Average charge states of heavy atoms in dilute hydrogen

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We measured the average electronic charge $\langle q \rangle$ of heavy ions with atomic numbers $Z=89$ through 116 traversing dilute hydrogen gas with velocities ranging from 1 to 2.6 times the Bohr velocity. We observe a strong linear dependence of the average charge on the velocity of the ions, and only a weak dependence on their atomic number Z. A more detailed examination of the experimental results shows that the $\langle q \rangle$ values reflect the influence of the $5f$ electron shell on the properties of these elements.

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I. INTRODUCTION

Studies investigating the electronic charges of heavy atoms moving in a gas medium immediately followed the discovery of nuclear fission, since the heavy atoms formed in nuclear fission are created with a high velocity and, consequently, with a high degree of ionization. The first experimental work concerning this problem was reported by Perfilov [1]; the first theoretical work was reported by Bohr [2] and Lamb [3].

Perfilov [1] determined the electronic charge of fission fragments by studying their deflection in a magnetic field. Fission fragments were obtained from the fission of uranium nuclei bombarded with neutrons. In the theoretical description of the average electronic charge of fission fragments, Bohr [2] assumed that a heavy atom moving rapidly through a rarified gas retains all of its electrons that have orbital velocities exceeding that of the ion relative to the medium. From this, using the Thomas-Fermi model for the structure of the atom, he obtained the well-known dependence of the mean electronic charge $\langle q \rangle$ of a heavy atom with atomic number Z on its velocity v:

$$\langle q \rangle = Z^{1/3} v / v_0 . \tag{1}$$

Here v_0 is the velocity (2.19×10^6 m/s) of the electron in Bohr's model of the hydrogen atom.

Independent of Bohr's work, Lamb [3] calculated the mean charge of fission fragments using energy considerations. He assumed that the 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."

The electronic charge of an ion changes due to the exchange of electrons as it collides with atoms of the traversed matter. The observed charge-exchange mechanism differs depending on whether the heavy ion is moving through a solid or through a dilute gas. In the first case, the mean time interval between two collisions is much shorter than in the second one; therefore, excited electronic states in the heavy ions significantly influence the process of ionization. Since electrons can be lost from the excited states of the ions with higher probability, the mean charges of heavy atoms traversing solids are significantly higher than in gaseous media. We use the term "mean charge" for the heavy ion's nuclear charge minus the average number of electrons it has under given conditions.

The interaction of two atoms involves two many-body systems, and the process of charge exchange can be complicated further by the possibility of multiple electron transfer in single collisions. These processes are so complex that today there is no reliable theory for predicting the mean charge of heavy ions moving through a dilute gas; practically all the available information on this subject has been obtained either empirically or by using very rough models. In fact, the same holds true for the interaction of heavy ions with solid targets (see, e.g., Refs. [4,5]).

There is renewed interest in the process of charge exchange by heavy ions traversing matter, stimulated by the development of new accelerators, heavy-ion sources, and new electromagnetic separators for the study of nuclear reactions induced by heavy ions (see, e.g., Ref. [6]). In practice, production of the evaporation residues (EVRs) in heavy-ion induced reactions is the only way to synthesize and study heavy atoms with atomic numbers above $Z=100$. We can take advantage of the fact that these products recoil from thin targets with well-defined velocities. Therefore, the application of, e.g., a gas-filled separator in these investigations needs accurate predictions of the charge states of very heavy ions attained through charge exchange. This, in turn, requires experimental information on the charge states of ions in gaseous media in order to develop reasonable models and a systematic organization of this information.

In the course of performing experiments for our heavy-element research program, we performed extensive measurements of the average charge states $\langle q \rangle$ for heavy ions with

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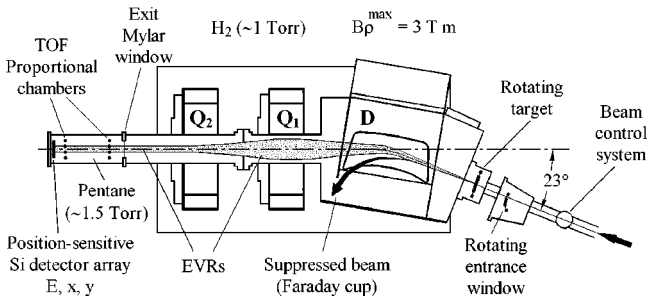

FIG. 1. Layout of the Dubna gas-filled recoil separator (the dipole magnet *D*, followed by the quadrupole doublet Q_1 and Q_2).

