Equilibrium Charge-State Distributions of Heavy Ions $(1-14 \text{ MeV})$

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Equilibrium charge-state distributions have been determined experimentally for chlorine and bromine ions with energies between 1 and 14 MeV traversing H_2 , He, N₂, O₂, Ar, and Kr gases and a carbon foil. In addition, a few measurements have been made using iodine beams traversing various targets —at 3, 10, and 12 MeV through carbon foil, at 8 MeV through $CO₂$, at 10 and 12 MeV through gold foil, and at 12 MeV through a Xe gas target.

I. INTRODUCTION

The charge of a fast ion moving through matter fluctuates as a result of electron loss and capture in collisions with the atoms of the target. After the ions have made a sufficient number of collisions with the target atoms an equilibrium distribution of charges is established mhich is dependent only on the velocity of the ions and the target material.

In this experiment equilibrium charge fractions have been measured for chlorine ions, at energies between 1 and 12 MeV, and bromine ions, at energies between 2 and 14 MeV, passing through six gases (H_2 , He, N₂, O₂, Ar, and Kr) and a carbon foil. The ions mere accelerated in the Robert J. Van de Graaff Laboratory's 3-MV tandem accelerator and the beams mere analyzed both eleetrostatically and magnetically before entering the experimental apparatus. This apparatus, as mell as the data collection and analysis, was identical to that described by Ryding, Wittkower, and Rose.^{1,}

This is the third in a series of papers describing the results of equilibrium-charge-fraction measure ments of heavy ions at high energies. Previous work using iodine and aluminum beams has already been published^{2,3}; some new results(with iodine) are included in the body of this paper. In addition, use has been made of earlier results to present some semiempirical predictions for the mean charge and distribution midth of a variety of heavy ions.

II. MATHEMATICAL DESCRIPTION

The mathematical formalisms of charge changing have been described extensively elsewhere. 4.5 The present description w1ll therefore be limited to those aspects which relate directly to the data obtained.

The charge-state composition of a beam passing through a gas may be described in terms of the fractions of the beam $F_i(\pi)$ in charge state i, where π refers to the number of target particles per cm

traversed by the beam $(\sum F_i(\pi) = 1)$. The variation of the fractions is described by a system of differential equations

$$
\frac{dF_i(\pi)}{d\pi} = \sum_j F_j(\pi) \sigma_{ji} - \sum_j F_i(\pi) \sigma_{ij}, \qquad (1)
$$

where σ_{ij} is the cross section for a charge change from ⁱ to j.

At large values of π , $dF_i(\pi)/d\pi \rightarrow 0$ and equilibrium conditions are reached. F_i will be used to designate these equilibrium values. It is not almays possible to describe the charge-state distribution of these equilibrium fractions by a simple mathematical function and it is often convenient to refer to the mean charge \overline{i} given by

$$
\vec{i} = \sum_{i} iF_{i} \tag{2}
$$

the distribution width or standard deviation

$$
\sigma^2 = \sum_i (\bar{i} - i)^2 F_i, \qquad (3)
$$

and the distribution asymmetry or skewness γ given by

$$
\gamma = \left[\sum_{i} \left(\overline{i} - i\right)^{3} F_{i}\right] / \sigma^{3} . \tag{4}
$$

III. RESULTS AND DISCUSSION

Sample measurements were obtained of chargestate fractions for both chlorine and bromine ions as a function of target thickness (see Fig. l of Ref. 3), in order to ensure charge equilibration at each energy. However, it mas not almays necessary to generate complete growth curves to obtain the equilibrium values F_i , and fractions which were in good agreement at two substantially different target thicknesses mere accepted.

Errors in the measured fractions were estimated from the repeatability of the data and from statis-

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tical errors associated with the pulse counting of each beam component. Errors involved in the determination of beam energy were negligible. For fractions F_i greater than $\sim 1\%$, the total estimated probable error is less than \pm 5%, whereas for fractions less than $\sim 1\%$, the probable error may be as large as $\pm 10\%$.

The measured charge-state fractions are presented in Table I (chlorine) and Table II (bromine) as a function of the projectile energy and target material. The mean charge, distribution width, and skewness are also listed. In Table III some equilibrium fractions for iodine beams in targets, which have not been published previously, are included.

