Letter

Exploring isospin-nonconserving effects in the upper fp shell with new mass measurements

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Nuclear mass measurements have recently been extended conspicuously to proton-rich region in the upper fp shell. The new data are utilized to study isospin symmetry breaking phenomena using Coulomb displacement energy (CDE) and triplet displacement energy (TDE) as probes. The new mass data, either measured for the first time or with greatly improved accuracy, removed several previously found anomalies in the systematical behavior in the fp shell. Remarkably, more regular odd-even staggering patterns can be established in both CDE and TDE, calling for a uniform explanation in terms of isospin-nonconserving (INC) forces across the sd, $f_{7/2}$, and upper fp shells. By extending the large-scale shell-model calculation [Phys. Rev. Lett. **110**, 172505 (2013)] to the upper fp-shell region, we found that, in order to describe the new data, the same INC force is required as previously used for the $f_{7/2}$ shell. Especially, we propose the $T = 1$ TDE for those triplet nuclei, that have pp, nn, and pn pairs on top of a common even-even $N = Z$ core, to be a good indicator for the isotensor component of isospin violating interactions, which is estimated here to be 150 keV.

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The nearly equal masses of proton and neutron as well as the nearly identical properties of nucleon-nucleon (NN) interactions lead to the elegant concept of isospin symmetry [1,2]. In nuclei, the Coulomb interaction and charge-dependent nucleon-nucleon interaction break this symmetry, giving rise to observable signatures. Especially for nuclei near the $N = Z$ line, effects of isospin-symmetry breaking (ISB) appear both in binding energies and excitation spectra [3–5]. Thus, investigation of very exotic proton-rich nuclei provides important testing ground for ISB, thereby enhancing our knowledge on nuclear forces and nuclear many-body effects far from stability.

There are various probes to address isospin-nonconserving (INC) forces in atomic nuclei [6,7]. Commonly used quantities are the mirror energy difference (MED) and triplet energy difference (TED), defined as excitation energy shift between mirror (analog) states. Extensive experimental studies have been performed to explore MED between mirror pairs in the sd, fp, and fpg shells [8–17]. Numerous TEDs for $A = 46$ [18], $A = 54$ [19], $A = 62$ [15], $A = 66$ [11] and $A = 70$ [12], $A = 74$ [20] triplets were thoroughly reviewed. Inspired by these spectroscopic data, ISB effects on MEDs and TEDs were investigated by the large-scale shell-model calculations [21,22].

Another category of suitable quantities are Coulomb displacement energy (CDE) and triplet displacement energy (TDE) determined from the masses of the ground states. CDE and TDE are, respectively, proportional to the linear ($\propto T_z$) and quadratic ($\propto T_z^2$) terms of the Wigner's isobaric multiplet mass equation (IMME) [23], where $T_z = (N - Z)/2$ is the z component of isospin T for a nucleus with Z protons and N neutrons. These terms are essentially related to the isovector and isotensor components of the charge violating interactions, and thus can potentially yield separate information on the charge symmetry and charge independence of the attractive NN interaction [24–26].

Detailed knowledge on CDE and TDE has further far-reaching implications in fundamental physics. For example, superallowed $0^+ \rightarrow 0^+$ β decays yield the most precise value for V_{ud}, the up-down element of the Cabibbo-Kobayashi-Maskawa (CKM) matrix, which is the key ingredient to test the CKM-matrix unitarity, a fundamental requirement of the electroweak standard model. To extract V_{ud} from experimental ft data, theoretical corrections must be applied to take account of unobserved ISB effects in the analog states of the parent and daughter nuclei [27,28]. For this purpose, experimental CDE and TDE [29] serve as a calibration for nuclear shell models utilized to calculate such corrections [28,30]. Reference [30] showed that anomalies observed in superallowed $0^+ \rightarrow 0^+$ β decay can only be explained by introducing INC forces into the model, which is achieved via fitting the experimental CDEs and TDEs. According to the

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2020 compilation of superallowed $0^+ \to 0^+$ β decays [31], these transitions are now known in the decays of nuclei up to ^{74}Rb in the upper fp shell, for which the ISB is the discussion topic of the present work.

