Total Ionization in Nitrogen by Heavy Ions of Energies 25 to 50 keV*

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The total ionization produced by ions stopped in nitrogen gas has been measured in the energy range 25–50 keV. The measurements were made using an ion accelerator for producing the incident particles and a cylindrical ionization chamber with a differentially pumped open window. The following ions were studied: H^+, He^+, C^+, N^+, O^+, and Ar^+. The average energy loss per ion pair, W, ranges from 34.6 eV for 25-keV protons to 115.2 eV for 25-keV argon ions. A calculation indicates that a large part of the variation of W with initial ion energy and atomic species may be attributed to the variation of the energy ultimately going into the kinetic energy of the atoms of the gas.

INTRODUCTION

THERE is presently only a small amount of information concerning the total ionization produced by heavy ions of initial energy less than 100 keV when they are stopped in a gas. We report here experimental results for ionization in nitrogen gas by heavy ions in the mass range 1–40 amu and the energy range 25–50 keV. The velocity of the incident ion at these energies is comparable to that of orbital electrons so that one may expect processes which are relatively unimportant at higher energies to contribute to the loss of energy by the primary particle. At these lower energies elastic scattering may be a particularly important process in the energy loss of the incident ion. We have calculated the fraction of the energy of the incident ion going ultimately into motion of the gas atoms[1] as a whole and find that this effect alone can account to a first approximation for the experimentally observed variation of W (average energy to produce an ion pair) with energy and type of incident ion.

EXPERIMENTAL METHOD

The experimental apparatus and technique were essentially the same as those described earlier.[2,3] An ion beam from a low-energy accelerator was analyzed magnetically and electrostatically and allowed to enter a gas-filled ionization chamber through a 0.0065-in.-diameter differentially pumped aperture. The chamber was operated alternately as an ionization chamber to measure the ionization current and as a proportional counter to measure the incident particle rate. Periodic alternation of these measurements served to reduce uncertainties due to the variations present in the incident particle rate.

The collection characteristics of the ionization chamber for nitrogen gas are shown by Fig. 1. This figure gives the relative ionization as a function of collecting potential for 50-keV protons, 50-keV N^+ ions, and 50-keV Ar^+ ions. These curves are representative of a number of curves taken for various ions, energies, and pressures. They exhibit regions of saturation of over 100 V where the collected ionization is constant to within around 0.4%.

The ionization chamber was supplied with nitrogen gas which had a stated purity of 99.99%. Periodically gas samples were taken from the chamber and analyzed by means of a mass spectrometer. This analysis showed the impurities in the nitrogen to be always less than 0.5%, with oxygen and water vapor as the principal contaminants. The amount of contamination given by the mass spectrometer should be taken as an upper limit of the actual contamination in the chamber, since the process of handling the sample between chamber and spectrometer very likely introduced more impurities than were originally in the chamber gas.

RESULTS

The results of the ionization measurements are presented in Tables I–VI and Fig. 2 for the following ions: H^+, He^+, C^+, N^+, O^+, and Ar^+. In Tables I–VI the following information is given for each measurement: (1) the initial energy E of the ion; (2) the ionization yield I in ion pairs per incident particle; (3) the average energy loss per ion pair in eV/ion pair ($W=E/I$); and (4) v_0/v, where $v_0=e^2/\hbar$ is the velocity

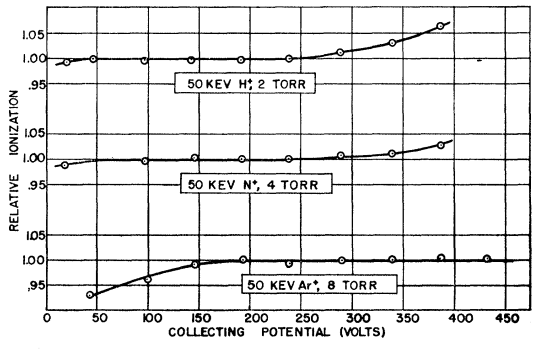

FIG. 1. Saturation curves for H^+, N^+, and Ar^+.

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[1] In subsequent discussion we will, for brevity, refer to gas *atoms*. For the case of polyatomic gases it is to be understood that these "atoms" are molecules.

