Eur. Phys. J. A **9**, 201–206 (2000)

THE EUROPEAN PHYSICAL JOURNAL A

New states in heavy Cd isotopes and evidence for weakening of the N = 82 shell structure

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Received: 13 July 2000 / Revised version: 22 September 2000
Communicated by J. Äystö

Abstract. A chemically selective laser ion source has been used in a β-decay study of heavy Ag isotopes into even-even Cd nuclides. Gamma-spectroscopic techniques in time-resolving event-by-event and multiscaling modes have permitted the identification of the first 2^+ and 4^+ levels in $^{126}Cd_{78}$, $^{128}Cd_{80}$, and tentatively the 2^+ state in $^{130}Cd_{82}$. From a comparison of these new states in $_{48}Cd$ with the $E(2^+)$ and $E(4^+)/E(2^+)$ level systematics of $_{46}Pd$ and $_{52}Te$ isotopes and several recent model predictions, possible evidence for a weakening of the spherical $N = 82$ neutron-shell below double-magic ^{132}Sn is obtained.

PACS. 27.60.+j 90≤A≤149 – 23.20.Lv Gamma transitions and level energies

Neutron-rich nuclei in the vicinity of the double-magic isotope $^{132}_{50}Sn_{82}$ have recently attracted renewed interest, triggered by the possibility of obtaining beams of very neutron-rich Ag, Cd and Sn nuclides with resonance ionization laser ion sources (RILIS) at the mass separator ISOLDE at CERN [1]. Apart from the importance to the single-particle structure outside the $Z = 50$, $N = 82$ core, the region "below" ^{132}Sn is of particular interest because of the possible existence of an anomalous behavior of the $N = 82$ shell closure when approaching the neutron drip-line (see, *e.g.*, [2,3]). After some initial theoretical papers in the 1970's [4,5] dealing with more general aspects of the weakening of the neutron shell structure near the drip-line, which are experimentally well established by now for $N = 20$ and $N = 28$ nuclei, evidence for such an effect at $N = 50$ was shown by Kratz *et al.* [6] for the decay of the r-process "waiting-point" isotope ^{80}Zn. For these light-mass magic neutron numbers, shell quenching obviously results in an increased collectivity and even setting in of strong quadrupole deformation. A weakening of the shell structure for $N = 50$ near the drip-line was also suggested by Haensel *et al.* [7], while an effect for $N = 82$ did not show up in their Hartree-Fock-Bogoliubov (HFB) calculations. A *spherical* quenching of the $N = 82$ magic neutron shell "below" ^{132}Sn was suggested by Kratz *et al.* [8] as an *"astrophysical request"* to properly reproduce the isotopic solar r-process abundances ($N_{r,\odot}$) in the $A \simeq 120$ mass region below the $N = 82$ shell closure. On the basis of self-consistent HFB calculations with the Skyrme force SkP, only shortly after, Dobaczewski *et al.* [9,10] predicted quantitatively that the well-known, pronounced spherical $N = 82$ (and $N = 126$) shell gap(s) near β-stability will, indeed, be reduced ("quenched") far from stability and may eventually disappear completely at the drip-line. An application of nuclear masses from the above HFB/SkP model around $N = 82$ in our r-process calculations [11] has resulted in a considerable improvement of the global $N_{r,\odot}$ fit, indicating a solution to a puzzle in r-process nucleosynthesis existing at that time. Given this success, in a next step we have replaced the exclusively spherical HFB/SkP masses by a mass model which takes into account both shell quenching and deformation; *i.e.* the ETFSI-Q approach [12] in our calculations of a canonical r-process (see, *e.g.*, [13, 14]). In the meantime, more than 60 different Skyrme parameterizations and various relativistic-mean-field

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(RMF) and relativistic-HF (RHF) approaches are being investigated (see, for example [15–19]; and the question of shell quenching is still discussed highly controversely. Restricting ourselves to Skyrme forces, some of the more recent parameterizations, such as SkI3 or SkI4, do make quantitatively different predictions on the trend of weakening of the neutron shell structure when approaching the drip-line, but still support the quenching assignment of SkP. Others, like SLy4, do not show a significant effect, and further ones, like SIII or SkM* do not yield shell quenching at all (see, *e.g.*, [2,3,17,20]). In this context, also the question whether for very neutron-rich isotopes pairing can still be treated in the BCS approximation, or pairing coupling between bound and continuum states becomes essential seems to have not yet been answered consentiently [2,19,21].