Another important implication is related to the comprehension of the stellar nucleosynthesis processes [32,33]. Precise masses especially in the vicinity of the waiting-point nuclei [34] are needed to locate the rp-process path and to infer properties of the underlying accreting neutron stars [35]. The CDE has been thought to be a reliable quantity for predictions of unknown masses on the proton-rich side of the $N = Z$ line [36]. Using CDE, one can not only derive nuclear masses, but also determine positions of unknown resonances, provided that level schemes of neutron-rich mirror nuclei are experimentally known [37,38]. Moreover, using TDE one can obtain precious information for $T_z = -1$ nuclei if experimental values exist for $T_z = 0$ and $T_z = +1$ nuclei belonging to the same $T = 1$ multiplets [39,40].

The CDE for mirror nuclei, defined as

$$\text{CDE}(A, T) = \text{BE}(T, T_{z<}) - \text{BE}(T, T_{z>}), \quad (1)$$

is an established measure of charge-symmetry breaking [3], where $\text{BE}(T, T_{z<})$ and $\text{BE}(T, T_{z>})$ are binding energies of the mirror partners having the smaller proton number ($Z_<$) and larger proton number ($Z_>$), respectively. Under the assumption of spherical nuclei, the CDE exhibits a linear dependence on $Z/A^{1/3}$ regardless of the shell structure [41–43]. This relationship could be taken as an initial estimate for CDE within an uncertainty of about 150 keV [44]. Hartree-Fock (HF) calculations could reproduce the experimental CDEs within an uncertainty of about 100 keV. Masses of proton-rich nuclei obtained from such calculated CDEs combined with known masses of the neutron-rich mirror partners have been used for mapping the proton drip line and in the rp-process calculations [45]. Influence of quadruple deformation on CDEs was also studied [46,47]. It is well known that due to the strong Coulomb repulsion in systems with all protons paired, CDEs exhibit odd-even staggerings with Z, which lead to larger CDEs for the mirror pairs having odd-$Z_>$ and even-$Z_<$ [48]. To emphasize the staggerings, ΔCDE is defined between nuclei with mass numbers A and $A + 2$,

$$\Delta\text{CDE}(A, T) = \text{CDE}(A + 2, T) - \text{CDE}(A, T). \quad (2)$$

The TDE for $T = 1$ triplets, a signature of charge-independence breaking, is defined as

$$\text{TDE}(A, T) = \text{BE}(T, T_{z<}) + \text{BE}(T, T_{z>}) - 2\text{BE}(T, T_z = 0). \quad (3)$$

To magnify the trend with increasing Z, ΔTDE for nuclei differing in mass number by four is defined as

$$\Delta\text{TDE}(A, T) = \text{TDE}(A + 4, T) - \text{TDE}(A, T). \quad (4)$$

In the 2013 work [10], two of us (K.K. and Y.S.) and collaborators carried out an investigation on the INC effects for masses $A = 42$–95 using CDEs and TDEs as probes. Large-scale shell-model calculations were performed by employing two modern effective interactions (GXPF1A and JUN45) for the corresponding mass regions with inclusion of the

Coulomb plus INC nuclear interaction. The INC interaction was applied only in the $f_{7/2}$ shell to understand the observed staggering in ΔCDE [10]. A clear conclusion was made that the INC forces are important for the $f_{7/2}$-shell nuclei.

