## Isospin Dependence in the Odd-Even Staggering of Nuclear Binding Energies

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The FRS-ESR facility at GSI provides unique conditions for precision measurements of large areas on the nuclear mass surface in a single experiment. Values for masses of 604 neutron-deficient nuclides  $(30 \le Z \le 92)$  were obtained with a typical uncertainty of 30  $\mu$ u. The masses of 114 nuclides were determined for the first time. The odd-even staggering (OES) of nuclear masses was systematically investigated for isotopic chains between the proton shell closures at Z = 50 and Z = 82. The results were compared with predictions of modern nuclear models. The comparison revealed that the measured trend of OES is not reproduced by the theories fitted to masses only. The spectral pairing gaps extracted from models adjusted to both masses, and density related observables of nuclei agree better with the experimental data.

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Significant progress has been achieved over the last years in constructing self-consistent mass models [1,2]. These models aim to reliably describe the properties of nuclei far off the valley of  $\beta$  stability, where the experimental information is scarce or even not available yet. For instance, in modeling the astrophysical *r* process of nuclear synthesis one needs precise knowledge of masses and halflives of very exotic nuclei and one has to rely on theoretical predictions since most of the nuclides involved have not even been produced in the laboratory yet. The predictions for these nuclides dramatically deviate for the different models [2]. Thus new experimental data on exotic nuclei and consequently better understanding of nuclear structure away from the valley of  $\beta$  stability is essential for further theoretical development.

Odd-even staggering (OES) of nuclear binding energies was detected in the early days of nuclear physics [3] and was explained by the presence of pairing correlations between nucleons in the nucleus [4]. Pairing contributes only little to the total nuclear binding energy but its influence on the nuclear structure is significant.

The common way to extract experimental information about the pairing correlations is to measure the value of the OES which approximates the pairing-gap energy ( $\Delta$ ) in the standard Bardeen-Cooper-Schrieffer (BCS) theory [5]. The latter quantity is connected with the strength of the pairing interaction (*G*):

$$\frac{2}{G} = \sum_{\nu} \frac{1}{\sqrt{(\varepsilon_{\nu} - \lambda)^2 + \Delta^2}},\tag{1}$$

where  $\varepsilon_{\nu}$  is the single-particle energy and  $\lambda$  is the chemical potential. The summation goes over all single-particle levels  $\nu$  below and above the Fermi energy. In order to evaluate this sum in local pairing functionals, a (smooth) cutoff in energy is usually implemented.

Neutron  $(\Delta_n)$  and proton  $(\Delta_p)$  pairing gaps are usually determined from finite-difference equations of measured masses [6], e.g., by the five-point formulas:

$$\Delta_n^{(5)} = -\frac{1}{8} [M(Z, N+2) - 4M(Z, N+1) + 6M(Z, N) - 4M(Z, N-1) + M(Z, N-2)], \qquad (2)$$

$$\Delta_p^{(5)} = -\frac{1}{8} [M(Z+2, N) - 4M(Z+1, N) + 6M(Z, N) - 4M(Z-1, N) + M(Z-2, N)], \qquad (3)$$

where M(Z, N) is the mass of an atom with Z protons and N neutrons.

The well-known parametrization  $\Delta \simeq 12/\sqrt{A}$  MeV [4] (A = N + Z) provides the average trend for nuclei close to stability. A dependence of the pairing strength on the neutron excess was suggested in [7]. It was later observed from the mass determination of exotic Dy-Hg isotopes that  $\Delta_p$  and possibly  $\Delta_n$  increase towards the proton dripline [8].

In this Letter we present new results on the OES obtained from our high-precision mass measurements compared with predictions of modern nuclear theories.

The experiment for direct mass measurements was performed at the FRS-ESR facility as continuation of a successful scientific program addressing basic nuclear properties of stored exotic nuclides [9-11]. Exotic nuclei were produced by projectile fragmentation of a (600-900) MeV/u <sup>209</sup>Bi primary beam in (4-8) g/cm<sup>2</sup> beryllium targets placed at the entrance of the fragment separator (FRS) [12]. The fragments were spatially separated in flight and injected into the cooler-storage ring ESR [13]. In the ESR, the velocity spread of the stored fragments was reduced by electron cooling to  $\delta v/v \approx 5 \times$  $10^{-7}$ . This condition provides an unambiguous relation between the revolution frequencies of the ions and their mass-to-charge ratios which is the basis for Schottky mass spectrometry (SMS). The time required for the electron cooling was about 10 s, which constrains the range of nuclides that might be investigated by this method. The SMS has reached the ultimate sensitivity by recording single ions stored in the ESR leading to a mass resolving power of more than  $2 \times 10^6$  (FWHM) [14,15].

