Schottky mass measurement of ^{208}Hg isotope: implication for the proton-neutron interaction strength around doubly-magic ^{208}Pb.

L. Chen,[1,2] Yu. A. Litvinov,[1,*] F. Bosch,[1] R. B. Cakirli,[3,4] R. F. Casten,[4,5] H. Geissel,[1,2] C. Kozhuharov,[1] W. R. Plaß,[1,2] C. Scheidenberger,[1,2] P. M. Walker,[6] H. Weick,[1] K. Beckert,[1] P. Beller,[1] D. Boutin,[2] L. Caceres,[1] J. J. Carroll,[7] R. S. Chakrawarthy,[8] D. M. Cullen,[9] I. J. Cullen,[6] B. Franzke,[1] J. Gerl,[1] M. Gorska,[1] G. A. Jones,[6] A. Kishada,[9] R. Knöbel,[1] S. A. Litvinov,[1] Z. Liu,[6] S. Mandal,[1] F. Montes,[10] G. Münzenberg,[1] F. Nolden,[1] T. Ohtsubo,[11] Z. Patyk,[12] Zs. Podolyák,[6] R. Propri,[7] S. Rigby,[9] N. Saito,[1] T. Saito,[1] M. Shindo,[13] M. Steck,[1] P. Ugorowski,[7] S. Williams,[6] M. Winkler,[1] H.-J. Wollersheim,[1] and T. Yamaguchi[14]

[1] GSI Helmholtzzentrum für Schwerionenforschung, Planckstraße 1, 64291 Darmstadt, Germany
[2] Justus Liebig Universität Giessen, Heinrich-Buff-Ring 16, 35392 Giessen, Germany
[3] Department of Physics, University of Istanbul, Istanbul, Turkey
[4] Institut für Kernphysik, Universität zu Köln, 50937 Köln, Germany
[5] Wright Nuclear Structure Laboratory, Yale University, New Haven, CO 06520-8124, U.S.A.
[6] Department of Physics, University of Surrey, Guildford, GU2 7XH, U.K.
[7] Youngstown State University, One University Plaza, Youngstown, OH 44555, U.S.A.
[8] TRIUMF, 4004 Wesbrook Mall, Vancouver, V6T 2A3, Canada
[9] University of Manchester, Oxford Road, Manchester, M13 9PL, U.K.
[10] Michigan State University, East Lansing, MI 48824, U.S.A.
[11] Niigata University, Niigata 950-2181, Japan
[12] Soltan Institute for Nuclear Studies, Hoża 69, 00-681 Warszawa, Poland
[13] University of Tokyo, Tokyo 113-0033, Japan
[14] Saitama University, Saitama 338-8570, Japan

Time-resolved Schottky mass spectrometry has been applied to uranium projectile fragments which yielded the mass value for the ^{208}Hg ($Z = 80$, $N = 128$) isotope. The mass excess value of ME=-13265(31) keV has been obtained, which has been used to determine the proton-neutron interaction strength in ^{210}Pb, as a double difference of atomic masses. The results show a dramatic variation of the strength for lead isotopes when crossing the N=126 neutron shell closure, thus confirming the empirical predictions that this interaction strength is sensitive to the overlap of the wave-functions of the last valence neutrons and protons.

Atomic nuclei are many-body systems in which the strong, weak and electromagnetic fundamental interactions play a major role by acting between the nucleons. The sum effect of these interactions is reflected in the total binding energy of the nucleus, which is directly connected with its mass [1]. Therefore, nuclear masses often provide hints to new nuclear structure effects. Indeed, shell structure and pairing correlations have been discovered through nuclear masses. Nuclear masses and binding energies are important, however, in a much wider domain. Examples occur in the studies of various nucleosynthesis processes in stars, in weak interaction physics relating to the unitarity of the Cabibbo-Kobayashi-Maskawa matrix, in tests of QED and in the determination of fundamental constants [2].