$Z=89$ through 116 and average velocities relative to the medium of a gas-filled separator of 1.0 to 2.6 times the Bohr velocity v_0. Many of the recoiling nuclei had never before been observed.

II. EXPERIMENTAL TECHNIQUE

In the present work, the atoms under study were obtained as evaporation residues of compound nuclei produced in complete-fusion nuclear reactions between beams of accelerated heavy ions and very heavy target atoms. The projectile particles were delivered by the U400 cyclotron of the Flerov Laboratory of Nuclear Reactions in Dubna. The mean charges of the recoiling complete-fusion reaction products were determined using the Dubna gas-filled recoil separator [7], see Fig. 1. The EVRs recoiling from a thin rotating target ($0.2-0.6$ mg/cm^2, depending on the studied reaction system) were separated in flight from beam particles, scattered nuclei and nuclear transfer-reaction products in the separator's dipole magnet, filled with hydrogen at a pressure of about 1 Torr. The dipole is followed by two quadrupole lenses for focusing the EVRs on the focal plane of the separator. The curvature radius of the separator is $\rho=1.8$ m, and the maximum magnetic rigidity of investigated atoms is $B\rho=3$ T m.

A rotating entrance window made of 1.5-μm Ti foils separates the gas-filled volume of the separator from the vacuum of the accelerator beam line. The hydrogen gas enters the separator volume between the last quadrupole lens and detection system and is pumped away at the target position, providing additional cooling of the target. A permanent gas flow (about 10 cm^3/min of hydrogen) through the system prevents the build-up of gaseous impurities that can affect the mean charge value of the heavy atoms. The separator was normally operated under hydrogen gas at a pressure of about 1 Torr.

A 1.5-μm Mylar exit window separates the detection module from the gas medium of the separator. The separated EVRs passed through a time-of-flight measurement system consisting of two multiwire proportional chambers placed in pentane at a pressure of about 1.5 Torr and were finally implanted in the detector installed in the focal plane of the separator. The focal-plane detector consisted of twelve silicon 40-mm-high$\times$10-mm-wide strips with position sensitivity in the vertical direction. We obtained horizontal (x) positions by recording the strip number for the reaction products, and vertical position signals (y) from the 4-cm-high resistive layers of the detectors. The detection system of the separator is described in more detail in Ref. [7].

After emerging from the target layer, the heavy atoms retain a relatively low number of electrons and thus have a correspondingly large electronic charge. Furthermore, the distribution of the ionic charge states is quite broad. Due to charge exchange in consecutive collisions with the gas atoms, the distribution of the electronic charges rapidly becomes narrower and the mean charge decreases to the equilibrium value $\langle q\rangle$.

In sequential collisions with the atoms of the medium the heavy atoms slow down and deviate from their primary direction. They move between the poles of a dipole magnet filled by dilute gas with gradually decreasing velocity along some average trajectory characterized by the mean curvature radius of the separator. Ions with mass number *A*, mean electronic charge $\langle q\rangle$, and velocity v, will be deflected in a magnetic field of flux density *B* following a trajectory with curvature radius ρ. The magnetic rigidity is related to the ionic charge as follows:

$$(B\rho)_0=0.0227A(v/v_0)/\langle q\rangle[\text{T m}]. \tag{2}$$

The mass number *A* was determined unequivocally in our experiments by establishing genetic links between position- and time-correlated signals from the implantation and subsequent α decay or spontaneous fission of mother nuclei and the decays of the known daughter nuclei. The velocity of the EVRs in the middle of the dipole magnet was calculated using reaction kinematics and energy losses from range tables for projectiles [8] and heavy nuclei [9].

The separated EVRs delivered to the separator's focal plane have a Gaussian-like distribution in the horizontal direction. Their actual magnetic rigidity can differ from the rigidity of the separator setting in a particular experiment. Experimental horizontal distributions of ^{252}No produced in the complete fusion reaction ^{206}Pb(^{48}Ca,2n) at two magnetic rigidities of the separator are shown in Fig. 2. The maxima of both distributions are shifted from the center of the detector array. In such cases the magnetic rigidity of the detected EVR can be estimated by correcting the rigidity of the separator $(B\rho)_0$ using the following expression:

$$(B\rho)_{\text{ion}}=(B\rho)_0(1+x/100D), \tag{3}$$

where $(B\rho)_0$ is the value of magnetic rigidity of the separator set for the given experiment, *D* is the dispersion, i.e., the shift of the maximum of the horizontal focal-plane distribution of EVRs per unit of $(B\rho)$ value. In our experiments *D* was determined to be about 7.5 mm per one percent change in $(B\rho)$. The shift x of the horizontal focal-plane distribution of ions relative to the middle of the detector array was calculated by fitting the experimental distribution with a Gaussian curve.

