Mean Charge of Ions ($5 \leq Z_1 \leq 26$) Emerging from Carbon Foils: Evidence for the Effect of Inner-Shell Vacancies

W. N. Lennard and D. Phillips
Solid State Science Branch, Atomic Energy of Canada Limited Research Company, Chalk River Nuclear Laboratories, Chalk River, Ontario K0J 1J0, Canada
(Received 8 April 1980)

The mean charge, $\bar{q}$, has been measured for projectiles $5 \leq Z_1 \leq 26$ exiting thin carbon foils at velocities $0.8 \leq v/v_0 \leq 1.0$. It is observed that $\bar{q}(Z_1)$ increases strongly with Z_1 with a broad peak occurring at $Z_1 \sim 15$. It is shown that much of the enhancement can be accounted for by post-foil Auger decay of projectile inner-shell vacancies.

PACS numbers: 34.50.Hc, 34.70.+e, 34.90.+q, 35.10.Hn

The mean charge, $\bar{q}$, of energetic heavy ions which penetrate through matter reaches an equilibrium value that is determined by competition between capture and loss of electrons. In general, $\bar{q}$ as measured downstream from solid targets ($\bar{q}_s$) is greater than after passage through gas targets ($\bar{q}_g$). Betz and Grodzins[1] (BG) proposed a model for $\bar{q}$ wherein substantial multiple excitation will occur within a solid but not in a gas; the major part of the difference $\Delta\bar{q} = \bar{q}_s - \bar{q}_g$ will then arise from deexciting Auger transitions *after* the projectile exists from the solid. Recent x-ray measurements[2-4] have revealed significant degrees of inner-shell excitation for S, Cl, and Ar projectiles inside solids. However, no direct evidence for the necessary Auger-electron yield has been found for 60-MeV I and 30-MeV Cu projectiles exiting thin solid targets.[5,6] Further, Baragiola, Ziem, and Stolterfoht[7] have shown, by measuring the downstream yield of projectile *LMM* Auger electrons for low-velocity Ar ions incident on carbon foils, that the BG model cannot account for more than ~25% of $\Delta\bar{q}$, the gas-solid difference. In general, the importance of inner-shell vacancies to post-foil mean charge values is not yet clearly established.

We report here measurements of the post-foil $\bar{q}$ values obtained for projectiles $5 \leq Z_1 \leq 26$ incident at low velocities, $v \sim v_0$, on thin carbon foils. These data, combined with recent Auger-electron-yield measurements, show that both the projectile K- and L-shell–vacancy fractions that exist within solids can have a strong influence on post-foil $\bar{q}$ values at low projectile velocities. Our results are the first evidence that changes in $\bar{q}$ as a function of Z_1 are not solely due to pronounced variations in ionization potentials as electron shells are filled.[8]

The experimental technique was similar to that described by Smith and Whaling.[9] A magnetically analyzed beam of ions from the Chalk River Nuclear Laboratories 2-MV high-voltage mass separator impinged on self-supporting carbon foils located in a large vacuum chamber at a working pressure of $\sim 3 \times 10^{-7}$ Torr. Incident projectile energies were in the range 16–25 keV/amu. The beam traversed in succession a collimator (1.5 mm×1.5 mm), a thin carbon foil (7 mm diam) positioned ~45 cm behind the collimator, an electroformed aperture (0.04 mm diam) located 36 cm downstream from the foil, a parallel-plate electrostatic analyzer (ESA) and an exist slit of adjustable width located 0.6 cm off the beam axis. The exit slit width could be varied to yield an ESA resolution, $\Delta E/E$, in the range (5–15)%. The ESA entrance aperture was positioned to accept particles scattered $< 0.1°$ from the forward direction. Ions were detected by a thin (1 mm) CsI(Tl) scintillator optically coupled to a photomultiplier. Another slit located on the beam axis permitted neutral particles to strike the same scintillator when a movable shutter was opened. The carbon foil thicknesses were measured by Rutherford backscattering of 1.6-MeV He^+ ions. The thicknesses were varied between 7 and 15 μg cm^{-2} to see that charge-state equilibrium was attained.[9-11] We did not observe any dependence on projectile charge state using incident Ne^+ and Ne^{++} beams, in agreement with earlier measurements.[10]

Charge-state spectra were recorded by measuring ion intensities as a function of the ESA voltage, which was ramped from 0–5 kV in a sawtooth manner with periods in the range 0.2–20 s. The voltage sweep (>100 cycles per measurement) provided for normalization to the beam current, although the latter was kept constant to within 15% during any measurement. A typical charge-state spectrum is shown in Fig. 1 for 744-keV $^{31}P^+ \to C(7\ \mu g\ cm^{-2})$. The exit velocity of the beam from the carbon foils was determined with use of recent heavy-ion stopping-power results.[12]