In this new experiment with SMS, 582 different nuclides were observed in the frequency spectra. From this set of nuclei, 117 were used for calibration. The achieved mass accuracy was typically 30  $\mu$ u which represents an improvement by a factor of 3 compared to our former experiments [10]. In addition, the masses of 139 nuclides were determined indirectly by means of known decay energies ( $\alpha$ ,  $\beta$ , or proton emission). The masses of 114 nuclides were obtained for the first time [14]. The measured masses cover a large area of neutron-deficient nuclides from krypton to uranium. All *directly measured* values (see [16]) have been included in the latest atomic mass evaluation [17].

The achieved experimental mass accuracy has allowed us to perform new systematical studies on nuclear pairing. The new data were combined with the data of Ref. [18] and precise values of OES for all even-Z isotopic chains in the region between the Z = 50 and Z = 82 closed shells were extracted. Only even-even nuclei were considered. The results obtained show that indeed the values of OES for both the protons and the neutrons increase towards the proton dripline, thus confirming earlier observations for a small number of nuclides [8]. Moreover, this is a general trend for all even-Z isotopic chains from tin to lead. The tin, tellurium, mercury, and lead isotopes were not considered for protons since the closed shells at Z = 50 and Z =82 have strong influence. Similarly, the nuclides with N =80, 82, 84, 124, 126, and 128 were excluded for neutrons. It is necessary to note that no such general trend of the OES was observed for isotopes below tin. For isotopes above lead the experimental information is still too scarce to draw a definite conclusion. The large number of newly obtained OES values allow us to perform quantitative comparisons with calculations. We will first compare the results with a macroscopic-microscopic model and then continue with up-to-date microscopic models.

The pairing-gap energies were extracted from the masses calculated with the finite-range liquid-drop model (FRLDM) [19]. In this model, the single-particle potential generated by the Yukawa interaction was used for the microscopic part. To obtain a maximal number of extracted OES values, the calculations were done using 2nd-order mass differences (three-point formulas [6]). The comparison with the experiment is shown for the isotopic chains of hafnium and tungsten in Fig. 1. It is clearly seen that the experimental isospin dependence of pairing-gap energies is not reproduced by the original FRLDM [19]. To improve this description, the BCS pairing part of the model has been adjusted to the new experimental data. Different from Ref. [19], a single-particle spectrum was generated with the deformed Woods-Saxon potential. The pairing strength G was parametrized with 2 constants for protons (p) and



FIG. 1. Comparison of the proton and neutron pairing-gap energies for even-even hafnium (upper panel) and tungsten (lower panel) isotopes derived with 2nd-order mass differences of experimental masses and from the predictions of the original FRLDM mass model [19] and with newly readjusted pairing strengths. The experimental values are taken from Refs. [16,18].

neutrons (n)  $G_{p(n)} = g_{0p(n)}/A + g_{1p(n)}(N-Z)/A^2$  [6,7]. The number of levels taken into account [see Eq. (1)] was equal to N for neutrons and Z for protons. All experimental OES values for even-even nuclides between Z =50 and Z = 82 were used for the adjustment. The best agreement between the values of OES derived from the calculated masses and experimental values was achieved with  $g_{0p} = g_{0n} = 20.80$  MeV and  $g_{1p} = -g_{1n} = 22.40$  MeV [14]. Note that the constants  $(g_{0p}, g_{0n})$  and  $(g_{1p}, g_{1n})$  converge to the same values, which is quite remarkable since this was not a constraint demanded in the analysis. The results obtained are labeled in Fig. 1 as readjusted FRLDM. The difference between the original FRLDM of Ref. [19] and the readjusted model is obvious. With this new pairing description the  $\sigma_{\rm rms}$  value for the prediction of nuclear binding energies between Z = 50 and Z = 82 closed shells has improved by about 25%.