III. RESULTS AND DISCUSSION

The average charge states of the EVRs with $Z=89$ through 116 were measured in the experiments aimed at the

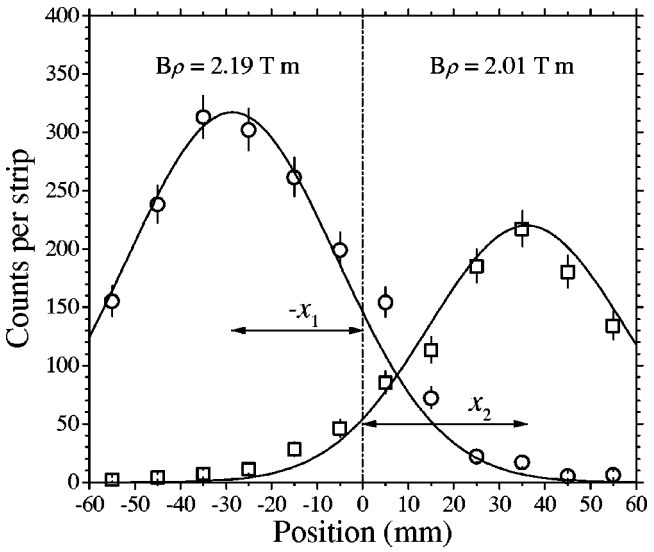

FIG. 2. Two experimental focal-plane horizontal distributions of ^{252}No corresponding to different magnetic rigidities $B\rho$ of the separator. Solid lines show the results of Gaussian-curve fitting of these distributions. The shifts of both distribution maxima from the center of the focal-plane detector are shown by arrows.

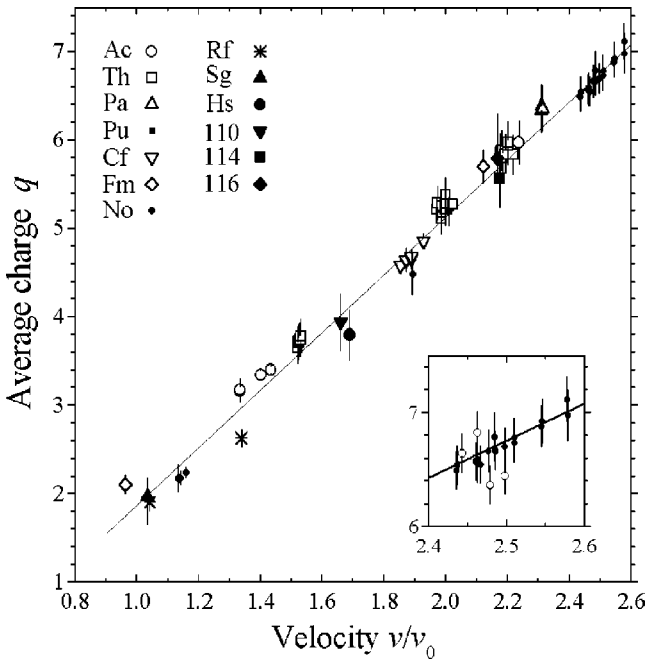

FIG. 3. Systematics of the measured equilibrium charge states of heavy atoms in dilute hydrogen and their dependence on v/v_0, where v is the velocity of the ion and v_0 is the Bohr velocity. The line shows a linear fit to the experimental data, see formula (4). The upper part of the systematics is given in the inset. Here the charges of No in hydrogen at the pressures of 0.5 and 1.5 Torr are shown by open circles.

synthesis of isotopes of heavy and superheavy elements [10,11] and in the reactions used for calibration of the detection system and the separator. Heavy EVRs were produced in the complete fusion reactions of projectiles ^{18}O, ^{22}Ne, ^{26}Mg, ^{34}S, ^{40}Ar, and ^{48}Ca with targets from ^{nat}Yb to ^{238}U, ^{242,244}Pu, and ^{248}Cm.

