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Equilibrium charge-state distributions for heavy ions exiting carbon foils

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It has been recently shown that equilibrium charge-state distributions (ECSD's) of heavy ions exiting carbon foils are well fitted by the χ^2 model, the Gaussian model, and the reduced χ^2 model in the low-, intermediate-, and high-velocity regions, respectively. A qualitative interpretation of these statistical distributions is given in the present work, and it is pointed out that the reduced χ^2 model is mathematically identical to the χ^2 model if the charge state i is replaced by $Z-i$ (Z is the projectile atomic number) in the probability density function. This equivalence is physically explained by the existence of a charge-state limitation in these two velocity regions. The author discusses also the validity of semiempirical relations giving the mean charge $\bar{i}$ and the standard deviation of the charge s in the different velocity domains. At sufficiently high velocities ($E/M \gtrsim 100$ keV amu^{-1}, $Z \lesssim 20$), experimental data available seem to indicate that $\bar{i}$ and s depend monotonically on Z and v, but this is not the case at lower velocities where shell effects play a major role. Finally, the author analyzes ECSD's observed for heavy ions, $59 \lesssim Z \lesssim 82$, at low energies ($E/M \lesssim 7$ keV amu^{-1}) and finds that these ECSD's are also very well fitted by the χ^2 model proposed previously for lighter ions, $Z \lesssim 26$, at $E/M \lesssim 20$ keV amu^{-1}. However, for these heavy ions, ECSD's are already Gaussian for $E/M \gtrsim 5$ keV amu^{-1}.

I. INTRODUCTION

The experimental and theoretical situation concerning equilibrium charge states and charge-changing cross sections of heavy ions in gaseous and solid media was thoroughly reviewed by Betz.[1] More recent data on the description of collisions of heavy ions in solid media can be found in Datz's review articles.[2] However, for the present, no theory allows the calculation of equilibrium charge-state distributions (ECSD's) for heavy ions in solids. In this work we will consider carbon-foil targets only. Previous analyses of ECSD's observed for heavy ions after passing through carbon foils[3-5] have shown that these distributions are well fitted using simple statistical models: (a) the χ^2 model[3,4] at low velocities ($v \lesssim 2\times10^8$ cm s^{-1}, $Z \lesssim 26$), (b) the reduced χ^2 model[5] at high velocities ($v > 3.6\times10^8 Z^{0.45}$ cm s^{-1}, $7 \le Z \le 36$), and (c) the Gaussian model at intermediate velocities.[3,4] These models allow the predictions of equilibrium charge-state fractions if the mean charge $\bar{i}$ and the standard deviation of the charge s can be precisely determined from empirical or semiempirical relations. Such relations have been found for heavy projectiles at high velocities.[5] At intermediate velocities no relation has been obtained until now for describing accurately the dependence of $\bar{i}$ and s on Z and v. For projectiles ($5 \le Z \le 26$) at low velocities ($v \simeq 2\times10^8$ cm s^{-1}), Lennard *et al.*[6] have recently observed that $\bar{i}$ and s exhibit oscillatory behavior as a function of Z.

The aims of this work are (a) to propose an interpretation for the different statistical distributions used in fitting ECSD's in the various velocity domains of heavy ions exiting carbon foils, (b) to discuss the possibility of predicting ECSD's of heavy ions in carbon, a possibility which depends principally on the existence of precise relations giving $\bar{i}$ and s as a function of v and Z, and (c) to verify the validity of the χ^2 model for ions with $Z > 26$ exiting carbon foils at low velocities.

II. VARIATION OF THE SHAPE OF ECSD'S WITH THE PROJECTILE VELOCITY

A. Analysis of argon data

The ECSD's of argon ions at the exit of a carbon foil have been measured in an extended energy range.[7-12] This is the reason why we choose this projectile for illustrating the behavior of ECSD's as a function of the ion velocity.

The mean charges $\bar{i}$ of ECSD's observed for argon ions in carbon foils are plotted in Fig. 1 as a function of the energy per nucleon of the emerging ions. Figure 1 shows a smooth increase of $\bar{i}$ with E/M.

