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Study of the average charge states of ¹⁸⁸Pb and ^{252,254}No ions at the gas-filled separator TASCA

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Abstract

The average charge states of ¹⁸⁸Pb and ^{252,254}No ions in the dilute helium gas were measured at the gas-filled separator TASCA. Hydrogen gas was also used as a filling gas for measurements of the average charge states of ²⁵⁴No. Helium and hydrogen gases at pressures from 0.2 mbar to 2.0 mbar were used. A strong dependence of the average charge states on the pressure of the filling gases was observed for both, helium and hydrogen. The influence of this dependence, classically attributed to the so-called "density effect", on the performance of TASCA was investigated. The average charge states of ^{252,254}No ions were also measured in mixtures of helium and hydrogen gases at low gas pressures around 1.0 mbar. From the experimental results simple expressions for the prediction of the average charge states of heavy ions moving in rarefied helium gas, hydrogen gas, and in their mixture are derived.

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Key words: Gas-filled separator, Average charges, Heavy ion, TASCA

1. Introduction

The knowledge on the charge state distribution of heavy ions 21 moving in gas-filled regions is an important issue of heavy-ion 22 3 beam related physics. This distribution depends on charge- 23 exchange collisions between the heavy ions and atoms of the 24 gas. Already decades ago, many experimental and theoretical 25 investigations have been performed on charge-exchange colli- 26 sions for light and medium heavy ions in rarefied gases [1]. As 27 results of these works many basic aspects of charge-exchange 28 collisions have been established. Recently, a renewed interest in 29 10 the understanding of average charge states of heavy ions in rar- 30 11 efied gases has been motivated by the production of heavy and 31 12 superheavy elements at gas-filled separators [2, 3, 4, 5, 6, 7, 8]. 32 13 The separation of different ions in gas-filled separators is 33 14 based on their different magnetic rigidities $B\rho = m\nu/qe$. B de- 34 15 notes the magnetic flux density, ρ the radius of curvature of the 35 16 ion trajectory, *m* and v the mass and velocity of the ion, respec- 36 17 tively, q its average (ionic) charge state and e is the elementary $_{37}$ 18

charge. The well prediction of the average charge states of the different heavy ions in the filling gas of the separator allows to define the further trajectories of them through the separator.

Several experimental studies of the average charge states of heavy ions have been performed at gas-filled separators to lay the basis for the correct prediction of average charge states of heavy and superheavy ions produced in fusion-evaporation reactions [9, 10, 11, 12, 13, 14]. Semi-empirical expressions were derived, based on a parameterization of the experimental data according to the theoretical underpinnings of charge-exchange collisions, such as an influence of the atomic shell structure of the heavy ions [9, 10, 11, 12, 13, 14]. These investigations were often performed at different pressures of the different facilities, which were, however fixed at a value which was evaluated to be optimum for the respective facility. The influence of a variation of the gas pressure has not been included in any of the above mentioned expressions.

A dependence of the average charge states on the gas pressure has been observed at the Dubna gas-filled recoil separator (DGFRS) [12]. As a possible explanation, a so called "density effect" was invoked, as it is well known from charge-exchange collisions [1]. However, an influence of this effect on the performance of gas-filled separators has not been investigated so far.

At present, gas-filled separators are typically filled with ei-

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ther pure helium (He) or pure hydrogen (H₂). Hydrogen seems 93 44 to provide better suppression of background originating from 94 45 target-like ions [12, 15]. However, as the average charge states 95 46 in pure H₂ are lower than in pure He, H₂-filled separators ne- 96 47

cessitate a stronger dipole magnet to bend the more rigid evap-48

oration residues (ERs). The use of a mixture of these two gases 97 49

allows combining the advantages of both gases, i.e., to retain a 50 good background suppression while keeping rather high aver- 98

51 age charge states of ERs. However, no data fig1nor formalism 99 52 to predict the average charge states of heavy ions in gas mix-100 53 tures exists. 54

Our present experimental work aimed at studying the aver-102 55 age charge states of heavy ions, more specifically ¹⁸⁸Pb and¹⁰³ 56 ^{252,254}No, in various rarefied gases and their mixtures at the¹⁰⁴ 57 gas-filled TransActinide Separator and Chemistry Apparatus¹⁰⁵ 58 (TASCA) [7, 16, 17, 18, 19] to investigate the influence of the¹⁰⁶ 59 "density effect" on ions of different atomic number, mass and 107 velocity. The measured data provide a basis for the prediction¹⁰⁸ 61 of average charge states in gas mixtures. 109 62

