Direct mass measurements of Cd isotopes show strong shell gap at N=82

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Abstract

A \(^{238}\)U projectile beam was used to create cadmium isotopes via abrasion-fission at 410 MeV/u in a beryllium target at the entrance of the in-flight separator FRS at GSI. The fission fragments were separated with the FRS and injected into the isochronous storage ring ESR for mass measurements. The Isochronous Mass Spectrometry (IMS) was performed under two different experimental conditions, with and without B\(_{2}\)-tagging at the dispersive central focal plane of the FRS. In the experiment with B\(_\rho\)-tagging the magnetic rigidity of the injected fragments was determined by an accuracy of \(2 \cdot 10^{-4}\). A new method of data analysis, using a correlation matrix for the combined data set from both experiments, has provided mass values for 25 different isotopes for the first time. The high selectivity and sensitivity of the experiment and analysis has given access even to rare isotopes detected with a few atoms per week. In this letter we present for the \(^{129,130,131}\)Cd isotopes mass values directly measured for the first time. The Cd results clearly show a very pronounced shell effect at N=82 which is in agreement with the conclusion from \(\gamma\)-ray spectroscopy of \(^{130}\)Cd and confirms the assumptions of modern shell-model calculations.

Keywords: In-Flight Separation, Storage Ring, Fission Fragments, Isochronous Mass Spectrometry, Shell Closure at N=82, \(^{129,130,131}\)Cd, Mass Models, Shell Model Calculations

1. Introduction

Accurate mass measurements reflect details of the evolution of nuclear structure and stability as well as the energy levels and spatial distributions of the bound nucleons \cite{1}. A first microscopic explanation of the observed shell structure and the corresponding magic numbers \(N=82,50\) of neutrons and protons, at which the nuclei have larger binding energies, refined the understanding of nuclear properties and manifested the application of the shell model. The advent and application of radioactive nuclear beam facilities \cite{4} and novel mass spectrometers \cite{5} have constantly enlarged the number of known isotopes with unusual proton-to-neutron ratios and thus explored the properties at the outskirts of the chart of nuclides and soon it became evident that the nuclear shell structure can change towards the driplines. Shell quenching, complete disappearance, or even new magic numbers have been theoretically predicted \cite{6,7} and observed in experiments \cite{8,9,10,11,12}. The best known examples, both theoretically and experimentally, are the \(N=20\) and \(N=28\) “islands of inversion” \cite{11,12,13,14,15,16} where the gain in correlation energy driven by quadrupole deformation is able to overcome the normal level population deduced from the standard spherical mean field. As a result the traditional \(N=20\) and \(N=28\) shell closures completely disappear. It has also been argued that such a shell quenching would occur for neutron-rich N=82 nuclei. This phenomenon was originally suggested based on Skyrme Hartree-Fock-Bogoliubov calculations \cite{16}. Almost simultaneously it was realized that the occurrence of an abundance trough around \(A \sim 115\) in r-process calculations \cite{17} could be cured using a mass model with a quenched shell gap far from stability \cite{18,19,20}. The abundance trough around \(A \sim 115\) is associated with a ‘saddle point behavior’ seen

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in the two-neutron separation energies for \( Z \approx 40 \) and \( N = 75-82 \) in several mass models related to a transition from deformed nuclei around \( N \sim 75 \) to spherical nuclei at \( N=82 \). In mass models with a quenched shell-gap like the quenched extended Thomas-Fermi model (ETFSI-Q) the deformation is greatly reduced and consequently the ‘saddle point behavior’ in the two-neutron separation energies disappears. However, it should be pointed that the ‘saddle point behavior’ and the quenching of the shell gap are not necessarily related as the first could also be associated to instabilities of mean-field models in regions of shape coexistence that require the inclusion of beyond-mean field correlations. It is interesting to notice that the ‘saddle point behavior’ is absent in the Duflo-Zuker shell-model inspired mass model and consequent does not result in an abundance trough around \( A \sim 115 \) despite of predicting a relatively large \( N=82 \) shell gap far from stability. Furthermore, the strong abundance troughs predicted in calculations were largely due to the use of the so called classical r-process approximation which assumes \( (n, \gamma) \leftrightarrow (\gamma, n) \) equilibrium and an instantaneous freeze-out of neutron captures. Nowadays, we know that the r-process occurs in a very dynamical environment with continuously changing astrophysical conditions and that the competition between neutron captures, beta-decays and fission has an important impact in the final abundances.

