ANOMALIES IN THE NUCLEAR CHARGE DEPENDENCE OF HEAVY ION MEAN CHARGE IN HELIUM AND AIR

L. A. PETROV, V. A. KARNAUKHOV, and D. D. BOGDANOV

Joint Institute for Nuclear Research

Submitted June 23, 1970

Zh. Eksp. Teor. Fiz. 59, 1926–1931 (December, 1970)

The mean charge of heavy ions, from krypton to tungsten, moving through air or helium at $\sim 10^9$ cm/sec is measured using a gas-filled mass separator. It is shown that the mean charge is nearly proportional to the velocity (e.g., for krypton). Deviations from proportionality can be well accounted for by the expression $\bar{e} = av + b$ with different signs of b for helium and air. For ions from Kr to Cs the dependence of the mean charge on nuclear Z obeys the formula $\bar{e} \sim Z^n$, where $n = 0.57 \pm 0.07$ in helium and $n = 0.6 \pm 0.1$ in air. The situation changes for the rare earths: here $\bar{e}$ no longer increases with Z. For terbium the mean charge is 15% lower than would be expected on the basis of an extrapolation. This anomaly is attributed to shell effects. Filling of the inner 4f shell evidently leads to a decrease of the cross section for electron loss. The relation between the directly measured charge and the effective charge that determines the energy lost by stopping is discussed.

WHEN ions pass through matter their equilibrium distribution among the most intense charged groups is well represented by a Gaussian distribution and is characterized by two parameters, the mean charge ($\bar{e}$) and the half-width. The mean charge of an ion having atomic number Z depends on the velocity and the medium; the half-width is practically constant: $\Delta e/\bar{e} \approx 40\%$. Up to the present time most of the experimentally known mean charges of ions passing through gases have pertained to ions with $Z < 20$.[1] For heavier ions information has been obtained only in individual cases, mainly for fission fragments.[2-6] In the present work we measured the mean charges of ions from Kr to W which passed through He and air at $\sim 10^9$ cm/sec.

An anomalous reduction of the mean charge was found in the case of the rare earths. For terbium this reduction is 15% below the value obtained by extrapolating from lighter elements.

EXPERIMENTAL PROCEDURE

We used a gas-filled magnetic separator similar to that described in detail in [7]. During the traversal of the mass separator the multiple charge-exchange collisions with molecules of the filling gas cause a continuous variation of the ionic charge, which fluctuates about an equilibrium value. The magnetic rigidity of the ion depends on the average charge e_N [obtained by averaging over all (N) collisions], which approaches $\bar{e}$ for sufficiently large N. By measuring the ionic distribution with respect to $H\rho$, we calculate $\bar{e}$ using the familiar equation

$$\overline{H}\rho = mvc/\bar{e}. \qquad (1)$$

The ionic distribution with respect to the average charge e_N is Gaussian, but its half-width is smaller by a factor $k\sqrt{N}$ than for the equilibrium distribution ($k < 1$):

$$\frac{\Delta e_N}{\bar{e}_N} \approx \frac{\Delta e_N}{\bar{e}} = \frac{1}{k\sqrt{N}}\frac{\Delta e}{\bar{e}}.$$

The dispersion of the average charge and the broadening of the pattern as the result of multiple ionic scattering on the gas molecules are the principal factors determining the resolution of the gas-filled mass separator. At our experimental pressures (2.7 Torr for He and 0.35 Torr for air) the resolution with respect to $H\rho$ was 3–5%.

We began with Kr^{84} and Xe^{132} ions accelerated to 102 and 147 MeV, respectively, using the U-300 cyclotron of the JINR; the energies of these ions were varied by means of filters. Energies and the distribution with respect to $H\rho$ were measured by means of a scanner that was equipped with a surface-barrier detector[8] and was located in the focal plane of the magnetic system.

Other heavy ions were derived from nuclear reactions induced by a 180–190-MeV Ar^{40} beam in different targets.1) The reaction products included ions of Mo^{90}, Ag^{103}, Sn^{110}, Cs^{125}, certain rare earth elements, and tungsten. Receiving momentum from the bombarding particles, these isotopes left the target, passed through the magnetic system, and were stopped in an aluminum collector located in the focal plane. The mean energy of the reaction products, determined from kinematics and the target thickness, was in the range 30–50 MeV ($v/v_0 = 3$–4) for all the investigated ions. The thicker targets (1.5–3.0 mg/cm^2) that were used to enhance the yields caused considerable spread of the energy, and therefore velocity, of the recoiling ions ($\Delta v/v \approx 0.3$). However, as was shown for Ag^{103}, an increase of target thickness from 1.7 to 4.5 mg/cm^2 does not affect the position of the maximum in the distribution over $H\rho$ (with 1% accuracy), while the half-width of the distribution increases from 3.5% to only 4%.[7] This result follows from the fact that the mean charge is nearly proportional to the velocity: $\bar{e} \approx f(Z)v$. This means that the velocity drops out of the expression for the magnetic rigidity of an "equilibrium" particle: $\overline{H\rho} \approx mc/f(Z)$.

