M_\lambda$ with tensor components given by [**?**]:

$$M_{\lambda\mu} = \sum_j \left[\frac{2}{\lambda+1}\left(g_l \mathbf{l}_j + g_s \mathbf{s}_j\right)\cdot\nabla_j\right]\left[r_j^\lambda Y_{\lambda\mu}(\hat{r}_j)\right]$$

where $\mathbf{l}_j$ and $\mathbf{s}_j$ are the orbital and intrinsic angular momenta of the jth nucleon, respectively. The values of g factors were taken from [**?**]. The free-space bare operators $M_{\sigma\lambda}$ must be renormalized into the valence space, which can be performed using the same ENO VS-IMSRG technique. After renormalization, the one-body operators of the free-space bare $M_{\sigma\lambda}$ become two-body operators in the valence space, defined by the corresponding two-body matrix elements. Using the valence-space OBTD and TBTD obtained from diagonalizing the effective Hamiltonian, the reduced transition probability is calculated using Eq. (2).

III. Calculations and Discussions

The ^{53}Fe $J^\pi = 19/2^-$ isomer represents the only case in which the highest multipole E6 transition has been observed [**?**, **?**, **?**, **?**]. In a recent experimental study [**?**], shell model calculations using two empirical interactions, GXPF1A and KB3G, were performed with model spaces consisting of either only the $0f_{7/2}$ orbital or the full fp shell. In empirical calculations of electromagnetic transitions, effective nucleon charges are employed, with different effective charges for different multipolarities determined by fitting experimental data. The problem is that only one E6 transition has been observed to date: the decay from the $19/2^-$ isomer to the $7/2^-$ ground state in ^{53}Fe. A single E6 transition datum cannot simultaneously determine both the proton and neutron effective charges.

In the present work, we performed ab initio calculations using the shell model with a full fp model space above the ^{40}Ca core. The valence-space shell model effective Hamiltonian, including the one-body single-particle energies of the valence particles, is obtained using the IMSRG evolution with the ENO approximation [**?**, **?**]. In ab initio calculations of multipole transitions, unique bare nucleon charges are used, meaning that the same bare charges are employed for different multipolarities of transitions.

In our calculations, two different versions of the chiral NN plus 3N interaction were used to test the dependence on the interaction. The potential labeled 1.8/2.0(EM) [**?**, **?**, **?**] employs the NN force (2NF) at N3LO [**?**] and 3NF at N2LO. This potential can globally reproduce the ground-state properties of nuclei from light- to heavy-mass regions, with the harmonic-oscillator (HO) basis frequency optimized at $\hbar\Omega = 16$ MeV [**?**, **?**]. We also used the recently developed interaction labeled NN+3N(lnl) [**?**, **?**], which employs N4LO 2NF and N2LO 3NF. For both the 1.8/2.0(EM) and NN+3N(lnl) potentials, the induced 3NF was properly included [**?**, **?**, **?**, **?**, **?**]. One major difference between the two potentials lies in their treatment of 3NF: the 1.8/2.0(EM) potential uses a nonlocal 3N regulator, whereas the NN+3N(lnl) potential employs both local and nonlocal (lnl) 3N regulators.

The basis space represents another important consideration. The present calculations begin with a spherical HO basis with $e = 2n + l \leq e_{\max}$ single-particle shells. The 3NF is limited to $e_1 + e_2 + e_3 \leq E_{3\max}$. For the 1.8/2.0(EM) and NN+3N(lnl) potentials, it has been demonstrated that basis truncations with both $e_{\max}$ and $E_{3\max}$ around 14 provide good convergence of VS-IMSRG calculations, particularly for ground-state energies up to medium-mass regions, with an optimized frequency around $\hbar\Omega = 16$ MeV [**?**, **?**, **?**, **?**]. However, spectroscopic calculations of heavier nuclei require larger basis spaces. For example, a recent study using the 1.8/2.0(EM) potential showed that $E_{3\max} = 24$ is required to achieve convergence of excited state calculations in the $A \approx 130$ mass region [**?**]. Another study on neutrinoless double-beta decay ($0\nu\beta\beta$) of $A \approx 130$ candidates showed that $E_{3\max} = 28$ is required to converge all operators [**?**]. Both studies nevertheless demonstrate that $e_{\max} = 14$ is sufficient for truncation of the single-particle basis. In the present work, we tested the convergence of calculations for the observed highest-multipole electromagnetic transition probabilities, for which no ab initio calculations have been performed previously.

[Figure 1: see original paper] shows the convergence of the E4, M5, and E6 reduced transition probabilities as functions of $e_{\max}$, $E_{3\max}$, and $\hbar\Omega$. We observe that for both potentials, the calculations converge well at $e_{\max} \geq 12$, $E_{3\max} \geq 16$, and $\hbar\Omega = 14 - 16$ MeV. Therefore, in the following calculations, we adopt $e_{\max} = 14$, $E_{3\max} = 24$, and $\hbar\Omega = 16$ MeV. This choice is similar to that used in calculations of excitation spectra [**?**] and $0\nu\beta\beta$ decays [**?**] in the mass-130 region.

The present calculations reproduce the excitation spectrum of ^{53}Fe. For simplicity, however, [Figure 2: see original paper] shows only a limited set of levels

associated with the $19/2^-$ isomer. The calculations reproduce the experimental data [**?**], with the calculation using the NN+3N(lnl) interaction showing better agreement. Both calculations yield the correct energy ordering of the $19/2^-$ level, which lies below the $13/2^-$ level. The lower energy of the $19/2^-$ level relative to those of the $17/2^-$ and $15/2^-$ levels naturally prevents M1 and E2 transitions from the $19/2^-$ state, leading to the formation of the $19/2^-$ metastable isomeric state.