Figure 3 shows the systematics of the measured equilibrium charge states of heavy atoms traversing dilute hydrogen vs the parameter v/v_0, where v is the velocity of the ions (assumed from the calculated recoil velocity of the EVR) and v_0 is the Bohr velocity. The uncertainties in the charge values are determined by uncertainties in measuring the projectile energies (1%), target thicknesses (5%), hydrogen pressure (4%), curvature radius and magnetic flux density (1.2% of the $B\rho$ value), dispersion (15%), and in establishing the maxima of position distributions of the detected EVRs. The latter depends on statistics and dominates in the case of a few produced atoms. Energy losses of EVR's in the target were considered to correspond to half of the layer thickness. The uncertainty in the velocity of atoms at the exit of the target was determined by the uncertainties in the target thickness and in knowledge of the actual position of the center of distribution of nuclei produced inside the target layer. In the case of poor statistics, the latter was set as 68% of the half of target thickness. In this case, the maximum of the position distribution of the EVRs on the detectors was calculated using a constant width of the distribution (standard deviation $\sigma = 3$ cm) that corresponded to the mean value observed in reactions with similar kinematics, and results of calculations using the ANAMARI code [12]. This code was developed for the simulation of the EVR trajectories in the separator, the corresponding spatial distributions, and the resulting collection efficiencies. The code takes into account reaction kinematics, energy losses and multiple scattering in the target and separator media, equilibrium charge states of the EVRs and the magnetic optical system of the separator.

As can be seen from Fig. 3, in the first approximation, the collected experimental data are fit well to a simple linear dependence

$$\langle q \rangle = 3.26 v/v_0 - 1.39. \quad (4)$$

This formula is in qualitative agreement with Bohr's model [2], which reflects the general dependence of charge state on an atom's velocity and a weak dependence on its atomic number, being proportional to $Z^{1/3}$ at constant velocity. In our case, however, the influence of the atomic number is of minor importance because the Z range studied corresponded to only $\pm 4.4\%$ of the mean $Z^{1/3}$ value. The formula allows one to estimate the average charge states of heavy atoms in hydrogen at a pressure of about 1 Torr with a precision of about 4% (mean value for all 73 measurements presented in Fig. 3), comparable to the accuracy of the measurements.

The new empirical formula can be compared with other systematics. Note that for the experiments aimed at the synthesis and study of very heavy nuclei with gas-filled separators, the accuracy of the charge state estimate is extremely important and should not be worse than 10%. For instance, in our experiments on the synthesis of superheavy nuclei [10,11] with a production rate of about one atom per month, a charge uncertainty of 8% would reduce the collection efficiency of atoms on the focal-plane detectors by only a factor

TABLE I. Experimental parameters for irradiations producing heavy recoil products.

Nuclide	Reaction	v/v_0	q_{exp}	q_{Bohr}	q_{Lamb}
^{254,255}No	^{238}U(^{22}Ne,5−6n)	1.14	2.19±0.13	5.3	2.4
^{252}No	^{206}Pb(^{48}Ca,2n)	1.89[a]	4.48±0.23	8.8	3.9
^{252}No	^{206}Pb(^{48}Ca,2n)	2.55	6.90±0.18	11.9	5.8
^{261}Rf	^{244}Pu(^{22}Ne,5n)	1.04	1.90±0.10	4.9	2.0
^{258,259}Rf	^{238}U(^{26}Mg,5−6n)	1.34	2.63±0.10	6.3	3.1
^{265,266}Sg	^{248}Cm(^{22}Ne,4−5n)	1.04	$1.97^{+0.21}_{-0.31}$	4.9	1.7
^{267}Hs	^{238}U(^{34}S,5n)	1.69	$3.80^{+0.27}_{-0.29}$	8.0	3.2
^{273}110	^{244}Pu(^{34}S,5n)	1.66	$3.94^{+0.32}_{-0.33}$	8.0	3.1
^{288,289}114	^{244}Pu(^{48}Ca,3−4n)	2.18	5.56±0.33	10.6	4.8
^{292}116	^{248}Cm(^{48}Ca,4n)	2.17	$5.79^{+0.50}_{-0.24}$	10.6	5.4

[a]Additional degrader was placed behind the target to reduce EVRs' energy.

of 2. The most recent empirical systematics for the same medium but for lighter atoms, Sm to Fm, was given in Ref. [13]:

$$\langle q\rangle = 2.6\times 10^{-7} v Z^{1/3} - 0.28$$

or

$$\langle q\rangle = 4.4\times 10^{-7} v Z^{1/3} - 3.92, \qquad (5)$$

for the parameter $vZ^{1/3}$ less than or greater than 2×10^{-7} m/s, respectively.