In any case, the idea of shell-quenching is worth further experimental study, apart from general nuclear-structure interest also because of its far-ranging consequences for astrophysics (see, *e.g.*, [13,22]). Therefore, mainly initiated by our own "astrophysical requests" from the 1993 Kratz *et al.* [8] paper, we have initiated a broad experimental program to explore exotic medium- to heavy-mass nuclei at different laboratories (*i.e.*, CERN/ISOLDE, JYFL/IGISOL, Mainz/SISAK, LLN/LISOL and GSI/FRS), using various production, separation and detection methods (for recent surveys, see, *e.g.*, [23–25]).

In a series of recent experiments with the General Purpose Separator (GPS) of ISOLDE, we have, for example, shown that due to the chemical selectivity introduced by RILIS very neutron-rich isotopes of $_{47}$Ag including the $N = 82$ r-process "waiting-point" nucleus 47-ms ^{129}Ag, and of $_{48}$Cd up to 95-ms ^{132}Cd$_{84}$, can be studied successfully in β-delayed neutron measurements (see, *e.g.*, [26, 24]). The present work concerns γ-spectroscopic studies of the decays of heavy odd-odd Ag nuclides into their even-even (ee) Cd daughters between $N = 78$ and the $N = 82$ shell closure.

In the current configuration of ISOLDE, the radioactive nuclides are produced through fission of ^{238}U in a thick UC$_2$-C target using the pulsed 1 GeV proton beam from the CERN Proton Synchrotron Booster (PSB) accelerator. The standard experimental setup for the pulsed RILIS at ISOLDE has been described in detail in previous publications [27,28]. In the present experiment, we have used RILIS with new microgating procedures [29]. These rely on the fact that the major fraction of the ionized Ag atoms diffuse out of the ion source shortly after a laser pulse. Using a fast beam gate to select a 20 μs time window, it was possible to suppress unavoidable, surface-ionized isobaric In and Cs background with minimal reduction of the Ag beam current. In the $A \simeq 130$ mass region, In is produced two and Cs five orders of magnitude stronger in ^{238}U-fission than the rare Ag activities sought for [30].

The decay of the radioactive Ag isotopes was investigated after implanting the atoms directly at the measuring position on a movable tape. The γ-ray detection system consisted of three high-resolution (FWHM=1.8 keV at 1.3 MeV) Ge-detectors with efficiencies of 54, 57 and 70%, respectively, positioned at 90, and 180 degrees to each other as well as a plastic ΔE-β-detector behind the source implantation spot. The Ge-detectors were shielded by 5 cm thick Pb cylinders. Singles spectra were taken in consecutive time-bins of 50 ms. Coincidence data were accumulated in event-by-event mode, in which each event included the time with respect to the preceding PSB pulse, the Ge-detector energies, and time-to-amplitude converter readings from the Ge-Ge and Ge-ΔE-β TACs. The tape was moved after each proton pulse to diminish the build-up of longer-lived isobaric In to Cs background at the measuring position.

As an example of the quality of data obtained, fig. 1 shows excerpts from the singles spectra of $A = 126$ (left part) and $A = 128$ (right part) in the energy region 550 keV to 950 keV, taken with one of the three Ge detectors. In both cases, the spectra were accumulated during operation of RILIS at different times after the PSB pulses. The respective upper spectrum was collected during the period from 50 to 200 ms, whereas the lower spectrum was taken during a later time-slice between 800 and 950 ms. Both γ-spectra appear to be rather clean in the energy region shown here, where the $2^+ \to 0^+$ and $4^+ \to 2^+$ transitions in ^{126}Cd and ^{128}Cd are expected according to level systematics. Identification of those γ-lines belonging to the decays of 95-ms ^{126}Ag and 58-ms ^{128}Ag was performed in two ways: i) by comparing spectra taken with ("laser-on") and without ("laser-off") RILIS operation, and ii) by following the decay, respectively growth-and-decay of short-lived activities in the "laser-on" spectra.