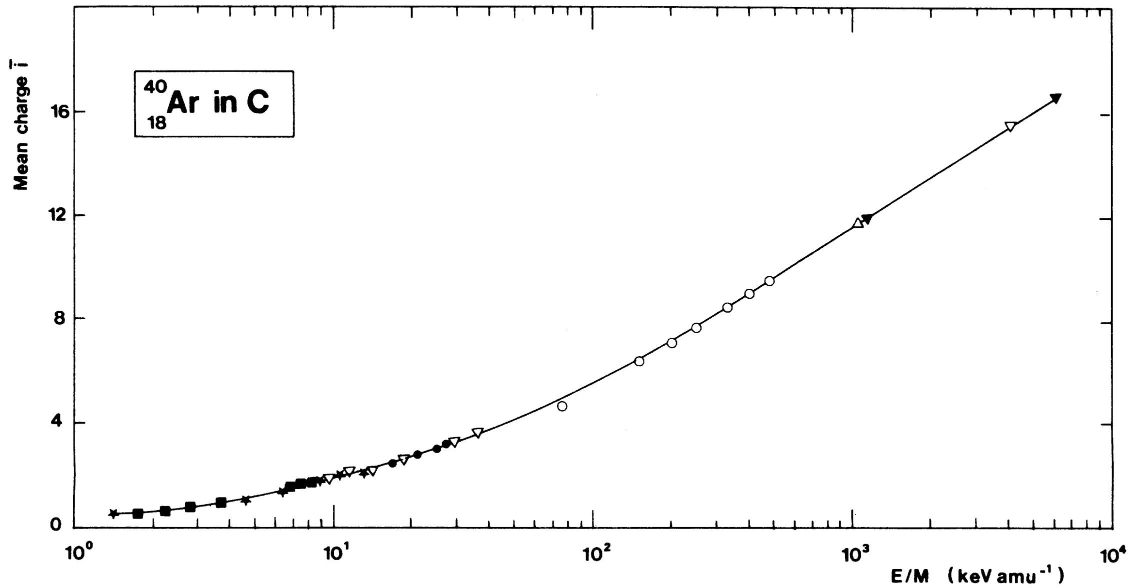

FIG. 1. Mean charges of observed ECSD's for argon ions having passed through carbon foils as a function of the energy per nucleon after the foil. Data are from Hvelplund *et al.* (Ref. 8) (★), Turkenburg *et al.* (Ref. 10) (□), Smith and Whaling (Ref. 7) (▽), Knystautas and Jomphe (Ref. 12) (○), Clark *et al.* (Ref. 11) (△), and Baron (Ref. 9) (▼). Solid curve is drawn to guide the eye.

In Fig. 2 observed values of the standard deviation s are plotted as a function of the energy per nucleon at the exit of a carbon foil for argon ions. Within the experimental errors, s varies smoothly with the beam energy; it first increases then reaches a plateau value for $30 \lesssim E/M$ (keV amu^{-1}) $\lesssim 200$ and finally decreases slowly at higher energies (notice the logarithmic E/M scale).

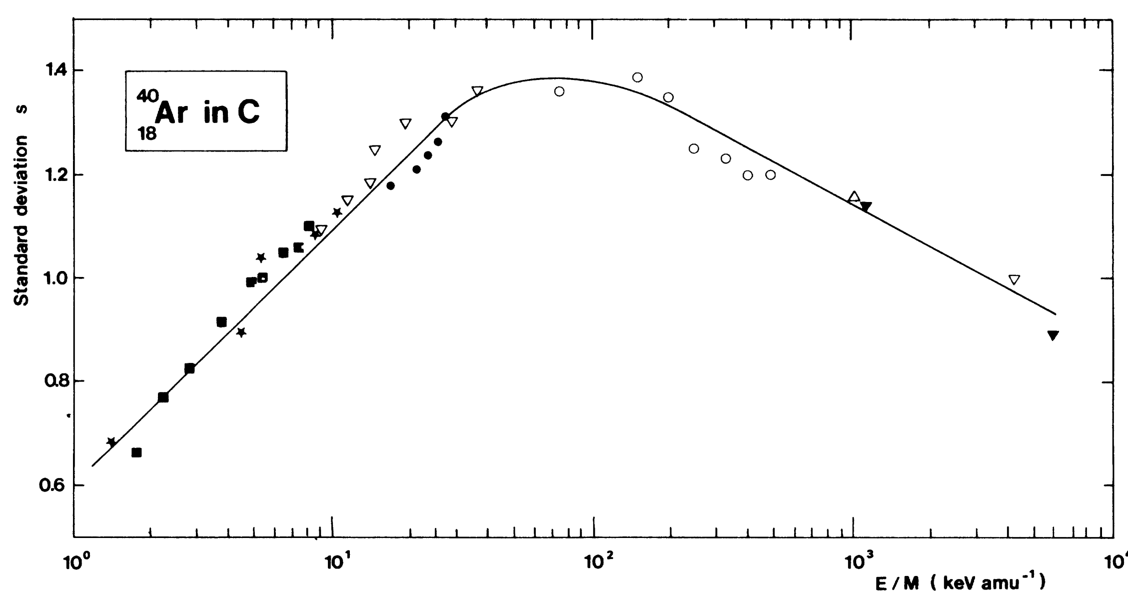

FIG. 2. Standard deviations of observed ECSD's for argon ions having passed through carbon foils as a function of the energy per nucleon after the foil. Data are extracted from the same references as those shown in Fig. 1. Solid curve is drawn to guide the eye.

Figure 3 shows observed and calculated ECSD's for argon ions in carbon foils for different emerging beam energies. Calculations have been made utilizing the most appropriate model, i.e., the χ^2 model[4] for $E/M \leq 36$ keV amu^{-1}, the Gaussian model[4] for $36 \leq E/M$ (keV amu^{-1}) ≤ 1160, and the reduced χ^2 model[5] for $E/M \geq 4120$ kev amu^{-1}. For these calculations we have employed the observed values of $\bar{i}$ and s. Very good agreement is obtained between observed and calculated distributions (see Fig. 3).

B. Interpretation of the different shapes of ECSD's in carbon

1. Symmetric distributions

The ECSD's of ions after passing through carbon foils result from many random phenomena: electron capture, electron loss, and excitation processes occurring in the solid which can be altered by exit surface effects and relaxation processes following emergence, principally autoionizing and Auger effects (see, for instance, Ref. 2). The final charge state can thus be represented as the sum of many random independent variables and is, by virtue of the "central-limit theorem",[13] asymptotically normally distributed (Gaussian model). Theoretical approaches made by Garcia,[14] and recently by Åberg and Goscinski,[15] have shown that ECSD's of heavy ions in foils are approximately Gaussian.

