Empirical relations for heavy-ion equilibrated charges and charge-changing cross sections in diluted H$_2$ with application

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Abstract. Highly ionized heavy evaporation residues (ERs) resulting from heavy-ion (HI) fusion–evaporation reactions are knocked out from solid targets to rarefied gas of gas-filled recoil separators. In gas, these ERs undergo charge-changing collisions on their way to a detection system. An equilibrium between the loss of charge (electron capture) and the gain of charge (electron loss) for ionized ERs allows one to use equilibrated charge-state distributions in trajectory calculations for ERs moving through a magnetic field. The distance from a target to the point where the ER charge-state distributions become to be the equilibrated one is essential for such calculations. This distance can be estimated in simulations based on electron capture and loss cross sections for energetic HIs. Reasonable approximations of available experimental data have provided these cross sections, which were applied to the Monte Carlo simulations of the ionic charge evolution for highly ionized heavy ERs. The results of these simulations demonstrate the setting of charge equilibration close to the target.

1 Introduction

Gas-filled recoil separators (GFRSs) are a valuable tool for the separation and studies of heavy evaporation residues (ERs) produced in complete-fusion reactions of heavy-ion (HI) projectiles with heavy target nuclei [1–8]. In these devices, ERs recoiling out of a thin target are efficiently separated in-flight from the primary HI beam particles and other unwanted products within a separation time of $\sim$1 μs. The separation (collection) efficiency for ERs of interest is essentially determined by their kinematics (angular and energy distributions) in combination with the focusing and bending capabilities of the ion-optical elements of the separator beamline.

For HI with atomic mass number $A$, ionic charge $q$ and velocity $v$ moving in a homogeneous magnetic field with magnetic flux density $B$, Larmor radius $\rho$ is determined by magnetic rigidity $B\rho$, which, in turn, is determined by the following relation:

$$B\rho = 0.0227A(v/v_0)/q = 0.144(EA)^{1/2}/q,$$  \hspace{1cm} (1)

where $B\rho$ is expressed in Tm, $v_0=2.187\times10^8$ m/s is the Bohr velocity, and $E$ is the HI energy in MeV. In a region filled with gas, HIs undergo atomic collisions, in which electrons can be either lost or captured, changing the charge states of HIs. If the mean free path of HIs between two charge-changing collisions is short enough compared with the length of its trajectory, i.e., the frequency of charge-changing collisions is large, while at the same time the energy loss due to collisions is negligible, the charge-state distribution will approach equilibrium conditions defined by mean charge $q_m$ and standard deviation $d$. Under these conditions, HIs can be assumed to closely follow the trajectory corresponding to their mean (equilibrated) charge value. According to the Bohr predictions [9], all electrons with orbital velocities smaller than $v$ are stripped, so the mean charge $q_m$ of HI with atomic number $Z$, as derived with the Thomas–Fermi model, can be expressed by

$$q_m = (v/v_0)Z^{1/3}.$$  \hspace{1cm} (2)

Eq. (2) is valid for the velocity range of $1 < v/v_0 < Z^{2/3}$, then Eq. (1) can be rewritten as

$$B\rho = 0.0227A/Z^{1/3}.$$  \hspace{1cm} (3)

Hence, corresponding trajectories are determined by the mass and atomic numbers only and are independent of the initial charge and velocity distributions.

In the present work, the emphasis is on the estimates of how far from a target the equilibrated charge-state distribution for ERs moving in rarefied H$_2$ is settled. These estimates are essential for a new generation of GFRSs intended to extract superheavy nuclei (SHN)
produced in complete-fusion reactions with accelerated HI beams [10]. For example, a new Dubna GFRS allows using relatively thick targets (∼q by exponential functions of HI charge state cross sections. These cross sections are approximated by taking into account single-electron capture and loss approximation of proximity to its equilibrated charge value changing process in gas is considered in the approx-

In practice, when dealing with GFRSs, HI charge-

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Fig. 1 Experimental data for the mean equilibrated charges of the low energy heavy ions [27,28] (different symbols) and for the mean charges of ERs obtained with DGFRS [26] (open squares) are shown as a function of reduced velocity $\left(\frac{v}{v_0}\right)Z^{1/3}$. The power function fit denoted as $q_m^{\text{fit}}$ is also shown (solid line with a shadow area corresponding to the 95% confidence band). See details in the text.

2 Mean equilibrated charges for heavy ions and ERs in H$_2$

In practice, when dealing with GFRSs, HI charge-

changing process in gas is considered in the approximation of proximity to its equilibrated charge value by taking into account single-electron capture and loss cross sections. These cross sections are approximated by exponential functions of HI charge state $q$ with the fixed value of $q_m$ used as a parameter [4,21–23]. Different approaches were used for the estimate of $q_m$. Thus in [4,22], the empirical systematics based on the data available at that time [24,25] were used. In another approach [3], the parametrization proposed in [21] and available experimental data were used to obtain the dependence of $q_m$ on $Z$ and $v/v_0$ in He gas. Later, the same data were used for new systematics [23], in which additional data were included.