 © 1980 The American Physical Society

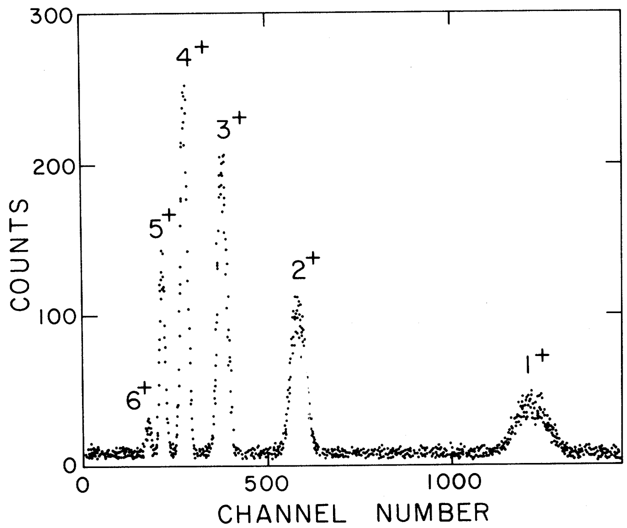

FIG. 1. Charge-state spectrum (excluding neutrals) for 744-keV $^{31}P^+$ incident on a carbon foil (7 μg cm^{-2}), corresponding to an exit energy 710 keV ($v/v_0 = 0.961$). The charge-state fractions are proportional to the heights of the appropriate peaks. The channel number is proportional to the voltage on the ESA deflection plates.

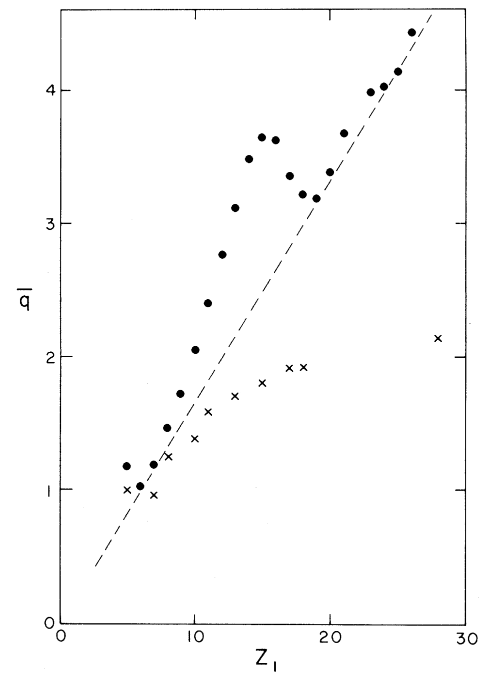

FIG. 2. The solid dots show the mean post-foil ionization, $\bar{q} = \Sigma q\varphi(q)$, measured for projectiles, Z_1, emerging from a thin carbon foil. The data are all for a common exit velocity, $v = v_0$. The crosses show gas target results ($Z_1 \to N_2, O_2$) taken from Ref. 14. The uncertainties in the present results are (1–3)%. See text for a description of the dashed curve.

The neutral fraction was measured directly by taking the ratio of yields alternately with and without high voltage on the ESA. Similarly, the negative-ion ($q = 1-$) fraction was measured relative to the $q = 1+$ fraction by reversing the ESA polarity. Significant $q = 1-$ fractions were observed only for $Z_1 = 6$, 8, and 9. The positions and widths of the charge-state peaks are inversely proportional to the charge state q, since the sweeping speed was constant. Thus, each charge-state fraction is proportional to the corresponding peak height, or to the peak area multiplied by q. The background arises from photomultiplier dark current.

In this Letter, we summarize our results through the mean charge, $\bar{q}(v) = \sum q\varphi(q,v)$ is the fraction of the beam emerging from the foil at velocity v and in charge state q as measured ~0.25 μs after exiting the foil. For each Z_1 (except $Z_1 = 22$), we have made measurements at a few velocities in the range $0.8 \leq v/v_0 \leq 1.0$ and interpolated the data (linearly) to obtain $\bar{q}$ values at any v in this region. The total uncertainty in $\bar{q}$ varies from 1% to 3%.