Dedicated filters can be constructed from mass differences to isolate specific nucleonic interactions. One such filter is the average interaction strength of the last proton(s) with the last neutron(s) denoted δV_{pn}. The proton-neutron interaction–being of fundamental interest for nuclear structure–has been intensively discussed in the last decades [3–14]. It has been shown that it is essential for the development of configuration mixing and for the onset of collectivity and deformation in nuclei [6], for changes of the underlying shell structure [7], and for the microscopic origins of phase transitional behavior in nuclei [7–9]. Being the largest along the N=Z line, the $p-n$ interaction can be related to Wigner's SU(4) symmetry [3]. The average value of the $p-n$ interaction strength can also be related to the nuclear symmetry energy [10]. Moreover, treatment of the nucleon-nucleon correlations finds similarities in interpreting other many-body systems: for example, odd-even staggering effects were observed in ultrasmall superconducting metallic grains [15]. This Letter will present a mass measurement for ^{208}Hg isotope, that provides one of the most important δV_{pn} values in the entire nuclear chart.

For even-even nuclei, the average $p - n$ interaction of the last two protons with the last two neutrons can be defined as [13]:

$$\delta V_{pn}(Z,N) = \frac{1}{4}[B(Z,N) + B(Z-2,N-2) \\ -B(Z,N-2) - B(Z-2,N)], \qquad (1)$$

where B is the binding energy of the nucleus. By assuming that the nuclear core remains essentially unchanged, δV_{pn}, by its definition, largely cancels the interactions of the last nucleons with the core. A given δV_{pn} value for an even-even nucleus refers to the average interaction of the $(Z-1)^{th}$ and Z^{th} valence protons with the $(N-1)^{th}$ and N^{th} neutrons.

The δV_{pn} values around the doubly-magic ^{208}Pb nucleus are a subject of extensive discussion in the last few years as, e.g., presented in Refs. [4, 5, 10, 14]. The $p-n$ interactions in this region have a characteristic and striking behavior that can be understood [4] in terms of the spatial overlaps of the last valence nucleons. Since the residual $p-n$ interaction is primarily short range and attractive, it should be strongest when the protons and neutrons are in orbits with greatest spatial overlap. The normal parity orbits in the shells of heavy nuclei have a generic sequence starting with high j (particle angular momentum), low n (principal quantum number) orbits at the beginning of a shell to low j, high n orbits at the end. For example, in the Pb region, the proton orbits just below Z = 82 are the $3s_{1/2}$ and $2d_{3/2}$ and the neutron orbits below N=126 are $3p_{3/2}$ and $3p_{1/2}$. Above Z=82 and N=126, the orbits for protons are $1h_{9/2}$ and $2f_{7/2}$ while, for neutrons, they are $2g_{9/2}$ and $1i_{11/2}$. Taking as a crude guide that the $p-n$ interaction scales inversely as $(\Delta l + \Delta n)$, it is clear that one expects large $p-n$ interactions when both protons and neutrons are below or both are above their respective shell closures, while one expects small interactions when one is above and the other below. This leads to an expected "crossing" pattern, which we will indeed see later. However, there is one major gap in the data for δV_{pn} in the Pb region. The $p-n$ interaction is only known for three of the four possible cases–there are no known values for protons filling just below Z=82 and neutrons just above N=126. The first point in this region is $\delta V_{pn}(^{210}\text{Pb})$. Three of the four masses needed for δV_{pn} in Eq. (1) are known. Only that for ^{208}Hg is not and the measurement of this mass is therefore of the utmost importance in understanding the $p-n$ interaction, its orbit dependence, and therefore its role in the evolution of nuclear structure.

The purpose of this Letter is to present the first direct mass measurement of the ^{208}Hg nucleus. This is critical as a test of our basic understanding of the dependence of the proton-neutron interaction on the spatial relations of the nucleonic orbitals, and therefore on the single particle levels in this region. It carries the added importance of relating directly to the quantum stabilization of nuclei beyond Pb which would otherwise be unbound due to the strong accumulation of repulsive Coulomb interactions. We will compare the empirical results with expectations based on the ideas outlined above. These results will also be a challenge to microscopic models of the structure of heavy nuclei such as Density Functional Theory [10].