2. Asymmetric distributions

It is clear that asymmetric ECSD's (with negative coefficient of skewness) observed at high velocities are due to the fact that the maximum charge state is equal to Z (see Fig. 3 and Ref. 5). At low velocities, where shell and surface effects are expected to play a major role, interpretation of observed asymmetric ECSD's (with positive coefficient of skewness) is not so obvious. In this case, however, the following observations indicate that the shape of ECSD's is also due to a limitation of the charge state (the maximum charge is equal to zero or minus one):

(a) The probability density function fitting ECSD's observed at high velocities is identical to that obtained at low velocities if we change the random variable i into $Z-i$. Indeed, it has been shown previously[4] that, in the low-velocity domain, the variable

$$t=c(i+2) \tag{1}$$

follows the χ^2 law with ν degrees of freedom.

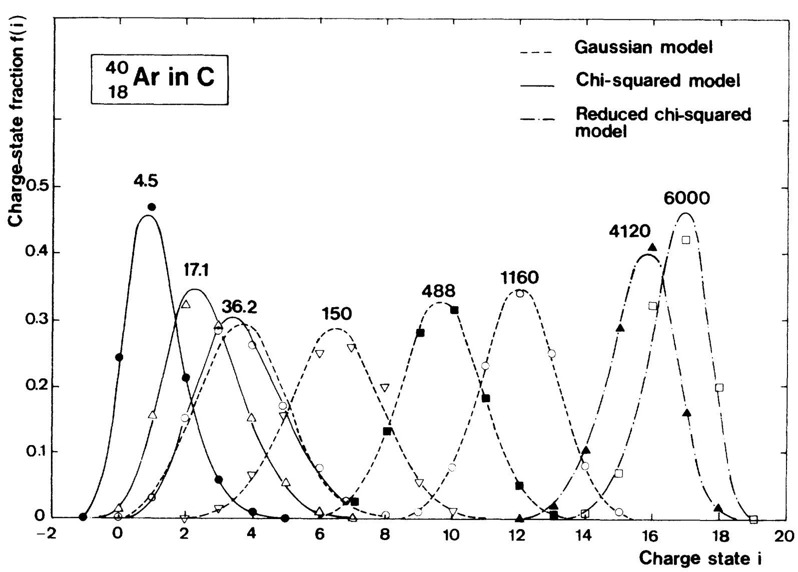

FIG. 3. Observed and calculated equilibrium charge-state fractions for argon ions and carbon foils at different energies per nucleon for the emerging ions (indicated in keV amu^{-1} above the corresponding curves). Data are from Hvelplund *et al.* (Ref. 8) (●, 4.5 keV amu^{-1}), Lennard *et al.* (Ref. 6) (△, 17.1 keV amu^{-1}), Smith and Whaling (Ref. 7) (○, 36.2 keV amu^{-1}), Knystautas and Jomphe (Ref. 12) (▽, 150 keV amu^{-1} and ■, 488 keV amu^{-1}), Baron (Ref. 9) (○, 1160 keV amu^{-1} and □, 6000 keV amu^{-1}), and Clark *et al.* (Ref. 11) (▲, 4120 keV amu^{-1}).

Then, the density of probability $f(i)$ of the charge state i is given by[4]

$$f(i)=c[2^{\nu/2}\Gamma(\nu/2)]^{-1}t^{\nu/2-1}e^{-t/2}\ ,\quad 0<t<\infty \tag{2}$$

where Γ is the gamma function and

$$c=2(\bar{i}+2)/s^2\ , \tag{3}$$

$$\nu=2(\bar{i}+2)^2/s^2\ . \tag{4}$$

In the high-velocity range, the density of probability of i is given by the same Eq. (2) where t, c, and ν are now given by the following relations[5]:

$$t=c(Z-i+2)\ , \tag{5}$$

$$c=2(Z-\bar{i}+2)/s^2\ , \tag{6}$$

$$\nu=2(Z-\bar{i}+2)^2/s^2\ . \tag{7}$$

(b) When the projectile can form a negative ion, ECSD's are Gaussian for smaller values of E/M than for nearby ions which cannot be found as negative ions. This situation is illustrated in Figs. 4 and 5 where ECSD's in carbon at low velocities, observed and calculated by the Gaussian and by the χ^2 model, have been plotted for ^{14}N and ^{23}Na ions, and for ^{16}O and ^{12}C ions, respectively. The ECSD's for ^{14}N and ^{23}Na projectiles which cannot form negative ions are better fitted by the χ^2 model than by the Gaussian model. In contrast, ECSD's for ^{16}O and ^{12}C projectiles are still symmetric and Gaussian at E/M as low as 6 keV amu^{-1} because finite fractions of negative ions exist for these projectiles.

