Analysis of charge-state distributions of heavy ions in carbon foils and gases

Y Baudinet-Robinet, P D Dumont and H P Garnir
Institut de Physique Nucléaire, Université de Liège, Sart Tilman,
B-4000 Liège, Belgium

Received 9 September 1977, in final form 1 November 1977

Abstract. The charge-state distributions of heavy ions after passing through carbon foils with an energy per nucleon \( E/M \leq 20 \text{ keV amu}^{-1} \), and through gases in the energy domain analysed, i.e. \( 10 \leq E/M (\text{keV amu}^{-1}) \leq 300 \), are very well fitted by our recent 'chi-squared model'. Charge-state distributions in carbon foils for \( E/M \geq 20 \text{ keV amu}^{-1} \) follow the Gaussian law surprisingly well. Charge-state distributions in carbon foils calculated using either the chi-squared model or the Gaussian model are in better agreement with experimental data than those calculated from the independent-electron model (binomial model).

1. Introduction

A general discussion of the charge-state equilibrium of ions having passed through solid or gaseous targets is given in several review articles (Northcliffe 1963, Betz 1972, etc). In particular, Betz (1972) reviewed very thoroughly the experimental and theoretical situation concerning equilibrium charge states and charge-changing cross sections of heavy ions \((Z > 16)\) with energies per nucleon larger than \(20 \text{ keV amu}^{-1}(v > 2 \times 10^8 \text{cm s}^{-1})\) in gaseous and solid media. The more recent results related to the nature of the states of penetrating ions in dense media have been reviewed by Datz (1976).

The main aim of the present work is to propose simple statistical models which fit closely the charge-state distributions observed in carbon foils and in gases. These semi-empirical models will be of great help in the understanding of mechanisms related to the passage of heavy ions through matter and in estimating charge-state distributions not investigated experimentally. The knowledge of charge-state distributions is important particularly in beam-foil spectroscopy. We are mainly interested in ions with \(5 \leq Z \leq 26\) accelerated at energies ranging from approximately 1 keV to 100 keV per nucleon.

Two simple statistical models have been proposed previously: the Gaussian model and the independent-electron model (binomial model). The Gaussian model was first developed by Dmitriev and Nikolaev (1965) for calculating charge-state distributions of fast ions \((v > 2.2 \times 10^8 \text{ cm s}^{-1})\) moving in solid and gaseous media. Betz (1972) showed that this model is valid for solid targets but that heavy gaseous strippers produce asymmetric charge-state distributions. Garcia (1973) has found a theoretical foundation for the Gaussian model taking a statistical approach engendered by descriptions of equilibrium conditions in gases and plasmas. The comparison between
observed charge-state distributions and distributions calculated using the Gaussian model, with the mean charge determined experimentally and the standard deviation of the charge given by the model, is made in Garcia's paper for iron and argon after going through carbon foils at 1.45, 0.957 and 0.664 MeV and for nitrogen at 0.567 MeV. The agreement is indeed encouraging for iron and argon but not for nitrogen. The binomial model was proposed by Dynfors et al (1975, 1976a) and Veje (1976) for describing charge-state distributions and beam-foil populations of heavy ions having passed through solids on the basis of an independent-electron model.

2. Charge-state distributions in carbon foils†

2.1. Mean and standard deviation

We have plotted in figure 1 the mean charges of the charge-state distributions observed for different ions in carbon foils as a function of the energy per nucleon for the emerging ions‡.

![Figure 1. Mean charges of observed charge-state distributions for different ions having passed through carbon foils as a function of the energy per nucleon after the foil. The data are from Hvelplund et al (1970) for N, O, F, Ne, Na, Mg, Al and Ar; from Smith and Whaling (1969) for Fe, N and Ar; from Girardeau et al (1971) for O and Ne; and from Bickel et al (1972) for B.](image)

There is a smooth dependence of \( \bar{i} \) on \( E/M \). At a given \( E/M \), \( \bar{i} \) increases generally with \( Z \) (exceptions are due to shell effects). We did not find simple accurate semi-empirical relations between \( \bar{i} \) and \( E/M \) or \( \bar{i} \) and \( Z \). The relation

\[
\bar{i} = a(E/M)^4
\]

