Projectile-charge dependence of the K x-ray emission cross section of carbon bombarded by megavolt nitrogen ions*

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The carbon K x-ray emission cross section of methane molecules bombarded by 1- to 5-MeV nitrogen ions has been measured for incident charges +2 to +5 under single-collision conditions. Only x rays with energies which penetrate the polypropylene window of a proportional counter are counted. The cross sections increase monotonically with ion energy. Above 2 MeV they also increase with ion charge, to a maximum of nearly 10^{-18} cm^2.

I. INTRODUCTION

When an ion with approximately 1-MeV energy per unit mass collides with a light target atom, the collision leaves the target atom with a number of vacancies distributed among its electron shells. The resulting ion may be studied by means of the x rays or Auger electrons which it emits. Measurements of the yield of x rays or Auger electrons provide data from which the probability of vacancy production may be determined, providing sufficient information is available on the state of ionization of the emitting ion. High-resolution spectral measurements, especially of x rays, provide information on the distribution of vacancies among the shells which, in addition to its intrinsic interest, is necessary for extraction of vacancy-production cross sections from yield data.

Interest in the vacancy-production cross sections has led to a number of experiments with high-energy ions under single-collision conditions. In such experiments, thin gas targets are used in order to gain control of the charge state of the incoming ion. In cases where K x rays were observed and the velocity V of the projectile is of the order of magnitude of the velocity u_K of electrons in the K shell of the target, it was soon discovered that the target x-ray yield increases rapidly with the charge state of the incident ion.[1-3] Subsequent measurements of the x-ray yield of[4] Cl → Ne and[5] Ar → Ne collisions produced x-ray cross sections as much as a factor of 10 above what is expected on the basis of Coulomb ionization theory[6] and the fluorescence yield ω_0 of the neutral atom. It was not known whether the discrepancy was associated with the increased fluorescence yield associated with highly ionized targets or with inadequacy of the Coulomb theory for cases in which $Z_1 > Z_2$ (where Z_1 and Z_2 are the atomic numbers of the projectile and target, respectively). X-ray cross sections measured for[7] Ne and[8] Ar with projectiles having $Z_1 < Z_2$ also display a dependence on the charge of the projectile.

The neon atom has been widely studied because it is monatomic, thus obviating all question of effects caused by the other atoms in the molecule,[9] and because it is observable with Si(Li) detectors and easily obtainable Bragg crystals. The results of a number of high-resolution studies of the satellites emitted by Ne bombarded by C, N, O, F, Cl, and Ar have been recently summarized,[10] and indeed the average fluorescence yield $\bar{\omega}$ can be as large as $10\omega_0$ for highly ionized projectiles. Although there is accordingly room for agreement with a Coulomb theory, measurements of the Auger-electron yield have cast doubt on the adequacy of this type of theory for collisions with $Z_1 \simeq Z_2$. (The Auger cross section is particularly useful, since it is approximately equal to the ionization cross section when the fluorescence yield is small, e.g., a maximum[10] of 0.16 for F^{+9} → Ne.) A dependence of the Auger yield on projectile charge state is, in fact, found.[11-13] However, the Auger cross section is nearly independent of the projectile charge i at all energies[13] when $i < Z_1 - 2$, and is approximately a linear function of i at 24 MeV for O → Ne and F → Ne when $i \geq Z_1 - 2$.[12] These results do not follow the i^2 dependence predicted by a simple Coulomb theory.

One possible explanation for these results is that a Coulomb theory applies, but that the charge producing the electric field has some other value than $i = Z_1 - n$, where n is the number of electrons on the projectile. In assessing the projectile-charge dependence of the Auger cross section for Cl → Ne collisions it has been suggested that the proper variable is the screened nuclear charge $Z_1 - \alpha n$, where α is an adjustable parameter.[14] In addition, measurements of x-ray satellite spectra have shown that the ionization produced

by projectiles having $Z_1 \geq i \geq Z_1 - 2$ (i.e., only K electrons) is a function of i rather than Z_1,[15] suggesting that the screened nuclear charge is active in this case, while for projectiles having L electrons the dependence only on i disappears.