For $A = 126$ (upper part of fig. 1), peaks at 652, 827 and 857 keV can be seen to be present only in the early spectrum and are attributed to ^{126}Ag decay. Intensity and γ–γ coincidence arguments suggest the line at 652 keV to represent the $2^+ \to 0^+$ transition in ^{126}Cd, and the line at 815 keV to be the $4^+ \to 2^+$ transition. An assignment as a second 2^+ level can be excluded since no γ-line at 1467 keV was observed, which would have been the cross-over ground-state (g.s.) transition of this 2^+ state. The pronounced peaks 909 and 926 keV, which show up in both, the early and the late spectrum, arise from the decay of longer-lived In isobars. Altogether, at least 8 γ-lines were assigned to ^{126}Ag decay, which could be placed in a (partial) level scheme containing levels up to 3.1 MeV with tentative spins up to 10^+. These data will be presented in detail in a forthcoming publication.

For $A = 128$ (lower part of fig. 1), only two relatively weak peaks at 645 and 784 keV were identified in the early spectrum, which could be assigned to the $2^+ \to 0^+$ and $4^+ \to 2^+$ transitions in ^{128}Cd due to their decay behavior. Again, a possible 1429 keV g.s.-transition from a hypothetical second 2^+ level could not be observed in the first time-slice. All other γ-lines are seen in both, the early and the late spectrum, and arise from the decay of the longer-lived In and Cs isobars. It is worth mentioning at this point, that those data as shown here are close to the

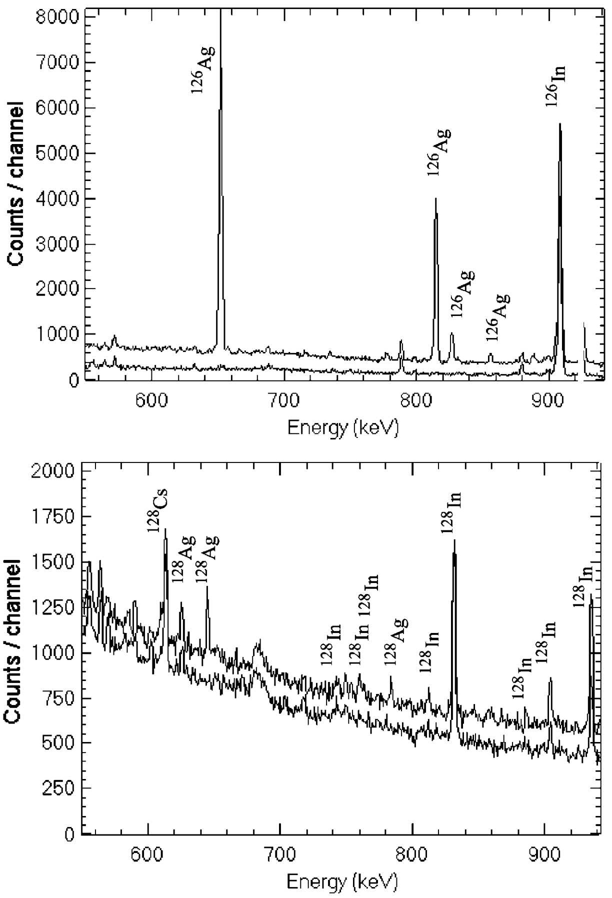

Fig. 1. Excerpts of "laser-on" γ-singles spectra for $A = 126$ (upper part) and $A = 128$ (lower part). For both masses, the upper spectrum has been taken just after closing the beam-gate, whereas the lower spectrum has been taken 750 ms later. The 95-ms ^{126}Ag and 58-ms ^{128}Ag peaks are only seen in the respective early spectrum.