2. General aspects of charge-exchange collisions 63

The charge state distribution of heavy ions passing through¹¹³ 64 a rarefied gas can be described in terms of the fractions of the¹ 65 heavy ions $F_i(x)$ in charge state *i*, where *x* refers to the number¹¹⁵ 66 of gas atoms or molecules per square centimeter traversed by¹¹⁶ 67 the heavy ion $(\sum_{i} F_i(x) = 1)$. The variation of the charge state¹¹⁷ 118 fractions is described by a system of differential equations

$$\int \frac{dF_i(x)}{dx} = \sum_{j,j\neq i} \left[\sigma_{ij} \cdot F_j(x) - \sigma_{ji} \cdot F_i(x) \right] \tag{1}$$

123 where, σ_{ij} and σ_{ji} are the cross-sections for electron capture 71 and loss processes [1]. At large values of x the variation of $\frac{1}{125}$ 72 the fractions decreases and may vanish completely, $\frac{dF_i(x)}{dx} \rightarrow 0._{126}^{125}$ 73 This means that the electron capture and electron loss processes 74 between the heavy ions and gas atoms compensate each other 75 and the fraction of each *i*-th charge state (hereafter: F_i) in the 76 heavy ions will not change anymore. This situation gives the 77 distributions of equilibrated charge states (average charge state) 78 of heavy ions. From this point of view the average charge state 79 is determined as: 80

$$\bar{q} = \sum_{i} q_i \cdot F_i \tag{2}^{133}$$

In principle, equations (1) and (2) allow to calculate the av-135 82 erage charge states of heavy ions as well as average charge136 83 state distribution. However, the knowledge on the electron cap-137 84 ture and loss cross-sections of heavy ions in rarefied gases is138 85 still scarce. First attempts to theoretically describe the aver-139 86 age charge state of heavy ions moving in a rarefied gas were₁₄₀ 87 performed in 1940 by Bohr [20] and Lamb [21] in studies of₁₄₁ 88 fission fragments. 142 89

Bohr assumed that a heavy atom moving rapidly through a143 rarefied gas retains all of its electrons that have orbital veloc-144 91 ities exceeding that of the velocity of the atom relative to the145 92

medium. Applying the Thomas-Fermi model for the structure of the atom, he then obtained the well-known dependence of the average charge state \bar{q} of a heavy ion with atomic number Z on its velocity v.

$$\bar{q} = (v/v_0) \cdot Z^{1/3}$$
 (3)

for a velocity range 1 < (ν/ν_0) < $Z^{2/3}$. Here, $v_0 = 2.193 \cdot 10^6$ m/s is the velocity of the electron in Bohr's model of the hydrogen atom.

In the work of Lamb the average charge states of fission fragments have been calculated using energy considerations. He assumed that a fragment moving through a rarefied gas with a velocity v "will be stripped down until the ionization potential of the next stage of ionization is greater than the kinetic energy of electrons bombarding the fragment with a velocity v" [21].

In principle, these relatively simple concepts of the average charge state are valid when the time between two subsequent collisions between the heavy ion and a gas atom or molecule is long enough that all excited electrons de-excite to the ground state. However, this is true only at low gas pressures with the number of collisions between the heavy ions and gas atoms or molecules being rare. The probability of collisions between excited heavy ions and species of the filling gas is not negligible in a wide range of gas pressures. This becomes more significant as the time between subsequent collisions becomes comparable to the lifetime of the excited states in the atomic shell. Heavy ions in excited atomic states are bound to more easily lose an electron in a collision with the medium, which leads to an increase of the average charge state with increasing gas pressure. Thus, the real average charge states of heavy ions (\bar{q}_r) in the gas will be different from the equilibrated ones (\bar{q}) and it can be determined as the sum of \bar{q} and a correction term of the so-called "density effect" ($\bar{q}_r = \bar{q} + \Delta q$).

The "density effect" has been well studied both experimentally and theoretically in the region of light and medium heavy ions (see [1] and reference therein). The maximum "density effect" was estimated by Bohr and Lindhard [22] and their results $(\Delta q = \bar{q}/5)$ showed that the real measured average charge states can deviate from the equilibrated ones by up to 20%.

In our simple representation of the correction term for the "density effect" we use an exponential function

$$\Delta q = C_0 \cdot \bar{q} \cdot e^{-\frac{\Delta r}{C_1}} \tag{4}$$

as an analogy to the radioactive decay law because this effect is related to the de-excitation of the electrons from excited states. C_0 is a constant which can in principle be directly determined to 0.2 according to the Bohr and Lindhard estimates. However, we keep this as a free parameter , which will be fixed by a fit procedure. C_1 is the average lifetime of excited states in the heavy ions, which we assume as a constant value and Δt is the time between two subsequent collisions where electrons of the heavy ions are excited. This time can be written as $\Delta t = kT/(\sqrt{2\pi}d^2\nu P)$, where k is the Boltzmann constant, T is the temperature, P is the pressure, d is the diameter of the atoms/molecules, and v is the velocity of the ion. We used