Given the relevance to both r-process nucleosynthesis and nuclear structure of the existence of a new ‘island of inversion’ approaching \( Z \sim 40 \) for \( N=82 \), there have been many experimental attempts to provide evidence for the quenching of the shell gap. It should be clarified that quenching is sometimes used as a synonymous of reduction of the shell gap. However, in the context of this letter we will use ‘quenching’ according to the most common usage of disappearance of shell effects pointing to an ‘island of inversion’ analogous to the well-studied cases around \( ^{32}\text{Mg} \) for \( N=20 \) and \( ^{42}\text{Si} \) for \( N=28 \). The experimental information about the evolution of the \( N=82 \) gap is so far mainly indirect.

The present Isochronous Mass Spectrometry (IMS) experiment and the previous Penning trap mass measurements for the tin isotopes yield direct information on the shell effect. We have applied IMS to determine the masses of \( ^{129,130,131}\text{Cd} \) isotopes. The cadmium and tin results provide a direct experimental evidence for the evolution of the \( N=82 \) shell gap south of \( ^{132}\text{Sn} \).

### 2. Experiment and data analysis

A 410 MeV/u \(^{238}\text{U}\) projectile beam was extracted from the synchrotron SIS-18 with an average intensity of \( 1 \times 10^{10} \) /spill and impinged on a 1032 mg/cm\(^2\) beryllium target at the entrance of the fragment separator FRS. Neutron-rich fission fragments created via abrasion-fission were separated in flight with the FRS applying pure magnetic rigidity (B\(p\)) separation with the standard ion-optical operation mode. The separation mode, without degraders, was enabled by the large mean velocity difference of the projectile fragments and fission products and the restricted angular acceptance of the FRS. Practically this means, a suitable B\(p\)-selection with the FRS can provide fission-fragment beams without significant contributions of projectile fragments. The ions of interest were injected into the Experimental Storage Ring ESR for IMS and at a mean velocity corresponding to the transition energy of \( \gamma_t = 1.41 \). The magnetic fields of the FRS and ESR were set for \(^{133,135,136}\text{Sn}\) ions in different runs, i.e., these isotopes were subsequently centered at the optical axis.

The ESR was operated in the isochronous mode without application of any cooling. This means that the velocity spread of the fragments is determined by the B\(p\) acceptance of the ion-optical system. In a previous publication we have demonstrated that for IMS experiments in addition to the revolution time of the stored ions the magnetic rigidity or velocity measurement is required, because the isochronicity is strictly realized only for a single mass-over-charge (m/q) value. In principle, this additional measurement is not required for Schottky Mass Spectrometry (SMS) because the relative velocity spread of the different stored and cooled ions can be as low as \( 10^{-7} \). Nevertheless, our refined SMS analysis has revealed that an additional influence of the mean velocity causes an observed correlation.

The method of IMS including B\(p\)-tagging can be illustrated by the simple first-order formula

\[
\frac{d(m/q)}{m/q} = \frac{\gamma^2 d(T)}{T} + (1 - \frac{\gamma^2}{\gamma_t^2}) \frac{d(Bp)}{Bp},
\]

where \( T, \gamma_t \) and \( \gamma \) are the revolution time, the transition energy, and the relativistic Lorentz factor, respectively.

The additional velocity \( v \) and magnetic-rigidity measurements in FRS-ESR IMS experiments require special methods due to the ESR operation with fast extracted ion bunches characterized by a width of (0.2 – 0.5) \( \mu \)s. Particle detectors inside the FRS would have severe problems to identify event-by-event the fragments and accurately measure \( v \) and \( Bp \). A B\(p\)-resolution of \( 10^{-4} \) or better is required to achieve a mass resolution of about 200 keV for m/q close to ideal isochronicity, details are presented in reference. In this context, one has to take into account that the FRS transmission is \( d(Bp)/(Bp) = 2\% \) and the corresponding ESR injection acceptance is roughly one order of magnitude less. Therefore, mechanical slits with a slit opening of \( \pm 0.5 \) mm placed at the central dispersive focal plane of the FRS were used in a pilot IMS experiment. The slits defined in this way the magnetic rigidity (B\(p\)-tagging) of each injected ion with an accuracy of \( 2 \times 10^{-4} \). The operation of the new isochronous Rare RI-Ring at RIKEN can easily implement additional B\(p\) and \( v \) measurements event-by-event, because of the DC beam from the cyclotron. IMS measurements have been performed with and without B\(p\)-tagging for the same
settings of the magnetic fields of the FRS and ESR. The revolution time of the circulating ions in the ring were measured with a time-of-flight (ToF) detector equipped with a thin carbon foil and two micro-channel-plate (MCP) branches placed in a homogeneous magnetic dipole field of about 8.4 mT. The created secondary electrons in the carbon foil were isochronously deflected onto the MCPs to generate timing signals at each turn. The signals were recorded with commercial digital oscilloscopes (Tektronix TDS 6154C, 40 GS/s, 15 GHz; LeCroy LC584AM, 4 GS/s, 1 GHz). The data sets of the two different experiments, with and without Bρ-tagging, were combined and analysed with the new correlation-matrix method \[40\]. In this work we used for the first time a variable correlation factor dependent on the measured \(\frac{m}{q}\). A detailed description of this novel analysis will be presented in a forthcoming publication \[10\]. The advantage of this new analysis is that we could include ions with very low statistics down to a few events of a single isotope. In this way we can now present more than 20 new mass values which were not included in previous IMS evaluations of the same experiments \[41, 36\].