The present measurements provide x-ray cross-section data for an atom simpler than neon in a region of projectile velocities such that $0.45 < V/u_K < 0.83$. The experiment is performed under single-collision conditions, but the carbon atom is present in the molecule CH_4, so that effects associated with the hydrogen electrons may possibly be present.[9] Previous measurements for collisions of high-energy heavy ions with methane are very few. The carbon K x-ray cross section for C, N, and Ne projectiles has been measured up to 150 keV.[16] The K Auger cross section appears to have been measured only for C ions up to 450 keV.[17]

II. EXPERIMENTAL PROCEDURE

Nitrogen ions created in an rf discharge ion source were accelerated by a 2.5-MeV Van de Graaff. Either singly charged N^+ ions were selected by an analyzing magnet to be sent into the collision region, or doubly charged N^{++} ions were chosen thereby enabling the ion energy to be increased to 5.0 MeV at a slightly reduced beam current. Either ion state, after selection by the magnetic analyzer, was sent through a gas-stripping cell about 6 ft long. Ions emerging in any one of the resulting charge states 1–5 were then selected by an electrostatic analyzer. Using this combination of magnetic and electrostatic analysis

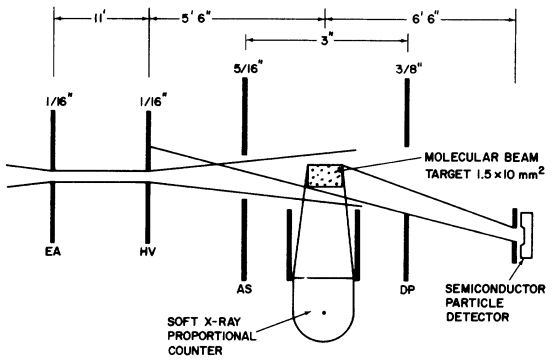

FIG. 1. Schematic diagram of experimental geometry (not to scale). Aperture separations and widths are given at the top of the figure. Ions of known mass, energy, and charge from a 3 MV accelerator (not shown) excite x rays in the methane gas of the molecular beam. Ions scattered from the carbon nuclei are monitored by a semiconductor particle detector placed at an angle of 8.8 mrad from the incident-beam direction.

of the ion beam, ions with a given atomic number, charge state, and energy could be selected.[18]

As depicted in Fig. 1, the ion beam was crossed with a thermal-velocity molecular beam of CH_4 and both scattered ions and carbon K x rays emerging from the crossing region were counted simultaneously. In such a case, the x-ray cross section is given by

$$\sigma_x = 4\pi \frac{X}{R}\left(\frac{d\sigma}{d\Omega}\frac{\Omega_R}{\Omega_x \epsilon_x}\right), \tag{1}$$

where X and R are the number of x rays and Rutherford scattered particles, respectively, detected during a run, Ω_x and Ω_R are the solid angles subtended by these detectors, $d\sigma/d\Omega$ is the differential cross section for screened Rutherford scattering toward the detector, suitably transformed from the center of mass to the laboratory coordinates, and ϵ_x is the efficiency of the proportional counter.

The molecular beam was created by a glass capillary effuser located at the center of an aluminum collision chamber, 10 in. high and 24 in. in diameter. The effuser was a glass matrix of many small holes, each 3 μm in diameter and 0.6 mm long, through which the target gas was forced.[19] The cross-sectional dimension of the molecular beam so formed was about 1.0 cm long in the direction of the ion beam and about 0.15 cm wide in the direction perpendicular to the ion beam. The beam was directed in a downward direction, first intersecting the ion beam, and then entering the mouth of a 6-in. oil-diffusion pump with a liquid-nitrogen-cooled baffle, which provided most of the pumping speed needed for the chamber plus the pumping speed needed to extract the target gas. A typical driving pressure of 6 Torr behind the effuser as measured with an oil manometer yielded a pressure of 0.94×10^{-2} Torr in the region of the ion beam. Outside the molecular beam, the pressure in the chamber was about 2×10^{-4} Torr. Further pumping was provided by a 6-in. sputter-ion pump and a 2-in. titanium evaporation pump located outside the differential pumping apertures labeled AS and DP in Fig. 1, and a 4-in. oil-diffusion pump located between collimating apertures EA and HV. As the molecular beam had a thickness of 3×10^{14} molecules/cm^2 and cross sections for charge transfer[20] are a maximum of 0.6×10^{-15} cm^2, the target was thin enough to ensure that impurity charge states were less than 18%. The performance of the entire pumping system was checked by using apparatus described elsewhere[21] to measure the charge-state distributions of 4-MeV ions passing through the electrostatic analyzer in charge state +3; with the effuser oper-

ating at typical pressure the population in charge states +2 and +4 totalled a few percent.