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the unit of mbar for the gas pressure *P* and the dimensionless₁₉₉ value of (ν/ν_0) for the velocity, as it is often practically used₂₀₀ at gas-filled separators. A value of $3 \cdot 10^{-10}$ m was taken for the₂₀₁ diameter of the heavy atoms and temperature was taken as $30^{\circ}_{.202}$ Applying the above mentioned constants and unit conversions₂₀₃ the real average charge states of heavy ions including the "den-₂₀₄ sity effect" will be: 205

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$$\bar{q}_r = \bar{q}(1 + C_0 \cdot e^{-10^{-11} \cdot \frac{4.61}{C_1 \cdot (v/v_0) \cdot P}})$$
 (5)₂₀

Here C_0 and C_1 are the above mentioned constants which²⁰⁸₂₀₉ will be fixed in a procedure. This expression provides the influence of the "density effect" relative to the equilibrated average²¹¹₂₁₀ charge state.²¹²

158 3. Experimental setup

215 The experiments were performed at the gas-filled recoil sep-216 159 arator TASCA [16, 17, 18]. A ⁴⁸Ca beam was delivered from₂₁₇ 160 a 14 GHz ECR ion source and accelerated by the linear accel-218 161 erator UNILAC to energies of 234.0 and 232.3 MeV. To make₂₁₉ 162 use of the highest presently available beam intensities at the₂₂₀ 163 UNILAC, TASCA features a windowless differential pumping₂₂₁ 164 system [16] and hence, no vacuum window was used. One222 165 lead (Z=82) isotope, ¹⁸⁸P, and two nobelium (Z=102) isotopes,₂₂₃ 166 252,254 No, were produced in the 48 Ca+ 144 Sm and 48 Ca+ 206,208 Pb₂₂₄ 167 fusion-evaporation reactions, respectively. Lead targets were₂₂₅ 168 produced in the lead sulfide (PbS) chemical form because of₂₂₆ 169 the higher melting point of this compound compared to metallic 170 lead [23]. The isotopic enrichment of ²⁰⁶Pb and ²⁰⁸Pb was 99%.₂₂₇ 171 The target material was evaporated onto $\approx 2 \,\mu$ m thick titanium 172 backing foils. Target thicknesses were ≈ 550 and $\approx 380 \,\mu g/cm_{228}^2$ 173 for the lead and samarium, respectively. All targets were cov-229 174 ered with a $10 \,\mu \text{g/cm}^2$ thick carbon layer in order to reduce ma-230 175 terial losses by sputtering. Three banana-shaped targets were₂₃₁ 176 mounted on the rotating target wheel system ARTESIA (A Ro-232 177 tating Target wheel for Experiments with Superheavy-element₂₃₃ 178 Isotopes at GSI using Actinides as target material). This tar-234 179 get wheel rotates at a frequency of 2000 rpm synchronized with₂₃₅ 180 the pulsed UNILAC beam (5 ms pulse length, 50 Hz repetition₂₃₆ 181 rate). Details about the target arrangement, its operation and the 182 target production can be found in [24, 25]. The isotope ${}^{188}\text{Pb}_{_{237}}$ 183 was produced at the beam energy of ≈ 219 MeV in the center-of-184 target which corresponds to the four neutron evaporation chan-185 nel from the compound nucleus ¹⁹²Pb. The chosen beam en-²³⁸ 186 ergies resulted in center-of-target energies of ≈218 MeV and 187 $\approx 216 \text{ MeV}$ [26] close to the maxima of the excitation functions₂₃₉ 188 of the two neutron evaporation channels from the compound 189 nuclei 254,256 No* [27]. 240 190

The magnetic system of TASCA consists of a dipole and₂₄₁ 191 a quadrupole doublet. Evaporation residues were separated₂₄₂ 192 from the primary beam and unwanted nuclear reaction products₂₄₂ 193 based on their differing magnetic rigidities, $(B\rho)_0$, in the gas-244 194 filled dipole [12]. Quadrupole doublet of the TASCA was op-245 195 erated in its ion-optical focusing mode so-called "High Trans-246 196 mission Mode" (HTM) [18]. To control the gas atmosphere in₂₄₇ 197 TASCA, commercially available MKS[®] gas-flow controllers²⁴⁸ 198

operated with a LabVIEW[®] program were used and permitted excellent control and a very high long-term stability of gas flow rates and, more importantly, of the gas pressure in the separator. This was achieved not only for an individual gas like He or H₂ but also for gas mixtures. Gases were always flowing from the rear end of the separator towards the target region.