Previous data which can be compared directly with the present results are sparse. The data of Almqvist et al., ⁶ who measured equilibrium fractions of chlorine beams in a carbon foil at 8 and 12 MeV, are substantially in agreement with the present results. On the other hand, a set of data presented by Litherland ${et}$ ${al.}, {^7}$ in oxygen gas at 10 MeV, shows considerable differences in both low- and high-charge-state fractions, although the fraction of the most probable charge state, the 5' in both cases, is in agreement with our value. No direct comparison with previous data can be made for incident bromine ions, although Moak et al.⁸ have obtained data at 15 MeV. An extrapolation shows that relatively small but significant differences exist both in this case and with iodine beams where a direct comparison can be made. These differences are due to a target-density effect⁹ induced by the large difference in the target cell lengths used; Moak et al. made use of a long gas cell so that the target density at equilibrium was \sim 10 times lower than in the present experiments.

The mean charge for both chlorine and bromine beams depends considerably on the choice of the target gas. The target gas giving the highest mean charge increases in mass with increasing energy, e.g., O_2 for a chlorine beam at 1 MeV. Ar at 6 MeV, and Kr at 12 MeV. By making use of our earlier iodine ion data² it may be observed that at a particular projectile velocity a given target gas gives the highest mean charge independently of the projectile species. As expected, the mean charge in a carbon foil is appreciably greater than in any of the gases at all energies. The distribution width is generally greatest in an oxygen gas target. This is true also for iodine beams in the same energy range. The maximum skewness is usually generated by collisions in the heaviest gas, krypton. Note the symmetry of distributions in a carbon foil due to the boil-off of Auger electrons from the excited atom as it leaves the foil. This process, itself statistical, tends to symmetrize the final distribution.¹⁰

By assuming that the distribution of ions among the more intense charge states is nearly Gaussian, Nikolaev⁵ derived a relationship between the distribution width and mean charge

$$
F_i \sigma \approx (2\pi)^{-1/2} e^{-(i-\bar{i})^2/2\sigma^2} \,. \tag{5}
$$

For the projectiles and targets used in the present studies $F_i \sigma$ is clearly observed to be a function of $(i - \overline{i})^2 / 2\sigma^2$, although not always nearly Gaussian owing to the complicating effect of the skewness γ . In Fig. 1 this feature is displayed by plotting $F_i \sigma$ vs $(\overline{i} - i)/\sigma$ for three projectile species passing through an oxygen gas target; a pure Gaussian distribution is shown for comparison.

IV. SEMIEMPIRICAL PREDICTIONS

Using the data available to them, several experimenters^{11, 12} have attempted to derive universal equations which would enable the charge-state distributions of all ions to be predicted. This is a formidable task in view of the limited available data and the complex nature of the interactions. In addition the present work indicates that at least three parameters $(\bar{i}, \sigma, \text{ and } \gamma)$ are required to give a reliable indication of a distribution in the energy range 1-20 MeV. Most semiempirical relationships have only attempted to predict the mean charge \bar{i} and have generally been based on masses and energies outside the present range of interest.

FIG. 1. $F_i \sigma$ plotted as a function of $(i - \overrightarrow{i})/\sigma$. This universal curve is independent of the projectile species and energy but does depend upon the target material, which in this case was oxygen gas.

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In the present work we are limited to values of $\overline{i}/\overline{z}$ less than 0.3 and the low-velocity approximation of Dimitriev and Nikolaev¹² (based on the Bohr criterion) is the only empirical prediction available. The simple Bohr theory predicts that \bar{i} varies as Z^{α}_{β} , where α ranges from $\frac{1}{3}$ to $\frac{2}{3}$ depending on the assumptions made in the statistical model of the atomic particle. Consequently, the following form was assumed:

$$
\overline{i} = Kv Z_{\rho}^{\alpha} \tag{6}
$$

where Z_{ρ} is the nuclear charge of the projectile, v is the velocity of the projectile, and K is a constant which depends upon the target material. Values of $\log_{10} i/v$ were then plotted against $\log_{10} Z_{\rho}$ for the chlorine, bromine, and iodine data. It was found that the points could be fitted quite well by a straight line of slope 0. 5. Consequently, the data were then plotted in the form $\overline{i}/Z_b^{1/2}$ vs v. The data were found to fit the empirical relationship

$$
\overline{i} = K v Z_p^{1/2} + C,\tag{7}
$$

where v is in units of 10^8 cm/sec and C is a constant which depends upon both the projectile and the target species. K was found to be 0.19 ± 0.02 for all projectile-target combinations in good agreement with Dimitriev's value of 0. 18 for an oxygen target and 0.16 for a helium target. However, in order to obtain a good fit to the experimental data, values of C ranging from $+0.10$ to -0.20 for gases, and 0.6 ± 0.05 for a carbon foil, were required. Representative data taken using an oxygen target are shown in Fig. 2, for which case $C \approx 0$. Equation (7) may be written in a more convenient form if it is assumed that the ratio of atomicweight to nuclear charge of an ion is approximately 2. 2, i. e.,

$$
A/Z_p \simeq 2.2 \tag{8}
$$

Then the mean charge given by

$$
\overline{i} \simeq 9.3KE^{1/2} + C \t{,} \t(9)
$$

where E is the projectile energy in MeV. It is interesting to note that, from Eq. (9), at a fixed energy the mean charge is roughly independent of the projectile species —^a prediction confirmed by the experimental results.