### 3. Results and discussion

The first check of the reliability of the new data analysis is the determination of the systematic error of the combined experiments. An average of 1600 events per reference mass could be applied in the analysis with the combined data sets. We determined the systematic error by using all reference masses and subsequently treat each of these masses as being unknown. The procedure can be illustrated by equation 2 and figure 1

\[
\sum_{i}^{n} \frac{(M_i - M_{i}^{\text{ref}})^2}{(\sigma_i^{\text{stat}})^2 + (\sigma_i^{\text{syst}})^2} = N_n
\]

where \(M_{i}^{\text{ref}}\) are the mass values and \(\sigma_i^{\text{ref}}\) the uncertainties of the reference nuclides. \(\sigma_i^{\text{stat}}\) and \(\sigma_i^{\text{syst}}\) are the statistical errors of the measured masses \(M_i\). \(N_n\) is the number of reference masses and \(\sigma_i^{\text{syst}}\) the systematic error. In this analysis 47 reference masses have been used \[42\]. The investigation covers the full m/q range of the present analysis. The mean value of the projected distribution is 1.29 keV. The deduced systematic error is 172 keV (standard deviation). For most of the new mass values the systematic error is the dominant contribution to the total error which results from the sum of the variances. The statistical error is determined by the uncertainty of the revolution time, which strongly depends on the number of turns for which the timestamps have been recorded. In the matrix method the statistical error for each nuclide was calculated by the square root of the diagonal elements of the inverse matrix. Details will be presented in reference \[40\].

A main motivation of the present mass measurements is the investigation the N=82 shell gap for Cd isotopes. We briefly review previous experiments and conclusions before presenting our results. Ref. \[43\] determined the beta-decay half-life of the \(^{130}\text{Cd}\) nuclide and its \(Q_\beta\) value. The mass of \(^{130}\text{Cd}\) in the Atomic Mass Evaluation 2012 (AME12) \[14\] is based on this \(Q_\beta\) value. It was concluded that the large measured \(Q_\beta\)-value of 8.32 MeV is an indication of shell quenching because the experimental value was in good agreement with the predictions of the ETFSI-Q mass model \[15\]. However, it is about 1 MeV larger than predicted by other mass models such as the Finite Range Droplet Model \[46\] or the Duflo-Zuker model \[23\]. A recent experiment \[47\] has determined a substantially shorter beta-decay half-life with more statistics for the \(^{130}\text{Cd}\) nuclides, which questions the determination of the \(Q_\beta\)-value in \[43\]. The measurement of the \(2^+\) excitation energy for \(^{126}\text{Cd}\) showed that their value was the smallest of the known N=80 isotones \[48\]. Again, it was interpreted as evidence of shell quenching. However, beyond-mean field calculations \[49\] showed that such a low excitation energy could be explained by the presence of prolate configurations in the wave function of \(^{126}\text{Cd}\). This may explain the measured small magnetic moment observed in \(^{127}\text{Cd}\) \[50\]. The same calculations predicted a much larger excitation energy of the \(2^+\) level in \(^{130}\text{Cd}\). This excitation energy has been measured to be 1.321 MeV \[51\] in agreement with shell-model calculations that assumed a N=82 shell closure. This fact together with the large excitation energy of the \(2^+\) state in \(^{126}\text{Pd}\) \[52\] (1.311 MeV) has been interpreted as evidence for a robust N=82 shell closure for both cadmium and palladium isotopes. Recently, a \(\gamma\)-spectroscopy experiment \[53\] has determined the \(3/2^-\) state in \(^{132}\text{Sn}\) corresponding to the \(p_{1/2}\) proton-hole state in \(^{132}\text{Sn}\). It is interesting to
Figure 2: Experimental shell effect. Difference of the measured mass values and the smooth Weizsäcker formula given by Eq. 3 for the elements Sn and Cd. The parameters used in the formula are presented in Table 1. The curves clearly show the extra binding energy due to the contribution of shell structure for the Sn and Cd nuclei in this mass region of N=82.