The separated and focused ERs were implanted into an (80×35) mm² large 16-strip position-sensitive silicon detector placed at the focal plane of TASCA (focal plane detector). The detector was cooled to temperatures ranging from -10°C to - 30°C. Its energy resolution (FWHM) was 50 keV for full energy α particles with an energy of 5.9 MeV, and the detection efficiency for α particles from implanted ERs was 55%. The detector and data acquisition system of TASCA as used in our experiments reported here was similar to the one at the veol-city filter SHIP (Separator for Heavy Ion Products) of the GSI, Germany [28].

The real average charge states (hereafter average charge state) were deduced from the measured distributions of ERs in the focal plane detector. The ERs were identified through their characteristic α decays [29]. The ER distribution in the focal plane is Gaussian-like in both, the horizontal and in the vertical direction. The actual magnetic rigidity can be determined from the position of the center of the horizontal distribution. If this is off-center on the detector indicating that the magnetic rigidity is different from the value preset at the separator and the real magnetic rigidity (hereafter magnetic rigidity) of the detected ERs can be estimated using the following expression

$$(B\rho)_r = (B\rho)_0 \cdot \left(1 + \frac{X}{100 \cdot D}\right) \tag{6}$$

where $(B\rho)_0$ is the value of magnetic rigidity of the separator set for the given experiment, and *D* is the dispersion at the focal plane of TASCA in unit of [mm] per one percent change in $(B\rho)_0$. In the HTM of TASCA the dispersion *D* is 0.9 mm [18]. The *X* is the shift of the center of ions distribution relative to the middle of the focal plane detector and it was determined by fitting the experimental data by Gaussian curve (see Fig. 1). The average charge states of the ERs can be found from the following expression [12] using the $(B\rho)_r$ value.

$$(B\rho)_r = 0.0227 \cdot \frac{A \cdot (\upsilon/\upsilon_0)}{\bar{q}_r} \tag{7}$$

where A is the atomic mass number of the ERs.

4. Results and discussions

4.1. Dependence of average charge states on the gas pressure

As an example, we show in Fig. 1 horizontal distributions of 252 No measured at different He pressures. For all measurements, the same magnetic settings centering ions with a magnetic rigidity of $(B\rho)_0=2.07$ Tm in the focal plane were used. At these settings, the 252 No ERs are centered in the focal plane at a He pressure of P=0.8 mbar. The experimental data clearly indicate that the position of the center of the distribution is a function of the gas pressure. At pressures lower than 0.8 mbar

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the distributions of ERs are shifting to higher magnetic rigidity, 249 indicating that their charge states become lower. At higher pres-250 sures, the situation is opposite. Ion optical simulations of the 251 ion trajectories in TASCA were performed using the TASCA 252 Monte-Carlo SIMulation (TSIM) code [30]. In the TSIM code, 253 the average charge states of heavy ions were estimated by using 254 the semi-empirical expression from [13]. This semi-empirical 255 expression was parameterized using the experimental data on 256 average charge states of heavy ions obtained at a He pressure of 257 0.66 mbar, as it was determined to be optimal for experiments 258 at the Berkeley Gas-filled Separator (BGS) [13]. The results 259 are shown in Fig. 1 as dashed lines. The calculation reproduces 260 well the distribution of ERs at a gas pressure of 0.5 mbar, which 261 is within the systematic uncertainty of $(B\rho)_0$ between BGS and 262 TASCA. However, the calculated distributions do not reflect the 263 shift of the centers of the distributions. It should be noted that 264 a shift of the distribution by the extent shown in Fig. 1 strongly 265 influences the transmission of the ERs through the separator. 266

The magnetic rigidity can be deduced from these distribu-267 tions using the expression 6. The results for ¹⁸⁸Pb, ²⁵²No and 268 ²⁵⁴No ions in He gas are shown in Fig. 2. The magnetic rigidi-269 ties of 254 No ions measured in H₂ are also shown in Fig. 2. 270 Error bars include uncertainties from the determination of the 271 center of the distribution (single strip width=5 mm) and from 272 the dispersion (15%). The magnetic rigidities are nonlinearly 273 increasing with decreasing gas pressure. Moreover, similar be-274 haviour of the magnetic rigidities depend on the gas pressure 275 are visible in Fig. 2. 276