The distribution over $H\rho$ for Mo^{90}, Ag^{103}, Sn^{110}, Cs^{125}, and Tb^{149} ions was obtained by measuring the distribu-

1)The target occupied the position of the source in the magneto-optical system.

tion of the characteristic γ-activity (but for Tb^{149} the α activity) of the collector in the focal plane. Isotopes were identified according to the γ energies, measured with a Ge-Li spectrometer, and the half-lives. In the cases of the rare earth isotopes, except Tb^{149}, and tungsten identification by means of radioactivity was difficult; the reaction products were registered with mica track detectors in the focal plane.[9]

EXPERIMENTAL RESULTS AND DISCUSSION

1. Figure 1 shows the velocity dependence of the mean charge for Kr^{84} in helium and air. In both cases the experimental points are well fitted by a straight line:

$$\bar{e} = av/v_0 + b. \qquad (2)$$

Here the coefficients are a = 2.45 ± 0.1 and b = − 0.8 ± 0.3 for helium, a = 2.45 ± 0.1 and b = 0.8 ± 0.3 for air. According to Bohr,[10] an ion moving through a medium retains only the electrons having orbital velocities that exceed the ionic velocity ($v_e > v$). In the Thomas-Fermi statistical model this criterion leads to a mean charge that is proportional to the velocity:[11]

$$\bar{e} = Z^{1/3} v / v_0 \qquad (3)$$

for velocities $v_0 < v < Z^{2/3} v_0$. This relation is correct only in first approximation; a deviation from proportionality for heavy ions was noted in [4,5], where the charges of fission fragments were measured and the relation $\bar{e} = f(Z) v^{1-X}$ was proposed for their mean charge. Our data show that this relation is not entirely suitable, since the value of X depends on the ionic velocity. We note that the effect of pressure on the equilibrium charge of heavy ions[3] cannot lead to appreciable nonproportionality. Also, it was shown in [4,7] that a reduction of the helium pressure induces a large deviation from proportionality of $\bar{e}$ and v.

It should not be surprising that (3), which is of qualitative character, is not consistent with the experimental results. A more rigorous relation is

$$\bar{e} = \bar{\nu}(v) \frac{v}{v_0}, \qquad (4)$$

where $\bar{\nu}(v)$, an effective quantum number in the Thomas-Fermi model, is an average over electron velocities from zero to v. We know that for the most weakly and the most strongly bound electrons ν is close to unity, but that in the intermediate region it has a flat maximum value that is near $Z^{1/3}$. In first approximation (4) agrees with (3), since $\bar{\nu}(v)$ is close to $Z^{1/3}$ for sufficiently high velocities. However, a more detailed analysis should allow for the slow decrease of $\bar{\nu}(v)$ with diminishing velocity. It can be shown that for a small velocity range (4) can be represented by the linear expression (2) with $b < 0$, as was observed experimentally in helium. When we go from helium to air the ionic charges are increased, especially for the lower velocities. The sign of b in (2) is then reversed.

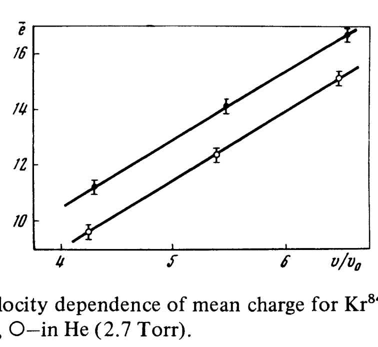

FIG. 1. Velocity dependence of mean charge for Kr^{84} ions: ●—in air (0.35 Torr), ○—in He (2.7 Torr).

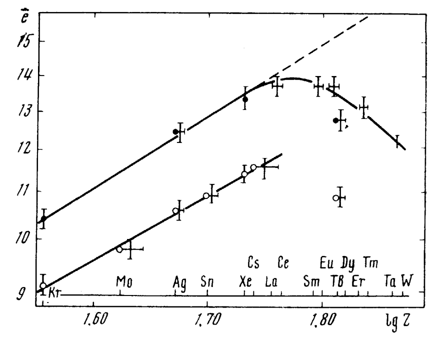

FIG. 2. Dependence of mean charge on Z ($v/v_0 = 4$): ●—in air (0.35 Torr), ○—in He (2.7 Torr).

