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

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Time-resolved Schottky mass spectrometry has been applied to uranium projectile fragments which yielded the mass value for the $^{208}\text{Hg}$ (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 $^{208}\text{Pb}$, 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 microscopic origins of phase transitional behavior in nuclei [7–9]. Being the largest along the N=Z line, the $\delta V_{pn}$ 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 $^{208}\text{Hg}$ isotope, that provides one of the most important δVpn 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]:

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

(1)

where $B$ is the binding energy of the nucleus. By assuming that the nuclear core remains essentially unchanged, $\delta V_{pn}$, by its definition, largely cancels the interactions of the last nucleons with the core. A given $\delta V_{pn}$ 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 $\delta V_{pn}$ 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 $\delta V_{pn}$ 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 $\delta V_{pn}$ in Eq. (1) are known. Only that for $^{208}\text{Hg}$ 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 $^{208}\text{Hg}$ 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 $^{208}\text{Hg}$ 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 $^{238}\text{U}$ 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 $4 \text{g/cm}^2$ $^{11}$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 pickup plates which are installed inside the ring aperture. The frequency spectrum at $30^{\text{th}}$ 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:

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

where $\alpha_p = (\Delta C/C)/(\Delta B_p/B_p)$ 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 $B_p$. The $\alpha_p$ is nearly con-
The mass excess $ME$ of the 229Tl isotope can be used for the calibration. The additional frequency peaks of 129I, 137Xe, and 221Rn ions are experimentally known [30] and can be used for the calibration. 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 208Hg 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 208Hg 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 $^{137}Xe^{52+}$, $^{137}Xe^{52+}$ 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 129Sn$^{49+}$, 200Ta$^{76+}$, 208Pb$^{79+}$, 208Pb$^{79+}$, 221Rn$^{84+}$, 229Pt$^{87+}$, 229Ra$^{87+}$, and 229Ac$^{87+}$ ions which lie close to the peak of 208Hg ions can be used for the calibration. The mass excess value of 208Hg 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 208Hg has been used to determine the $\delta V_{pn}$ value for 210Pb using Eq. (1). This value and those for other nuclei in the 208Pb region are shown in Figs. 2 and 3. Figure 2 shows $\delta V_{pn}$ 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 208Pb 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 208Hg, the $\delta V_{pn}(^{210}Pb)$ point is the first $\delta V_{pn}$ value for the region $Z \leq 82, N > 126$, that is, the lower-right quadrant in Fig 3. The $\delta V_{pn}$ value of 210Pb is 165.2(10) keV which is about 2.5 times smaller than the $\delta V_{pn}(^{208}Pb)$ 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 $^{208}Pb_{126}$ 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 $\delta V_{pn}$ value. In contrast, 210Pb 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 $\delta V_{pn}$ value. The observed drop is such that, as seen in Fig. 2, the $\delta V_{pn}$ value for 210Pb is comparable to those in the other asym-
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.

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