However, no conclusion about INC forces could be drawn for the upper fp-shell and heavier nuclei [10]. Even for the $f_{7/2}$ shell, some puzzles remained. An apparent change in the staggering phase of the experimental ΔCDE/Z for $T = 1/2$ at $A = 69$ was left as an open question [10]. The calculations could reproduce the smooth trend of the TDEs for $A = 4n + 2$ in lighter masses, where n is an integer number, as well as a sudden decrease at $A = 58$ and the corresponding drop in ΔTDE at $A = 54$. However, the TDE analysis for the sequence of $A = 4n + 2$ nuclei was stopped at $A = 58$ because no experimental masses were available. Furthermore, the TDE of $A = 4n$ could not be determined due to the lack of experimental masses of $T_z = -1$ nuclei.

Recently, plenty of experiments on mass measurements of proton-rich nuclei have been performed. The masses were either measured for the first time or with greatly improved accuracy. To mention a few, masses of neutron-deficient gallium isotopes, $^{60-63}$Ga, were measured with the TITAN multireflection time-of-flight mass spectrometer [49]. At the experimental Cooler-Storage Ring (CSRe) in Lanzhou, masses of $T_z = -3/2$, -1, and $-1/2$ nuclei were measured [50–52] with high precision by an upgraded method of isochronous mass spectrometry (IMS). This novel technique, named Bρ-defined IMS [53], greatly advanced mass measurements of neutron-deficient nuclei toward the heavier mass region, allowing for a considerable extension of our knowledge on systematic behavior of CDE and TDE to the upper fp shell.

In Fig. 1, CDEs and TDEs starting from $A = 20$ are illustrated together with a simplest estimate for the nuclear Coulomb energy by assuming nucleus as a uniformly charged sphere [3]. The corresponding values were determined utilizing the masses from Atomic Mass Evaluation 2012 (AME2012) [54], and 2020 (AME2020) [55], as well as the latest experimental input. One observes in Fig. 1(a) that the calculated Coulomb energy generally overestimates the CDEs for $T = 1/2$. These large deviations from the data, by more than 1 MeV on average, were qualitatively explained by the exchange effect due to the Pauli principle, which keeps the protons apart and thus weakens the Coulomb repulsion [56]. There have been many attempts to resolve this discrepancy known as the Nolen-Schiffer anomaly [57–59].

With the Coulomb energy as a mean value, an odd-even zigzag pattern of ΔCDE/Z is seen in Fig. 1(b). The pattern exhibits oscillations with a common phase, but at $A = 69$, the mass data from both AME2012 and AME2020 suggest a phase reversion. This phase reversion was noticed in Ref. [10] and discussed in detail in Ref. [47]. Notably, the shell-model calculation in Ref. [10], no matter with or without the INC interaction in the $f_{7/2}$ shell, could not substantiate this perceived phase reversion. It was speculated [10] that the anomaly might be attributed to the cited mass value of ^{69}Br in Ref. [54] being for an isomer rather than the ground state. In AME2020, with the proton separation energy determined from the β-delayed proton emission of ^{69}Kr experiment [60], the mass excess of
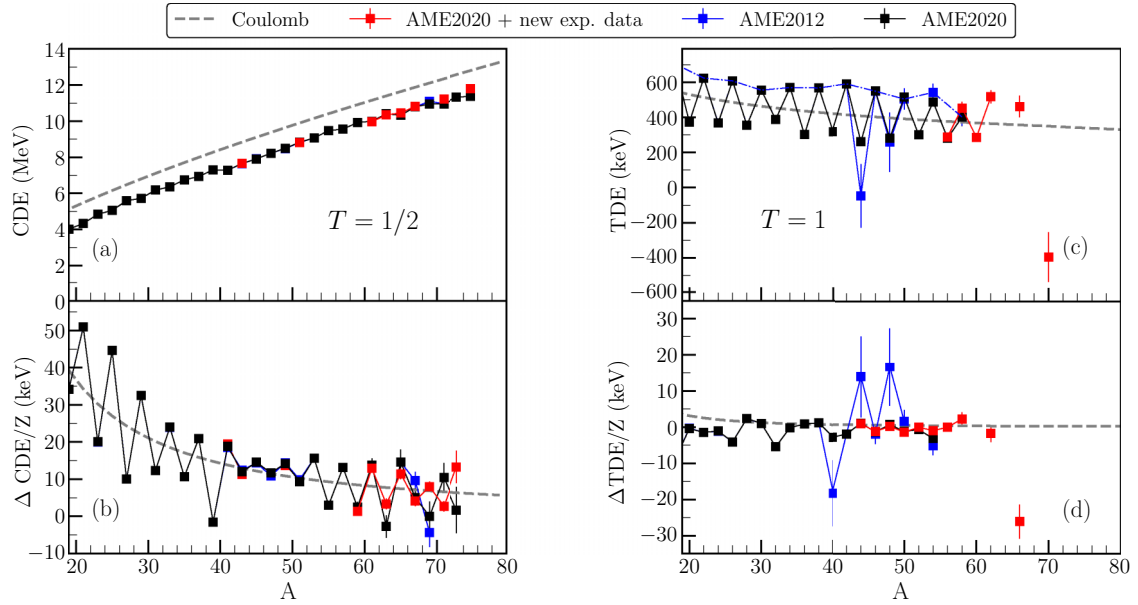
FIG. 1. Experimental (a) CDE, (b) CDE differences between A and $A + 2$ nuclei (shown as ΔCDE/Z), (c) TDE, and (d) TDE differences between A and $A + 4$ nuclei (shown as ΔTDE/Z). For comparison, Coulomb energies calculated assuming nuclei as uniformly charge spheres [3] are also shown.