The average charge states of ²⁵²No and ²⁵⁴No ions in He gas 277 as a function of the velocity are shown in Fig. 3. In a first step, 278 the velocity of the ions inside the target was estimated from 279 fusion-evaporation kinematics. Using the SRIM code [26], the 280 velocity at the exit of the target was estimated, and finally, the 281 velocity at the center of the dipole magnet, after having trav-282 elled through 603 mm of gas was estimated using the stopping-283 power tables of [31]. Error bars show the sum of uncertainties 284 from the deduced magnetic rigidities, the magnetic flux den-285 sity $(B\rho)_0$ (0.8%), and the velocity $\upsilon (\approx 2\%)$. The total uncer-286 tainty of the velocities was deduced from the uncertainties of 287 the beam energy (0.2%) and the thicknesses of the Ti backing₃₀₆ 288 foil (10%) and PbS target (10%), respectively. The variation of $\frac{1}{307}$ 289 velocities in Fig. 3 is due to different beam energies and target 290 thicknesses. The average charge states measured at constant gas 291 pressure (0.8 mbar) show a linear dependence on the velocity, as 292 predicted by expression 3. However, the average charge states³⁰⁹ 293 at higher and lower gas pressures deviate significantly from the 294 results measured at 0.8 mbar. 295 310

From the results presented above the existence of an effect₃₁₁ 296 which is related solely to the gas pressure follows. Such a be-312 297 haviour of the average charge states has been observed earlier₃₁₃ 298 at the DGFRS [12] and was explained by the so-called "density₃₁₄ 299 effect" [1], as mentioned in [12]. The average charge states can₃₁₅ 300 be described using expression 5 as a function of $1/[(\nu/\nu_0)P]_{.316}$ 301 The average charge states are shown in Fig. 4 as a function₃₁₇ 302 of $1/[(v/v_0)P]$. However, in our representation of the "density 303 effect" we used the magnetic rigidities which has a smaller rel-304 ative uncertainties compared to the real average charge states³¹⁸ 305

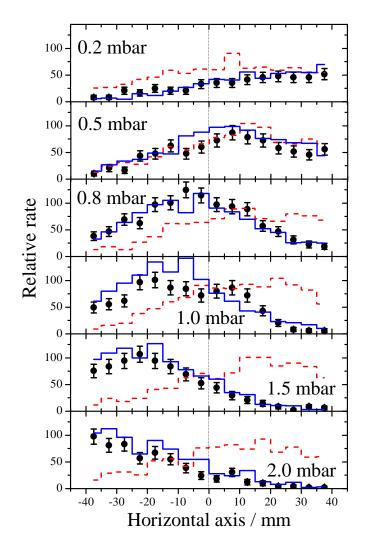


Figure 1: Experimental distributions of 252 No at the focal plane detector depending on the He gas pressure. The same magnetic rigidity of $(B\rho)_0=2.07$ Tm was used for TASCA in HTM. Only statistical error bars are shown. Lines are the calculated distributions by the TSIM code [30] using the average charge state predictions from [13] (dashed) and including the "density effect" (solid). See text for details.

(see Fig. 2 and Fig. 4). Similar to \bar{q}_r the reverse magnetic rigidity can be written as a function of $1/[(v/v_0)P]$ combining expressions 5 and 7:

$$\frac{1}{(B\rho)_r} = \frac{1}{0.0227 \cdot A \cdot (\nu/\nu_0)} \cdot \bar{q} (1 + C_0 \cdot e^{-10^{-11} \cdot \frac{4.61}{C_1 \cdot (\nu/\nu_0) \cdot P}}) (8)$$

where A, (ν/ν_0) , and \bar{q} are atomic mass number, velocity, and equilibrated average charge state of heavy ions, respectively. The reverse magnetic rigidities of measured heavy ions are shown in Fig. 5a as a function of $1/[(\nu/\nu_0)P]$. The results of fits for each ion are also shown in Fig. 5a by dashed lines.

Results of the fitted curve for ¹⁸⁸Pb, which will be used later to provide a general expression for the correction of the "density effect" in the existing semi-empirical expression is given below:

$$\frac{1}{(B\rho)_r} = 0.593 + 0.0652 \cdot e^{-\frac{4.61}{1.8 \cdot (\nu/\nu_0) \cdot P}}$$
(9)

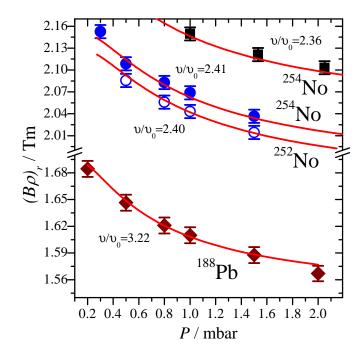


Figure 2: Dependence of magnetic rigidities of lead and nobelium evaporation residues on the gas pressure. Full and open circles denote the magnetic rigidities of 252 No and 254 No in He gas, respectively. Full rectangles denote the magnetic rigidities of 254 No in H₂ gas. The curves are representing the results of an estimation. See text for details.