For the estimates of $q_m$ in diluted H$_2$, the first inspection was performed for the scaling based on the large body of experimental data obtained for HIs passed through different gas media [15]. The absolute uncer-
tainty of this fit is $\Delta q_m^{\text{SG}} = 0.48$, and the relative uncertainty is determined by the relation $\Delta q_m^{\text{SG}}/Z = 2.6\%$. For the heaviest ions with $100 \leq Z \leq 118$, the first value leads the uncertainties in $q_m^{\text{SG}}$ corresponding to 16.6 and 6.0% for Fm ($Z = 100, x = 0.0871$) and Og ($Z = 118, x = 0.187$) ions, respectively ($x$ is the scaling parameter used in [15]). The direct application of the relation for the relative uncertainty gives us the respective estimates of 260 and 309% for the same ions. Figure 1 in [15] shows that experimental points at $x \approx 0.2$ (the main area of interest to GFRS) are scattered within $0.04 < q_m^{\text{SG}}/Z < 0.16$. The SG scaling predicts the value of $\approx 0.078$. These estimates can be compared to those obtained in the experiments [26], which correspond to 4.8 and 5.9% for the same Fm and Og ions. One may suggest that the scaling presented in [15] is unsuitable in practice when dealing with GFRSs (see also considerations below).

Different scalings of the available $q_m$ data for Br and heavier ions [27,28] together with those obtained using

In general, ERs escaping a target have a broad charge-state distribution. In addition to the ‘solid’ equilibrated component (see, for example, [13–15]), there is a non-equilibrated one with charge states much higher than those inherent in the equilibrated charges [16–20]. This non-equilibrated component corresponds to excited nuclear states that strongly affect the ionization of inner atomic shells owing to the conversion of nuclear transitions in ERs. Vacancies formed in the conversion of inner shells of ionized ERs lead to the Auger cascades, which significantly increase the ion charges of ERs over the expected equilibrium magnitudes. Thus, reliable electron capture and loss cross section data for ERs over the expected equilibrium magnitudes. Thus, reduced velocity ($\left(\frac{v}{v_0}\right)Z^{1/3}$). The direct application of the relation for the relative uncertainty gives us the respective estimates of 260 and 309% for the same ions. Figure 1 in [15] shows that experimental points at $x \approx 0.2$ (the main area of interest to GFRS) are scattered within $0.04 < q_m^{\text{SG}}/Z < 0.16$. The SG scaling predicts the value of $\approx 0.078$. These estimates can be compared to those obtained in the experiments [26], which correspond to 4.8 and 5.9% for the same Fm and Og ions. One may suggest that the scaling presented in [15] is unsuitable in practice when dealing with GFRSs (see also considerations below).

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Dubna GFRS (DGFRS) [26] were tested, and the best one (the least reduced $\chi^2$ value) was obtained for $q_m$ being a power function of reduced velocity $(v/v_0)Z^{1/3}$. In Fig. 1, these data are shown together with the approximated mean charge values obtained in the DGFRS, which is the position shift of the maxima for the ERs horizontal distribution in the focal plane of GFRS. The relation $X_m$ determines fine-tuning of ion-optical elements of GFRS. Inaccuracy in the choice of $q_m$ leads to the shift of maxima $X_m$ for ER distributions in the focal plane of GFRS. The relationship $q_m^{fit}/q_m = 1 + X_m/(100D)$ determines this shift, in which $q_m^{fit}$ is the mean charge estimated with some approximation ($q_m^{fit}$ or $q_m^{fit}$ in our case), $D$ is the dispersion of GFRS, which is the position shift of the maximum for the ERs horizontal distribution in the focal plane of GFRS. Inaccuracy in the choice of $q_m$ leads to the shift of maxima $X_m$ for ER distributions in the focal plane of GFRS. The relationship $q_m^{fit}/q_m = 1 + X_m/(100D)$ determines this shift, in which $q_m^{fit}$ is the mean charge estimated with some approximation ($q_m^{fit}$ or $q_m^{fit}$ in our case), $D$ is the dispersion of GFRS, which is the position shift of the maximum for the ERs horizontal distribution in the focal plane of GFRS.
3 Charge-changing cross sections and their approximations

As mentioned in Sect. 2, single-electron capture and loss cross sections, as exponential functions of the difference between HI charge state $q$ and fixed value of $q_m$, were used to consider a charge-changing process inside a gas. The equilibrated charge distribution was assumed with a Gaussian shape, mean value $q_m$, and standard deviation (width) $d$ [4,21,22]. Appropriate exponential functions were written using these values. Effective parameters of the exponential functions for capture and loss cross sections $\sigma_{q,q-1}$ and $\sigma_{q,q+1}$, respectively, are interconnected [21,22] if $q_m$ and $d$ are used. The lasts could be taken from the empirical systematics [21,24,25]. Parameters of the $\sigma_{q,q-1}$ function were derived with a fit to the cross sections given by the analytical formulae [31,32] and applied in respective simulations for $^{252}$No and $^{259}$Rf ERs passing through H$_2$ [23] and for $^{58}$Ni ions passing through N$_2$ [22].