Nevertheless, the experimental average charge values of atoms of No and heavier, with velocity $v/v_0 > 1.5$, are about 9% lower than predicted by the systematics [13]. These estimates are in even greater disagreement with our data at lower velocities, systematically exceeding the measured values by 27% on average.

Table I presents experimental values of the average charges states q_{exp} of the heavier atoms (No through element 116) traversing dilute hydrogen with the indicated velocities. Reactions used to produce the atoms under consideration are given in the second column. The last two columns give the charge values calculated in model approaches by Bohr [formula (1)] and Lamb, respectively. In the latter case we took the calculated ionization potentials of No, Rf, and Sg from Refs. [14–16], respectively. For the atoms of Hs through element 116 we started from the known ionization potentials of their chemical analogs, i.e., Fe, Ni, Pb, and Te [17], and employed a simple linear interpolation to estimate charge values, similar to formula (6) of Ref. [18]. However, in contrast to Ref. [18], kinetic energies of the electrons were calculated in Lamb's approach [Ref. [3], point (c)], which gives a kinetic energy value four times less than that given in Ref. [18] [formula (5)].

Comparison of the experimental and calculated average charge values shows that both theoretical approaches can provide only a qualitative description for the heavier atoms, as was already shown for lighter atoms. Calculations with Bohr's formula result in values twice as high as those from the experiment. Lamb's approach shows better agreement. For instance, calculated and measured charges for Rf (at $v/v_0 = 1.04$) and Sg agree within experimental uncertainties. Experimental values for No (at $v/v_0 = 1.14$) and Rf (at $v/v_0 = 1.34$) are lower than the calculated ones by 10 and 18 %, respectively, while other charge measurements for atoms of No through element 116 with $v/v_0 > 1.6$ exceed the calculated values by 7–21%.

Note that in making theoretical estimates we used calculated ionization potentials or those of chemical analogs. However, one can hardly expect considerable improvement of these estimates, even when using more ''correct'' values. A drawback of both theoretical models is that they do not take into account the properties of the medium, although it is evident from the experiment that average charge states vary for different gases and show dependence on the gas pressure as well.

Most of the data were obtained at the hydrogen pressure of 1 Torr. We also carried out a few experiments at 0.5 and 1.5 Torr with ^{252}No produced in the ^{206}Pb(^{48}Ca,2n) reaction (see the inset in Fig. 3). Solid circles show data for No taken at 1 Torr, the results obtained at other pressures are shown by open circles. These data points deviate from the set measured at 1 Torr; however, they still lie within experimental uncertainties. Increasing the gas pressure by 50% results in an increase of the charge by about 2%, and reducing the pressure by a factor of 2 leads to about a 5% decrease of the charge. This observation can be apparently explained by the so-called ''density effect.'' Evidently, excited states in a heavy ion can significantly influence the probabilities for electron capture and loss, since electrons are stripped more easily from an excited level than from the ground state. Higher gas pressure means a shorter average time between consecutive ion collisions with atoms of matter. This results in a higher collision probability of a still excited heavy ion with the next gas atom and thus in its higher average charge state. The experimental and theoretical aspects of the density effect were reviewed by Betz in Ref. [4].

The measured average charge values show approximately a linear dependence on the velocity of the ions (see Fig. 3), in agreement with Bohr's formula. According to this model, the average charge state of the atom depends weakly on its atomic number, being proportional to $Z^{1/3}$ at constant velocity. Lamb's theory gives a different dependence of charge states on Z: ionization potentials should vary as the electrons populate atomic levels and reach maximum values for the closed shells. Accordingly, one could expect oscillations in average charge states of atoms with different Z traversing a gas at a constant velocity. The ions that have to lose electrons from the closed shells should show lower average charge states, compared with the higher-Z neighbors. Such a deviation from the monotonic trend in charge states was first observed for the atoms of rare-earth elements moving through helium and air [19], and was later studied experimentally [18]. A maximum in the dependence of $B\rho/A$ on Z in the vicinity of Hf ($Z=72$) with respect to the smooth curve proportional to $Z^{-1/3}$, and the respective minimum of the value q/v, was observed in Ref. [18]. This observation was explained by the gradual filling of the $4f$ electron shell (and the increase of the binding energy of the next electron to be removed) and further population of the $5d$ shell. A similar effect could be expected [18] to occur near Rf ($Z=104$),