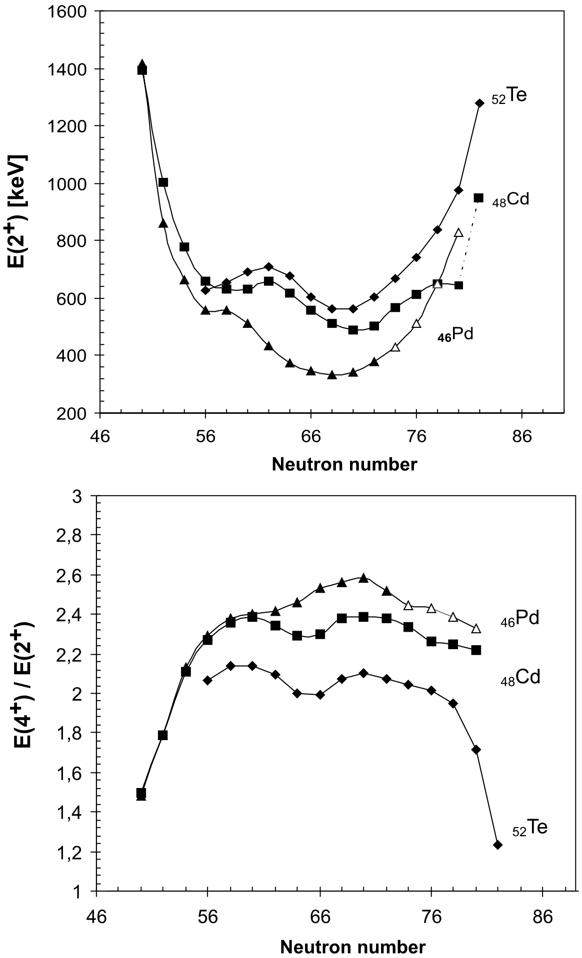

Fig. 2. Level systematics of even-even $_{46}$Pd, $_{48}$Cd and $_{52}$Te isotopes. Upper part: Energies of the first 2^+ states; lower part: ratio of 4^+ to 2^+ energies. The values for ^{120}Pd$_{74}$–^{126}Pd$_{80}$ (open triangles) are from the IBM-2 calculations of [31]. For discussion, see text.

present limit that can be achieved with RILIS and GPS-ISOLDE. They represent the results from about 20000 collection cycles, taken after each PSB pulse, requiring a total beam-time of about 16 hours. However, due to the duty-cycle necessary to minimize the dominating In and Cs isobaric activities, the effective measuring time for the γ-spectrum of the first time-slice shown in fig. 1 was only of the order of 30 min.

Therefore, for $A = 130$ we could only obtain a very first evidence for a few γ-lines that decay with a half-life shorter than the 0.3 s and 0.5 s ^{130}In isomers. One of them at an energy of 957 keV, which is only observed in the first time-bin and decays with an estimated half-life of about 50 ms, is tentatively attributed to ^{130}Ag decay and may represent the $2^+ \rightarrow 0^+$ transition in neutron-magic ^{130}Cd$_{82}$. A second "short-lived" γ-line at 1395 keV ($T_{1/2} \leq 200$ ms), which is seen in the first two spectra, might be the lowest 1^+ level in 165-ms ^{130}Cd decay to ^{130}In. The position of this $[\nu g_{7/2} \pi g_{9/2}]$ two-quasi-particle (2QP) state strongly fed in Gamow-Teller decay of ^{130}Cd is expected around 1.4 MeV from both level systematics and our calculations within the QP Random Phase Approximation (QRPA). In any case, both assignments have to be considered speculative, and will have to be confirmed in a future experiment using RILIS in combination with the High-Resolution Separator (HRS) of ISOLDE.

The above new information on the first 2^+ and 4^+ levels in ^{126}Cd$_{78}$ and ^{128}Cd$_{80}$, together with the indication of the rather low-lying 2^+ in ^{130}Cd$_{82}$, can now be included in the known level systematics in the $Z_{\text{magic}} = 50$ Sn region. The upper part of fig. 2 summarizes the present knowledge of the first 2^+ energies ($E(2^+)$) in the ($Z_{\text{magic}}-2$)=48 Cd isotopes, together with the data of the($Z_{\text{magic}}-4$)=46 Pd and the ($Z_{\text{magic}}+2$)=52 Te isotones as a function of neutron number between the two magic shells $N = 50$ and $N = 82$. The first observation is that the trend of the $E(2^+)$ of the Cd and the Te isotopes is quite similar in the mid-shell region up to 2–3 neutron-pairs outside both closed shells, whereas the $E(2^+)$ of the Pd isotones (experimental data up to $N = 72$, and recent IBM-2 predictions [31] beyond) show a slightly different pattern. Regarding the Cd isotopic chain near both shell closures, it is immediately evident that the proton-rich nuclides below $N = 56$ exhibit a rather steep increase of the 2^+ energies towards $N = 50$ with $E(2^+) = 1395$ keV (very similar to the trend of the light Pd isotopes), whereas the neutron-rich Cd nuclei beyond $N = 74$ exhibit only a moderate increase of the $E(2^+)$ towards $N = 82$ shell closure. The first 2^+ energy of $N = 80$ ^{128}Cd is even slightly lower than that in $N = 78$, then rising by only 300 keV to $E(2^+) = 957$ keV at $N = 82$. In contrast to this trend, the corresponding $E(2^+)$ of the heavy Te isotones and the predicted (IBM-2) values of the Pd isotones show a steady increase of the $E(2^+)$ towards $N = 82$, rather resembling the slopes of the proton-rich Cd and Pd nuclides towards $N = 50$.