3. *Comparison with ECSD's in gaseous targets*

It has been pointed out previously[16] that asymmetric ECSD's observed in gaseous targets are very well fitted by a χ^2 model with three parameters. We want to emphasize that this statistical model has not the same physical interpretation as the χ^2 model (with two parameters) describing ECSD's in carbon targets. Indeed, ECSD's in heavy gases are still asymmetric for very high velocities at which the fraction of neutrals is equal to zero. Moreover, it is well known that physical processes occurring during the passage of ions through matter are very different according to whether the target is a solid or a gas.[1,2]

III. PREDICTION OF ECSD'S

A. High-velocity region

All the ECSD's of heavy ions ($7\le Z\le 36$) in carbon at high velocities

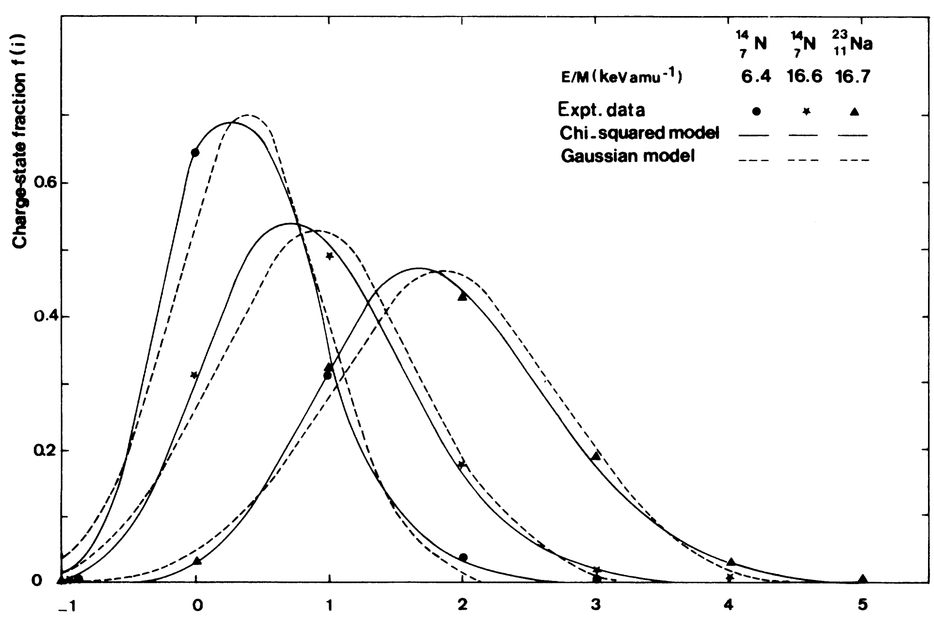

FIG. 4. Equilibrium charge-state fractions observed and calculated by the Gaussian model and by the χ^2 model, for N and Na ions in carbon foils. Data are from Hvelplund *et al.* (Ref. 8) for N at 6.4 keV amu^{-1} and Na at 16.7 keV amu^{-1}, and from Lennard *et al.* (Ref. 6) for N at 16.6 keV amu^{-1}.

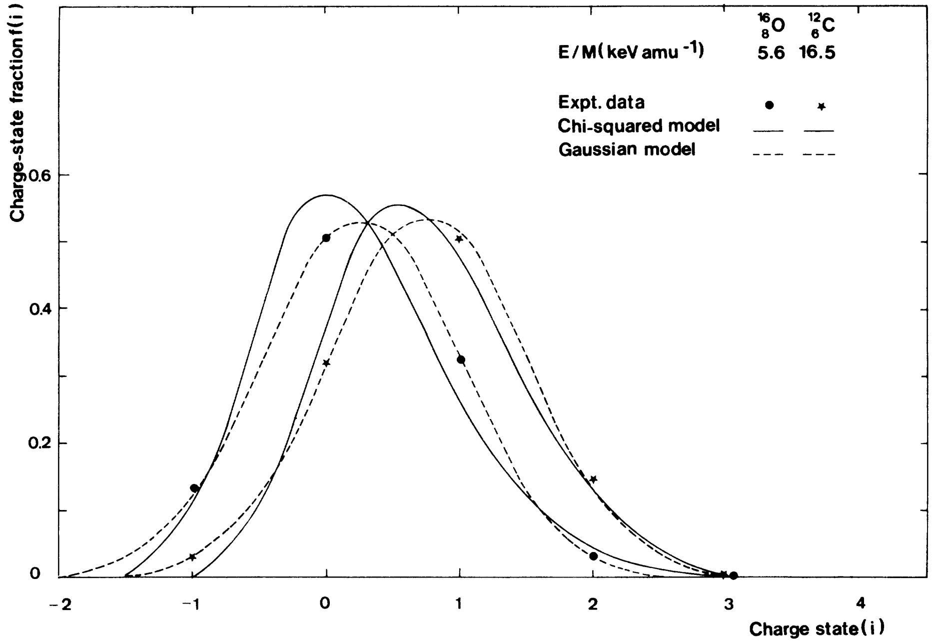

FIG. 5. Equilibrium charge-state fractions observed and calculated by the Gaussian model and by the χ^2 model for O and C ions in carbon foils. Data are from Hvelplund *et al.* (Ref. 8) for O at 5.6 keV amu^{-1}, and from Lennard *et al.* (Ref. 6) for C at 16.5 keV amu^{-1}.

$(v > 3.6\times 10^8 Z^{0.45}\ \mathrm{cm\,s^{-1}})$,

available in the literature, are very well fitted by the reduced χ^2 model we proposed recently.[5] More precisely, charge-state fractions $f(i)$ are given by Eqs. (2), (5), (6), and (7) of Sec. II which depend on the two parameters $\bar{i}$ and s. These parameters can be accurately determined using simple empirical relations depending only on v and Z (see Ref. 5). These results indicate that shell effects seem not very important at these high energies.