note that these new data are in perfect agreement with the predictions of the Duflo-Zuker monopole Hamiltonian [55] which is the base for the Duflo-Zuker mass model. The new experimental data has allowed to determine all relevant proton single particle states showing that no proton subshell closures are expected for light N=82 isotopes pointing to a significant reduction of the N=82 shell gap in analogy to the reduction observed in isotopes heavier than N=82. This reduction does not necessarily mean a quenching of the shell gap. It is well known that shell gaps reach a maximum value for magic numbers of protons and neutrons [56].

Figure 2 shows the difference of the experimental mass values for Sn and Cd isotopes and the prediction of the liquid drop model [1, 57]. The liquid drop parameters have been deduced from a fit to the tabulated values of the Atomic Mass Evaluation 2012 (AME12) [44]. The liquid-drop binding energy $B_{LD}$ has the form

$$B_{LD} = b_{coul} A - b_{surf} A^{2/3} - \frac{1}{2} b_{sym} \frac{(N - Z)^2}{A} - \frac{3}{5} \frac{Z^2 e^2}{R_c} - \left\{ \begin{array}{ll}
0, & \text{e-e nuclei} \\
2 b_{pair} A^{-1/2}, & \text{e-o or o-e nuclei} \\
2 b_{pair} A^{-1/2}, & \text{o-o nuclei}
\end{array} \right. \quad (3)$$

with $R_c = 1.24 \text{ fm} \cdot A^{1/3}$.

In figure 2 the differences between experimental mass excess (ME) and the corresponding liquid-drop values are plotted. The ME is the difference between the atomic mass and the corresponding mass number, both expressed in atomic mass units. The liquid-drop values are calculated with equation 3 and the parameters in table 1. The strong extra binding energy for the neutron-rich Cd isotopes is clearly observed at N=82, it amounts to roughly -14 MeV. The error bars for $^{129,130}$Cd are well within the symbol size whereas for $^{131}$Cd we have a large statistical error because only two events have been recorded with few number of turns. The experimental results in figure 2 clearly demonstrate that the tin and cadmium isotopes are characterized by the same difference of shell corrections at N=82. This observation contradicts reference [15] and clarifies the situation of the different statements in references [51, 58, 53].

The experimental ME values for $^{129,130,131}$Cd nuclei and the errors are presented in table 2. Our directly measured mass value for $^{130}$Cd atoms is 600 keV lower than the value deduced from $Q_β$ measurements [43]. This difference could be caused by a contribution of an excited state in the $β$-decay experiment. In principle, also in our measurements for $^{129,131}$Cd nuclei unresolved isomers could contribute [53, 54] whereas in $^{130}$Cd the lifetime is too short to interfere in this experiment. However, in this case the observed strong shell effect for Cd isotopes at N=82 would be even stronger for pure ground-state masses. The nearly equal shell effect for Sn and Cd is a striking feature of the present experimental results.

Other established results from mass measurements to discuss the evolution of nuclear structure and shells are the nucleon separation energies and their derivatives. The one-neutron separation energy $S_n$ and the pairing gap energy $\Delta_n^{(3)}$ is defined by:

$$S_n = -M(Z, N) + M(Z, N - 1) + M(0, 1) \quad (4)$$

$$\Delta_n^{(3)} = (−1)^N \frac{1}{2} [S_n(Z, N) - S_n(Z, N + 1)] \quad (5)$$

$A$ and $Z$ are the mass and proton numbers, respectively.

In the upper panel of figure 3 the shell evolution is manifested by our experimental (indicated in red) or AME12 one-neutron separation energies and their derivative near N=82. In the lower panel, we display an estimation of the gap representing the single particle spectrum (3-mass formula) which in principle is the derivative of the $S_n$-values. Both presentations demonstrate a strong shell effect at N=82 for Sn and Cd isotopes in complete agreement with the characteristics of figure 2. Again it is a striking experimental observation that Sn and Cd isotopes are governed by the same strong shell gap at N=82. The magnitude of

| $b_{coul}$ | $b_{surf}$ | $b_{sym}$ | $b_{pair}$ |
|-----------|-----------|-----------|-----------|
| 15.747    | 17.903    | 47.494    | 12.822    |

Table 1: Fitted liquid-drop parameters. The parameters, in units of MeV, for the Weizsäcker formula have been obtained by a fit to the mass values of reference [14].
Figure 3: In the upper panel experimental one-neutron separation energies ($S_n$) are shown for Cd and Sn isotopes. In the lower panel the corresponding pairing-gap energies are presented.