As far as the $E(4^+)/E(2^+)$ ratios are concerned (see lower part of fig. 2), the first observation is that for all these three isotopic chains there is a similar trend between neutron numbers $N = 56$ up to $N = 74$, however with different magnitudes of the energy ratios. The ($Z_{\mathrm{magic}}-4$) Pd isotopes have the highest values lying between 2.4 and 2.6; the ($Z_{\mathrm{magic}}-2$) Cd ratios vary between 2.3 and 2.4; and the ($Z_{\mathrm{magic}}+2$) Te isotones lie with about 2.0 to 2.1 lowest. As for the $E(2^+)$, also the $E(4^+)/E(2^+)$ ratios of the heaviest Cd isotopes tend to deviate from the Te ratios, the latter decreasing from about 2.0 at $N = 76$ to 1.7 at $N = 80$, whereas the Cd values remain nearly constant at 2.25.

These similarities and differences in the $_{46}$Pd, $_{48}$Cd and $_{52}$Te isotopic chains are now discussed in terms of $B(E2)$ and β_2 (quadrupole deformation) values which can be deduced from the $E(2^+)$ with the relations $B(E2) \sim E(2^+)^{-1}$ and $\beta_2 \sim B(E2)^{1/2}$ given by Raman *et al.* [32]. Figure 3 shows the experimental and calculated $B(E2)$ values in units of $\mathrm{e^2fm^4}$ for the neutron-rich $_{46}$Pd, $_{48}$Cd, $_{50}$Sn and $_{52}$Te isotones beyond $N = 60$. It is clearly evident from this figure, that compared to the (near-) spherical proton-magic Sn isotopes, all three, the four- and two-proton-hole Pd and Cd isotopes as well as the two-proton-particle Te nuclei, have considerably higher $B(E2)$ values indicating that sizeable collectivity persists close to the $N = 82$ magic shell. Slightly beyond neutron mid-shell (*i.e.* at $N = 68$–70), the isotones of all three elements exhibit the maximum collectivity with $B(E2) \simeq 7100\,\mathrm{e^2\,fm^4}$ for Pd, and $B(E2) \simeq 5500\,\mathrm{e^2\,fm^4}$ for Cd. The values are decreasing towards higher neutron numbers. Significant differences between the Cd and Te isotope chains, where still experimental $E(2^+)$ values exist, begin to show up beyond $N = 74$, where both Cd and Te isotones have roughly the same $B(E2) \simeq 4500\,\mathrm{e^2\,fm^4}$. Whereas the $B(E2)$ values of heavy Te isotopes then continue to decrease to 3500 $\mathrm{e^2\,fm^4}$ at $N = 78$ and 2900 $\mathrm{e^2\,fm^4}$ at $N = 80$, the values of the corresponding four- and two-neutron-hole Cd isotones remain practically constant at $B(E2) \simeq 3900\,\mathrm{e^2\,fm^4}$. With the relation of [32] given above, this $B(E2)$ value converts into a quadrupole deformation parameter $\beta_2 \simeq 0.12$, clearly showing that below the $Z = 50$ magic number, in the Cd isotopes some kind of collectivity persists up to the neutron shell closure. And neutron-magic ^{130}Cd with its tentative $E(2^+) = 957$ keV would in addition be

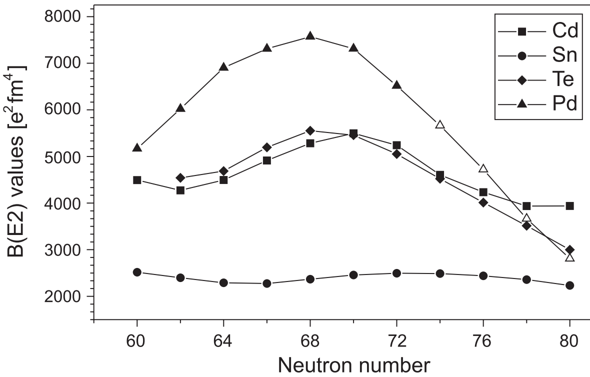

Fig. 3. $B(E2)$ values of neutron-rich $_{48}$Cd, $_{50}$Sn and $_{52}$Te isotopes, deduced from the $E(2^+)$ with the relation given by Raman *et al.* [32].

indicative of a larger quadrupole polarizability in this ($Z_{\mathrm{magic}}-2$) isotope than in its ($Z_{\mathrm{magic}}+2$) neutron-magic isotone ^{134}Te. It will be interesting to see if future $B(E2 : 0^+_{\mathrm{g.s.}} \to 2^+_1)$ measurements via medium-energy Coulomb excitation will support our conclusions.