[2] J. A. Phipps, J. W. Boring, and R. A. Lowry, Phys. Rev. **135**, A36 (1964).

[3] R. A. Lowry and G. H. Miller, Phys. Rev. **109**, 826 (1958).

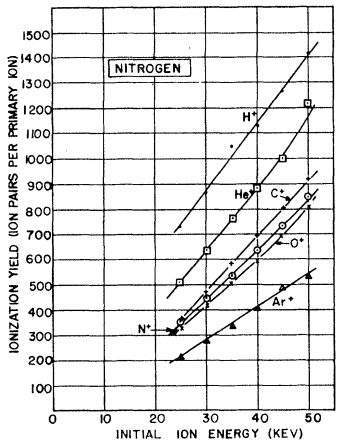

FIG. 2. Ionization yield for various ions.

of electron in the first Bohr orbit of hydrogen, and v is the initial velocity of the incident ion. All measurements were made at a chamber pressure of 4 Torr. The principal sources of the uncertainties in the ionization yield listed in Tables I–VI were statistical uncertainties in the yield as averaged from individual ion current and counting rate determinations, and uncertainty in the background current of the electrometer used to measure the ionization current. To provide a comparison of the results for the various ions with equal velocities, the tabulated values of W have been plotted in Fig. 3 as a function of v_0/v.

DISCUSSION OF RESULTS

As shown in Fig. 3, the ionization results for nitrogen gas generally follow the pattern that was seen in argon.[2] For a given ion, W is seen to increase as the energy or velocity is decreased (except for H^+), while for a given initial ion velocity, W is found to increase slightly as the mass of the ion increases. Some differences, however, should be noted between the general patterns of the results for argon and nitrogen. First, the pattern of increasing W with increasing ion mass for a given initial ion velocity appears to hold for all of the measurements in nitrogen gas, whereas in argon gas the value of W for the low-velocity argon ions was found to be less than that for the nitrogen ions of equal initial velocity. The second difference to be noted between the results for argon and nitrogen is that the increase of W in going from protons to He^+ ions is much larger for nitrogen gas than for argon. A third difference between the results for argon and nitrogen is that the value of W for protons in argon (around 27 eV/i.p.; i.p.=ion pair) is slightly larger than the quoted values of

TABLE I. Ionization yield for H^+ in nitrogen.

Ion energy keV	I Ion pairs per primary ion	W eV/i.p.	v_0/v
25.0	722±5	34.6±0.3	1.00
30.0	866±4	34.6±0.2	0.916
35.0	1049±7	33.4±0.2	0.846
40.0	1130±6	35.4±0.2	0.792
45.0	1269±25	35.5±0.7	0.750
50.0	1418±9	35.3±0.3	0.709

TABLE II. Ionization yield for He^+ in nitrogen.

Ion energy keV	I Ion pairs per primary ion	W eV/i.p.	v_0/v
25.0	511±4	48.9±0.5	2.00
30.0	638±4	47.0±0.3	1.84
35.0	767±4	45.6±0.3	1.70
40.0	881±5	45.4±0.3	1.59
45.0	1009±4	44.6±0.2	1.49
50.0	1216±5	41.1±0.2	1.41

TABLE III. Ionization yield for C^+ in nitrogen.

Ion energy keV	I Ion pairs per primary ion	W eV/i.p.	v_0/v
25.0	372±2	67.2±0.4	3.47
30.0	470±2	63.7±0.4	3.16
35.0	578±3	60.6±0.3	2.93
40.0	692±3	57.8±0.3	2.74
45.0	801±3	56.2±0.3	2.58
50.0	913±4	54.7±0.3	2.46

TABLE IV. Ionization yield for N^+ in nitrogen.

Ion energy keV	I Ion pairs per primary ion	W eV/i.p.	v_0/v
25.0	352±2	71.1±0.4	3.74
30.0	447±2	67.2±0.4	3.41
35.0	540±2	64.8±0.3	3.16
40.0	637±3	62.8±0.3	2.96
45.0	736±3	61.1±0.3	2.79
50.0	849±4	58.8±0.3	2.64

TABLE V. Ionization yield for O^+ in nitrogen.