The experiment has been performed at the GSI Helmholtzzentrum für Schwerionenforschung in Darmstadt, Germany, where the combination of the heavy-ion synchrotron SIS [16], the in-flight fragment separator FRS [17], and the ion storage-cooler ring ESR [18] provides unique experimental conditions for nuclear structure studies of exotic nuclei stored in an ultrahigh vacuum ($\sim 10^{-11}$ mbar) [19, 20]. The mass of the ^{208}Hg nucleus has been measured with time-resolved Schottky Mass Spectrometry [21, 22] as part of a program on direct mass measurements of neutron-rich uranium projectile fragments in which the masses for about 30 neutron-rich nuclides have been obtained for the first time [23].

The primary beam of ^{238}U projectiles accelerated by the SIS to an energy of 670 MeV/u. The intensity of the primary beam was about $2 \cdot 10^9$ particles per spill. The exotic nuclides were produced via the projectile fragmentation in a 4 g/cm^2 ^{9}Be production target placed in front of the FRS facility. The primary beam and the produced fragments emerge from the target as highly-charged ions with no or very few bound electrons. The neutron-rich fragments were separated in flight and injected into the storage ring ESR. The injection into the ESR was optimized with the primary beam and the magnetic fields of the FRS-ESR facilities were fixed at the constant magnetic rigidity of 7.9 Tm throughout the experiment. In the ESR the fragments were stored and electron cooled. The time required for the electron cooling restricts the nuclides that can be accessed with the SMS. For low intensity fragment beams this time is of the order of a few seconds [24].

The electron cooling process compresses the phase-space volume of stored beams and the initial longitudinal velocity spread is reduced to typically $\Delta v/v \approx 5 \times 10^{-7}$. By selecting the cooler voltage we define the velocity of the merged electrons and thus the velocity of the cooled ions. In order to cover the range of stored fragments from neodymium to uranium, the cooler voltages were varied from 190 kV up to 220 kV in steps of 2 kV.

The stored ions circulate in the ESR with frequencies of about 2 MHz. The frequencies were measured by means of time-resolved SMS [21, 22]. Stored highly-charged ions induce at each revolution signals on the capacitive pick-up plates which are installed inside the ring aperture. The frequency spectrum at 30^{th} harmonics of the revolution frequencies of the ions has been analyzed by means of the fast Fourier transform which yields Schottky frequency spectra. The frequency bandwidth of 320 kHz was simultaneously measured, which is sufficient to cover the entire frequency acceptance of the ESR. From each injection of the ions into the ESR several revolution spectra accumulated over about 20 seconds have been obtained. The data acquisition and the data analysis are similar to the ones described in detail in Ref. [22].

The revolution frequencies of the cooled stored ions f are related to their mass-over-charge ratios m/q by the following expression:

$$\frac{\Delta f}{f} = -\alpha_p \frac{\Delta(m/q)}{(m/q)}, \qquad (2)$$

where $\alpha_p = (\Delta C/C)/(\Delta B\rho/B\rho)$ is the momentum compaction factor of the ring which characterizes the relative variation of the orbit length C per relative variation of the magnetic rigidity $B\rho$. The α_p is nearly con-

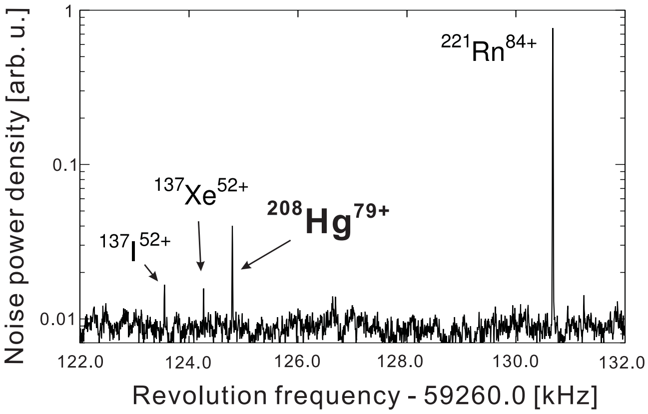

FIG. 1: A 10 kHz part of the measured revolution frequency spectrum. For this spectrum the electron cooler voltage and current were set to U_c=198 kV and I_c=400 mA, respectively. A peak of hydrogen-like ^{208}Hg^{79+} ions is at about 125 kHz. The masses of ^{137}I^{52+}, ^{137}Xe^{52+} and ^{221}Rn^{84+} ions are experimentally known [30] and can be used for the calibration.

stant over the entire ring acceptance and in this experiment it was approximately 0.19. The intensities of the frequency peaks are proportional to the corresponding number of stored ions, which is often used for half-life measurements [25–27].