In any case, none of the recent global models — neither the macroscopic-microscopic nor the self-consistent (deformed) microscopic theories [33–35] — predict such quadrupole collectivity near the $N = 82$ shell closure. They all agree in that for the heavy Pd, Cd and Te isotopes, there is a sudden drop of deformation around $N \simeq 72$ reaching sphericity ($\beta_2 < 0.03$) already at $N \simeq 76$. Hence, based on our new Cd results, we would also predict sizeable polarizability for the heavy ($Z_{\mathrm{magic}} - 4$) Pd isotopes, in clear contrast to the above mass models as well as the extensive series of IBM-2 calculations of Kim *et al.* [31]. In this study, the Tokyo-Köln collaboration has been able to achieve excellent fits to the existing structures of the ee-Pd nuclides from $N = 56$ up through $N = 70$. The authors also report predictions for both the $B(E2)$ values and structures for the heavier Pd nuclides. These predictions include a steady decrease in $B(E2)$ values by a factor of three going from $N = 72$ through $N = 80$ and a corresponding increase in the energy of the first 2^+ state from $\simeq 400$ keV for ^{118}Pd$_{72}$ up to $\simeq 750$ keV for ^{126}Pd$_{80}$.

In a recent RMF calculation of β_2 parameters, Lalazissis *et al.* [36] show values below 0.1, decreasing with increasing neutron number for the $N = 76$, 78 and 80 Pd, Cd and Te nuclides. The trend they predict for the Pd isotopes is quite in line with the energies and $4^+/2^+$ ratios calculated by Kim *et al.* [31]. Hence, the expectation is that the heavy Cd and Pd nuclides will show smoothly increasing 2^+ energies approaching the $N = 82$ closed shell in the same manner as observed for the Te isotopes and for the departure from $N = 50$ shown by Cd and Pd (see fig. 2).

The question whether this effect would be called a shell quenching is not so obvious. That is, does the observed divergence between the structures of the Te and Cd at $N \geq 76$ signal a difference in the underlying shell gap at $N = 82$ for ($Z_{\mathrm{magic}} - 2$) relative to ($Z_{\mathrm{magic}} + 2$), or is it due to a weakening of (ideal) sphericity with a slight onset of quadrupole collectivity below ^{132}Sn? In terms of the $N = 82$ shell gap, at least the expected trend is clear. The difference between the last occupied neutron shell below $N = 82$ (the $\nu d_{3/2}$ according to experiment, or the $\nu h_{11/2}$ as predicted by most theories) and the first occupied orbital above $N = 82$ (the $\nu f_{7/2}$) should shrink with decreasing Z [9,10,2,20]. When using SkI3 or SkI4, this effect is roughly 350 keV per proton-pair removed from $Z = 54$ down to $Z = 44$; however, with SLy4 it is about a factor 4.5 less [20]. Neutron excess should supply an extra lowering of the $E(2^+)$ with decreasing Z. To our knowledge, none of the present self-consistent microscopic models — neither HF or HFB nor RMF or RHB theories — has calculated the energies of low-lying quadrupole states in the Sn region with a full quantal treatment. Also classical shell-model codes seem not to be able to reproduce the new data below and near-beyond ^{132}Sn. Therefore, we acknowledge in this context, that P.-G. Reinhard has provided us with his semi-classical estimates of the $E(2^+)$ energies of neutron-rich Cd, Sn and Te isotopes using different Skyrme parameterizations, such as SkT6, SkI4 with an improved spin-orbit coupling, and a more recent deformed SMF theory with self-consistent cranking [20]. But even these microscopic Skyrme-HF calculations do not produce a satisfactory reproduction of our experimental observations. The two first parameterizations predict the $E(2^+)$ of $N = 76$ to $N = 82$ Cd isotopes to lie above the Te isotones, with smoothly increasing energies between about 2 and 3.5 MeV; whereas the latter Skyrme force does shift the Cd energies below the $E(2^+)$ of the Te isotopes, but still shows too steep an increase at too high excitation energies towards $N = 82$.

