Equilibrium charge-state distributions for 2-20-MeV argon ions in carbon

Émile J. Knystautas and Michèle Jomphe

Laboratoire de l'accélérateur Van de Graaff, Département de physique, Université Laval, Québec G1K 7P4, Canada

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Equilibrium charge-state distributions have been measured for argon ions in thin carbon foils between 2 and 20 MeV. Equilibration takes place in foils at least as thin as $5 \mu g/cm^2$. Our measurements link up smoothly with other work at lower and at higher energies. Several models have been applied to postdict the data. It was found that while the mean charge can be quite accurately determined from empirical models, such as the Gaussian one, or even from extrapolation on Z, serious discrepancies arise with respect to the distribution widths. As a result, charge fractions which are off the peaks of the distributions can be in error by factors of 2 or more, rendering their measurement essential for the purposes of the present study. Evidence of ionic LM shell structure has also been observed in this work.

I. INTRODUCTION

The advent of high-energy accelerators for heavy ions has spurred renewed interest in the charge-changing processes of ions passing through gaseous and solid media. In addition, knowledge of ionic-charge distributions are used in energyloss calculations. The principal motivation for the present study, however, was to aid in the identification of emission lines arising from argon ions excited by their passage through thin carbon foils. By comparing the intensity variation of an unknown line as a function of incident energy with the equilibrated charge distributions at various energies, the unknown line can often be assigned to a given charge state as a first step in its identification.

As will be seen later, although various empirical models, or even extrapolation, can be used to obtain quite satisfactory values for the average charge, the details of the measured distributions are such that the relative proportions of several charge states may be in error by factors of 2 or more compared with model values. Consequently, for the purposes of beam-foil spectroscopy mentioned above, these distributions must be measured.

Work in this area prior to 1973 is subsumed in an exhaustive data compliation by Wittkower and Betz.¹ Betz has also written a comprehensive review article² on collision processes in, and models for, equilibrium charge distributions. The present measurements in argon fill a gap between those made below 1.5 MeV and those made above 40 MeV.

II. EXPERIMENTAL CONDITIONS

Thin self-supporting homemade carbon foils were interposed between the 90° analyzing magnet and the beam-switching magnet of an 8-MV CN Van de Graaff accelerator. Beams of singly, doubly, and triply charged argon ions were produced in a modified rf source³ in the accelerator terminal and impinged on the foil target after being collimated. The stripped ions were chargeselected by the beam-switching magnet, where they were deviated by 15°, and detected in a Faraday cup.

Beam-defining slits at the entrance to the Faraday cup were adjusted to be narrow enough to separate cleanly all measurable charge states, yet wide enough to ensure a saturated signal (which indicated that *all* ions of a given charge were being detected). Additional precautions were taken to remove spurious signals due to secondary electrons and leakage currents. These have been described previously.⁴ During scans of switchermagnet current, the Faraday cup current was digitized and recorded online by a PDP-15 computer.

Two to three runs were made at each energy and the measurements averaged. In addition, a run was made at the highest energy (19.5 MeV) with a thinner foil (~ 5 μ g/cm²) to ascertain whether charge equilibration was already attained.

III. RESULT AND DISCUSSION

Measured equilibrium charge fractions, mean charges, distribution widths, and skewnesses as a function of incident ion energy are listed in Table I. The former are plotted in Fig. 1.

Target thicknesses were all $20 \pm 4 \ \mu g/cm^2$ (measured by the air-equivalent stopping power for 5.5-MeV α particles) except for a run made at 19.5 MeV with a $5 \pm 1 \ \mu g/cm^2$ target which served to verify that equilibration had already taken place in the thinner foil. That this is so is not surpris-

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E(MeV)	\overline{q}	đ	s	F_2	F_3	F_4	F_5	F ₆	FŢ	F ₈	F_9	F ₁₀	F 11	F ₁₂	F ₁₃
3	4.82	1.36	0.18	0.028	0.144	0.259	0.254	0.206	0.084	0.024	0.002				
6	6.53	1.39	-0.13		0.013	0.064	0.157	0.247	0.258	0.199	0.055	0.008	0.001		
8	7.23	1.35	-0.20		0.002	0.021	0.085	0.175	0.264	0.295	0.122	0.031	0.004		
10	7.81	1.25	-0.20			0.006	0.032	0.104	0.234	0.338	0.214	0.061	0.010	0.001	
13	8.58	1.23	-0.13				0.008	0.042	0.118	0.306	0.302	0.173	0.044	0.007	
16	9.17	1.20	0.0					0.015	0.057	0.210	0.332	0.256	0.106	0.021	0.002
19.5	9.67	1.20	0.0					0.003	0.026	0.133	0.282	0.315	0.184	0.050	0.008
			-					-							

TABLE I. Equilibrium charge-state distributions of argon ions in carbon foils. Symbols are explained in the text.

ing. Baron⁵ has found that for velocities below 0.05 c, nonequilibrium distributions do not show up in even the thinnest available foils (~2 μ g/cm²). Judging by the reproducibility of different runs under the same conditions, the errors in the charge fractions F_q were estimated to be approximately 5%, i.e., $F_q \pm 0.05 F_q(F_q > 0.1)$ and 10% for $F_q < 0.1$. As expected, no dependence of the F_q on incident ion charge was observed.