Going to microscopic models, pairing-gap energies were calculated from predictions of nuclear mass calculations of several self-consistent mass models, which are SkyrmeHartree-Fock (SHF) calculations plus pairing treated in the BCS (HF + BCS) formalism (MSk7 force) [20] and three models where pairing is treated with the Bogoliubov (HFB) approach with different pairing cutoff parametrizations (BSk1, BSk2) [21,22] and including density-dependent pairing (BSk3) [23]. The results calculated with 4th-order mass differences [Eqs. (2) and (3)] are presented for even hafnium isotopes in Fig. 2. It is clearly seen that the general trend is *not* reproduced by any of the models. A comparison with four other recent HFB models where the implementation of different effective masses, with and without density-dependent pairing (BSk4-BSk7) [24], showed similar results as those of Fig. 2.

Another branch of self-consistent mean-field models are relativistic mean-field (RMF) models employing finiterange (FR) meson fields or point couplings (PC). For this study, we employed three of the best parametrizations available, namely, NL-Z2 [25] and NL3 [26] for the finite-range variant and PC-F1 [27] for the point-coupling model. In contrast to the mass models described above,





FIG. 2. Comparison of the proton  $(\Delta_p)$  and neutron  $(\Delta_n)$  pairing-gap energies for even-even hafnium isotopes derived from fourth-order differences of experimental and of calculated masses. Hartree-Fock plus pairing treated with the BCS formalism (MSk7) and three HF-Bogoliubov models with different cutoff parametrizations (BSk1 and BSk2) and density-dependent pairing (BSk3) were used. The experimental values are taken from this work (full symbols) and from Ref. [18] otherwise.

FIG. 3. Comparison of the proton  $(\langle \Delta \rangle_p(uv))$  and neutron  $(\langle \Delta \rangle_n(uv))$  pairing-gap energies for even-even hafnium nuclides calculated with several RMF and Skyrme-Hartree-Fock models (see text). These models were adjusted not only to the nuclear binding energies but also to form-factor related observables. The experimental points were derived from measured masses using Eqs. (2) and (3). The full symbols represent our new experimental values; the others are from Ref. [18].

these RMF forces are adjusted to both energy and formfactor related observables (e.g., rms radii, diffraction radii, surface thicknesses, etc.), and are meant to describe both kinds of observables. Furthermore, we performed calculations with the SHF forces SkI3 and SkI4 [28] with extended spin-orbit terms, which-similar to the RMF forces-have been adjusted to both masses and density related observables. In both the RMF and the SHF models, we employ BCS pairing with a density-independent  $\delta$ force. As a first approach, the pairing gaps have been estimated from the single-particle spectrum with *uv*-weighted single-particle gaps [29] ( $v^2$  are the occupation probabilities), which circumvents the uncertainties related to the calculation of odd-even systems. These quantities constitute a measure of the pairing contribution to the OES. As discussed in Ref. [29], these results need to be carefully interpreted due to polarization effects and the nonpairing-type contributions to the OES.

It is striking that the RMF and SHF calculations in Fig. 3 give very similar results: the general trend of the rising pairing-gap energies is reproduced. However, some local discrepancies are observed, as, e.g., close to  $T_z = 5$ , which can be related to the N = 82 closed shell. Although the models in Fig. 2 in general have much higher predictive power for the nuclear masses [2], the difference in the description of the experimental OES data is obvious. This result has not been expected and demonstrates the need for a better understanding of both the roles of the various observables and adjustment protocols as well as the procedure of calculating OES within these frameworks.

All models tested in this Letter take into account the nuclear deformation which is essential here since most of the nuclei investigated are deformed. Moreover, the observed general trend of OES has the same slope and magnitude for nearly all isotopic chains from xenon to platinum. Use of the 2nd, 3rd, or 4th-order mass differences to disentangle the mean-field contributions to the OES and pairing-gap energies has been intensively discussed in the literature [30-34]. Our overall conclusions, as checked in different analyses, are not changed if 3, 4, or 5-mass formulas are used. Since the mass number changes within an isotopic chain, volume effects might contribute to the observed isospin dependence of the OES.

With the new data available, it became possible to examine the OES predictions of different theories. The new results are helpful for a better description of the pairing in exotic nuclei, which is mandatory for a *reliable* theory.

An important future aspect is whether the observed trend of the pairing-gap energies persists for nuclides with even greater neutron excess. A recent experiment has been performed at the FRS-ESR to measure masses in the Yb-Pb region on the neutron-rich side of the chart of nuclides. Measurements of very exotic neutron-rich nuclides, which cannot be produced with the present facility, are foreseen within the FAIR project [35].

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