X rays were detected by a proportional counter with a window which was about $\frac{3}{4}$ in. in diameter and which was located about $1\frac{13}{16}$ in. from the scattering center. A very thin window of stretched polypropylene was chosen, since it seemed to have the best transmission-to-gas-permeability ratio of any material tried. The thin polypropylene was supported on fine nickel mesh, about 78% transparent, which was in turn supported on brass vanes with about $\frac{1}{8}$ in. spacing. The presence of the carbon in the polypropylene window introduces an absorption edge near the diagram line of carbon. Satellite lines, arising from carbon atoms with higher ionized states than a single K-shell ionization, may be sufficiently shifted in energy that they fall in the highly absorbing region of the polypropylene. No estimate can be made at this time as to how much satellite radiation is absorbed by the window. However, when protons were used to bombard N_2 gas flowing through the effuser, nitrogen radiation was visible, even though its energy is such that it is strongly absorbed by the window. This suggests that the window was highly irregular in thickness, and that there were areas of the window that were so thin as to transmit nitrogen or carbon satellite x rays.

The average efficiency of the counter for carbon K x rays was determined by a separate measurement of the yield produced by a 1.0-MeV protons incident upon methane. For this case σ_x may be taken as the product of the cross section for Auger-electron emission $\sigma_A = 1.08 \times 10^{-18}$ cm^2 measured by Toburen[22] and the fluorescence yield $\omega_K = 2.2 \times 10^{-3}$ found by Langenberg and van Eck.[23] The unknown parameter in Eq. (1) is then ϵ_x, which was deduced to be 0.043.

Utilizing windows as thin as those used in this experiment introduced a residual-gas background in the collision region due to the counter gas permeating through the film. As a precaution, the customary gas mixture of 90% argon and 10% methane (P-10) was abandoned since the presence of the argon in the collision region would have caused a high background in the x-ray count rate. The argon L x ray, with a much higher cross section than the carbon K emission, would have been detected by the proportional counter at nearly the same energy as the carbon $K\alpha$ x ray. The relatively poor resolution of the counter would not have resolved these different emissions. To alleviate this problem, pure methane was used as a counter gas, and the voltage on the detector anode was increased accordingly to provide the necessary gain.

To accumulate the x-ray counts, the pulse-height spectrum from the counter, after suitable amplification, was sorted by a multichannel analyzer. A spectrum from some relatively early measurements is shown in Fig. 2, for N ions incident on CH_4. The energy scale was established by bombarding CH_4 with protons; the resulting peak was centered in channel 12. The figure shows no evidence for nitrogen x rays, which would come in approximately channel 17. The background when the effuser was shut off contains x-ray counts from residual gas atoms, unexplained discharges with high voltage on in the proportional counter, and pickup of fast transients by the preamplifier used; the latter two rates being beam independent. The total number of x rays that were accumulated for a given number of incident ions were obtained by summing the counts in those channels that represented the carbon peak. No evidence for appreciable intensity of nitrogen x rays (relative to the intensity of carbon x rays) would have been obtained by summing the counts in those channels that represented a nitrogen peak.

Scattered ions were counted with a semiconductor particle detector, approximately $\frac{1}{8}$ in. in