For the common exit velocity $v = v_0$ (~25 keV/amu), our results are shown as solid circles in Fig. 2. Where our data overlap other measurements (see Refs. 9–11, 13, and 14), the agreement is always better than 5% except for $Z_1 = 9$ where our values are systematically 15% larger than those of Ref. 10. The crosses are interpolations of gas target data ($Z_1 \to N_2$, O_2) taken from Ref. 14. We observe that $\bar{q}$ for projectiles emerging from (solid) carbon targets increases strongly with Z_1 and that there is a pronounced peak centered at $Z_1 \sim 15$. These new data do not correlate with Z_1-dependent effects expected on the basis of ionization potentials.[8] Our data also confirm that post-foil $\bar{q}$ values are unrelated to the effective charge values that parametrize stopping powers, although the Z_1 dependence of stopping powers in carbon exhibits an oscillatory behavior.[12]

In summary, there is no theory which describes $\bar{q}$ at kiloelectronvolt energies. Previous semiempirical estimates, discussed in detail in Ref. 8, have yielded $\bar{q} \propto Z_1^{1/3}$ and $\bar{q} \propto Z_1^{1/2}$. Clearly these prescriptions are inadequate in describing the present data.

In an effort to understand these new data, we reconsider the BG suggestion that post-foil Auger deexcitation can dramatically influence the observed mean ionization. This interpretation is supported by recent Auger-electron–yield measurements together with derived absolute inner-shell–vacancy fractions, f_i ($i=K,L$), for some of the projectiles studied here. Projectiles having equilibrium inner-shell vacancies (K or L shell) within the solid will increase their charge by one unit after exiting the foil, i.e., $\Delta\bar{q}_A = f_i$, where $\Delta\bar{q}_A$ represents the contribution to $\bar{q}$ due to post-foil Auger decay. Assuming that the radiative decay of the inner-shell vacancies can be neglected, we obtain $\Delta\bar{q} = \Delta\bar{q}_A$.

Baragiola, Ziem, and Stolterfoht[7] find for Ar ions that the fraction of projectiles with L vacancies is $f_L(\mathrm{Ar}) = 0.24$ at $v = v_0$. Similar absolute Auger emission measurements by Schneider *et al.*[15] with thin carbon foils yield $f_L(\mathrm{P}) = 0.63 \pm 0.19$, again at $v = v_0$. Thus we expect that $\bar{q}$ for P and Ar will contain contributions from the two-electron Auger process of 0.63 and 0.24, respectively. As Z_1 decreases from 18 to 11, the L-vacancy fraction increases for constant velocity projectiles.[16] However, the Auger process will shrink in importance as a decay mode as the number of available M-shell electrons also decreases. For example, for Mg ($Z_1 = 12$), the L-vacancy fraction may be very large indeed but few ions will have ≥ 2 M-shell electrons since $\bar{q} \sim 2$. Consequently, the Auger process can make only a small contribution to $\bar{q}$. For $Z_1 > 18$, $f_L(Z_1)$ is decreasing for $v \sim v_0$ and here $\Delta\bar{q}_A$ is again small.

The dashed line in Fig. 2 is the simplest representation for $\bar{q}(Z_1)$ in the absence of a post-foil Auger contribution with the conditions that (i) $\bar{q}(Z_1 = 0) \to 0$ and (ii) $\Delta\bar{q}_A \sim 0$ for $Z_1 \sim 20$–26. The enhancements observed at $Z_1 = 15$ and 18 relative to this line are ~ 1.1 and ~ 0.2, respectively, i.e., of the same magnitude as $\Delta\bar{q}_A$ allowed by the L-vacancy–fraction measurements. Further comparisons would appear to be unjustified in the absence of a theory for $\bar{q}$.

For $Z_1 \leq 10$, K-vacancy fractions are responsible for post-foil Auger deexcitation. Schneider *et al.*[15] have also measured projectile KLL Auger yields and deduced values of f_K for projectiles ($Z_1 = 5$–8) emerging from carbon foils at $v = v_0$. They find $f_K(B) = 0.19$, $f_K(C) = 0.035$, $f_K(N) = 0.013$, and $f_K(0) = 0.0025$. Here also we expect $\Delta\bar{q}_A = f_K$, as long as $\bar{q}$ is small (thus guaranteeing that $\omega_K \ll 1$). We therefore predict that $\bar{q}(B) - \bar{q}(C) \sim 0.15$ at $v = v_0$. Our data show that $\bar{q}(B)$ exceeds $\bar{q}(C)$ by 0.15 ± 0.04, in good agreement with the predicted difference.

For the limited velocity region studied here, $0.8 \leq v/v_0 \leq 1.0$, we observed that the $\bar{q}(v)$ data were represented accurately by linearly increasing functions of velocity for all Z_1. These results then indicate that the enhancement of the broad peak at $Z_1 \sim 15$ increases with decreasing projectile velocity. Since the post-foil Auger measurements[7,15] show that f_L values for both P and Ar ($Z_1 = 15$ and 18) have saturated for projectile velocities well below $v = v_0$, our velocity-dependent $\bar{q}$ results further support the interpretation that the broad peak originates from an inner-shell–vacancy effect.