3.1 Testing approximations for electron capture cross sections

Compatibility between $\sigma_{q,q-1}$ obtained for low energy HIs passed through H$_2$ [21,33,34] with the same values resulted in the approximation [30] was checked to get an idea of their applicability for simulations of the charge-changing process in the lightest gas. One can remind that the approximation [30] is based on the notion of a classical cross section introduced by Bohr and Lindhard [32]. The same test for the applicability of the empirical formula [31] was also performed. The results of both of these inspections are shown in Figs. 4 and 5.

Besides the approximations [30,31], a single universal scaling rule was later proposed with the use of reduced capture cross section $\sigma_{q,q-1}/q^{0.76}$ as a function of reduced energy $E/q^{0.4}$ [35]. This scaling is ‘extremely well defined for $E/q^{0.4} > 5$ keV/nucleon and includes all projectile charge states,’ as was mentioned in this work. According to Fig. 1 in [35], perfect data scaling is demonstrated, but for HIs not heavier than Fe. Thus, the applicability of this scaling rule has to be also tested for possible usage in simulations. In Fig. 6, the result of applying the scaling rule [36] to the same cross section data is shown. As one can see, this scaling gives appreciably worse results than those proposed in [31,32], as shown in Figs. 4 and 5.

Note that in the subsequent application of any approximation, it should be considered that initial charge-state distributions for heavy ERs are assumed to be very broad, as mentioned in Sect. 1. For example, for $^{252}$No ERs produced in the $^{206}$Pb($^{48}$Ca,2$n$) reaction and knocked out from a relatively thin target with the mean energy of 39 MeV, the initial charge states cover the range of +10 $\leq q \leq$ +80, as will be further considered in Sect. 4. Under these conditions, the appropriate reduced energies are in the regions of $42 \geq E_r \geq 13$, $31 \geq E_r \geq 7.2$, and $62 \geq E_r \geq 27$ for the $E_r$ scaling proposed in [31,32] and [36], respectively (see Figs. 4–6). In the following, an exponential function of the type $\sigma_r = a_r \exp(b_r E_r)$ was used to fit a large set of experi-
mental single-electron capture cross sections, with $\sigma_r$ and $E_r$ being a reduced cross section and reduced energy, respectively, according to three different definitions proposed in [31,32,36]. Single-electron capture cross sections thus obtained, which correspond to the regions of reduced energies mentioned above, vary in the ranges of $(6.9\pm0.4) \leq \sigma_{q,q-1} \leq (323\pm12)$, $(6.0\pm0.4) \leq \sigma_{q,q} \leq (147\pm9)$, and $(4.7\pm0.5) \leq \sigma_{q,q+1} \leq (131\pm8)$ in the units of $10^{-16}\text{cm}^2$. The indicated errors correspond to the 95% confidence level, as shown in Figs. 4, 5, and 6. The transition from scaling [31] to [36], corresponds to the degradation in the quality of fits (increase in $\chi_r^2$ values) leading to an increase in relative uncertainties of the cross sections.

From now on, the exponential approximation for $\sigma_{q,q-1}$ cross sections with the reduced energy $E_r = E/q^1/7$ [31] and fitted parameters $a_c = 8.77 \pm 0.29$, and $b_c = -0.0614 \pm 0.0013$ will be utilized in simulations of charge-changing process for heavy ERs.

3.2 Empirical approximations of B. Franzke [37]

In 1981, B. Franzke proposed two empirical formulae for single-electron capture and loss cross sections [37]. They are a power function of $q/q_m$ with the positive and negative index of power $p$ for $\sigma_{q,q-1}$, and $\sigma_{q,q+1}$, respectively. The value of $p$ depends on the ratio $q/q_m$. Good agreement between experimental data and calculated values was shown for the loss cross sections of 1.4 MeV/nucleon U ions colliding with N$_2$. Simultaneously, no comparison for capture cross sections was presented, and thus testing both of these formulae was of interest for their possible application.

According to Eq. (1) presented in [37], the electron capture cross section, omitting some factors, is approximated by a function of $(q/q_m)^p$, where $p$ depends on HI charge $q$ ($p = 4$ for $q \leq q_m$ and $p = 2$ for $q \geq q_m$). For low energies (in a non-relativistic approximation), reduced cross section $\bar{\sigma}_{q,q-1}$ transformed from Eq. (1) in [37] becomes a function of $(q/q_m)^p$ and can be written as

$$\bar{\sigma}_{q,q-1} = 230.5E^2\sigma_{q,q-1}/q_m^2/Z^{1/2},$$

where $\sigma_{q,q-1}$ is the experimental cross section expressed in the units of $10^{-16}\text{cm}^2$, and $E$ is the HI energy in MeV/nucleon. An attempt was made to get the data scaling with the reduced cross section values determined by Eq. (4) as a function of $q/q_m$ and the same $\sigma_{q,q-1}$ data presented in Figs. 4, 5, and 6. Such data repre-
The same as in Figs. 4, 5, and 6 but for reduced capture cross sections \( \tilde{\sigma}_{q,q-1} \) according to Eq. (4), which is a function of \( q/q_{m}^{p} \), where \( q_{m}^{p} \) is a power function fitted to the \( q_{m} \) data shown in Fig. 1. See the text for details.