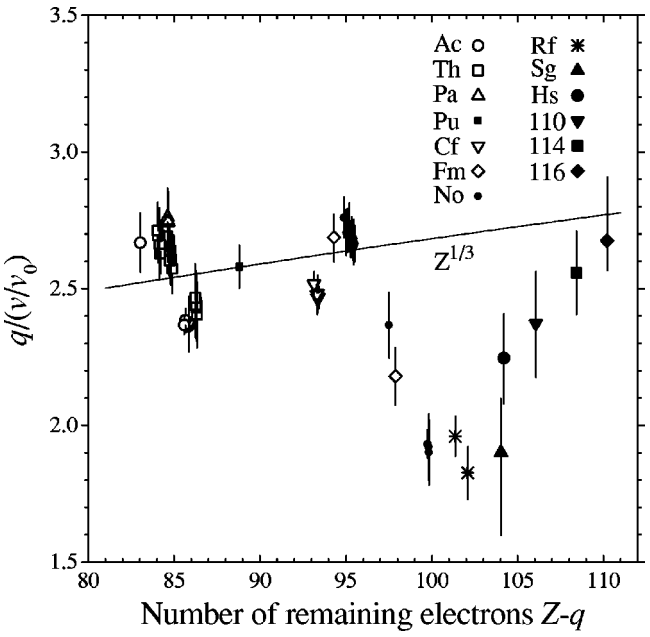

FIG. 4. Measured average charge states (scaled by the value of v/v_0) vs the number of remaining electrons in the atoms. The line proportional to $Z^{1/3}$ is drawn to guide the eye.

based on the analogy of the $4f$-$5d$ and $5f$-$6d$ shells in lanthanides and actinides, respectively. The same effect of atomic shell structure on the average charges was studied in Ref. [20] for light atoms with $Z \leqslant 18$.

Figure 4 shows average charge states measured in the present work (scaled by the value of v/v_0) vs the number of residual electrons in an ion. A considerable disagreement with the general trend $\langle q \rangle/(v/v_0) \propto Z^{1/3}$ is observed as we approach Rf. The gradual population of the $5f$ shell and the subsequent population of the $6d$ shell can explain this effect. Our observation agrees with isoelectronic sequences known from atomic physics.

IV. CONCLUSIONS

Using the Dubna gas-filled recoil separator we measured the mean charge states of heavy atoms with $Z=89$ through 116, traversing hydrogen at 1 Torr in the velocity range of 1 to $2.6\,v/v_0$. In the first approximation, the average charges follow an empirical linear dependence on the velocity, in agreement with Bohr's theory.

The pressure of the gas medium influences the ionization-recombination process resulting in an increase in the average charge states at higher gas pressure. Such a variation could be explained by the so-called ''density effect'': an increase of the gas pressure results in a shorter mean free path for the ion, a higher collision probability for an excited ion with gas atoms, and a correspondingly higher average charge of an ion, as the probability of losing an electron from an excited state is larger than from the ground state.

A more detailed examination of the charge states of ions with different atomic numbers at similar velocities reveals deviations from the general monotonic trend $[\langle q \rangle/(v/v_0) \propto Z^{1/3}]$. A considerable effect is observed in our experiments when the number of residual electrons in the ions approaches 104. The gradual population of the $5f$ shell and the corresponding increase in the ionization energy of the next electron to be removed, and the subsequent population of the $6d$ shell can explain the effect. Our observations agree with isoelectronic sequences known from atomic physics.

The interaction between a heavy ion and atoms of a medium is so complex that model calculations reproduce only the gross trends in the charge state variations. The existing theories still need to be refined for more quantitative and accurate predictions of average charges. So far, only semiempirical formulas and systematics can be used in practice for calculating average charges.

From the viewpoint of an experimental application, the set of measured data permits the construction of a reliable empirical systematics for the mean charge states of heavy atoms traversing hydrogen. The charge states estimated from these systematics have proved to be accurate enough for us to choose properly the magnetic parameters of the Dubna gas-filled recoil separator. The numerous experiments performed employing this setup resulted in the production of the heaviest new nuclides with $Z=106$ through 110 and the investigation of their decay properties, and recently in the synthesis of the new superheavy elements with $Z=114$ and 116.

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