^{69}Br was updated to be $-46260(40)$ keV, that is by 150 keV more bound than the one in AME2012. However, the phase revision at $A = 69$ still remains and the reversed phase persists for the heavier masses of ^{73}Rb and ^{75}Sr [see black squares in Fig. 1(b)]. It is remarkable that by applying the new experimental mass values, the previously suggested phase revision at $A \geqslant 69$ disappears. The change is due to the new experimental mass excess of ^{71}Kr of $-46056(24)$ keV, which is by 270 keV less bound than the value in AME2020. Furthermore, the phase at $A = 73$ was rectified by the new mass of ^{75}Sr, which is by 420 keV less bound than the AME2020 value derived from the Q value of the β decay of ^{75}Sr [61].

The TDEs for $T = 1$ starting from $A = 20$ are shown in Fig. 1(c), and compared to the Coulomb energy estimate [3]. With the new mass data, the TDEs for $A = 4n + 2$ are conspicuously extended from $A = 58$ (see Ref. [10]) to $A = 70$ and those for $A = 4n$ are now presented up to $A = 60$. The estimated Coulomb energy serves as a smoothly changing average, around which TDEs oscillate, thereby dividing data into two branches either above or below it. Within error bars of the last few data points, the TDEs of $A = 4n + 2$ ($A = 4n$) lie, on average, roughly by 150 keV above (below) the Coulomb energy prediction.

The blue dash-dotted line in Fig. 1(c) connects the $A = 4n + 2$ TDEs determined by the AME2012 mass data and is shown to emphasize the sudden drop at $A = 58$. This sudden drop seemed to suggest a termination of the odd-even oscillation in TDE [10]. Now with the updated mass data, the zigzag trend continues. This change is due to the updated masses of ^{54}Ni and ^{58}Zn. Compared to AME2012, the new mass of ^{54}Ni is by 65 keV more bound and that of ^{58}Zn is by 52 keV less bound, which result in a decrease in TDE at $A = 54$ and an increase at $A = 58$, thus retaining the smooth trend for overall TDEs of the $A = 4n + 2$ branch.

A large, negative TDE at $A = 70$ is observed, which deviates strongly from the general trend in Fig. 1(c). This anomaly was discussed in detail by analyzing the systematics of b and c coefficients of the quadratic IMME [62], and was attributed to the presently adopted mass of ^{70}Br in AME2020.