For single-electron loss cross sections, an empirical formula proposed by the author (Eq. (2) in [37]) was also tested. It seems more convenient for practical routine simulations than more elaborated approaches [38,39]. For low energies (in a non-relativistic approximation), reduced electron loss cross section \( \tilde{\sigma}_{q,q+1} \), according to Eq. (2) in [37], can be written as

\[
\tilde{\sigma}_{q,q+1} = 1.3239E^{1/2}q_{m}^{2}\sigma_{q,q+1}/10^{Y},
\]

where \( \sigma_{q,q+1} \) is expressed in the units of \( 10^{-16} \) cm\(^2\), and \( Y = (0.71 \log Z)^{3/2} \) is the parameter deduced from calculated binding energies for the outermost electrons [37]. As for capture cross sections, the reduced loss cross section is a function of \( (q/q_{m})^{p} \), in which index \( p \) depends on \( q \) (\( p = -2.3 \) for \( q \leq q_{m} \) and \( p = -4 \) for \( q \geq q_{m} \) [37]).

Unfortunately, there is a lack of available data on \( \sigma_{q,q+1} \), corresponding to HIIs colliding with H\(_{2}\) [21], which have appropriate masses and energies. For further analysis, the data for U ions at rather high energy of 1.4 MeV/nucleon [37] and Fe ions at \( E \leq 0.357 \) MeV/nucleon [40] were added to the data set compiled in [21]. The experimental cross sections [21,37,40] were converted into reduced ones using Eq. (5), and such scaling was tested with the empirical \( q_{m} \) values resulted in the approximations shown in Figs. 1 and 2. The \( \tilde{\sigma}_{q,q+1} \) values were plotted as a function of \( q/q_{m} \), and fitted with a power function \( \sigma_{pl}(q/q_{m})^{p_{pl}} \), as shown in Fig. 8. The best fit to the data (the least \( \chi^{2} \) values) was obtained with the \( q_{m}^{p_{pl}} \) approximation shown in Fig. 1. The cross section data for iodine ions at \( E < 0.04 \) MeV/nucleon (see [21] and Fig. 8) were ignored in the
fit since they significantly deviated from the general dependency $\sigma_{q,q+1} = f(q/q_m)$ observed for the tested $q_m$ values. Parameter values obtained with the power function fit to the $\sigma_{q,q+1}$ data using the $q_m^{pl}$ values are the following: $a_{pl} = 0.153 \pm 0.010$ and $b_{pl} = -2.59 \pm 0.15$.

For $^{252}$No at the highest initial charge state $q \simeq +80$, the $\sigma_{q,q+1}$ value thus obtained corresponds to $3 \cdot 10^{-16}$ cm$^2$, which is less than the respective $\sigma_{q,q-1}$ value by several magnitude orders. The loss cross sections become comparable with the capture ones at the charge states from +3 to +10, i.e., in the vicinity of the equilibrated charge. At these charge states, the electron loss cross sections vary in the region of $(15 \pm 4) \geq \sigma_{q,q+1} \geq (0.68 \pm 0.07)$ in the units of $10^{-16}$ cm$^2$ with the errors corresponding to the 95% confidence level, as shown in Fig. 8.

### 3.3 New empirical approximations for charge-changing cross sections

Scaling of experimental data according to the ratio of $q/q_m$ could be the right way to a general description of charge-changing cross sections, bearing in mind its quite proper application to the estimates of $\sigma_{q,q-1}$ (see Sect. 3.2). In this context, it was revealed that a good scaling for $\sigma_{q,q-1}$ could be obtained with $q/q_m$ earlier used as an argument. A power function and the exponential function using $q/q_m$ were used for the data fit. According to $\chi^2_r$ criteria, the exponential function using the $q/q_m$ ratio showed better results than those obtained with the power one for any choice of $q/q_m$.

In Fig. 9, the electron capture cross section data [21,34,35] are shown as a function of $q/q_m$ corresponding to the best data scaling. Parameter values obtained with this approximation vary in the region of $(10.3 \pm 0.5) \leq \sigma_{q,q-1} \leq (116 \pm 12)$ (in the units of $10^{-16}$ cm$^2$) and with errors corresponding to the 95% confidence level, as shown in Fig. 9. As one can see, these estimates somewhat differ from the previous ones obtained with approximations using scalings according to [31, 32, and 36] (see Sect. 3.1). Simple scaling was also found for the single-electron loss cross sections expressed with ratio $\sigma_{q,q+1}/\sigma_{q,q-1}$ as a function of $q/q_m$ in applying this approach to the data [21,37,40]. As in the previous cases, two variants with the $q_m^{pl}$ and $q_m^{lf}$ mean charges were tested. The best scaling was obtained with the $q_m^{pl}$ charges, according to the results of fits using the exponential function

$$a_{el} \exp(b_{el} x)$$

and power one $a_{pl} x^{b_{pl}}$ for $x = q/q_m$. The result of the power function fit, corresponding to the least $\chi^2_r$ value, is shown in Fig. 10. Parameter values for this function are as follows: $a_{pl} = 0.345 \pm 0.036$, $b_{pl} = -4.32 \pm 0.19$. As in the previous case, dealing with the $^{252}$No reduced cross sections (see Fig. 8), $\sigma_{q,q+1}$ values at the highest initial charge states are much less than the respective ones for $\sigma_{q,q-1}$. The $\sigma_{q,q+1}$ values become comparable with the $\sigma_{q,q-1}$ ones in the equilibrated charge vicinity, i.e., at the charge states from +3 to +10. At these charge states, according to the approximation, $\sigma_{q,q+1}$ vary in the region of $(2.5 \pm 1.2) \geq \sigma_{q,q+1} \geq (0.55 \pm 0.09)$ (in the units of $10^{-16}$ cm$^2$) and with errors corresponding to the 95% confidence level shown in Fig. 10). These estimates correspond to the $\sigma_{q,q-1}$ approximation shown in Fig. 4.