Ion energy keV	I Ion pairs per primary ion	W eV/i.p.	v_0/v
25.0	325±2	77.0±0.5	4.01
30.0	423±2	71.0±0.5	3.66
35.0	502±3	69.8±0.4	3.39
40.0	597±3	67.0±0.4	3.16
45.0	693±3	65.0±0.3	2.98
50.0	818±5	61.1±0.4	2.83

TABLE VI. Ionization yield for Ar^+ in nitrogen.

Ion energy keV	I Ion pairs per primary ion	W eV/i.p.	v_0/v
25.0	217±1	115.2±0.7	6.33
30.0	280±1	107.3±0.6	5.76
35.0	337±2	103.8±0.6	5.34
40.0	409±2	97.9±0.6	5.00
45.0	473±3	95.1±0.6	4.73
50.0	534±2	93.6±0.5	4.48

26.4 eV/i.p. for 5-MeV alpha particles[4] and 26.7 eV/i.p. for 1.8-MeV protons,[5] while our value of around 35 eV/i.p. for protons in nitrogen is less than the quoted values of 36.4 for the high-energy alpha particles[4] and 36.7 for the high-energy protons.[5] Because of the fact that W for our low-energy protons in nitrogen was less than the high-energy value, a special effort was made to compare the results for 50-keV protons in nitrogen to those for 50-keV protons in argon. To this end, several determinations were made for each of these cases, with the results consistently agreeing with those quoted above. In the results for protons in nitrogen there is a slight indication of an increase of W with increasing energy, but the size of this effect is about equal to the uncertainties in the measurements.

The measurements for helium ions in nitrogen indicate an increase of W with decreasing energy. Although these measurements are for energies considerably less than those of Jesse[6] and those of Schaller, Huber, and Baumgartner,[7] the general pattern seems consistent with their results. Jesse found that W for alpha particles increased from 36 eV/i.p. at 9 MeV to 38 eV/i.p. at 1 MeV, while Schaller *et al.* obtained a fairly constant value of 41 eV/i.p. for helium ions in the energy range 270–630 keV.

INTERPRETATION

In the past few decades, under the particular influence of the work of Bohr,[8] there has been considerable development in a theoretical understanding of the relative importance of the processes involved in the penetration of atomic particles through matter. Specific reference is made here to the article by Lindhard, Nielsen, Scharff, and Thomsen.[9] In this section we employ their development in an analysis of the above data for ionization in nitrogen and of the recently published data[2] for ionization in argon.

An ion may lose energy in its passage through a gas by inelastic collisions with the atoms of the gas, resulting in excitation and ionization and by elastic collisions where the kinetic energy lost by the incident ion is transferred to kinetic energy of the recoil atom. In each of these processes the secondary particles (electrons from ionization processes and the recoil atoms from elastic collisions) may lose their energy in subsequent collisions. The inelastic processes may be termed electronic processes since they involve interaction between the incident particle and the electrons of the

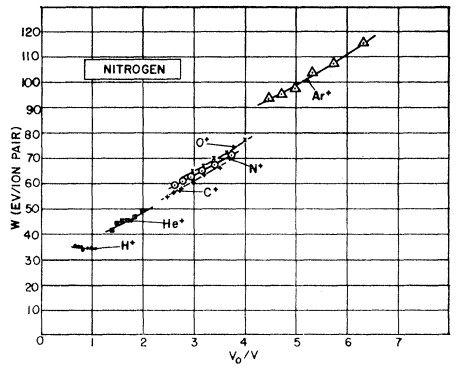

FIG. 3. Comparison of W values of different incident ions at various initial velocities v. $v_0=e^2/\hbar=2.2\times10^8$ cm/sec.

gas atoms. The elastic collisions, however, result in the transfer of energy to motion of entire gas atoms. Because of the relatively small mass of electrons, it is unlikely that a significant amount of the energy that originally went into electronic processes will ultimately end up in atomic motion. On the other hand, some of the energy going into kinetic energy of recoil atoms may ultimately go into electronic processes. It is convenient to divide the initial energy (E) of the incident particle into that energy ultimately going into electronic processes (η) and that energy finally going into atomic motion (ν).