Table 2: Measured mass excess values ($ME$) of Cd isotopes. The statistical ($\sigma_{stat}$) and the total ($\sigma_{total}$) errors are tabulated. The systematic error is 172 keV for all the new masses measured. In addition, the extrapolated (#) values [44] and the $^{130}$Cd mass deduced from $Q_\beta$-measurement [43] are listed.

| isotope | $ME$ [keV] | $\sigma_{stat}$ [keV] | $\sigma_{total}$ [keV] | $ME_{AME12}$ [keV] | counts |
|---------|------------|-------------------|-------------------|------------------|-------|
| $^{129}$Cd | -63145 | 17 | 173 | -63509(196)# | 18 |
| $^{130}$Cd | -62131 | 41 | 177 | -61534(164) | 5 |
| $^{131}$Cd | -55583 | 925 | 941 | -55331(196)# | 2 |

the shell gap is within the experimental uncertainties the same.

A comparison of the experimental values for Cd isotopes with different theoretical mass models is presented in figure 4. The models are based on microscopic-macroscopic descriptions [46, 59, 60, 47], the Hartree-Fock-Bogoliubov (HFB) theory [61], and the shell-model inspired model of Duflo-Zuker [23].

Figure 4: Comparison of experimental mass values for Cd with different theoretical predictions.

In general, the comparison with theoretical predictions clearly demonstrates that the agreement with experimental results is getting worse towards the shell closure at $N=82$ and of course in the n-rich region, where no experimental results were available before. There are basically 3 different regions in this comparison. In the first region for $N\leq81$ the theories are relatively similar in their trend but all predict too low mass values (over-binding). At the shell the predictions are quite unreliable and beyond $N>82$, where no experimental data exists, they widely scatter. The upper panel show the described comparison for Cd isotopes and the lower panel the corresponding results for Sn isotopes. The masses of the presented Sn isotopes have been measured with Penning traps with higher resolving power and should not have influenced by unresolved isomeric states. The predictive power of the different models can be quantitatively characterized by the $\sigma_{rms}$ values. They are listed in table 3 for Sn and Cd isotopes in the experimentally known range of AME12 and for Cd isotopes presented in figure 4.

The comparison with theories over a larger $Z$ range for $N=82$ isotones is presented in figure 5 for the one-neutron gap, $\Delta_{1n}(Z, 82) = S_n(Z, 82) - S_n(Z, 83)$. Experimental $S_n$ values deduced from mass measurements exist only above cadmium. The figure clearly shows the large gap measured for $^{130}$Cd that is within the experimental uncertainties of similar magnitude than the one for $^{132}$Sn. No theoretical model is able to reproduce such a large gap for Cd and Sn isotopes. Looking at the evolution of the one-neutron gap and averaging the odd-even staggering one observes that only the ETFSI-Q and HFB27 models predict a strong reduction towards $Z=40$.

The new data for Cadmium isotopes is not consistent with an early quenching of the $N=82$ shell gap as claimed before by several publications [42, 48]. However, it cannot
nounced shell effect at N=82 which is assumed by modern shell-model calculations. The experimental values for the shell corrections are roughly the same for Sn and Cd isotopes.

The goal of future IMS experiments will be to measure the lower elements (40 < Z < 48) at and near the N=82 shell. Only the mass values of the lower elements will be decisive for the observation of possible shell quenching as it is proposed in several theoretical models. However, experimentally this interesting region can only be accessed by the next generation exotic nuclear beam facilities [62, 63, 64, 65].

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Table 3: Comparison of measured data with models. RMS deviations for Sn and Cd isotopes for different theoretical models are presented. In this comparison the new IMS values and the tabulated experimental values of AME12 are included. $\sigma_{rms,1}, \sigma_{rms,2}$ represent the values for the cases of all experimentally known Cd isotopes and the Cd isotopes in the mass range as shown in figure 4, respectively.

|         | $\sigma_{rms,1}$ [keV] | $\sigma_{rms,2}$ [keV] |
|---------|------------------------|------------------------|
| FRDM    | 619                    | 1537                   |
| HFB-27  | 523                    | 775                    |
| DZ28    | 315                    | 958                    |
| ETFSI-Q | 342                    | 512                    |
| ETFSI-1 | 479                    | 1200                   |
| WS3     | 396                    | 709                    |

Figure 5: One-neutron gap (see text) for N = 82 isotones.