The empirical scalings considered in this section and Sect. 2 can be used to estimate the equilibrated mean charge, single-electron capture, and loss cross sections in rarefied H$_2$ for low energy HIIs and ERIs produced in fusion–evaporation reactions. The results of these approximations are summarized in Table 1.

The approximations presented in Table 1, being applied to $^{252}$No ionized ERIs at their charge states on the assumed region’s edge, showed a significant spread in the cross section values, as mentioned above. In Fig. 11, single-electron capture and loss cross sections are shown as functions of charge states $q$ assumed for
and power functions presented in Table 1. As one can see in Fig. 11, empirical mean equilibrated charges $q_{m}^{e}$ and $q_{m}^{f}$ used in these approximations differ from those corresponding to $\sigma_{q,q-1} = \sigma_{q,q+1}$, that is also in Fig. 10. Some explanations of this difference one can find in [21].

### 4 Monte Carlo simulations of charge-changing process for ERs

The ERs knocked out from relatively thin targets by a HI beam have well-determined energy distributions and forward peaked angular distributions, as the results of their straight-forward production in the complete fusion–evaporation reactions. These distributions can be obtained with Monte Carlo (MC) simulations considering the evaporation of light particles from compound nuclei formed in the reaction and processes of stopping and multiple scattering of ER atoms inside a target [41].

In Fig. 12, the energy, angular and charge distributions for $^{252}\text{No}$ produced in the $^{206}\text{Pb}(^{48}\text{Ca},2n)$ reaction are shown. The first two were obtained as described earlier [41]. The input charge distribution will be considered below. The energy and charge distributions will be used for the MC simulations of charge distributions for No ions escaped a target, whereas the angular distributions are given for reference.

According to the previous considerations [4, 22, 23], the distance of HI between successive charge-changing collisions is determined as

$$ l = -\lambda \ln P, $$

where $\lambda$ is the mean free path between two charge-changing collisions and $P$ is the probability for HI to survive flight path $l$ without any collisions (a random number between 0 and 1). The mean free path $\lambda$ is related to the total charge-changing cross section $\sigma_{tot} = \sigma_{q,q-1} + \sigma_{q,q+1}$ according to the relation:

$$ \lambda = 1/(n\sigma_{tot}), $$

Presented parameter values correspond to $E$ in MeV/nucleon and $Y = [0.71 \log(Z)]^{3/2}$. $\sigma_{q,q-1}$ and $\sigma_{q,q+1}$ are obtained in the units of $10^{-16}$ cm$^2$.
Single-electron capture and loss cross sections, $\sigma_{q,q-1}$ and $\sigma_{q,q+1}$, respectively, for the 39 MeV $^{252}\text{No}$ ions moving in H$_2$ are shown as a function of ion charge state $q$ (lines with a shadow area corresponding to the 95% confidence band). The cross section scalings, which are shown in Figs. 4 and 8, and Figs. 9 and 10, were used for the calculations presented in the upper and bottom panel, respectively. See details for the scaling functions in Table 1 where $n$ is the molecular density of a gas in cm$^{-3}$. For the $\sigma_{q,q-1}$ and $\sigma_{q,q+1}$ cross sections, the suitable approximations obtained in Sect. 3 can be used. The multiple electron capture and loss processes are neglected. The absolute cross section values for $^{252}\text{No}$ ions moving in rarefied H$_2$ are shown as a function of their charge states in Fig. 11. Simple estimates with these cross section values and Eq. (7) show that for the H$_2$ molecular density of 3.3·$10^{-16}$ cm$^{-3}$ at the pressure of 1 Torr, $\lambda$ increases from 2.5·$10^{-4}$ cm ($q = +80$) to 7.5·$10^{-2}$ cm ($q = +5$), as $\sigma_{\text{tot}}$ decreases from 3·$10^{-14}$ to 3·$10^{-16}$ cm$^2$. Cross sections shown in Fig. 11 will be further used in simulations of the charge state distributions for No ERs.

In Fig. 13, the evolution of the input charge states for $^{252}\text{No}$ ions knocked out from the 0.4 mg/cm$^2$ $^{206}\text{Pb}$ target into the H$_2$ gas under the pressure of 1 Torr is shown. Mean charges $q_m$ and corresponding mean penetration depths $L_m$ were obtained with the statistical analysis of respective distributions resulted in MC simulations. The charge-changing cross sections shown in Fig. 11 (upper panel) and the energy distribution (Fig. 12) were used in these simulations. Mean charges are shown as a function of $L_m$ increasing with the number of the charge-changing collisions (starting from the first one). As one can see, at $L_m \gtrsim 1.5$ cm, $q_m$ becomes equal to 5.75 irrespective of the input charge state. This value is achieved with the number of the
collisions becoming higher as the input charge state increases. Note that the mean charge value, corresponding to \( \sigma_{q,q+1} = \sigma_{q,q-1} \), is equal to 5.65 (see Fig. 11), slightly different from the \( q_m \) value obtained in simulations. Such a difference is the result of different slopes in \( \sigma_{q,q+1}(q) \), and \( \sigma_{q,q-1}(q) \) cross sections [21].