In a very recent publication by Korgul *et al.* [37], new OXBASH calculations are presented that are able to fit the observed levels in the $N = 84$ isotones, including the rather low-lying $E(2^+)$ in ^{134}Sn and ^{136}Te. In a previous (unmodified) OXBASH calculation prior to the experimental studies, the 2^+, 4^+ and 6^+ levels of ^{134}Sn were predicted a factor of 1.6 too high [38]. While the new calculations can reproduce the observed levels, that fall at 726, 1045, and 1247 keV, respectively, the fit is accomplished *only* by including an unexplained factor of 0.6 in the crucial, low-energy diagonal matrix elements. Although there is no obvious direct relationship between the anomalous behavior observed in ^{134}Sn$_{84}$ and the departure of the 2^+ energies in $N < 82$ Cd isotopes from the expected trends, both of these experimental observations serve to highlight the difficulties in using theoretical models developed near the valley of stability to describe nuclear properties in increasingly neutron-rich nuclides.

With regard to the possible mechanisms for the quenching of the $N = 82$ closed shell at $Z < 50$, we note the discussion presented by Walters [39]. He shows the existence of large monopole shifts in the positions of the $\nu p_{3/2}$ and $\nu p_{1/2}$ levels, both for the $N = 82$ to $N = 126$ region as well as for the $N = 20$ to $N = 50$ region. In particular, the $\nu p_{1/2}$ orbital lies at the top of the $N = 126$ closed shell for ^{207}Pb and the $\nu h_{9/2}$ orbital is 3.4 MeV deep in this nucleus. In ^{133}Sn, however, they are nearly degenerate at 1656 keV and 1561 keV, respectively. Continuation of this trend could drive the $\nu p_{3/2}$ and $\nu p_{1/2}$ orbitals even below the $\nu f_{7/2}$ shell, thus resulting in a narrowing of the $N = 82$ gap.

Given this still inconclusive situation, one may ask whether there exist other experimental indications for a weakening of the $N = 82$ shell strength in the ^{132}Sn region. And there are, indeed, several signatures, which may each by itself not be convincing, but in their cumulative effect seem to strengthen the shell quenching picture. The change of the ordering of low-j and high-j neutron SP states beyond $N = 82$ and in particular the surprisingly low-lying $\nu p_{3/2}$ and $\nu p_{1/2}$ states in ^{133}Sn [40,23] are consistent with the prediction of a reduction of the l^2-term in the Nilsson Hamiltonian for quenched shells [9,10]. Recent mass measurements around ^{132}Sn [41] appear to be best reproduced with mass models that contain quenched neutron shell gaps (see, *e.g.*, fig. 4 in [23]). Furthermore, our recent QRPA calculations to reproduce half-life and delayed neutron emission data of $N \simeq 82$ $_{47}$Ag and $_{48}$Cd decays into their β-decay daughter nuclei seem to require a reduced $N = 82$ shell gap with reduced pairing, and in adition a certain “hindrance” of the GT SP-model strength [24,42]. Finally, as already mentioned above, also the low-lying $E(2^+)$ in ^{134}Sn as well as the extended set of excited states in $N = 84$ ee-nuclei from the very recent paper of [37] indicate reduced pairing beyond ^{132}Sn.

Summarizing, we have presented γ-spectroscopic information on new levels in very neutron-rich ee-Cd isotopes which extend the systematics of the first 2^+ and 4^+ states up to the $N = 82$ shell closure. We suggest that our results may be interpreted as one of the first signatures of a weakening of the $N = 82$ shell structure already one proton-pair below ^{132}Sn. However, further experimental as well as theoretical studies are definitely needed, in particular when considering the astrophysical consequences ranging from a precise knowledge of the r-process matter flow through the $A \simeq 130$ $N_{r,\odot}$ peak up to reliable predictions of the Th-U-Pu r-process chronometers to determine the age of our Galaxy (see, *e.g.*, [13,43,44]).

We would like to thank in particular P.-G. Reinhard for providing us with his unpublished results on SHF calculations in the ^{132}Sn region, and we appreciate helpful discussions with him as well as with A. Brown, J. Dobaczewski, K. Heyde and J. Rikovska Stone. Furthermore, support from various national foundations, in particular the German BMBF (06MZ864) and DFG (463RUS17/40) and the U.S. Department of Energy is gratefully acknowledged.

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