Lindhard *et al.*[9] have derived a basic integral equation for any physical additive quantity that would result from the passage of an atomic particle through matter. They consider that portion of the energy of the incident ion ultimately going into electronic processes, and that portion finally dissipated as atomic motion as two such quantities.

Under the assumptions of an inverse square potential for the elastic (or nuclear) scattering cross section and of a stopping cross section for electronic processes proportional to the square root of the energy[10] of the incident ion,[11] the cumulative energy transferred to atomic motion is found to be

$$\nu(E)=E_c\{-12+6[1+2(E_c/E)^{1/2}]\ln[1+(E/E_c)^{1/2}]\}, \quad E<E_c, \quad Z_1=Z_2, \quad (1)$$

[4] W. P. Jesse and J. Sadauskis, Phys. Rev. **90**, 1120 (1953); T. E. Bortner and G. S. Hurst, *ibid.* **93**, 1236 (1954).
[5] H. V. Larson, Phys. Rev. **112**, 1927 (1958).
[6] W. P. Jesse, Phys. Rev. **122**, 1195 (1961).
[7] L. Schaller, P. Huber, and E. Baumgartner, Helv. Phys. Acta **36**, 2 (1963).
[8] N. Bohr, Kgl. Danske Videnskab. Selskab, Mat. Fys. Medd. **18**, No. 8 (1948).
[9] J. Lindhard, V. Nielsen, M. Scharff, and P. V. Thomsen, Kgl. Danske Videnskab. Selskab, Mat. Fys. Medd. **33**, No. 10 (1963).
[10] J. Lindhard and M. Scharff, Phys. Rev. **124**, 128 (1961).
[11] The electronic stopping cross section is taken as

$$S_e=8\pi e^2a_0Z_1^{7/6}Z_2Z^{-1}vv_0^{-1},$$

and the nuclear stopping cross section for an sth power potential as

$$S_n=\frac{4\pi}{s-1}\left[\left(\frac{2Z_1Z_2e^2}{M_0v^2}\right)^2a_s^{2s-2}\frac{3s-1}{8s^2}\right]^{1/s}\frac{M_1M_2}{(M_1+M_2)^2}E,$$

with the inverse square potential ($s=2$) stopping cross section approximated by

$$S_n^0=(\pi^2/2.7183)e^2a_0Z_1Z_2Z^{-1/3}M_1(M_1+M_2)^{-1},$$

where $a_0=\hbar^2/m_ee^2$, v is velocity of the incident ion, $v_0=e^2/\hbar$, M_0 is the reduced mass, $a_s=0.8853\ a_0\ Z^{-1/3}$, and M_1 and M_2 are the masses of the incident ion and gas atom.

TABLE VII. Average value of W' obtained for nitrogen gas.

Ion	$W'=\frac{\eta\ (\text{calc})}{I\ (\text{expt})}$ eV/ion pair
	(Initial ion energy 25–50 keV)
Hydrogen	32.8
Helium	34.1
Carbon	31.9
Nitrogen	30.9
Oxygen	31.0
Argon	32.0

TABLE VIII. Average value of W' obtained for argon gas.

Ion	$W'=\frac{\eta\ (\text{calc})}{I\ (\text{expt})}$ eV/ion pair	Energy range (keV)
Hydrogen	25.8	8 –50
Helium	23.3	25 –50
Carbon	24.5	25 –50
Nitrogen	25.9	12.5–50
Oxygen	24.6	25 –50
Argon	21.1	25 –100

where E is the energy of the incident ion and E_c is the energy at which the electronic and nuclear stopping cross sections are equal. In our case E_c is given by

$$E_c=0.5174Z^{3/4}Z_1^{-1/3}A_1^{3}(A_1+A_2)^{-2},\quad (\text{keV})$$

where $Z^{2/3}=Z_1^{2/3}+Z_2^{2/3}$, A is the mass number, and the subscripts 1 and 2 refer to the ion and the gas atoms, respectively.

Equation (1) is valid only at energies where E is less than E_c but of the same order of magnitude. However, in this analysis, we assume the region of validity to be as indicated, i.e., $E<E_c$. Equation (1) assumes, also, that the incident ion is of the same atomic species as the atoms of the stopping medium ($Z_1=Z_2$).