The shapes of the fitting curves are very similar which in-346 319 dicates that the "density effect" is influencing relatively to the 320 equilibrated average charge states of heavy ions, which are de-347 321 termined from the type of the colliding heavy ion and gas. 322 Based on this feature we made an attempt to parameterize the₃₄₈ 323 "density effect" for various heavy ions, various gases, and var-349 324 ious velocities. As we mentioned in section 1 typical semi-350 325 empirical expressions for the prediction of the average charge₃₅₁ 326 states are filled to data taken at a single gas pressure, which₃₅₂ 327 is different for the different expressions. Therefore, it is rea-328 sonable to find a correction term to the already existing semi-329 empirical expressions, which takes into account the pressure₃₅₅ 330 dependency i.e., the "density effect". 331 356

The magnetic rigidities of all measured ions at₃₅₇ 332 $(1/[(\upsilon/\upsilon_0)P])^{fix}=0.42$, which corresponds to $\Delta t=20 \text{ ps}_{,_{358}}$ 333 were used for the normalization of the measured data. 334 359 Normalized reverse magnetic rigidities are shown in Fig. 5b₃₆₀ 335 as a function of $1/[(v/v_0)P]$. These values are well lying on one 336 single exponential curve. The data were fitted by the following 337 normalized function: 338

$$(10)_{362}^{362} = C_2 \cdot (1 + C_0 \cdot e^{-10^{-11} \cdot \frac{4.61}{C_1 \cdot (v/v_0) \cdot P}})$$

where $(B\rho)_r^{fix}$ is the magnetic rigidity estimated by fit func-³⁶⁵ tions for each ion (as an example: expression 9 for ¹⁸⁸Pb) at the³⁶⁶ given $(1/[(v/v_0)P])^{fix}$. C_i (i=0,2) are the constants. The results³⁶⁷ of the fit and its parameters are shown in Fig. 5b. The magnetic³⁶⁸ rigidities estimated with this function are shown by the solid³⁶⁹ lines in Fig. 2. The corresponding average charge states can be³⁷⁰

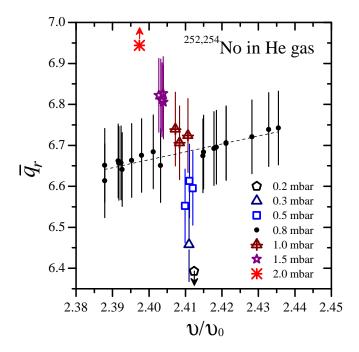


Figure 3: Measured average charge states of ^{252,254}No ions depend on their velocity (expressed in units of the Bohr velocity). The dashed line shows a linear fit of experimental data measured at a gas pressure of 0.8 mbar. Arrows show the lower (up) and higher (down) limits for average charge state values. See text for details.

found using the following expression.

$$\bar{q}_r = \bar{q}_r^{fix} \cdot (C_2 \cdot (1 + C_0 \cdot e^{-10^{-11} \cdot \frac{4.61}{C_1 \cdot (v/v_0) \cdot P}}))$$
(11)

where \bar{q}_r^{fix} is the average charge state at the chosen $(1/[(\nu/\nu_0)P])^{fix}$ value (see above).

The results of estimated average charge states of these ions are shown in Fig. 4. The influence of the "density effect" is well describing for all cases.

The expression 11 can be used for other separators which use different parameterized expressions for the prediction of the average charge states at different gas pressures. The estimation of the "density effect" at other separators can be done by correcting expression 11. Let's assume that there is a semi-empirical expression at a gas filled separator and it gives the $\bar{q}_{s.emp}$ at the optimal gas pressure P_{sep} of separator. In this case the average charge state can be found as:

$$\bar{q}_r = \bar{q}_{s.emp} \cdot \frac{0.614}{(B\rho)_{sep}} \cdot (0.96 + 0.1 \cdot e^{-\frac{2.09}{(\nu/\nu_0) \cdot P}})$$
(12)

where 0.614 and is the value of the reverse magnetic rigidity at $(1/[(\nu/\nu_0)P])^{fix}=0.42$ for ¹⁸⁸Pb estimated by expression 9. $(B\rho)_{sep}$ is the estimated reverse magnetic rigidity at the given gas pressure P_{sep} and velocity (ν/ν_0) of heavy ions by expression 9 at the particular separator. This simple expression can be used to estimate the influence of the "density effect" at various gas-filled separators.