† The beam which emerges from the target is observed essentially in the forward direction.
‡ Most of these values may be obtained from the tabulation of charge-state distributions by Wittkower and Betz (1973). In this tabulation, however, in a few cases, the energy of the ions is given before the foil and not after the foil and must be corrected: for instance this is the case for the data of Girardeau et al (1971).
where $a$ and $A$ are constants which depend smoothly but not regularly on $Z$, is valid in a limited energy domain and may be used for interpolation purposes. For example, for oxygen ions with energy between 10 and 30 keV amu$^{-1}$ the relation is

$$i = 0.10[E/M\text{ (keV amu}^{-1}\text{)}]^{0.84}. \quad (2)$$

The observed values of the standard deviation, $s(i)$, which characterises the width of the distribution, are plotted in figure 2 against the energy per nucleon for different ions. The relation

$$s(i) = b(E/M)^B \quad (3)$$

where $b$ depends on $Z$, and $B$ is nearly constant ($B \approx 0.13$ for most of the ions analysed) holds for $E/M$ lower than a limit which depends on $Z$. In particular for oxygen ions with energy between 5 and 30 keV amu$^{-1}$, $s(i)$ is given by

$$s(i) = 0.585[E/M\text{ (keV amu}^{-1}\text{)}]^{0.13}. \quad (4)$$

The standard deviation $s(i)$ does not increase regularly with $Z$ at a given velocity (see figure 2).

![Figure 2. Standard deviations of observed charge-state distributions for different ions having passed through carbon foils as a function of the energy per nucleon after the foil. The data are extracted from the same references as those shown in figure 1.](image)

2.2. Probability density function

2.2.1. General description. We have tried to fit all the observed charge-state distributions with a few simple statistical functions. It turned out that all the symmetric distributions are fitted surprisingly well using a Gaussian (or normal) law and in all the asymmetric cases the distribution of $t = c(i + 2)$, where $c$ is a parameter depending on energy, follows with a good approximation the chi-squared law with $v$ degrees of freedom ($\chi^2_v$) where $v$ depends on energy (Baudinet-Robinet et al 1977). We note that the term $+2$ in the new variable $t = c(i + 2)$ is justified by the fact that a few ions can emerge from the foil with the charge $i = -1$ but never with the charge $-2$; hence the probability density must be equal to zero for $i = -2$. If we use $t = c(i + a)$, where $a$ is a free parameter, the agreement between observed and calculated distributions is better for $a = 2$ than for $a \neq 2$. 
Let us first recall some properties of the $\chi^2$ distribution. The density of probability, $g(t)$, of a $\chi^2$ variable, $t$, is given by

$$g(t) = [2^{v/2}\Gamma(v/2)]^{-1} t^{v/2-1} e^{-t/2} \quad 0 < t < \infty.$$  

(5)

The mean $\bar{t}$ and the variance $s^2(t)$ of this distribution are given by

$$\bar{t} = v$$  

(6)

$$s^2(t) = 2v.$$  

(7)

The maximum of the distribution occurs for

$$t = v - 2 = \bar{t} - 2.$$  

(8)

We now introduce $t = c(i + 2)$ as a $\chi^2$ variable, we deduce from equations (6) and (7):

$$c = 2(i + 2)/s^2(i)$$  

(9)

$$v = 2(i + 2)^2/s^2(i).$$  

(10)

c and $v$ can be determined from these relations if $\bar{t}$ and $s^2(i)$ are known. The density of probability of $i$, $f(i)$, is then given by

$$f(i) = cg(t).$$  

(11)

The maximum of the charge-state distribution occurs at

$$i = i - (2/c).$$  

(12)

The coefficient of skewness $\gamma$ characterising the asymmetry of the distribution is given by

$$\gamma = \frac{\int (i - \bar{i})^3 f(i) \, di}{(s(i))^3} = \frac{2^{3/2}}{v^{1/2}}.$$  

(13)

We have calculated the probability density functions in the Gaussian model and in the chi-squared model using the observed values of $\bar{t}$ and $s(i)$. Note that these two models depend on two parameters. We have also calculated the charge-state fractions in the binomial model: we adjusted the parameter $\alpha$ which gives the probability that an electron is transferred from the back of the foil to a bound state outside the core (see Veje 1976) in order to match the mean of the distribution with the observed mean value. If $n$ is the maximum number of electrons outside the closed core, $\alpha$ is given by

$$\bar{t}_{\text{obs}} = n(1 - \alpha).$$  

(14)