The value of the penetration depth thus obtained, corresponding to setting a charge equilibration, can be compared to a similar one observed in an experiment. For instance, the 13.9 MeV Br ions with charge states +6, +7, +8, and +10 achieved a charge equilibration at the H\(_2\) target thickness of \( W_t \gtrsim 4.5 \times 10^{16} \) mol/cm\(^2\) (see Fig. 3 in [42]). The value is close to the one obtained in these simulations (\( W_t \gtrsim 4.9 \times 10^{16} \) mol/cm\(^2\)), corresponding to \( L_m \gtrsim 1.5 \) cm at the H\(_2\) molecular density of 3.3 \( \times 10^{16} \) cm\(^{-3}\).

In further consideration, it was assumed that the reliable initial charge-state distribution for ionized \(^{252}\)No ERs escaping the \(^{208}\)Pb target is the composition of the ‘normal solid’ component and of the non-equilibrated one. The ‘normal solid’ component corresponds to the equilibrated charge-state distribution for HIs passed through a solid [13–15], whereas the non-equilibrated component has charges which are much higher than ‘normal’ ones [16–20] (see Sect. 1). The analysis of charge-state distributions for ERs of different masses and velocities showed that the number of charge states for the non-equilibrated component \( N_{neq} \) exceeds those for the equilibrated one \( N_{eq} \) by 2 to 4. Mean charges \( q_m^{neq} \), and standard deviations \( d_{neq} \) of the non-equilibrated component also exceed the same parameters of the equilibrated one \( q_m^eq \) and \( d_{eq} \) within the same factor [19,20]. In subsequent simulations for \(^{252}\)No ERs escaping the \(^{208}\)Pb target, it was assumed that \( N_{neq} = 2N_{eq} \). Parameters of distributions were assumed to be as those: \( q_m^{neq} = 2.5q_m^eq \) and \( d_{neq} = 2.5d_{eq} \).

In Fig. 14, the evolution of the charge distribution for the ionized \(^{252}\)No ERs knocked out from the 0.4 mg/cm\(^2\) \(^{208}\)Pb target into the H\(_2\) gas under the pressure of 1 Torr is shown. It starts from the initial charge and energy distributions for ERs (Fig. 12). Its evolution is further considered in the same way as was done above for fixed input charges, using charge-changing cross sections similar to those shown in Fig. 11 (upper panel). In Fig. 14, charge–penetration depth distributions are shown for the definite number of the charge-changing collisions for No ERs, as the respective smoothed contour maps. The distributions of the penetration depths in the charge-changing regime are close to the similar ones for distances from the target, taking into account a preferred movement of ERs in a forward direction (see Fig. 12). Note that the energy losses of No ERs along the penetration depths obtained in simulations are negligible at the respective H\(_2\) pressure. As one can see, after 60 collisions, the charge distribution becomes close to the equilibrated one with the mean charge approaching the value obtained above (see Fig. 13). The number of collisions is a little more than the one obtained at \( q_{inp} = +50 \) due to higher charge states in the non-equilibrated component of the input distribution. Its presence also leads to the two-humped distribution of penetration depths for ERs with fully equilibrated charges achieved approaching 80 collisions (see Fig. 14).

A quantitative analysis of thus obtained charge distributions for different charge-changing cross sections and target thicknesses was performed with double-Gaussian fitting the simulated data. The values of fitted parameters as functions of the number of collisions \( n_{col} \) leading to the charge-state changes are shown in Fig. 15. As one can see in the figure, charge equilibration in gas is established at \( n_{col} \gtrsim 20 \) for the initial distribution corresponding to the ‘solid’ equilibrated-charge component. The parameters of \( q_m \) and \( d \) become such as those that would be expected for the charge equilibration. A similar equilibration for the initial charge distribution corresponding to the non-equilibrated component occurs at \( n_{col} \gtrsim 60 \). According to this consideration, the non-equilibrated component in the charge distribution is still resolved at \( n_{col} = 62 \) with the probability of \( 1 - N_{eq}/(N_{eq} + N_{neq}) \approx 0.07 \). It could not extract from the simulations corresponding to \( n_{col} \geq 63 \) since the parameter values of \( q_m \) and \( d \) become close to those for the equilibrated component that dominates in these simulations.

The effect of the target thickness on the charge equilibration of ERs is tested with the same charge-changing cross sections, as in the previous case. Increasing target thickness is essential not only for a new DGFRS [11,12], as mentioned in Sect. 1, but for any GFRS bearing in mind the future experiments on the synthesis of SHN with \( Z > 118 \). It is one way to raise the production yield...
Fig. 14 The evolution of the charge distribution for the ionized $^{252}$No ERs knocked out from the 0.4 mg/cm$^2$ $^{206}$Pb target at their moving through the H$_2$ gas. The charge state–penetration depth distributions are shown as the respective smoothed contour maps corresponding to the definite number of the charge-changing collisions (increasing from 20 to 80).