Experiments [**?**, **?**, **?**, **?**] observed that the $19/2^-$ state decays to lower $11/2^-$, $9/2^-$, and $7/2^-$ states via high-multipole E4, M5, and E6 transitions, respectively. Higher multipole γ decays have lower transition probabilities, which can produce long-lived isomers. Experimental observations [**?**, **?**, **?**, **?**] of rare high multipole γ decay in ^{53}Fe provide a unique laboratory for studying electromagnetic transitions with high multipolarity.

compares the calculated reduced probabilities of the observed highest-multipole transitions E6, M5, and E4 with experimental data [**?**]. The NN+3N(lnl) potential provides an improved result for the M5 transition probability that is closer to the data. However, the calculated E6 transition probability differs from the experimental data by approximately a factor of three, although the experimental uncertainty is large for this transition. The present VS-IMSRG calculation approximates operators truncated at the two-body level via IMSRG(2) [**?**], whereas a recent study has developed IMSRG with operators truncated at the three-body level, labeled IMSRG(3) [**?**, **?**]. Another study indicated that IMSRG combined with the generator coordinate method can capture more collective correlations in deformed nuclei [**?**]. In Refs. [**?**, **?**], it was found that inclusion of two-body currents can improve calculations of magnetic dipole moments (transitions). While consideration of these factors could improve the calculations, such an investigation lies beyond the scope of the present work and will be addressed separately.

In empirical shell model calculations, effective nucleon charges compensate for missing correlations. The proton and neutron effective charges are determined by fitting corresponding experimental data to electromagnetic transitions, with different effective charges used for different multipolarities. Unfortunately, only one E6 experimental data point is currently available, which is insufficient to determine both proton and neutron effective charges for the E6 transition.

presents similar results for E2 and M1 transitions in ^{53}Fe, with data from [**?**]. Using bare nucleon charges, we also calculated the ^{53}Fe E2 and M1 reduced transition probabilities available experimentally. We observe that the observed E2 and M1 transition probabilities can be reproduced reasonably well (all are well within 2σ of the experimental values). Nevertheless, improvements in IMSRG and the inclusion of two-body currents should also enhance calculations of low-multipole transitions.

To better understand the long-lived $19/2^-$ isomer of ^{53}Fe, we examine the state configurations. [Figure 3: see original paper] shows the configurations of the

$7/2^-$, $9/2^-$, $11/2^-$, and $19/2^-$ states obtained from the calculations. We see that the $0f_{7/2}$ component dominates these states, which can be understood through simple three-hole (3h) coupling $(f_{7/2})^{-2}(f_{7/2})^{-1}$ with $J = 19/2$ being the highest spin obtainable in 3h coupling. As shown in [Figure 3: see original paper], the $19/2^-$ isomer has a purer $0f_{7/2}$ component, with fewer occupations in the higher $0f_{5/2}$ and $1p_{3/2,1/2}$ orbitals. The pronounced dominance of the $0f_{7/2}$ component for both neutron and proton orbits arises from the $N(Z) = 28$ shell gaps on either side of the $f_{7/2}$ orbit. However, we find that the effects of the higher $0f_{5/2}$ and $1p_{3/2}$ orbitals are not negligible.

IV. Summary

High-multipole electromagnetic transitions are of particular interest in nuclear structure studies, providing a sensitive test of theoretical models. To date, the highest multipole transition is the electric hexacontatetrapole (E6) transition from the $J^\pi = 19/2^-$ isomer to the $7/2^-$ ground state in ^{53}Fe, accompanied by M5 and E4 transitions to the first $9/2^-$ and $11/2^-$ excited states, respectively. A recent experiment [**?**] provided clear detection of these highest-multipole transitions with improved precision for the transition rates. While shell-model calculations with empirical interactions were performed in that experimental study [**?**], empirical calculations cannot definitively determine the E6 transition probability because the empirical shell model uses effective charges determined by fitting experimental data. With only one observed E6 transition to date—from the $J^\pi = 19/2^-$ isomer to the $J^\pi = 7/2^-$ ground state in ^{53}Fe—an experimental E6 transition rate cannot simultaneously determine both proton and neutron effective charges.

In this study, we performed ab initio calculations of the highest multipole electromagnetic transitions observed from the $19/2^-$ isomer of ^{53}Fe using two different versions of chiral two- plus three-nucleon interactions. Using the valence-space in-medium similarity renormalization group, the free-space interaction matrix was decoupled to form a low-momentum valence space, obtaining the valence-space effective interaction for shell-model calculations. The convergence of the excitation spectrum and electromagnetic transitions of ^{53}Fe has been thoroughly tested. The present ab initio calculations reproduce the experimental spectrum well. The $19/2^-$ isomerism arises from its lower energy compared to the $17/2^-$ and $15/2^-$ levels, which prevents natural E2 and M1 transitions from the $19/2^-$ state. Using the same decoupling method, free-space bare operators of electromagnetic transitions evolve into the valence space. With unique bare nucleon charges, we can reproduce the measured highest multipole E6, M5, and E4 transition strengths. The low multipole E2 and M1 transitions observed in this nucleus can also be reproduced using bare nucleon charges. Moreover, we provide the configurations of the $7/2^-$, $9/2^-$, $11/2^-$, and $19/2^-$ states, revealing the structure of the $J^\pi = 19/2^-$ isomer.

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Note: Figure translations are in progress. See original paper for figures.