The obtained parameterized expressions are useful for the estimation of average charge states of heavy ions passing through

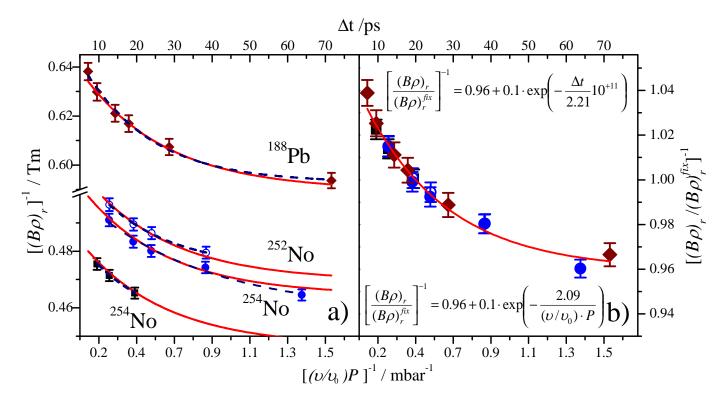


Figure 5: Dependence of reserve magnetic rigidities a) and b) normalized reverse magnetic rigidities of lead and nobelium evaporation residues on $1/[(v/v_0)P]$ (bottom horizontal axis) i.e., time between the consequence collisions Δt (top horizontal axis). The dashed curves represent the results of a fit of data plotted in a). The solid curves represent the results of a fit of data plotted in b). Normalized fit functions with fitted parameters are given in b). See text for details.

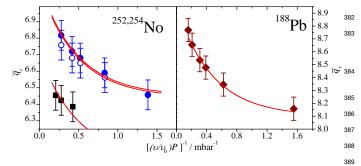


Figure 4: The average charge states of ²⁵²No and ²⁵⁴No ions depend on³⁹⁰ $1/[(\nu/\nu_0)P]$. left) Full and open circles denote average charge states of ²⁵⁴No₃₉₁ and ²⁵²No in He gas, respectively. Full rectangles denote the average charge states of ²⁵⁴No in H₂ gas. right) Full symbols denote the average charge states of ¹⁸⁸Pb in He gas. The curves represent the results of an estimation. See text³⁹³ for details.

TASCA. Results of TSIM calculations are shown in Fig. 1.397 371 The average charge states of heavy ions in the TSIM calcula-398 372 tion were estimated using the parametrization from [13], which399 373 neglects the "density effect" (dashed lines) and with expression400 374 12, which includes the "density effect" (solid line) relative to401 375 the estimated average charge states from [13]. The calculations402 376 based on expression 12 describe the experimental distributions403 377 of ERs over a wide pressure range well. The results also well₄₀₄ 378 describe the width of the ER distributions. As mentioned be-405 379 fore, the "density effect" originates from excitations of elec-406 380 trons in the atomic shells and, therefore, it has no influence407 381

on the collision kinematics (angular spread) between the heavy ions and atoms of the gas.

4.2. The average charge states of heavy ions in gas mixtures

The average charge states of 254 No ions were measured in mixtures of He and H₂. The ratio of the numbers of He to H₂ atoms ($\nu = n_{He}/n_{H_2}$) were established by controlling the gas flow rate of each gas. The total pressure of these mixtures was measured. The average charge states i.e., the magnetic rigidities, were measured at gas mixture ratios of ν =1, 2, 3, and 4 at various pressures. The results are shown in Figs. 6 and 7 as a function of $1/[(\nu/\nu_0)P]$.

First of all, the "density effect" is also observed in gas mixtures. The estimated average charge states/magnetic rigidities of 254 No ions in pure He and pure H₂ are included in Fig. 6 and Fig. 7 for reference (dashed lines). The average charge states/magnetic rigidities of 254 No ions traveling through gas mixtures lie in between these curves, and their absolute values depends on the ratio of the two gases.

Let us now consider the formalism of charge-exchange collisions in terms of charge state fractions $F_i(x)$ (see Sect. 2). In the case of pure He (H₂) gas, the equilibrated charge states are reached at large values of *x*, and their distribution is determined by fractions $F_i^{He}(x)$ and $F_i^{H_2}(x)$). In the case of different types of gas atoms, each fraction of the *i*-th charge state of heavy ions will have a probability $p(He) = n_{He}/(n_{H_2} + n_{He})$ and $p(H_2) = n_{H_2}/(n_{H_2} + n_{He})$ to collide with He atoms or H₂

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molecules, respectively, with $p(He) + p(H_2)=1$. Then, the fraction of the *i*-th charge state of heavy ions $(F_i^m=1)$ in gas mixtures of He and H₂ will be determined by

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$$F_i^m = F_i^{He} \cdot p(He) + F_i^{H_2} \cdot p(H_2).$$
(13)

Assuming the equilibration of charge states in gas mixtures, the following expression can be derived from expressions 3 and 9:

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$$\bar{q}^m = \bar{q}^{He} \cdot p(He) + \bar{q}^{H_2} \cdot p(H_2)$$
 (14)