Finally, since f_L and f_K are known to be sensitive functions of target material (Z_2),[4,17] it would be informative to make $\bar{q}$ measurements for fixed Z_1 (e.g., $Z_1 = 15$, where $\Delta\bar{q}_A = 0.63$ in carbon) on different targets. Good vacuum conditions are necessary to prevent a Z_2 effect from being obscured by surface hydrocarbon contamination. Such data for $Z_1 = (8, 9)$ and $Z_2 = (26, 27$, and $28)$ bear on the origin of the transient magnetic fields which act on nuclei slowing down in ferromagnetic materials.[18]

Our results represent the first evidence that the Z_1 dependence of $\bar{q}$ for low-velocity heavy ions is nonmonotonic with Z_1, and that the structure is not simply correlated with ionization potentials. Although the latter effects are expected and indeed observed, e.g., for the rare earths, $Z_1 = 58$–72,[19] the present data strongly suggest that the post-foil Auger decay of inner-shell vacancies makes a much larger contribution to $\bar{q}$ than that caused by shell effects, as evidenced by the enhancement observed at $Z_1 \sim 15$ in carbon. While our results have shown that the magnitude of $\Delta\bar{q}_A$ can be pronounced in some instances, they do not quantitatively support the BG model. The conclusion reached by Baragiola, Ziem, and Stolterfoht[7] for the particular case $Z_1 = 18$ in carbon has been shown here to be of more general validity: The gas-solid difference in measured $\bar{q}$ values *cannot* be attributed entirely to projectile inner-shell–vacancy fractions within the solid target. The strong, almost linear, increase of $\bar{q}$ with Z_1 even after subtraction of $\Delta\bar{q}_A$ leaves much of the gas-solid difference as yet unexplained.

[1]H. D. Betz and L. Grodzins, Phys. Rev. Lett. 25, 211 (1970).

[2]H. D. Betz, F. Bell, H. Panke, G. Kalkoffen, M. Welz, and D. Evers, Phys. Rev. Lett. 33, 807 (1974).

[3]F. Hopkins, Phys. Rev. Lett. 35, 270 (1975).
[4]L. C. Feldman, P. J. Silverman, and R. J. Fortner, Nucl. Instrum. Methods 132, 29 (1976).
[5]S. Datz, B. R. Appleton, J. A. Biggerstaff, T. S. Noggle, and H. Verbeek, in *Proceedings of the Sixth International Conference on Atomic Collisions in Solids, Amsterdam, The Netherlands, 1975, Abstracts of papers*, edited by F. W. Sars and W. F. van der Weg (North-Holland, Amsterdam, 1976), p. 162.
[6]S. Datz, B. R. Appleton, J. R. Biggerstaff, M. G. Menendez, and C. D. Moak, Bull. Am. Phys. Soc. 18, 662 (1973).
[7]R. A. Baragiola, P. Ziem, and N. Stolterfoht, J. Phys. B 9, L447 (1975).
[8]H. D. Betz, Rev. Mod. Phys. 44, 465 (1972).
[9]P. L. Smith and W. Whaling, Phys. Rev. 188, 36 (1969).
[10]P. Hvelplund, E. Laegsgaard, J. O. Olsen, and E. H. Pedersen, Nucl. Instrum. Methods 90, 315 (1970).
[11]P. Hvelplund, E. Laegsgaard, and E. H. Pedersen, Nucl. Instrum. Methods 101, 497 (1972).
[12]D. Ward, H. R. Andrews, I. V. Mitchell, W. N. Lennard, R. B. Walker, and N. Rud, Can. J. Phys. 57, 645 (1979).
[13]W. S. Bickel, H. Oona, W. S. Smith, and I. Martinson, Phys. Scr. 6, 71 (1972).
[14]A. B. Wittkower and H. D. Betz, At. Data 5, 113 (1973).
[15]D. Schneider, N. Stolterfoht, D. Ridder, H. C. Werner, R. J. Fortner, and D. L. Matthews, IEEE Trans. Nucl. Sci. 26, 1136 (1979).
[16]R. J. Fortner and J. D. Garcia, Phys. Rev. A 12, 856 (1975).
[17]W. N. Lennard and I. V. Mitchell, Nucl. Instrum. Methods 132, 39 (1976).
[18]K. Dybdal, J. S. Forster, and N. Rud. Phys. Rev. Lett. 43, 1711 (1979).
[19]L. A. Petrov, V. A. Karnaukhov, and D. D. Bogdanov, Zh. Eksp. Teor. Fiz. 59, 1926 (1970) [Sov. Phys. JETP 32, 1042 (1971)].