Using Eq. (1) in the integral equations of Lindhard *et al.*,[9] we obtain the corresponding energy transfer to the atomic motion when the incident ion is of a different species from the atoms of the medium:

$$\nu(E)=2E_{1c}\left\{\xi_1-\ln(1+\xi_1)-\frac{1}{2a}\left[\frac{\xi_1^2}{2}-\xi_1+\ln(1+\xi_1)\right]\right.$$
$$\left.+\frac{3}{10a^2}\left[\frac{\xi_1^3}{3}-\frac{\xi_1^2}{2}+\xi_1-\ln(1+\xi_1)\right]\cdots\right\},\quad E<E_c,\quad (2)$$

where E_{1c} is the energy at which the electronic and nuclear stopping cross sections are equal for the two atomic species considered,

$$\xi_1=(E/E_{1c})^{1/2},\quad \delta=4M_1M_2(M_1+M_2)^{-2},$$

and

$$a=(E_c/\delta E_{1c})^{1/2}.$$

Again, Eq. (2) is applicable only in the energy range of the incident particles where $\delta E/E_c$ is less than unity.

In the above we have assumed the nuclear stopping cross section at low energies to be constant. At higher energies, however, any approximation to this cross section should decrease with increasing energy. If we call E_0 the energy at which the cross section ceases to be assumed constant, and employ an inverse $\frac{4}{3}$ power potential for energies above E_0, so that E_0 becomes

$$E_0=0.252(M_1+M_2)M_2^{-1}Z_1Z_2Z^{1/3},\quad (\text{keV}),$$

we obtain the energy ultimately going into atomic motion as

$$\nu(E)\approx(1+C_1)E_{1b}\ln\left(\frac{E_{1b}+E}{E_{1b}+E_0}\right)$$
$$-\tfrac{2}{3}E_{1b}\left\{\left(\frac{\delta E}{E_c}\right)^{1/2}-\left(\frac{E_{1b}\delta}{E_c}\right)^{1/2}\tan^{-1}\left(\frac{E}{E_{1b}}\right)^{1/2}\right.$$
$$\left.-\left(\frac{\delta E_0}{E_c}\right)^{1/2}+\left(\frac{\delta E_{1b}}{E_c}\right)^{1/2}\tan^{-1}\left(\frac{E_0}{E_{1b}}\right)^{1/2}\right\}$$
$$-\frac{3C_2E_c^{1/4}E_{1b}^{3/4}}{\sqrt{2}\delta^{1/4}}\left\{-\tfrac{1}{2}\ln\left(\frac{\xi^{1/2}+\sqrt{2}\xi^{1/4}+1}{\xi^{1/2}-\sqrt{2}\xi^{1/4}+1}\right)\right.$$
$$\left.+\tan^{-1}\left(\frac{\sqrt{2}\xi^{1/4}}{1-\xi^{1/2}}\right)\right\}+\nu_1(E_0)+C_3,\quad E>E_0,\ Z_1\neq Z_2,$$

where

$$E_{1b}=(E_0E_{1c})^{1/2},$$
$$\xi=E/E_{1b},\quad \xi_0=E_0/E_{1b},$$
$$C_1=\nu'(E_0)[1+(E_0/E_{1b})],$$
$$C_2=\left[\frac{2}{3}\left(\frac{\delta E_0}{E_c}\right)^{1/4}-\frac{2}{9}\left(\frac{\delta E_0}{E_c}\right)^{3/4}+\cdots\right],$$
$$C_3=\frac{3C_2E_c^{1/4}E_{1b}^{3/4}}{\sqrt{2}\delta^{1/4}}\left\{-\tfrac{1}{2}\ln\left(\frac{\xi_0^{1/2}+\sqrt{2}\xi_0^{1/4}+1}{\xi_0^{1/2}-\sqrt{2}\xi_0^{1/4}+1}\right)\right.$$
$$\left.+\tan^{-1}\left(\frac{2\xi_0^{1/4}}{1-\xi_0^{1/2}}\right)\right\},$$

$\nu(E_0)$ is Eq. (2) evaluated at E_0, and $\nu'(E_0)$ is the derivative with respect to the energy, evaluated at E_0. Equation (3) is derived under the assumption that the energy of the recoil atoms is less than E_0 so that Eq. (1) is valid for the recoil atoms. This assumption is valid under the conditions of the experiments reported here. Because the nuclear cross sections are strongly peaked in the forward direction, Lindhard *et al.*[9] assumed in the development of Eq. (1) that the energy transferred during a collision is small compared with the energy of the incident particle. The identical approximation has been assumed in the derivation of Eq. (3), and an attempt was made to keep the three

equations, Eqs. (1), (2), and (3), to the same order of approximation.