Fig. 15 Parameter values of double-Gaussian fitting the charge distributions are shown as functions of the number of collisions, which are the results of charge-changing simulations for the ionized $^{252}$No ERs knocked out from the $^{206}$Pb target of the thickness of 0.4 and 1.0 mg/cm$^2$ (filled and open symbols, respectively). Circles and squares plot the results obtained with the charge-changing cross sections shown in the upper panel of Fig. 11 for the equilibrated (eq.) and non-equilibrated (neq.) components, respectively. Triangles plot similar results for the cross sections shown in the bottom panel of the figure of SHN, which can be synthesized in fusion–evaporation reactions with sub-pb cross sections, as estimated in theory (see, for example, [43,44] and Refs. therein). Figure 15 shows the changes in charge distribution parameters for the ionized $^{252}$No ERs knocked out from the 1.0 mg/cm$^2$ $^{206}$Pb target. Simulations were performed in the same way as the previous ones, but using the ER energy and charge distributions obtained for the 1 mg/cm$^2$ target (see Fig. 12). Similar parameter values were obtained comparing with the previous ones corresponding to the 0.4 mg/cm$^2$ target. Thus, after 80 collisions, all $^{252}$No ERs with the equilibrated charges occupy the same 0–10 cm distance from the target. One can state that the target thickness difference has a minor effect on the charge equilibration of the ionized ERs in the rarefied H$_2$ gas.
According to Fig. 11, electron capture cross sections for No ions with $q \gtrsim +20$ (the approximation with the scaling [31]) are about three times higher than those, corresponding to the similar one presented in Fig. 9. Simultaneously, electron loss cross sections for the same $q$ values, which correspond to the approximation of the reduced values [37], are about three times lower than those obtained with the $\sigma_{q,q+1}/\sigma_{q,q-1}$ ratio presented in Fig. 10. The effect of the cross section values on the equilibration of $^{252}\text{No}$ charge distributions and corresponding penetration depths was also tested comparing the results of previous simulations with those using the pair of cross sections shown in the bottom panel of Fig. 11. The evolution of the charge distribution for $^{252}\text{No}$ ERs knocked out from the 1.0 mg/cm$^2$ $^{206}\text{Pb}$ target was considered in the same manner as in previous cases, but with this pair of the cross sections. The results of the analysis of charge distributions obtained in these simulations are also shown in Fig. 15. As one can see, no significant differences were obtained for the parameter values. One can state that difference in the charge-changing cross sections for $q \gtrsim +20$ has a minor effect on the charge equilibration of ionized ERs in the rarefied H$_2$ gas. In any case, the equilibrated charge distribution is maintained somewhat between 60 and 80 charge-changing collisions.

The two-humped (bimodal) character of the distributions for the penetration depths (see Figs. 14 and 15) does not allow to obtain a direct connection between the penetration depth and the charge distribution evolving to the equilibration. In Fig. 16, the distributions of penetration depths are shown for the charge distributions close to the equilibration (60 collisions) and those corresponding to the charge equilibrium (80 collisions). Simulations were performed for the 1 mg/cm$^2$ target with the two pairs of the charge-changing cross sections shown in the upper and bottom panel of Fig. 11. As one can see, after 80 collisions, all $^{252}\text{No}$ ERs with the equilibrated charges occupy a distance of 0–12.5 cm from the target irrespective of the cross sections used in simulations.

At the end of this section, one can state that the difference in charge-changing cross sections and target thickness lead to minor changes in the final equilibrated charge distributions for heavy ERs produced in fusion–evaporation reactions. Despite their differences, the equilibrated charge distribution occupies 0-12.5 cm from a target. The mean charge is approaching the value determined by the equality of $\sigma_{q,q+1} = \sigma_{q,q-1}$. A relatively large number of collisions is caused by the equilibration process of the non-equilibrated component in the initial charge-state distribution. High capture cross sections for high charge states of the non-equilibrated component cause relatively small penetration depths into the rarefied gas for ERs escaped from a target. The mean value of the penetration depths, corresponding to the re-charging of the charge-equilibrated ERs, varies from 0.08 to 0.12 cm with about the same standard deviations (the values depend on the charge-changing cross sections used in simulations). These circumstances allow one to use the equilibrated charge-state distribution in trajectory simulations for heavy ERs passing through magnetic elements filled with rarefied H$_2$ gas.