B. Intermediate-velocity region

In Figs. 6(a) and 6(b), available values of $\bar{i}$ and s have been plotted, respectively, as a function of the atomic number of the projectile for different values of E/M ranging from 23 to 1000 keV amu^{-1}. We considered only data for $Z \leq 26$ because of the scarcity of data for heavier projectiles. Figure 6 shows that the oscillatory behavior observed at 23 keV amu^{-1} by Lennard *et al.*[6] seems to disappear at $E/M > 100$ keV amu^{-1}. However, more data are needed, especially for $Z > 19$, to confirm this statement. Within the experimental errors, values of s are independent of the projectile velocity for $100 \lesssim E/M$ (keV amu^{-1}) $\lesssim 500$, $Z \leq 18$. The decrease of s for $E/M = 1000$ keV amu^{-1} is due to the fact that the projectile can be completely stripped at this high velocity.[5] In summary, for sufficiently high velocities ($E/M > 100$ keV amu^{-1}, $Z \lesssim 20$), $\bar{i}$ and s could be estimated from relations depending smoothly on Z and v. However, more experimental work needs to be done to establish these relations accurately. When $\bar{i}$ and s are known, charge-state fractions $f(i)$ are easily calculated by the Gaussian model

$$f(i) = [1/(2\pi s^2)^{1/2}]e^{-(i-\bar{i})^2/2s^2} . \tag{8}$$

C. Low-velocity region

At low velocities ($v \lesssim 2\times 10^8$ cm s^{-1}, $Z \leq 26$) where the Gaussian model is no longer valid,[4] there is a monotonic dependence of $\bar{i}$ and s on v for a given projectile (see Figs. 1 and 2 of this work and Figs. 1 and 2 of Ref. 4). However, the dependence of $\bar{i}$ and s on Z, at a given velocity, exhibits oscillatory behavior deducible from Figs. 1 and 2 of Ref. 4 and clearly shown by Lennard *et al.*[6] at $v \simeq 2\times 10^8$ cm s^{-1} for 21 projectiles, $5 \leq Z \leq 26$ (see Fig. 6, data at 23 keV amu^{-1}). Therefore, in this low-velocity

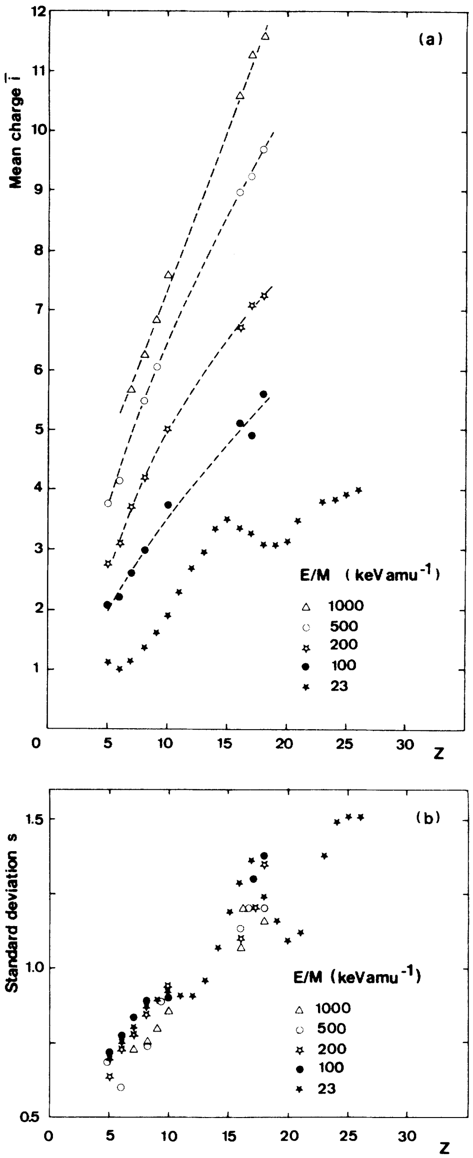

FIG. 6. Observed values of the mean charge (a) and of the standard deviation of the charge (b) for ECSD's in carbon as a function of Z, at different energies per nucleon for the emerging ions. Data at 23 keV amu^{-1} are obtained by interpolating the results of Table I in Ref. 6; other data are obtained by interpolating the results given in the tabulation of Wittkower and Betz (Ref. 19) except the following data: B at 100 keV amu^{-1} (Ref. 17), at 200 and 500 keV amu^{-1} (Ref. 18); Ne at 1000 keV amu^{-1} (Ref. 9); and Ar (data extracted from Figs. 1 and 2 of this work). Dashed lines (a) are drawn to guide the eye.

region, interpolation and extrapolation of $\bar{i}$ and s values to nearby ions are risky. However, if $\bar{i}$ and s are known from interpolation or extrapolation of data for the same ion at other energies, ECSD's can be calculated by the χ^2 model, at least for ions with $Z \leq 26$.[4] Charge-state fractions $f(i)$ are given by Eq. (2) of Sec. II where t, c, and v are expressed by Eqs. (1), (3), and (4). ECSD's observed for ions with $Z > 26$ at low velocities have not been analyzed previously[4] and will be discussed in Sec. IV.