The Coulomb estimate in Fig. 1(d) predicts very small ΔTDEs for lighter nuclei and the values approach zero with increasing A. Experimental data, with only small scatter in the light mass region, do not deviate much from this trend. Especially in the range of $A = 44$–62, the new ΔTDE data follow precisely the Coulomb energy prediction. The previously found irregular values at $A = 40$, 44, and 48, determined with the data from AME2012, now converged to the Coulomb estimate. Also the small drop at $A = 54$, due to the sudden drop in TDE at $A = 58$ when data of AME2012 were used, disappears.

The experimental CDE and TDE values determined with the new masses can be found in the Supplemental Material [63] and Refs. [49–51,64]. The data beyond $A = 58$, TDE for $T = 1$ and CDE for $T = 1/2$, permit us to extend the ISB study to the upper fp shell. We have performed shell-model calculations with the same nuclear INC interaction as in the $f_{7/2}$ orbit [10]. Figures 2(a) and 2(b) show the calculated CDEs for isospin $T = 1/2$. The calculations reproduce all experimental data reasonably well. The odd-even oscillation in CDE between A and $A + 2$ nuclei, described in ΔCDE/Z, are reproduced correctly. The oscillations can be understood by the difference between the proton- and neutron-pairing gaps due to the Coulomb force [10]. Nevertheless, except that the INC forces lead to a reduction in staggering amplitudes of CDEs for $A = 41$–53, no significant differences can be seen in the calculations with and without INC. Finally at $A = 73$, the present experimental value of ΔCDE/Z for $T = 1/2$ deviates from the calculation by approximately 6 keV
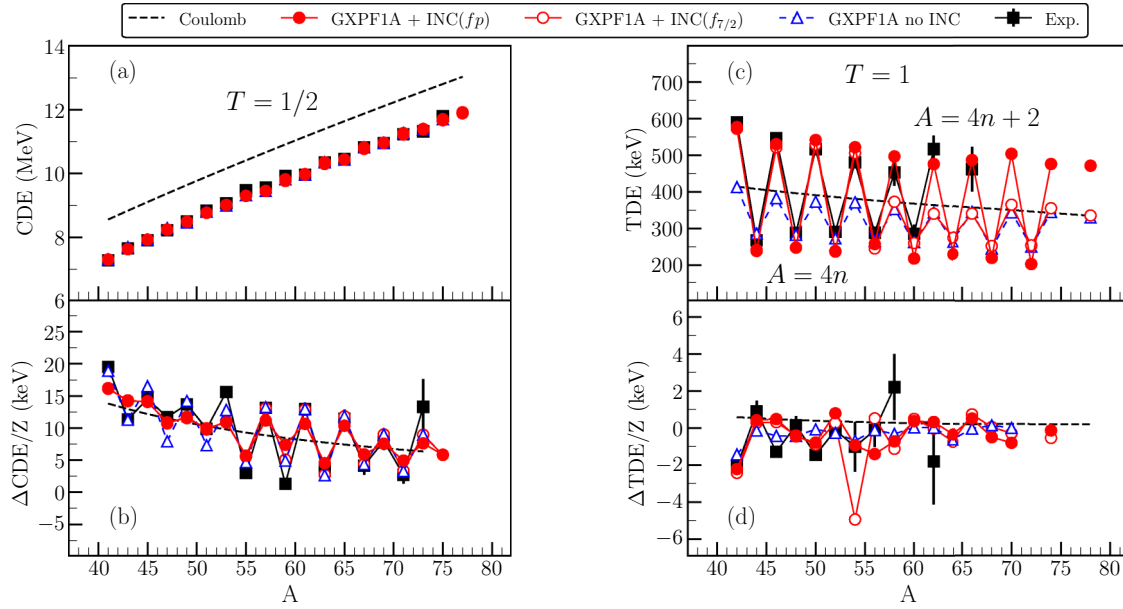
FIG. 2. Experimental (a) CDE for $T = 1/2$ and (b) CDE differences between A and $A + 2$ nuclei (shown as ΔCDE/Z), (c) TDE for $T = 1$ and (d) TDE differences between A and $A + 4$ nuclei are compared with the shell-model calculation. For comparison, estimated Coulomb energies [3] are also shown.