Small-angle multiple scattering can be a significant problem for heavy ions especially at low energies. No measurements of this effect were made in the present study. However, based on two recent works, such scattering was judged to be unimportant here. In the first one, Hooton *et* al.⁶ measured the half-angle distribution for the neighboring-Z chlorine ions in carbon foils of comparable thickness to be of order of 0.1° to 0.2° at 13 and 21.8 MeV. At the other end of our energy scale (1 and 2 MeV), Cardon *et al.*⁷ recently reported measurements for the much heavier krypton ions to be 0.05 and 0.03 rads, respectively, although in somewhat thinner foils ($6.5 \pm 1.0 \ \mu g/cm^2$).

A plot of the mean charge \overline{q} vs energy/nucleon (Fig. 2) shows a smooth behavior from the lowenergy Cal Tech data,⁸ through the present work at intermediate energies, to the two points obtained on high-energy European machines.^{5,9} Here again, as is often the case, the empirical expression due to Nikolaev and Dmitriev,¹⁰

$$\frac{\bar{q}}{Z} = \left[1 + \left(\frac{v_1 Z^{0.45}}{v}\right)^{1/0.6}\right]^{-0.6}, \qquad (1)$$



FIG. 1. Equilibrium charge distributions of argon ions in carbon between 2 and 20 MeV. The curves are drawn to guide the eye.



FIG. 2. Mean charge \overline{q} as a function of incident ion energy. The solid line is from the empirical expression of Nikolaev and Dmitriev (Ref. 10).

where v is the incident ion velocity and $v_1 = 3.6$, $\times 10^8$ cm/sec, gives a very good approximation to the mean charge over a wide range of energies.

On the other hand, the distribution widths d defined by

$$d = \left(\sum_{i} (q_{i} - \bar{q})^{2} F_{q_{i}}\right)^{1/2}$$
(2)

appear to be more difficult to predict empirically. The simple expression of Betz and Schmelzer,¹¹

$$d = 0.27 \ Z^{\overline{2}} \tag{3}$$

gives a constant width of 1.15, independent of energy. This is clearly not borne out by the experiment, as can be seen by the comparison with our data in Fig. 3. On the other hand, the expression given by Nikolaev and Dmitriev¹⁰

$$d = 0.5 \left\{ \overline{q} \left[1 - \left(\frac{\overline{q}}{Z} \right)^{1/k} \right] \right\}^{1/2}, \qquad (4)$$

where k = 0.6, significantly underestimates the measured distribution widths, especially at lower energies. As a result, there can arise substantial discrepancies for those charge states that are away from the peak of the distribution. This is



FIG. 3. Distribution widths d as a function of incident energy.

unsatisfactory for the purposes mentioned in Sec. I.

The skewness parameter² has been included for completeness only in Table I, and no significance is attached to the small values obtained, except insofar as it confirms the observation that solids tend to produce more symmetrical distributions than do gaseous targets.²

A number of models have been proposed to account for the shape of equilibrium charge-state distributions, with a view to obtaining universal parametrized empirical expressions which could then be used to predict such distributions for arbitrary values of Z and incident ion energy. The Gaussian model of Nikolaev and Dmitriev¹⁰ is one of the most widely used, and appears to be more than adequate for the mean charge \bar{q} . A physical justification of the Gaussian model has been suggested by Garcia.¹² However, as was seen above (Fig. 3), serious discrepancies can arise when it is used to predict the distribution widths.

Another empirical attempt to reproduce the shapes of the distributions is the "chi-squared" model of Baudinet-Robinet *et al.*^{13,14} Its principal advantage is to account for asymmetrical distributions, such as are found with gaseous targets.



FIG. 4. Plots of the ratios of adjacent equilibrium fractions F_{q+1}/F_q vs q. The nonlinearity is a sensitive indicator of departure from a Gaussian shape. The discontinuity in slope at q=8 is due to the L-M shell transition.

In solids, its range of application is for energies below $\sim 20 \text{ keV}/\text{amu}$. Thus, we have not applied this model to our data.

The independent-particle model, due to Dynefors et al.¹⁵ and Veje,¹⁶ combinatorially relates final charge-state probabilities for the exiting ions to the probability of capture of electrons from the downstream foil surface into unoccupied states. The model has been applied with some success to measurements at lower energies where only valence electrons are involved.¹⁶ At the higher energies used in the present work, three subshells, 2p, 3s, and 3p, come into play and the unambiguous assignment of a distinct capture probability to each of the three subshells by fitting to the data becomes most unwieldy. A satisfactory application of the independent-particle model to cases where core vacancies play a dominant role must await the calculation of theoretical capture probabilities.

Figure 4 displays a plot of F_{a+1}/F_q vs q for various incident energies. Such a plot is a sensitive indicator of departures from the usual symmetrical Gaussian shape expected with solid targets.¹⁷ The discontinuity in slope observed at q=8 is attributed to ionic shell structure, arising from depletion of the M shell. Moak *et al.*¹⁸ have observed a similar effect for 100 and 140 MeV bromine ions.

IV. CONCLUSIONS

We have measured equilibrium charge-state distributions of 2-20-MeV argon ions in carbon foils. Equilibration already has taken place in the thinnest foils used (~ 5 μ g/cm²). The effect of ionic shell structure is observed in these distributions. While the mean charge can be reliably obtained in this region on the basis of the Gaussian model, it was found that the detailed distributions could not be satisfactorily obtained by this or any other current model, rendering their measurement necessary.

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