5 Summary

The charge-changing process in rarefied H$_2$ gas was considered for ionized heavy evaporation residues (ERs) produced in the fusion–evaporation reactions in the present work. It was assumed that ERs knocked out from a target by heavy-ion (HI) projectiles have a broad charge-state distribution. Besides their differences, the equilibrated component contains a large charge range from 0-12.5 cm to a target. The mean charge is approaching the value determined by the equality of $\sigma_{q,q+1} = \sigma_{q,q-1}$. A relatively large number of collisions is caused by the equilibration process of the non-equilibrated component in the initial charge-state distribution. High capture cross sections for high charge states of the non-equilibrated component cause relatively small penetration depths into the rarefied gas for ERs escaped from a target. The mean value of the penetration depths, corresponding to the re-charging of the charge-equilibrated ERs, varies from 0.08 to 0.12 cm with about the same standard deviations (the values depend on the charge-changing cross sections used in simulations). These circumstances allow one to use the equilibrated charge-state distribution in trajectory simulations for heavy ERs passing through magnetic elements filled with rarefied H$_2$ gas.
cades, which significantly increase the ion charges of ERs over the equilibrated ones. Transformation of both these components into the equilibrated one in rarefied H$_2$ may occur at large distances from the target due to the number of electrons' captures that lead to charge equilibration.

Mean equilibrated charges of HIs in rarefied H$_2$ gas were estimated using two empirical formulae derived in the present work using available experimental data. The first one was the result of a power function fitting the HI data [27,28] and the data obtained with the Dubna gas-filled recoil separator (DGFRS) for the heaviest ERs [26]. These mean equilibrated charges corresponded to the $q_{eq}$ values. The second formula was derived using a linear function fitting the DGFRS data only [26] relating to the $q_{eq}$ equilibrated charges. Comparing the $q_m$ values obtained with these formulae showed their mutual agreement within ±10% at the velocities of $1.8 \leq v/v_0 \leq 3.0$.

A way to charge equilibrium is determined by single-electron capture and loss cross sections $\sigma_{q,q-1}$ and $\sigma_{q,q+1}$, respectively, which are changed with the charge states of ERs moving in gas. Several empirical approaches to the estimates of charge-changing cross sections [31,32,36,37] were examined to choose the best one describing the HI data at the energies of 0.035–1.4 MeV/nucleon [21,33,34,36,39]. The exponential function fitting the $\sigma_{q,q-1}/q$ data against the $E/q^{1/2}$ values (scaling proposed in [31]) was making the best among others according to $\chi^2$ criteria. For the electron loss cross sections, the empirical formula proposed by Franzke [37] revealed a rather good scaling of low energy data using the $q/q_{eq}$ values as an argument. Reduced electron loss cross sections $\sigma_{q,q+1}$ in the dependence on $q/q_{eq}$ were well fitted with a power function.

New empirical formulae for the $\sigma_{q,q-1}$ and $\sigma_{q,q+1}$ cross sections were also proposed, which disclosed a rather good agreement with experimental data according to the $\chi^2$ criteria. Thus, an appropriate scaling was obtained for the $\sigma_{q,q-1}$ data in the dependence on $x = q/q_{eq}$. The available data were well fitted with the exponential function in the form of $a \exp[b/(x + c)]$. A quite good approximation was also obtained for the $\sigma_{q,q+1}/\sigma_{q,q-1}$ ratios displayed as a function of $q/q_{eq}$. The cross section ratios were well fitted with a power function.

In the application of the obtained formulae to the ionized $^{252}$No ERs produced in the $^{206}$Pb($^{48}$Ca,2$n$) reaction, the approximations proposed for the $\sigma_{q,q-1}(q)$ cross section showed a difference corresponding to a factor of 3 for the calculated values at $q \gtrsim 20$. A similar mutual disagreement was also obtained for the $\sigma_{q,q+1}(q)$ cross section approximations. For the $q \ll q_m$ charge states, $\sigma_{q,q+1}(q)$ values thus obtained differed from each other even more. However, this difference is not critical in the initial stage of the equilibration process for highly ionized ERs.

Monte Carlo simulations, similar to those used earlier [4,22,23], but based on the approximations obtained for charge-changing cross sections, allowed one to get an idea of the rapidity of the equilibration process for initially highly ionized ERs. By the example of $^{252}$No, these simulations showed that the ‘solid’ equilibrated charge component with the initial $q_m \simeq 24$ becomes the equilibrated one in rarefied H$_2$ gas after ~30 charge-changing collisions. It has $q_m \simeq 6$ and stretches for 0–5 cm from a target. Much higher charge states of the non-equilibrated component become equilibrated after ~65 collisions at the same penetration depth from the target. The charge equilibration of ionized heavy ERs poorly depends on the target thickness. Such fast (short-range) equilibration allows one to use mean equilibrated charges in simulations of ERs transmission through gas-filled magnetic-optical systems.

A similar approach could be applied to the consideration of charge equilibration for ionized ERs in rarefied He gas, bearing in mind many gas-filled separators working with He [2–5,7,8,23]. In this case, distances between successive charge-changing collisions for HIs do not differ so much from those estimated for H$_2$. Indeed, an exponential approximation to the $\sigma_{q,q-1}$ data [21,31,45] scaled according to [31] gives us cross section values of $(3.5–160) \times 10^{-16}$ cm$^2$ for the initial $^{252}$No charge states between +10 and +80. These cross sections lead to the respective distances between charge-changing collisions in the range of $(8.7–0.19) \times 10^{-2}$ cm for the He atomic density of $3.3 \times 10^{16}$ cm$^{-3}$ at the pressure of 1 Torr. Data on $\sigma_{q,q+1}$ for HIs passed through He should be similarly considered and approximated, bearing in mind their importance for the charge-changing cross section estimates in the vicinity of the ER equilibrated charge.

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