(about 1.3 σ), which may suggest an improvement of the mass of ^{75}Rb.

In contrast, significant effects due to the INC interaction are found in TDEs for $T = 1$. As shown in Fig. 2(c), for $A = 42$–78, experimental TDE data indicate large oscillations between the $A = 4n + 2$ and $A = 4n$ data branches. The calculated results (filled circles) with the INC interaction for the full fp shell are in excellent agreement with the data. When the INC interaction is switched off, the TDEs for the $A = 4n + 2$ branch decrease drastically, while those for $A = 4n$ are hardly affected. The distinct response to the INC force from the two branches can be interpreted as follows. As triplet nuclei in a TDE of $A = 4n + 2$ branch can be understood as putting pp, nn, and pn pairs on top of a common even-even $N = Z$ core, the $T = 1$ TDEs for $A = 4n + 2$ triplets are considered as good indicator of the isotensor force acting among them. As shown in Fig. 2(c), the prediction of the isotensor force of 165 keV (= $\beta_{pp} + \beta_{nn} - 2\beta_{pn}$, see Ref. [63,65,66]) agrees well with the average difference of $\sim$150 keV between the calculated TDEs with and without the INC interaction included. If the INC for the upper fp shell is switched off, the TDEs drop suddenly at $A = 58$, consistent with the previous result in Ref. [10].

With the calculated TDE value, 503 keV at $A = 70$, together with the known masses of ^{70}Se and ^{70}Kr (see Ref. [63,64]), the mass excess of ^{70}Br can be deduced to be $-51877(70)$ keV. This value agrees with the prediction from the systematic study [62] of $-51934(16)$ keV.

We can thus conclude that the INC interaction is significant for $A = 4n + 2$ TDEs, and the inclusion of INC in the upper fp shell in shell-model calculations turns out to be as important as in the $f_{7/2}$ orbit. The TDE for $T = 1$ is a good indicator of the isotensor INC force, which is estimated to be 150 keV from the present study in the $A = 42$–78 region.

Figure 2(d) shows the calculated ΔTDE/Z for both branches of data points. In consistence with the data, they are close to zero and vary only slightly in the entire mass region. The ΔTDE/Z for $A = 4n + 2$ may be regarded as another quantity to underline the (nearly) A-independent upward shift of TDEs due to the INC effect.

Little is currently known about the INC effects in the heavier mass regions toward ^{100}Sn. An initial investigation of CDEs and TDEs was carried out with the JUN45 interaction in the fpg model space including the $g_{9/2}$ orbit [10]. The study was incomplete because no INC forces were included. Based on the present analysis, we may expect that the INC effects would still be important and the discussed TDE oscillations in this work would continue to nuclei in the $A = 80$–100 region.

In summary, the new mass data allow us, for the first time, to make a significant step toward understanding the isospin symmetry breaking in the upper fp shell. For CDEs, the regular odd-even staggering phase of the CDE differences persists up to $A = 73$, rectifying the previously found phase inversion at $A = 69$. For TDEs, a general odd-even staggering pattern up to $A = 62$ was observed. The new data refuted several anomalies in TDEs suggested before. Detailed shell-model calculations, employing one uniform nuclear INC interaction for the entire fp shell, provided an excellent description of the experimental CDE and TDE values. The largest change in TDE due to the INC effect for $A = 4n + 2$ triplet nuclei, having pp, nn, and pn pairs on top of a common even-even $N = Z$ core, is estimated to be 150 keV for the isotensor component of isospin violating interactions. Finally, the astonishingly regular behavior of the charge symmetry and charge independence of the attractive NN interactions discussed here may justify an extension of the INC concept up to the ^{100}Sn region.

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