2. We shall now consider how the mean charge depends on the atomic number of the ion at a given velocity. According to Born and Lindhard,[10,11] we can expect $\bar{e} \sim Z^{1/3}$. A power dependence on Z was also obtained in [12], where it was assumed, for better agreement with experiment, that an ion keeps electrons having velocities $v_e > \gamma v$, where γ is a slowly varying coefficient. To calculate v_e a statistical model of the ion was used, singling out either the least tightly bound electron or the outermost electron, and obtaining 1/3 and 2/3, respectively, as the corresponding power of Z. In their experiments with fission fragments Cohen and Fulmer[4] concluded that $\bar{e} \sim Z^{1/3}$ describes the results satisfactorily only for the light fragment group passing through helium. The charge ratio of light and heavy fragments is more consistent with $\bar{e} \sim Z^{0.6}$.

Figure 2 shows our data on mean charges of heavy ions having velocity $v/v_0 = 4$ in helium and air. The open and closed circles pertain to the cases when a definite isotope was registered in the focal plane. The uncertainty of Z in these cases is associated with the fact that a given isotope could be formed either directly in the reaction or through β^+ decay of isobars with higher Z. Crosses without circles pertain to data obtained by registering ions in the focal plane using mica track detectors. Mean values of Z and A were obtained by analyzing the yields of different nuclear reactions in the given isotope region:[13] the accuracies were ±0.5 and ±1.0%, respectively.

In the Kr–Xe region the points lie about straight lines representing $\bar{e} \sim Z^n$, with n = 0.57 ± 0.07 in helium and n = 0.6 ± 0.1 in air. A drastic change occurs in the rare earth region, where the mean charge ceases to increase and even manifests a tendency to decrease. This anomalous behavior occurs both in air and in helium. For terbium $\bar{e}$ is about 15% smaller than the value obtained by extrapolation of the formula that is valid in the Kr–Xe region.

The observed anomaly appears to be associated with shell effects. We know that in the rare earths a deep-lying 4f shell begins to be filled. We can assume that this process is accompanied by a reduction of the cross section σ_e for electron loss. On the other hand, we have charge equilibrium when σ_e equals the cross section σ_c for electron capture. Since $\sigma_c \sim e^2$, where e is the ionic charge,[11] the decrease of σ_e shifts the equilibrium toward a lower charge.

3. It is of interest to determine whether the observed anomaly for the mean charge also appears in the specific ionization of heavy ions. We begin by considering the more general question of the relation between the directly measured mean charge $\bar{e}$ and the effective charge Z* that determines the stopping loss. The theoretical situation is not clear, although it was noted in [11] that the effective charge Z* can exceed $\bar{e}$ appreciably because in close collisions the electronic shells of interacting particles will overlap. Pierce and Blann,[14] who analyzed the experimental specific ionization of heavy ions from S^{32} to I^{127}, concluded that Z* and $\bar{e}$ are close in gaseous media.[2)] This conclusion would be supported by a demonstration that Z* and $\bar{e}$ depend in an identical manner on the kind of slowing gas.

For the ionization loss of heavy ions we use the relation

$$\left(\frac{dE}{dx}\right)_{Z,A,v} = Z^{*2} f(Z_1, A_1, v), \tag{5}$$

where Z, Z_1, A, and A_1 are the atomic numbers and mass numbers of the ion and slowing medium. Following [14], to derive Z* we use the specific ionization ratio of a heavy ion and proton with the same velocity:

$$\frac{(dE/dx)_{Z,A,v}}{(dE/dx)_{p,v}} = \frac{(Z^*)^2}{(Z_p^*)^2} = (Z^*)^2, \tag{6}$$

where Z_p^*, the effective charge of a hydrogen "ion," is practically unity for energy > 0.25 MeV.[15] Specific ionization data were taken from [14] for heavy ions and from [16] for protons. Using (6), we obtain effective charges equal to 9.60 ± 0.25 and 11.2 ± 0.3 for Br^{79} at 31.5 MeV and I^{127} at 50.5 MeV ($v/v_0 = 4$), respectively, slowed in helium at ≈ 200 Torr. Our data (allowing for the pressure effect) yield $\bar{e}$ (Br^{79}) = 9.8 ± 0.3 and $\bar{e}$ (I^{127}) = 11.7 ± 0.3. Z* and $\bar{e}$ coincide within the limits of experimental error.

Table I shows how the effective charge for I^{127} ions depends on the stopping gas (argon, nitrogen, or helium). Averaging over the energies yields $(Z_N^*/Z_{He}^*)^2 = 1.19 \pm 0.04$ and $(Z_{Ar}^*/Z_{He}^*)^2 = 1.29 \pm 0.05$. The effective charge increases in heavier gases. The mean charge behaves similarly, but to a somewhat greater degree; our data yield $(\bar{e}_N/\bar{e}_{He})^2 = 1.38 \pm 0.08$, while Cohen and Fulmer[4] obtained $(\bar{e}_{Ar}/\bar{e}_{He})^2 = 1.45$. Thus the possible difference between Z* and $\bar{e}$ does not exceed 10%.