The unambiguous isotope identification of frequency peaks is based on a pattern recognition algorithm and is discussed in detail in Ref. [28]. Several tens of nuclides are simultaneously present in the measured spectra. The production cross-section of ^{208}Hg in this reaction has been estimated with EPAX2 formula [29] and amounts to only 1.5 μbarn. The ^{208}Hg nuclide has been observed only in one injection into the ESR where it was present in a hydrogen-like charge state. A part of the revolution frequency spectrum with ^{208}Hg^{79+} ions is illustrated in Fig. 1. The peaks of ions with experimentally known masses [30] are used to calibrate the frequency spectrum. In this case the frequency peaks of ^{137}I^{52+}, ^{137}Xe^{52+} and ^{221}Rn^{84+} ions can be used for this purpose.

Several tens of injections of the ions into the ESR have been analyzed for each setting of the electron cooler, i.e. cooler voltage U_c and current I_c. The time-resolved SMS enables the possibility to correct for overall frequency drifts in time due to instabilities of, for example, magnet power supplies. Therefore, the frequencies from all measured spectra of the same cooler setting can be combined together [31]. The advantage of this procedure is that already within the 10 kHz spectrum illustrated in Fig. 1 the additional frequency peaks of ^{129}Sn^{49+}, ^{200m}Au^{76+}, ^{208}Tl^{79+}, ^{208}Pb^{79+}, ^{221}At^{84+}, ^{229}Fr^{87+}, ^{229}Ra^{87+}, and ^{229}Ac^{87+} ions which lie close to the peak of ^{208}Hg^{79+} ions can be used for the calibration. The mass excess value of ^{208}Hg has been determined and amounts to $ME = -13265(31)$ keV. The details of the data fitting and error propagation can be found in Ref. [23]. The obtained ME value agrees within the error bars with the value from the systematics -13100(300) keV [30], where the new mass value is 165 keV more bound.

The new precisely measured mass value of ^{208}Hg has been used to determine the δV_{pn} value for ^{210}Pb using Eq. (1). This value and those for other nuclei in the ^{208}Pb region are shown in Figs. 2 and 3. Figure 2 shows δV_{pn} values for several isotopic sequences as a function of neutron number. Figure 3 shows the same results in a complementary way in the form of a color-coded plot for this portion of the nuclear chart. Here the vertical and horizontal lines at $N = 126$ and $Z = 82$ mark the double shell closure at ^{208}Pb and divide this region into four quadrants that can be labeled hole-hole (lower left where both types of nucleons are below their respective closed shells), particle-hole (upper left), particle-particle (upper right), and hole-particle (lower right).

Using the present mass measurement of ^{208}Hg, the $\delta V_{pn}(^{210}$Pb) point is the first δV_{pn} value for the region $Z \leq 82$, $N > 126$, that is, the lower-right quadrant in Fig 3. The δV_{pn} value of ^{210}Pb is 165.2(10) keV which is about 2.5 times smaller than the δV_{pn} (^{208}Pb) value of 426.8(5) keV. This sudden decrease is signified by the blue box (labelled also with "2") just after the junction of the shell closure lines.