We adopted this choice of $\alpha$ except for sulphur for which we have used the value of $\alpha$ explicitly given by Veje (1976). Values of $\alpha$ for other elements could be also deduced from curves given in Veje's paper; since, however, these values have been obtained by the method of trial and error, it seemed to us more justifiable to adjust $\alpha$ to obtain agreement between the observed and calculated mean values of $i$.

We shall now review the results obtained from the analysis of many charge-state distributions of heavy ions in carbon foils.
2.2.2. Applications to experimental data

(i) Boron. Bickel et al. (1972) have measured charge-state distributions for boron ions with $20 \leq E/M \text{ (keV amu}^{-1}) \leq 154$. All the results are closely approximated by a Gaussian function. In figures 3(a) and (b) we give distributions calculated from different approximations (Gaussian model, binomial model and Zaidins's calculations (see Bickel et al. 1972, Marion and Young 1968), together with the distributions observed for $E/M = 21$ and 109 keV amu$^{-1}$ respectively. Recently, To and Drouin (1976) have measured charge-state distributions for boron ions in carbon for incident energies between 2 and 7 MeV and found good agreement with the Gaussian model.

(ii) Carbon. Charge-state distributions have been measured at energies ranging from 369 to 1450 keV by Smith and Whaling (1969) and from 1 to 5 MeV by Girardeau et al. (1971). In this whole energy domain the Gaussian model is in excellent agreement with the experimental data and gives better results than the binomial model.

(iii) Nitrogen. For $E/M$ larger than 20 keV per nucleon, the normal distribution fits very well the observed data of Hvelplund et al. (1970), Smith and Whaling (1969) and Girardeau et al. (1971). At lower energies the distributions are asymmetric and very well fitted using the binomial model as well as the chi-squared model.

(iv) Oxygen. All the charge-state distributions observed by Hvelplund et al. (1970), and by Girardeau et al. (1971) for $5.6 \leq E/M \text{ (keV amu}^{-1}) \leq 300$, are closely fitted by a Gaussian function. Charge-state distributions observed and calculated using
the binomial and normal approximations are given in figure 4 for different values of the ion energy. The binomial model gives poor fits especially at low energy where it is expected to work well since the core excitations are very small.

Figure 4. Observed (Hvelplund et al 1970) and calculated charge-state fractions for oxygen ions and carbon foils at different energies for the emerging ions. Calculations have been made using the Gaussian model and the binomial model with \( n = 4 \) (\( z = 0.93 \), 0.81 and 0.60 at 5.6, 11.6 and 29.9 keV amu\(^{-1} \) respectively).

(v) **Fluorine.** The charge-state distributions observed by Hvelplund et al (1970) for \( E/M \) larger than 20 keV amu\(^{-1} \) are close to a Gaussian; for lower energies the chi-squared model is superior to other models. Distributions calculated using the chi-squared model and the binomial model are compared to the observed distributions for \( E/M \) equal to 4.8 and 9.9 keV amu\(^{-1} \) in figures 5(a) and (b), respectively.

(vi) **Neon and argon.** For these elements the binomial model with \( n = 6 \) is known to work well at low energy (Veje 1976). For Ne at \( E/M \) larger than 10 keV per nucleon, we found that the normal distribution fits very well the distributions observed by Hvelplund et al (1970) and by Girardeau et al (1971). At lower energy, the observed fraction of neutral atoms is larger than that calculated from the chi-squared model. This is also the case for argon at \( E/M \) lower than about 2.5 keV per nucleon. This abnormal behaviour is due to the extremely high stability of the neutral atoms for these closed-shell elements at low energy. It is only in these low-energy cases that the binomial model gives better agreement with the experimental data than the Gaussian or chi-squared models. Charge-state distributions have been measured for argon by Hvelplund et al (1970), Smith and Whaling (1969) and Turkenburg et al (1975). All these distributions are closely fitted using the chi-squared model provided that \( E/M \) is larger than 2.5 keV per nucleon. Comparison between the different models is shown in figure 6: for 4.5 and 36.2 keV amu\(^{-1} \) argon ions.