Table I

$E(I^{127})$, MeV	75	60	50	40	32
$(Z_{Ar}^*/Z_{He}^*)^2$	1.18±0,09	1.27±0.10	1.31±0.10	1.37±0.10	1.32±0.10
$(Z_N^*/Z_{He}^*)^2$	1.18±0,09	1.21±0,09	1.19±0,09	1.19±0,09	1.18±0,09

Table II shows the correlation between the stopping powers and the anomalies for the equilibrium charges

Table II

	Helium	Air
$\frac{(dE/dx)_{Tb}}{(dE/dx)_I}$	0.97±0,07	0.99±0.07
$(\bar{e}_{Tb}/\bar{e}_I)^2$	0.94±0,07	0.93±0.07

in the rare earth region. These data pertain to Tb^{149} and I^{127} at 17.4 MeV; the stopping power of Tb^{149} was taken from [17]. According to (6), except for a loss due to nuclear collisions we obtain

$$\frac{(dE/dx)_{Tb}}{(dE/dx)_I} = \frac{(Z_{Tb}^*)^2}{(Z_I^*)^2}. \tag{7}$$

The agreement, within error limits, of the ratios of stopping powers and squared equilibrium charges for Tb^{149} and I^{127} furnishes additional evidence that Z* and $\bar{e}$ are close. (We note that the agreement is improved by allowing for nuclear collisions in dE/dx.) The anomaly in $\bar{e}$ is repeated in the effective charge. If Z* varied according to $Z^* \sim Z^{1/2}$ for the rare earths, the specific ionization of Tb^{149} and I^{127} ions would differ by more than 20%.

In conclusion, the authors wish to express their deep gratitude to G. M. Ter-Akop'yan for his large contribution to the development of the gas-filled mass separator, and to N. Shadieva for assistance in the work done with mica detectors.

[1] V. S. Nikolaev, Usp. Fiz. Nauk **85**, 679 (1965) [Sov. Phys.-Usp. **8**, 269 (1965)].
[2] N. O. Lassen, Phys. Rev. **69**, 137 (1946).
[3] N. O. Lassen, Mat.-Fys. Medd. Dan. Vid. Selsk. **26**, No. 5 and No. 12 (1951).
[4] B. L. Cohen and C. B. Fulmer, Phys. Rev. **109**, 94 (1958).
[5] P. Armbruster, D. Hovestadt, H. Meister, and H. J. Specht, Nucl. Phys. **54**, 586 (1964).
[6] H. D. Betz and G. Hortig, Phys. Lett. **22**, 643 (1966).
[7] I. Bacho, D. D. Bogdanov, Sh. Dorotsi, V. A. Karnaukhov, L. A. Petrov, and G. M. Ter-Akop'yan, Prib. i Tekh. Éksp. **2**, 43 (1970).
[8] V. A. Karnaukhov, L. Rubinskaya, G. Ter-Akop'yan, V. Titov, and V. A. Chugreev, JINR Preprint R13-4454, Dubna, 1969.
[9] A. Kapuszik, V. P. Perelygin, S. P. Tretiakova, and N. H. Shadieva, Proc. Intern. Conf. on Corpuscular Photography, Florence, 1966.
[10] N. Bohr, Phys. Rev. **58**, 654 (1940) and **59**, 270(1941).
[11] N. Bohr and J. Lindhard, Mat.-Fys. Medd. Dan. Vid. Selsk. **28**, No. 7 (1954).
[12] J. H. M. Brunings, J. K. Knipp, and E. Teller, Phys. Rev. **60**, 657 (1941).
[13] G. Kumpf and V. A. Karnaukhov, Zh. Eksp. Teor. Fiz. **46**, 1545 (1964) [Sov. Phys.-JETP **19**, 1045 (1964)].
[14] T. E. Pierce and M. Blann, Phys. Rev. **173**, 390(1968).
[15] W. Booth and I. S. Grant, Nucl. Phys. **63**, 481 (1960).
[16] W. Whaling, in Handbuch der Physik, S. Flügge, Ed., Springer-Verlag, Berlin-Göttingen-Heidelberg, Vol. 34, 1958, p. 193.
[17] J. Gilat and J. M. Alexander, Phys. Rev. **136**, B1298 (1964).

Translated by I. Emin
218

[2)] The effective charges Z* are close in solid and gaseous media; the equilibrium charges are considerably larger for ions emerging from a solid medium. This result appears to be associated with the "shaking off" of electrons at the interface of two media. [14]