The $^{208}_{82}$Pb$_{126}$ nucleus is in the symmetric hole-hole region, where both protons and neutrons fill low j high n orbits below the closed shell, and therefore it should (and does) have a very large δV_{pn} value. In contrast, ^{210}Pb has two extra valence neutrons which occupy orbits just above the 126 closed shell, giving an asymmetric particle-hole case where one expects a low δV_{pn} value. The observed drop is such that, as seen in Fig. 2, the δV_{pn} value for ^{210}Pb is comparable to those in the other asym-

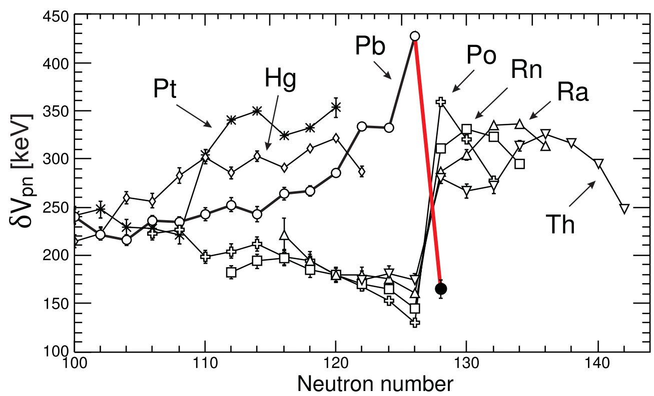

FIG. 2: (Color online) Experimentally known δV_{pn} values for isotopic chains below and above the $Z = 82$ proton closed shell. Only even-even nuclei are considered. If not shown the error bars are within the symbol size. The newly obtained δV_{pn} value for ^{210}Pb is emphasized by a filled symbol and the sharp drop from ^{208}Pb to ^{210}Pb is highlighted.

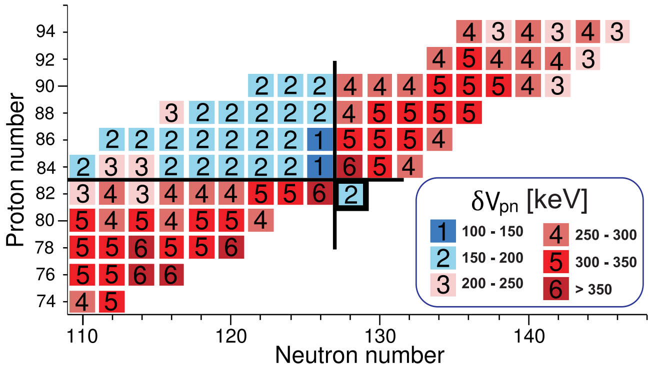

FIG. 3: (Color online) Experimentally known δV_{pn} values on the chart of nuclides. Only even-even nuclei are considered. Four quadrants are defined by the $Z = 82$ proton and $N = 126$ neutron shell closures. The diagonal symmetry in the δV_{pn} values for lower-left and upper-right quadrants is clearly seen and can be explained by the "symmetry" in the quantum numbers of the single-particle orbitals occupied by the valence nucleons (see text). The δV_{pn} value for ^{210}Pb (indicated with black square) is the first experimental result in the lower-right quadrant. It has a similar magnitude as the δV_{pn} values in the upper-left quadrant.

metric group (upper left quadrant of Fig. 3 with $Z > 82$, $N \leq 126$). This drop vividly validates the sensitivity of the $p - n$ interaction to spatial overlaps of orbits in a crucial region and completes the evidence for a crossing pattern across closed shells. Furthermore, it shows that such measurements can provide indirect information on the underlying single particle level structure.

In summary, we have presented the first measurement of the mass of ^{208}Hg which allows the extraction of the empirical $p - n$ interaction of the last nucleons in ^{210}Pb. This is the first value known in the lower right quadrant of the nuclear chart surrounding the ^{208}Pb doubly magic shell closure. As such the new result confirms the expectations based on the spatial overlap of the valence orbits in this region and their relation to very general properties of shell structure near stability. In turn this methodology can be used to study the possible quenching of the known shells and to search for new shell closures in nuclei far from stability where the generic sequencing of orbits within major shells may be significantly different than near stability.