(vii) **Sodium.** Hvelplund et al (1970) have measured charge-state distributions in carbon for sodium ions at energies ranging from 3.9 to 20.9 keV amu\(^{-1} \). All these distributions are fitted very well using the chi-squared model (see figure 3 of Baudinet-Robinet et al 1977). The binomial model gives bad results even at energies as low
Charge-state distributions of ions in carbon and gases

Figure 5. Observed (Hvelplund et al 1970) and calculated charge-state fractions for fluorine ions and carbon foils at (a) 4.8 keV amu\(^{-1}\), (b) 9.9 keV amu\(^{-1}\) for the emerging ions. Calculations have been made using the binomial model with \( n = 5 \) (\( x = 0.95 \) and 0.87 at 4.8 and 9.9 keV amu\(^{-1}\) respectively) and the chi-squared model (\( c = 8.1 \) and 7.9; \( v = 18.1 \) and 20.7 at 4.8 and 9.9 keV amu\(^{-1}\) respectively).

Figure 6. Observed and calculated charge-state fractions for argon ions and carbon foils at 4.5 and 36.2 keV amu\(^{-1}\) for the emerging ions. Calculations have been made using the binomial model with \( n = 6 \) (\( x = 0.81 \) and 0.38 at 4.5 and 36.2 keV amu\(^{-1}\) respectively) and the chi-squared model (\( c = 7.8 \) and 6.3; \( v = 24.3 \) and 35.9 at 4.5 and 36.2 keV amu\(^{-1}\) respectively).
as 3.9 keVamu⁻¹. This is explained (Veje 1976) by the fact that for this element core excitations have to be taken into account even at energies around 100 keV.

(viii) **Magnesium and aluminium.** Charge-state distributions in carbon for these elements have been measured by Hvelplund *et al* (1970) for 3.3 ≤ *E/M* (keV amu⁻¹] ≤ 20. All these data are closely approximated by a normal distribution which gives much better results than the binomial model (except for Al at 3.3 keV amu⁻¹ where the two models are competitive). It is mentioned in Veje (1976) that Mg data can be well fitted by a three-electron system (*n* = 3) although Mg has only two electrons outside the core. We point out here the fact that different statistical distributions can sometimes be in agreement with the experimental data because they have, for particular values of their parameters, the same shape; for instance the binomial distribution tends as *n* increases towards the normal distribution very quickly if \( \alpha \approx 0.5 \). However, they correspond to completely different interpretations of the physical process.

(ix) **Sulphur.** For this element the binomial model with *n* = 6 is claimed (Veje 1976, Dynefors *et al* 1976b) to be in agreement with the experimental data (Berry *et al* 1970, Dynefors *et al* 1976b). The lowest energy at which the charge-state distribution was measured (by Berry *et al* 1970) is 1 MeV. In figure 7, a comparison is shown between this observed distribution and the results of the Gaussian and binomial models. The normal distribution is in closer agreement with the data than the binomial distribution. The Gaussian model also fits quite well the distributions observed at higher energy (Ambros and Kleinfeld 1972) as can also be seen in figure 7 for 4.6 MeV sulphur ions.

![Figure 7. Observed and calculated charge-state fractions for sulphur ions and carbon foils at 1 and 4.6 MeV incident energies. Calculations have been made using the Gaussian model at 1 and 4.6 MeV and the binomial model with *n* = 6 (\( \alpha = 0.34 \)) at 1 MeV.](image)

(x) **Chlorine.** Charge-state distributions observed by Turkenburg *et al* (1975) and Wittkower and Ryding (1971) for chlorine ions in carbon are in agreement with the Gaussian model for *E/M* ≥ 30 keV amu⁻¹ and with the chi-squared model for *E/M* ≤ 10 keV amu⁻¹. No measurements have been made in the intermediate energy domain. Examples of fits are given in figure 8 for different energies of the chlorine...
Charge-state distributions of ions in carbon and gases

Figure 8. Observed and calculated charge-state fractions for chlorine ions and carbon foils at different energies for the emerging ions. (The data of Wittkower and Ryding (1971) correspond to an energy of 1 MeV before the foil.) Calculations have been made using the chi-squared model ($c = 6.3, 5.1, 5.9$ and $5.8; y = 17.5, 18.2, 23.9$ and $31.2$ at $0.09, 0.21, 0.33$ and $1$ MeV respectively) and the Gaussian model at $1$ MeV.

ions. The binomial model with $n = 5$ does not give such an agreement with the experimental data even at low energies.