IV. ANALYSIS OF ECSD'S FOR HEAVY IONS ($Z > 26$) AT LOW VELOCITIES

A. Shape of ECSD's

Very few ECSD's in carbon have been measured for heavy ions ($Z > 26$) at low velocities ($v < 2\times10^8$ cm s^{-1}). Heinemeier *et al.*[20] have measured these distributions for Pr, Gd, Lu, Hg, Tl, and Pb ions at beam energies ranging from 300 to 1000 keV. The ECSD's have been obtained for U ions at 2 and 4 MeV,[19] and recently for Kr ions at 1 and 2 MeV.[21]

In Fig. 7 the fraction of neutrals $f(0)$ has been plotted as a function of Z for ECSD in carbon available in the literature[19,20,22] at E/M equal to 5 keV amu^{-1}. Figure 7 shows that $f(0)$ tends to decrease with Z. Fluctuations of $f(0)$ around the general trend are probably due to shell effects important at these low velocities (see also Sec. III) and to experimental errors. For the heavier projectiles studied ($59 \leq Z \leq 82$), $f(0)$ is smaller than 0.07 while for the lighter projectiles $f(0)$ is generally much larger, at $E/M = 5$ keV amu^{-1}. Thus, ECSD's for heavy ions ($Z \geq 59$) are expected to be Gaussian at lower velocities than those required for

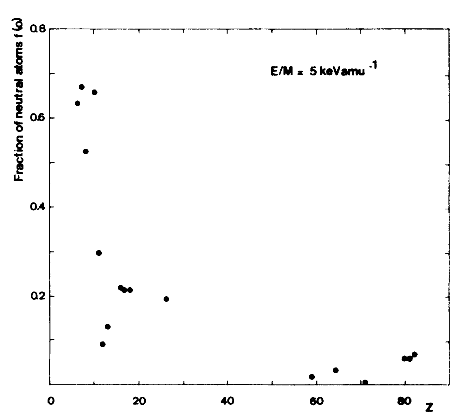

FIG. 7. Observed fractions of neutral atoms, $f(0)$, as a function of the projectile atomic number at E/M equal to 5 keV amu^{-1}. $f(0)$ values are obtained by interpolating or extrapolating the data given in Refs. 19, 20, and 22.

lighter ions ($Z \leq 26$) (see Sec. II).

In Fig. 8 ECSD's observed[20] and calculated by the χ^2 model and by the Gaussian model are plotted for Pr projectiles in carbon at 2.6 and 6.9 keV amu^{-1}. The χ^2 model fits the data very well at 2.6 keV amu^{-1}, but at 6.9 keV amu^{-1} the distribution is already Gaussian.

We have verified that all ECSD's measured[20] for $59 \leq Z \leq 82$ at $E/M \lesssim 5$ keV amu^{-1} are well fitted by the χ^2 model. Examples of such fits for Tl ions, for different values of E/M, are given in Fig. 9. Distributions calculated by the Gaussian model are also given for comparsion. It is clear from Fig. 9 that the χ^2 model works better than the Gaussian model for $E/M < 5$ keV amu^{-1}. Let us note that in Figs. 8 and 9 all the distributions are calculated using the observed values of $\bar{i}$ and s.

B. Determination of $\bar{i}$ and s

The major problem in the prediction of ECSD's in this low-velocity region is the determination of $\bar{i}$ and s. Values of $\bar{i}/Z^{1/2}$ and s (available for ions with $Z > 26$ at low velocities) as a function of the emerging ion velocity are plotted in Figs. 10 and 11, respectively. There is a monotonic dependence of $\bar{i}$ and s on v for a given projectile. Small fluctuations of s around the general trend appearing in Fig. 11, for a given ion, are probably due to statistical uncertainties more important for s than for $\bar{i}$ as already mentioned. Just as for lighter ions, monotonic dependence of $\bar{i}$ and s on Z at a given velocity cannot generally be found. However, examination of the values observed for the three rare earths analyzed (Pr, Gd, and Lu)[20] shows that the relation

$$\bar{i}/Z^{1/2} = 0.33v \tag{9}$$

fits the data very well (see Fig. 10). As pointed out in Ref. 20, rare earths can be expected to behave similarly because their ionization potentials show only small variations and Eq. (9) is probably a good approximation for all the rare earths. Let us note that this relation has been proposed for approximating the mean charges for heavy ions in carbon foils in the range $\bar{i}/Z < 0.3$.[1] Figure 10 shows that Eq. (9) is not valid for the other heavy projectiles studied, i.e., Hg, Tl, Pb, and U. Analysis of observed values of s for Pr, Gd, and Lu[20] leads to the following relation for approximating the standard deviations for ECSD's of the rare earths:

$$s = Z^{-1.26}(118v + 94)\ , \tag{10}$$

where v is in 10^8 cm s^{-1}.