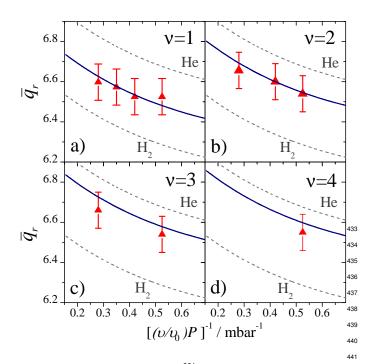


Figure 6: The average charge states of 254 No ions depending on pressures of₄₄₂ the gas mixtures ν =1, 2, 3 and 4. Experimental values are shown by triangles. Solid curves represent the estimated average charge states from expression (7. Dashed curves represent the average charge states dependence of 254 No ions on the pressure of pure He and H₂. See text for details.

where \bar{q}^{He} and \bar{q}^{H_2} are the equilibrated average charge states⁴⁴⁴ 416 of No ions in pure He and H₂ at a given pressure P, respec-445 417 tively. This expression is just a weighted mean value of the 446 418 two equilibrated average charge states in pure He and pure H_2 .⁴⁴⁷ 419 However, this expression cannot be used directly to obtain the 448 420 average charge states, as no correction terms for the "density⁴⁴⁹ 421 effect" are included. To this end, let us make the assumption 450 422 that the ratio $\Delta q/\bar{q}$ is constant for He and H₂ at pressures P^{451} 423 within (0.2-2.0) mbar based on results from previous subsec-452 424 tion (marked by dashed curves in Fig. 6 and Fig. 7). Then the⁴⁵³ 425 454 following expression will be valid: 426 455

$$\bar{q}_{r}^{m} = \bar{q}_{r}^{He} \cdot p(He) + \bar{q}_{r}^{H_{2}} \cdot p(H_{2})$$
(15)⁴⁵⁶

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The average charge states estimated using expression 15 for₄₅₈ gas mixtures are shown with solid lines in Fig. 6. The average₄₅₉ charge states at various pressures of pure He and H₂ gases were₄₆₀ estimated using the parameterized expressions from the previ-₄₆₁ ous subsection. The estimated values describe well the average₄₆₂

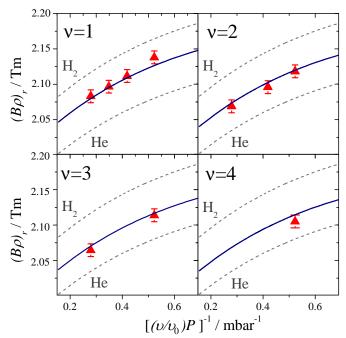


Figure 7: Same as Fig. 6 but for magnetic rigidities. See text for details.

charge states of ²⁵⁴No ions in the gas mixtures over a wide range of gas pressures.

The corresponding magnetic rigidities can also be estimated using expressions 7 and 15. The results are shown in Fig. 7. The magnetic rigidities are also well predicted within a gas pressure range of (1-2) mbar. In the case of gas mixtures, the width of the charge state distribution can be different from that in pure gases due to the different types of ion-atom collisions. However, the observed width is between the values of the pure gases.

5. Summary and Conclusion

The average charge states and magnetic rigidities of ¹⁸⁸Pb, ²⁵²No and ²⁵⁴No ions produced in the fusion-evaporation reactions ⁴⁸Ca+¹⁴⁴Sm and ⁴⁸Ca+^{206,208}Pb were investigated at the gas-filled recoil separator TASCA. The gases He, H₂, and their mixture in different ratios were used as filling gas. Measurements were performed at different pressures ranging from 0.2 to 2.0 mbar. In all gases, we observed a strong dependence of the average charge states and magnetic rigidities on the gas pressure. This dependence was attributed to the so-called "density effect". Experimental data were discussed in a simple analytical way and corresponding simple an analytical expression were used for the parameterizations of the data. The results of parameterized expression well describes the behaviour of the average charge states and magnetic rigidities of ¹⁸⁸Pb, ²⁵²No and ²⁵⁴No ions in pure He and H₂ gases. This effect should be taken into account to predict more precisely the average charge states of heavy ions traveling through dilute gases in gas-filled recoil separators. The ion-optical calculations of the TASCA separator with the "density effect" included describe the experimental

data very well. This is essential to obtain reliable estimates of 463 the transmission of the recoil separator for ERs. The average 464 charge states of heavy ions in gas mixtures were systematically 465 measured for the first time. Using the results from the case of 466 pure He and H₂ gas and also presenting a simple analytical dis-467 cussion, an expression for the determination of average charge 468 states in a gas mixture was proposed. Results reported in this 469 paper can be used for a better planning of future experiments 470 at gas-filled recoil separators and also for other experimental 471 setups which using the dilute gases at pressures around 1 mbar. 472

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