(xi) Iron. Smith and Whaling (1969) have measured charge-state distributions in carbon foils for iron ions with $3.4 < E/M$ (keV amu$^{-1}$) $< 25.9$. The data are in very good agreement with the chi-squared model for $E/M < 17$ keV amu$^{-1}$ and with the Gaussian model for $E/M \geq 20$ keV amu$^{-1}$. The distributions observed at different energies cannot be well fitted by the binomial model. Let us mention that the distributions observed at $3.4, 10.1$ and $20.6$ keV amu$^{-1}$ could agree with a ‘binomial model’ where the number $n$ of electrons outside the core would be equal to $2, 8$ and $12$ respectively (see end of §2.2.2 (viii)).

3. Charge-state distributions in gases

It is well known that equilibrium charge-state distributions in gases heavier than helium are asymmetric (Betz 1972, Wittkower and Ryding 1971). We have obtained good fits to these asymmetric distributions with the chi-squared model developed in §2.2. Examples of such fits are given in figure 4 of Baudinet-Robinet et al (1977) for chlorine ions in $O_2$ gas at $1, 4$ and $10$ MeV incident energies. Charge-state fractions for iodine ions in $O_2$ gas observed by Ryding et al (1969), and calculated using the chi-squared model, are given in figure 9 for different incident energies. In table 1 we give charge-state fractions for chlorine ions in argon gas observed by Wittkower and Ryding (1971) and calculated from the chi-squared model for $1$ and $8$ MeV incident energies. The agreement between observed and calculated distributions varies slightly with the ion energy (as for distributions in carbon foils) but is surprisingly good.
Figure 9. Equilibrium charge-state fractions for iodine ions in O$_2$ gas measured and calculated using the chi-squared model at different incident energies on the target (\(c = 6.6, 3.9\) and \(3.9\); \(v = 26.3, 24.7\) and \(31.9\) at \(1.05, 6.0\) and \(12.0\) MeV respectively).

In the whole energy domain analysed for charge-state distributions in gases heavier than helium \((10 \leq E/M\) (keV amu$^{-1}$) \(\leq 300)\) the chi-squared model is superior to the Gaussian model.

The equilibrium charge-state model in gases allows the direct deduction of the ratios of capture and loss cross sections if only single-electron capture and loss are considered (see for instance Betz 1972). Let us denote the relative number of ions with charge \(i\) by \(F_i\) and the cross section of the process whereby an ion with charge \(i\) is converted into an ion with charge \(k\) by \(\sigma_{i,k}\). We have:

\[
\frac{F_{i+1}}{F_i} \approx \frac{\sigma_{i,i+1}}{\sigma_{i+1,i}}.
\]

Table 1. Charge-state fractions, \(F(i)\), for 1 and 8 MeV chlorine ions in argon gas observed (Wittkower and Ryding 1971) and calculated using the chi-squared model \((c = 9.0\) and \(14.2; v = 35\) and \(107\) at 1 and 8 MeV respectively).

<table>
<thead>
<tr>
<th>(1) MeV</th>
<th>(8) MeV</th>
</tr>
</thead>
<tbody>
<tr>
<td>(i)</td>
<td>(F_{\text{obs}})</td>
</tr>
<tr>
<td>0</td>
<td>0.035</td>
</tr>
<tr>
<td>1</td>
<td>0.321</td>
</tr>
<tr>
<td>2</td>
<td>0.417</td>
</tr>
<tr>
<td>3</td>
<td>0.180</td>
</tr>
<tr>
<td>4</td>
<td>0.040</td>
</tr>
<tr>
<td>5</td>
<td>0.0059</td>
</tr>
<tr>
<td>6</td>
<td>0.0015</td>
</tr>
<tr>
<td>7</td>
<td>—</td>
</tr>
<tr>
<td>8</td>
<td>—</td>
</tr>
<tr>
<td>9</td>
<td>—</td>
</tr>
<tr>
<td>10</td>
<td>—</td>
</tr>
</tbody>
</table>
From equations (5), (11), (15), (9) and (10) we deduce:

\[
\frac{\sigma_{i,t+1}}{\sigma_{i+1,t}} \approx \frac{g(t+c)}{g(t)} \left( \frac{t+c}{t} \right)^{\frac{t^2}{2} - 1} \exp\left(-\frac{c}{2}\right) = \left(\frac{i + 3}{i + 2}\right)^{\frac{(i + 2)^2}{2s(i)^2} - 1} \exp\left[-(i + 2)/s^2(i)\right].
\]

(16)

For charge-state distributions in gases of heavy ions with velocity larger than \(2.2 \times 10^8\) cm s\(^{-1}\), semi-empirical relations exist which relate the average equilibrium charge \(\bar{i}\) and the velocity \(v\), and charge \(Z\) of the ion (Betz 1972, Wittkower and Ryding 1971). Semi-empirical relations have also been found for the standard deviation \(s(i)\) as a function of \(Z\) and \(\bar{i}\). However, as emphasised in Betz (1972) ‘it should be kept in mind that despite the generally smooth behaviour of \(\bar{i}\), all these semi-empirical estimates are useful mainly for interpolation purposes, and that extrapolation beyond the investigated ranges of both \(Z\) and \(v\) must still be regarded as risky’. If \(\bar{i}\) and \(s(i)\) can be determined, charge-state fractions for ions in heavy gases may be calculated to a good approximation using the chi-squared model.

4. Conclusions

Systematic analyses of charge distributions of heavy ions measured in carbon foils have revealed that the distributions for all ions except argon, with energy higher than about 20 keV per nucleon, are closely approximated by Gaussian curves. For a few elements the Gaussian model is already valid at low energies (for oxygen at 4.8 keV amu\(^{-1}\), neon at 13.5 keV amu\(^{-1}\), aluminium and magnesium at 3.3 keV amu\(^{-1}\)). For argon, on the contrary, the charge-state distributions are still asymmetric at energies as high as 36 keV amu\(^{-1}\). All the non-Gaussian distributions are fitted very well by the newly proposed chi-squared model. Better agreement with the experimental results is obtained using either the Gaussian model or the chi-squared model than the binomial model. In the energy domain analysed (1 \(\leq E/M\) (keV amu\(^{-1}\)) \(\leq 100\)) we did not find precise semi-empirical relations for the mean and standard deviation of the charge. Approximate values for these parameters may be obtained by interpolating existing data for the same ion at other energies or for nearby ions. However, this is somewhat risky because of shell effects, as shown in figures 1 and 2. Hence, charge-state distributions in carbon foils may generally be calculated to a good approximation using the Gaussian model (for \(E/M \geq 20\) keV amu\(^{-1}\)) or the chi-squared model (for \(E/M \leq 20\) keV amu\(^{-1}\)).

Charge-state distributions observed in gases heavier than helium are in good agreement with the chi-squared model in the whole energy domain analysed (10 \(\leq E/M\) (keV amu\(^{-1}\)) \(\leq 300\)). Thus, charge-state fractions in gases may be predicted if the mean and standard deviation of the charge can be obtained from existing semi-empirical relations (see §3).

Acknowledgments

We thank Professor C Mahaux for reading the manuscript. We are grateful to Professor L Winand for his interest in this work and to Mrs C Leroux for carefully drawing the figures. This work was supported by the ‘Fonds National de la Recherche Scientifique’, the ‘Université de Liège’ and the ‘Institut Interuniversitaire des Sciences Nucléaires’.
References

Betz H D 1972 Rev. Mod. Phys. 44 465–539
Datz S 1976 Nucl. Instrum. Meth. 132 7–17
Dynefors B, Martinson I and Veje E 1975 Phys. Scr. 6 71–2
———1976a Phys. Scr. 13 308–12
Marion J B and Young F C 1968 Nuclear Reaction Analysis. Graphs and Tables (Amsterdam: North-Holland) pp 34–47
Wittkower A B and Betz H D 1973 Atom. Data 5 113–66