C. Conclusion

All the ECSD's in carbon observed for heavy ions ($59 \leq Z \leq 82$) at low energies are well fitted by the χ^2 model for $E/M \lesssim 5$ keV amu^{-1} and by the Gaussian model for $E/M > 5$ keV amu^{-1}. The mean charge and the standard deviation of the

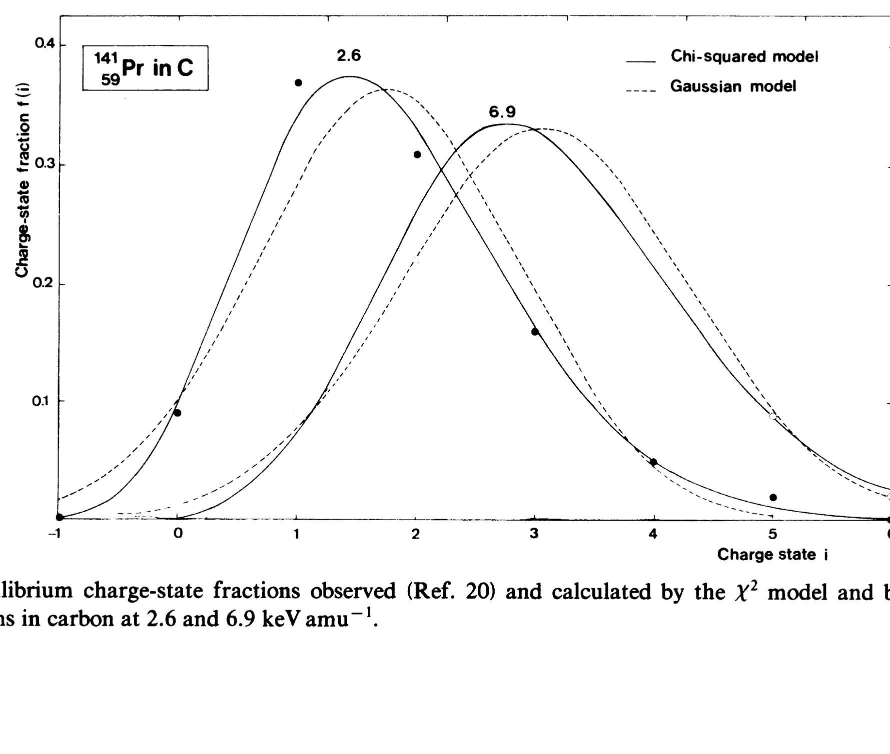

FIG. 8. Equilibrium charge-state fractions observed (Ref. 20) and calculated by the χ^2 model and by the Gaussian model, for Pr ions in carbon at 2.6 and 6.9 keV amu^{-1}.

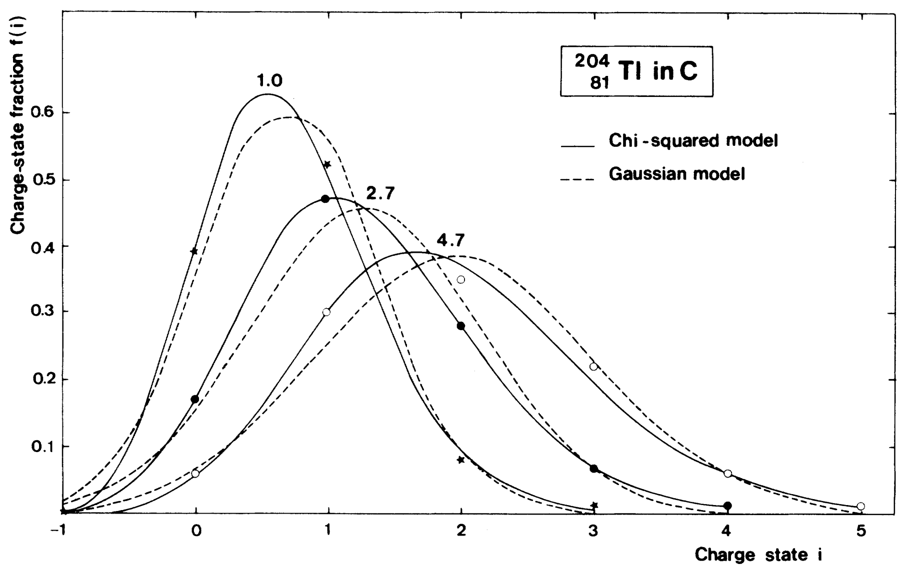

FIG. 9. Equilibrium charge-state fractions observed (Ref. 20) and calculated by the χ^2 model and by the Gaussian model, for Tl ions in carbon at 1.0, 2.7, and 4.7 keV amu^{-1}.

charge at a given velocity do not depend monotonically on Z and these quantities cannot generally be predicted with good accuracy from semiempirical relations. However, for the rare earths, $\bar{i}$ and s can be estimated by Eqs. (9) and (10) and equilibrium charge-state fractions $f(i)$ may be predicted by Eq. (8) or Eqs. (1)—(4), in the energy range 300—1000 keV.

V. CONCLUSIONS

We discuss the following conclusions.