The authors are grateful to K. Blaum, D. S. Brenner, K. D. Groß, O. Klepper, W. Nazarewicz, Yu. N. Novikov, T. Radon and Th. Stöhlker for fruitful discussions. This work is supported in part by the German DFG through a Mercator Guest Professorship at the University of Köln (Ko. 142/112-1), the U.S. Department of Energy under Grant DE-FG02-91ER-40609, the Turkish Atomic Energy Authority (TAEK) Grant OUK120100-4, and the Association of Helmholtz Research Centers under Contract VH-NG-033. Support from Brookhaven Technology Group, U.S.A., and STFC and AWE plc, U.K., is greatly acknowledged.

* Electronic address: `y.litvinov@gsi.de`

[1] A. Bohr and B. R. Mottelson, *Nuclear structure* (World Scientific, Singapore, 1998).
[2] K. Blaum, Phys. Rep. **425**, 1 (2006).
[3] P. Van Isacker, D. D. Warner, and D. S. Brenner, Phys. Rev. Lett. **74**, 4607 (1995).
[4] D. S. Brenner, R. B. Cakirli, and R. F. Casten, Phys. Rev. **C 73**, 034315 (2006).
[5] R. B. Cakirli, D. S. Brenner, R. F. Casten, and E. A. Millman, Phys. Rev. Lett. **94**, 092501 (2005).
[6] I. Talmi, Rev. Mod. Phys. **34**, 704 (1962).
[7] K. Heyde, P. Van Isacker, R. F. Casten, and J. L. Wood, Phys. Lett. **B 155**, 303 (1985).
[8] P. Federman and S. Pittel, Phys. Lett. **B 69**, 385 (1977).
[9] P. Federman and S. Pittel, Phys. Lett. **B 77**, 29 (1978).
[10] M. Stoitsov, R. B. Cakirli, R. F. Casten, W. Nazarewicz, and W. Satula, Phys. Rev. Lett. **98**, 132502 (2007).
[11] J. Dobaczewski, W. Nazarewicz, J. Skalski, and T. Werner, Phys. Rev. Lett. **60**, 2254 (1988).
[12] Y. Oktem, R. B. Cakirli, R. F. Casten, R. J. Casperson, and D. S. Brenner, Phys. Rev. **C74**, 027304 (2006).
[13] J. Y. Zhang, R. F. Casten, and D. S. Brenner, Phys. Lett. **B 227**, 1 (1989).
[14] R. B. Cakirli and R. F. Casten, Phys. Rev. Lett. **96**, 132501 (2006).
[15] F. Braun, J. von Delft, D. C. Ralph, and M. Tinkham, Phys. Rev. Lett. **79**, 921 (1997).
[16] K. Blasche, D. Böhne, B. Franzke, and H. Prange, IEEE Trans. on Nucl. Sci. **Ns-32**, 2657 (1985).
[17] H. Geissel et al., Nucl. Instr. Meth. **B 70**, 286 (1992).
[18] B. Franzke, Nucl. Instr. Meth. **B24/25**, 18 (1987).
[19] F. Bosch et al., Int. J. Mass Spectr. **251**, 212 (2006).
[20] B. Franzke, H. Geissel, and G. Münzenberg, Mass Spectr. Rev. **27**, 428 (2008).
[21] Yu. A. Litvinov et al., Nucl. Phys. A **734**, 473 (2004).
[22] Yu. A. Litvinov et al., Nucl. Phys. **A 756**, 3 (2005).
[23] L. Chen, Ph. D. thesis, Universität Giessen (2008).
[24] Yu. A. Litvinov et al., Hyp. Int. **132**, 281 (2001).
[25] Yu. A. Litvinov et al., Phys. Lett. B **573**, 80 (2003).
[26] T. Ohtsubo et al., Phys. Rev. Lett. **95**, 52501 (2005).
[27] Yu. A. Litvinov et al., Phys. Rev. Lett. **99**, 262501 (2007).
[28] T. Radon et al., Nucl. Phys. A **677**, 75 (2000).
[29] K. Sümmerer and B. Blank, Phys. Rev. C **61**, 34607 (2000).
[30] A. H. Wapstra, G. Audi, and C. Thibault, Nucl. Phys. A **729**, 129 (2003).
[31] H. Geissel et al., Nucl. Phys. A **685**, 115c (2001).