B. Distribution Width σ

An empirical relationship for σ has also been derived from the present results. Dimitriev and Nikolaev¹² suggested the following simple relation:

$$
\sigma = \sigma_0 Z_p^n \,. \tag{10}
$$

TABLE I. (continued)

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FIG. 2. Function $\overline{i}/Z_b^{1/2}$ vs projectile velocity for a number of different projectiles passing through an oxygen target. Iodine data taken from Ref. 2. Single selenium , data point derived from unpublished data taken by the authors.

Logo was therefore plotted against $\log Z_{b}$ using the present results and it was found that a value of $n=0.5$ best fitted the data. Values of $\sigma/Z_h^{1/2}$ were then plotted as a function of velocity and are shown in Fig. 3 for an oxygen target.

It can be seen that at higher velocities σ is essentially constant and from this work the following approximate relationship may be derived:

$$
\sigma = \sigma_0 Z_p^{1/2} \tag{11}
$$

for $v > 3.5 \times 10^8$ cm/sec. Values of σ_0 for all projectiles were determined to be $\sim 0.17 \pm 0.02$ in a H_2 target, 0. 20 ± 0. 03 in He, 0. 27 ± 0. 03 in N₂, O₂,

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 1 G. Ryding, A. B. Wittkower, and P. H. Rose, Phys. Rev. 185, 129 (1969).

- ${}^{2}G.$ Ryding, A. B. Wittkower, and P. H. Rose, Phys. Rev. 184, 93 (1969).
- 3G. Ryding, A. Wittkower, G. H. Nussbaum, A. C. Saxman, R. Bastide, Q. Kessel, and P. H. Rose, Phys. Rev. B $1, 1081$ (1970).
	- ⁴S. K. Allison, Rev. Mod. Phys. 30, 1137 (1958).
- $5V.$ S. Nikolaev, Usp. Fiz. Nauk 85, 679 (1965) [Sov. Phys. Usp. 8, 269 (1965)].
- 8E. Almqvist, C. Broude, M. A. Clark, J. A. Kuehner, and A. E. Litherland, Can. J. Phys. 40, 954 (1962).
- ${}^{7}A$. E. Litherland, E. Almqvist, H. R. Andrews,
- C. Broude, and J. A. Kuehner, Bull. Am. Phys. Soc.

FIG. 3. Function $\sigma/Z_b^{\frac{1}{2}}$ vs projectile velocity for a number of different projectiles passing through an oxygen target. Iodine data taken from Ref. 2. Single selenium data point derived from unpublished data taken by the authors.

Ar, Kr, and a carbon foil.

C. Distribution Skewness γ

No semiempirical fits have been obtained for γ which is extremely sensitive to the accuracy of the data.

V. CONCLUSION

In conclusion, it can be stated that although the semiempirical relationships of Dimitriev and Nikolaev appear to hold in the range of their applicability for a variety of ion species and target materials, no real understanding of the fundamentals behind these relationships can be obtained without measurements of the fundamental parameters-the charge-changing cross sections themselves. Measurements of this type are in progress at this laboratory.

8, 75 (1963); H. R. Andrews, E. Almqvist, C. Broude, M. A. Eswaran, J. A. Kuehner, and A. E. Litherland, Tandem Quart. 1, 1 (1963).

 C^8 C. D. Moak, H. D. Lutz, L. B. Bridwell, L. C. Northcliffe, and S. Datz, Phys. Rev. 176, 427 (1968).

- ⁹G. Ryding, H. D. Betz, and A. Wittkower, Phys. Rev. Letters 24, 123 (1970); H. D. Betz, *ibid.* 25, 903
- (1970). 10 H. D. Betz and L. Grodzins, Phys. Rev. Letters 25 ,
- 211 (1970).
- ¹¹H. D. Betz, G. Hortig, E. Leischner, C. Schmelzer, B. Stadler, and J. Weihrauch, Phys. Letters 22, ⁶⁴³ (1966).
- 12 I. S. Dimitriev and V. S. Nikolaev, Zh. Eksperim. i Teor. Fiz. 47, 615 (1964) [Sov. Phys. JETP 20, 409 (1965)].