(a) It has been shown that the shapes of ECSD's

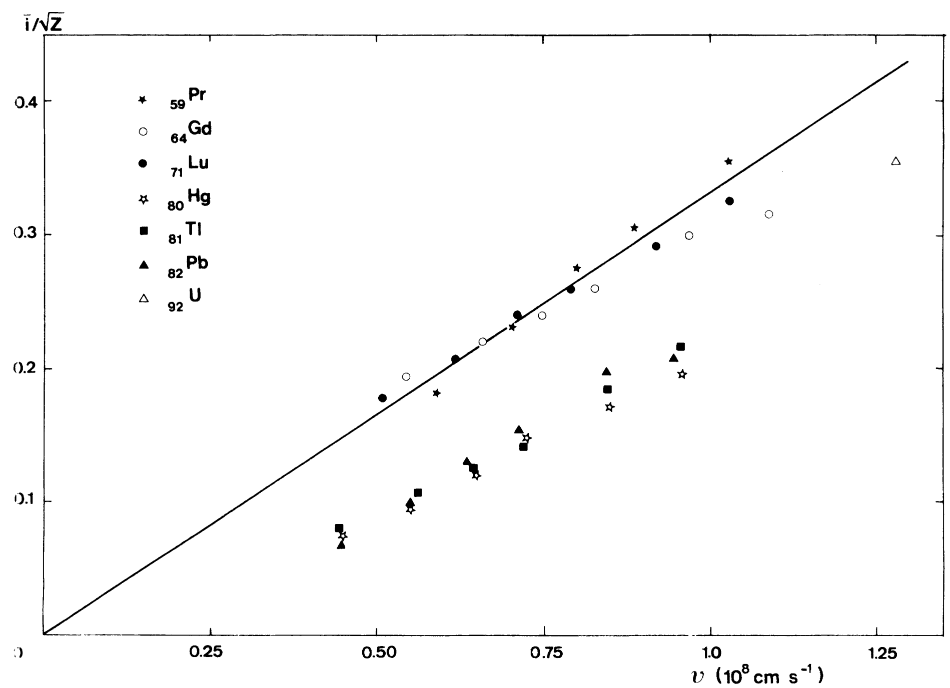

FIG. 10. Ratios of observed equilibrium mean charges to $Z^{1/2}$ for heavy ions ($59 \leq Z \leq 92$) having passed through carbon foils as a function of the ion velocity after the foil. All the data come from Ref. 20 except the datum for uranium which comes from Ref. 19. Straight line represents the relation $\bar{i}/Z^{1/2}=0.33v$.

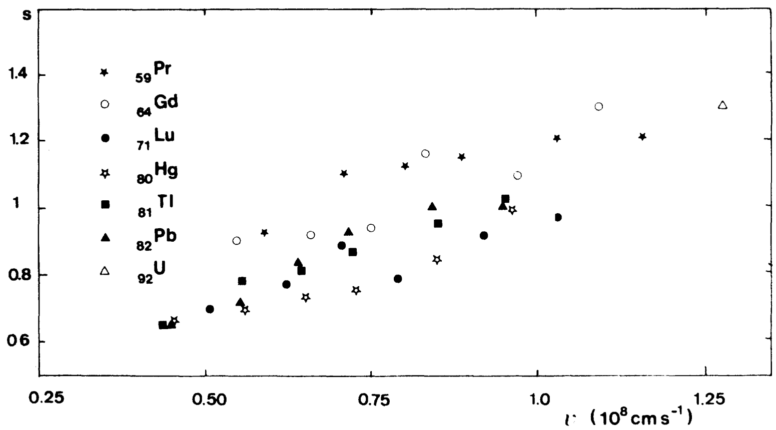

FIG. 11. Standard deviations of ECSD's observed for heavy ions ($59 \leq Z \leq 92$) having passed through carbon foils as a function of the ion velocity after the foil. Data are extracted from the same references as those shown in Fig. 10.

observed for heavy ions at the exit of a carbon foil, at low and high velocities, are due to the fact that the maximum number of electrons which can be captured or lost by the projectile is limited. The principal argument leading to that conclusion is that the statistical function fitting ECSD's observed at high velocities is identical to that obtained at low velocities if the random variable i is changed into $Z-i$.

(b) The ECSD's for heavy ions in carbon can be predicted by previously proposed models if the mean charge $\bar{i}$ and the standard deviation of the charge s can be estimated.[4,5] Examination of data for $\bar{i}$ and s have revealed that these quantities vary smoothly with v for a given projectile and that the dependence of $\bar{i}$ and s on Z at a given velocity is monotonic only for sufficiently high velocities ($v > 4.4 \times 10^8$ cm s^{-1}, $Z \lesssim 20$). Accurate relations for estimating $\bar{i}$ and s for heavy projectiles at high velocities ($v > 3.6 \times 10^8 Z^{0.45}$ cm s^{-1}, $7 \leq Z \leq 36$) have been given previously.[5] For intermediate velocities ($v > 4.4 \times 10^8$ cm s^{-1}) it should be possible to obtain such relations but more data are needed. Finally, at low velocities ($v < 4.4 \times 10^8$ cm s^{-1}, $Z \lesssim 20$) shell effects play a major role; $\bar{i}$ and s are oscillating functions of Z, at a given velocity, and so cannot be estimated from simple relations.

(c) Systematic analyses of ECSD's in carbon observed for ions with $59 \leq Z \leq 82$, at very low velocities [$0.4 \leq v(10^8$ cm s$^{-1}) \leq 1.2$][20] have disclosed that these distributions are well approximated by the χ^2 model for $E/M \lesssim 5$ keV amu^{-1} and by the Gaussian model for $E/M > 5$ keV amu^{-1} [Let us recall that ECSD's for lighter ions ($Z \leq 26$) are generally Gaussian only for $E/M \gtrsim 20$ keV amu^{-1}.] As for lighter ions, $\bar{i}$ and s cannot generally be determined from universal relations in this low-velocity range. However, for rare earths, shell effects are less important and $\bar{i}$ and s can be estimated from simple functions of Z and v proposed in this work.

ACKNOWLEDGMENT

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