The value of ν as calculated from Eq. (3) may be subtracted from the initial ion energy E to give the energy lost to electronic processes

$$\eta = E - \nu .$$

Calculated values of η along with the experimentally observed ionization values may be used to obtain the average energy going into electronic processes for each ion pair produced. We call this energy W' $(=\eta/I)$ and tabulate its values in Table VII for the nitrogen data and in Table VIII for the argon data.[2]

It is interesting to note that by subtracting the energy lost to atomic motion the resulting W value W' is nearly constant for the various ions in a given gas and is approximately the same as the W value found experimentally at high ion energies (where ν is relatively small).

We may make a further comparison to illustrate the constancy of W'. We assume a fixed W' for a given gas and calculate W from the relation

$$W = E/I = W'E/\eta ,$$

where the values of η are those obtained from the calculations. The dashed curves in Fig. 4 show the comparison of W calculated in this manner with the experimental values for nitrogen gas which are the solid curves. Figure 5 gives the same comparison for argon gas. It is seen that the energy and mass dependence of the experimental curves are represented rather well by the calculations based on a constant value for W'. It should be emphasized that these calculations employ approximate stopping cross sections so that the absolute values of W' may vary somewhat from the values calculated here. Nevertheless, the calculations do illustrate that even though the measured values of W are far from constant in this energy range, the average energy required to produce an ion pair is quite constant if the energy loss to atomic motion is taken into account.

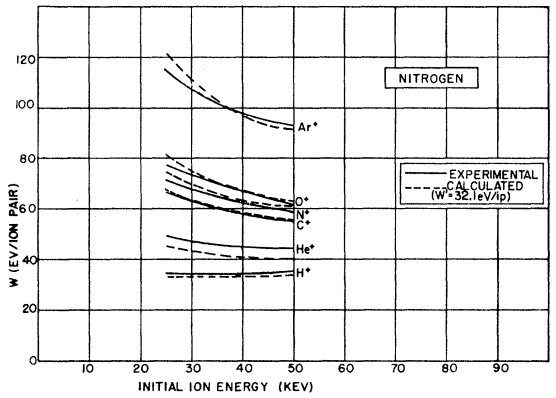

FIG. 4. Comparison of experimental values of W in nitrogen with values calculated for constant W'.

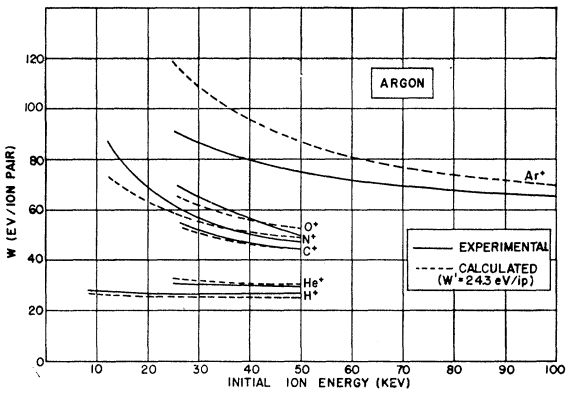

FIG. 5. Comparison of experimental values of W in argon with values calculated for constant W'.

In the calculations of ν for the various ions in nitrogen gas, the binding energy of the nitrogen molecule was neglected, i.e., we assumed that the ion was passing through a monatomic nitrogen gas. This is justified on the grounds that the molecular binding energy is small compared with the energy of the recoil atom. Lindhard *et al.*[9] have used the Thomas-Fermi model of the atom to obtain numerical solutions for η and ν for the case where the incident ion is of the same atomic species as the gas. For argon ions in argon gas, one may obtain from their results a W' between 30 and 32 eV/i.p. The fact that these values for W' are larger than the values calculated above merely reflects the uncertainty in the absolute value of the nuclear stopping cross section.