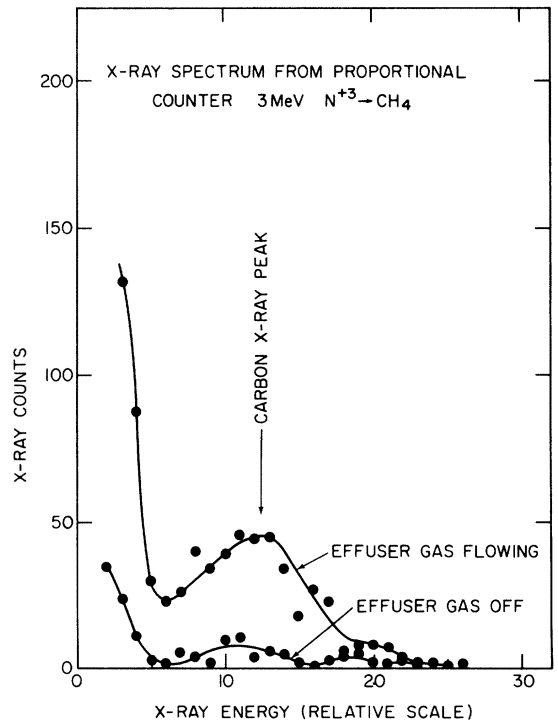

FIG. 2. X-ray spectra accumulated with molecular beam flowing and without molecular beam, for +3 nitrogen ions at 3 MeV. Typically, pulses with heights between 5 and 23 on the horizontal scale were summed to obtain the total number of counts X used for calculating the cross sections.

diameter, located $\frac{11}{16}$ in. away from the center of the ion beam at a distance of 78 in. from the scattering center. The differential pumping aperture DP (see Fig. 1) served as an antiscattering collimator to prevent ions scattered from collimator HV from reaching the particle detector. Since the angle by which the detected ions were scattered was small, electron-screening corrections to the usual Rutherford scattering formula were necessary. The tables of Everhart[24] and Bingham[25] were used to determine the screened Rutherford cross sections for nitrogen ions at all energies as well as for the 1.0-MeV protons that were used during the counter efficiency measurements. Values of the screening length a and the collision diameter b were computed according to the prescriptions given, and no adjustment was made to account for variation of a with ion-charge state.

A contaminant beam of unknown origin and $\frac{1}{4}$ the energy of the projectiles was present for ions incident in charge +2.[18] These ions were reduced to less than 10% of the main beam by adjustment of vertical electrostatic deflection plates used in combination with the quadrupole focusing magnets. A single-channel analyzer was used to select amplified pulses from the semiconductor detector from only the higher-energy group. As the x-ray cross sections decline with velocity, it was concluded the x rays produced by these ions would produce a systematic error of less than 10% in the cross sections for +2 projectiles.

The dependence of the x ray and scattered-particle yield upon the effuser driving pressure was measured. For both particles and x rays, the number of counts per incident ion was a linear function of effuser pressure, as expected.[19] In general, there was a negligible amount of x rays detected by the counter when the effuser gas flow was stopped. (The amounts were not negligible before a 1-in.-long aluminum tube, $\frac{3}{4}$ in. in diameter, was attached to the front of the detector so that there was no line of sight from the counter window to either of the apertures labeled AS and DP in Fig. 1.) This demonstrates that x rays detected were due to the effuser gas flow, and essentially none were due to methane gas at atmospheric pressure permeating the thin counter window, or to beams striking the walls of the chamber and associated apparatus. (The main beam was stopped in a Faraday cup located behind the semiconductor particle detector.) However, the particle detector counted some scattered ions even when no gas was coming through the effuser. This was probably due to a small amount of scattering from the aperture labeled AS in Fig. 1. This aperture was of such a diameter that no ions could pass through the preceding collimators and strike it; however, ions scattered once by the previous collimator could be scattered a second time by aperture AS into the Rutherford detector. For this reason, all data to be presented were taken with the effuser on and with the effuser off, and the slope $\Delta X/\Delta R$ of the graph of X plotted as a function of R was used in place of the ratio X/R in Eq. (1). Values of $\Delta X/\Delta R$ were accurate to 5%, with nearly all uncertainty coming from the counting statistics of ΔX.

III. RESULTS

Figure 3 and Table I give the x-ray cross sections as a function of ion energy for several incident-ion-charge states. For comparison the carbon K x-ray cross sections for nitrogen bombarding solid carbon[26] are also plotted in Fig. 3. The upward trend of the cross sections as the ion

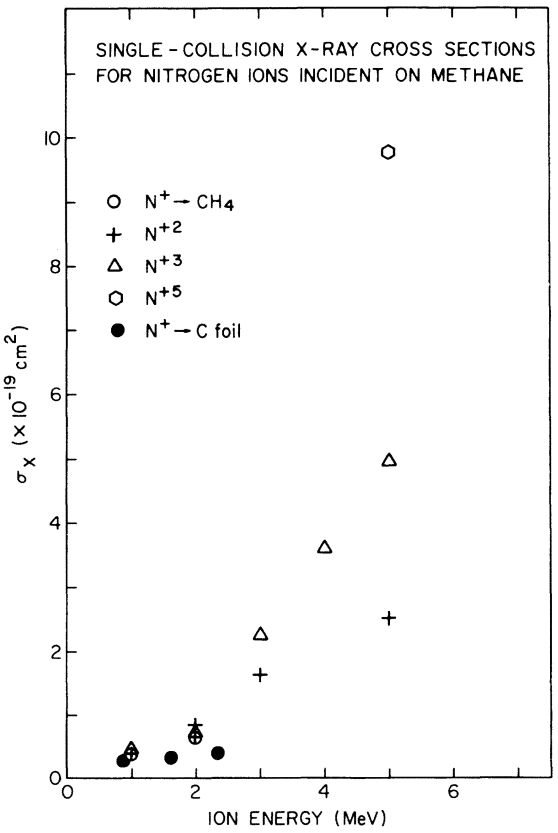

FIG. 3. Carbon K x-ray cross sections for methane molecules bombarded with nitrogen ions as a function of the energy of the ion, with the charge of the incident ion as a parameter. For comparison, the measurements of Ref. 26, made using a carbon foil target and a Bragg crystal spectrometer, are plotted as filled circles.

TABLE I. Carbon K x-ray-emission cross sections σ_x for methane molecules bombarded by nitrogen ions.

Ion energy (MeV)	Ion charge	σ_x (10^{-20} cm^2)
1	1	4.0
1	2	3.8
1	3	4.3
2	1	6.2
2	2	6.8
2	3	7.2
3	2	16.4
3	3	22.6
4	3	36
5	2	25
5	3	50
5	5	97

energy increases is apparent. Such an increase is expected for $V < u_K$. More significantly, there is a marked increase in the x-ray yield as the incident charge is increased, at least for energies above 2 MeV. At 5 MeV the cross section for charge +5 ions is approximately double that for charge +3 ions. From Fig. 3 it would appear that between 2 and 3 MeV there is an energy threshold for the charge-state dependence to become effective. Below this value, the cross sections are relatively independent of incident ionic charge, while at 5 MeV they are a linear function of ionic charge for the three charge states measured.

IV. DISCUSSION

Some observations can be made on the consistency of the observed charge-state dependence with theory. As for Al → Al and Ni → Ni,[27] the best theory for production of K vacancies in C → C collisions in solids seems to be a molecular-orbital theory involving close approach of the $2p\sigma$ and $2p\pi$ levels of the united atom.[26,28] In terms of this theory, the increase in x-ray production with projectile charge would be explained in terms of an increased number of vacancies in the $2p\pi$ orbital. It has been predicted that this number is $\frac{1}{3} N_v$ in low-velocity asymmetric collisions,[29] where N_v is the number of vacant states in the $2p$ level of the isolated nitrogen projectile. It has also been shown that deviations from $\frac{1}{3} N_v$ set in at high velocities, possibly because of long-range interactions between levels in the L shells of the approaching collision partners.[30] This interaction between L shells could, in principle, depend on projectile charge. However, the $2p$ level in the nitrogen projectile of the present collisions is probably completely vacant, or at most occupied with one electron. The greatest fractional change that could be produced in the number of vacancies is therefore probably much smaller than the observed fractional variation in x-ray yield with projectile charge. It may, therefore, be speculated that the increased yield is associated with a rapid increase in the fluorescence yield for target atoms left with less than three electrons, just as is found for neon.[10] The fluorescence yield for carbon does rise from 0.0026 in the neutral state to approximately 0.01 for various configurations of the three-electron ion, and to unity for a two-electron ion with one K electron.[26] The observed increase in yield may thus be associated with ionization of the L shell of the target atom, rather than with the primary K-vacancy production.

Theories[31-34] of a type other than the molecular-orbital theory may eventually be required to explain projectile-charge effects. Because of the possible attenuation of x rays with energies above the K absorption edge of carbon in the proportional counter window, the present data cannot be relied upon for exact comparison with theory. However, they do show that there is a large dependence on the charge of the projectile at the higher energies measured.

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