K-Shell Ionization of Carbon by Fast Protons*

L. H. Toburen
Battelle, Pacific Northwest Laboratories, Richland, Washington 99352
(Received 28 February 1972)

The cross sections for K-shell ionization of carbon have been measured for incident protons with energies from 0.3 to 2.0 MeV. These cross sections were obtained from Auger-electron yields measured for methane, ethane, ethylene, and acetylene gas targets assuming that each K-shell vacancy results in the emission of an Auger electron. The molecular surroundings of the carbon atom were found to influence the K-Auger-electron yield; e.g., for 1.0-MeV-proton impact, the carbon-K-shell ionization cross section was 1.00×10^{-18} cm^2 for acetylene and 1.10×10^{-18} cm^2 for ethane. These differences are larger than the estimated uncertainty in the relative values of the measured cross sections (5%). The estimated uncertainty in the absolute value of the cross sections is 20%. A comparison of our Auger-electron yields with published x-ray production cross sections was used to estimate the fluorescent yield of carbon. The fluorescent yield determined from these data was found to vary with proton energy from 0.0016 at 0.3 MeV to 0.0025 at 2.0 MeV.

INTRODUCTION

Previous measurements of the cross sections for ionization of the K shell of carbon by protons have been based on x-ray yield determinations.[1,2] Large uncertainties must be assigned to the ionization cross sections obtained in this way due to large uncertainties in the fluorescent yield which is used to convert from x-ray production cross sections to ionization cross sections. For example, fluorescent yields reported for carbon vary from $\omega_K = 0.007$[1] to $\omega_K = 0.0009$.[3] The two most recent measurements, with quoted uncertainties of less than 20%, differ by a factor of 3 ($\omega_K = 0.00113 \pm 0.00024$[4] and $\omega_K = 0.0035 \pm 0.0004$[5]). As was pointed out by Glupe and Mehlhorn,[6] these large uncertainties in the fluorescent yield for elements of low atomic number are reflected as small uncertainties in the Auger yield; $A_K = 1 - \omega_K$, where A_K and ω_K are the Auger and fluorescent yields, respectively. It is, therefore, much more accurate to deduce the K-shell ionization cross sections for low-Z elements from Auger-electron production cross sections than from x-ray production cross sections. For the case of carbon, the fluorescent yield is sufficiently small that x-ray fluorescence can be neglected completely with an error of less than 1%.

In this paper we report measurements of carbon K-shell ionization cross sections based on Auger-electron yields measured for protons on various hydrocarbon gases. Results were obtained for proton energies between 0.3 and 2.0 MeV. Several different target gases were used in order to determine the extent to which the K-shell ionization cross sections determined in this way were affected by the molecular surroundings of the carbon atom. The measured cross sections are compared to calculated values and a comparison of our Auger-electron yields with the x-ray yields of Khan *et al.*[1] is used to estimate the fluorescent yield for carbon.

EXPERIMENTAL METHOD

The basic experimental technique has been described in detail previously.[7] A proton beam from a Van de Graaff generator is directed through a differentially pumped target cell. Electrons ejected in proton-molecule collisions pass through a slit in the target cell, are energy analyzed by a cylindrical-mirror electrostatic analyzer, and are detected by a continuous channel electron multiplier. Electrons can be detected at angles between 20° and 130°; these limits are fixed by the physical size of the apparatus. Cross sections differential in both electron energy and emission angle are measured directly by this method. Integration with respect to emission angle yields cross sections differential in electron energy, and by integration with respect to both electron energy and emission angle the total ionization cross section is obtained. An illustration of the cross sections differential in electron energy for 1.0-MeV protons on hydrogen and ethane is shown in Fig. 1. The results for hydrogen are compared with theoretical values calculated using the Bethe formulation of the Born approximation[8] and using binary-encounter theory.[9] Excellent agreement is obtained between calculated and measured cross sections for electron energies greater than approximately 50 eV. For incident proton energies as high as 1.0 MeV and for a simple target such as hydrogen, it is assumed that the Born approximation and binary-encounter theory will provide reliable estimates of the cross sections for electron ejection so long as the electron energy is not too low. The agreement of measured and calculated cross sections for electron energies above 50 eV is, therefore, evidence of the accuracy of the absolute val-

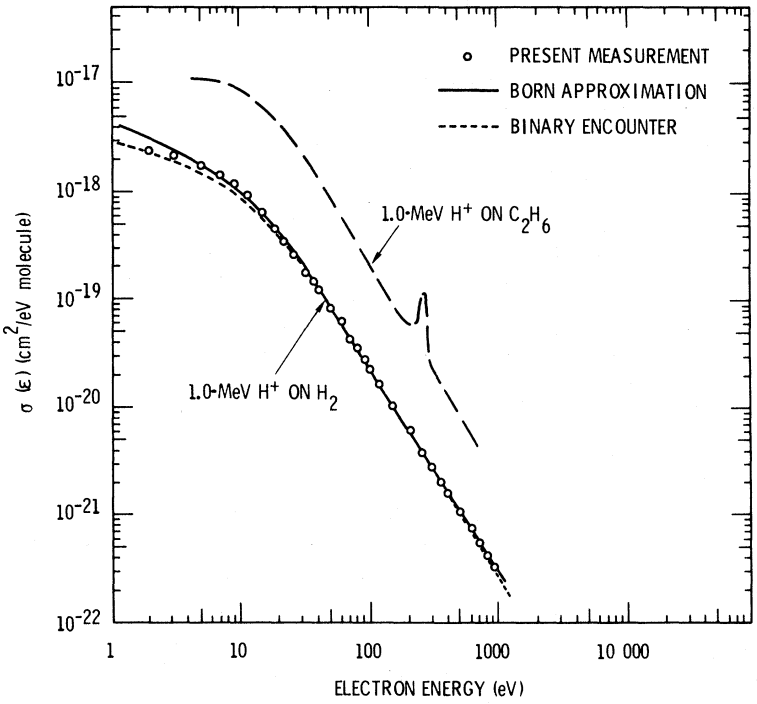

FIG. 1. Cross sections differential in electron energy for electron emission from hydrogen and from ethane by 1.0-MeV protons.

ues of the measured cross sections. The uncertainty in the absolute value of the cross sections for these continuum or "knock-on" electrons is estimated as ±20%. This estimate is based on the propagation of uncertainties estimated in the determination of detection efficiency, solid angle, gas pressure, and other experimental parameters from which the cross sections are calculated and appears to be a conservative estimate when comparisons are made with previous measurements[10] and with calculated cross sections.[10,11]

The electron emission spectrum for 1.0-MeV protons on ethane is shown in Fig. 1, primarily to indicate the similarity in the shape of the continuum-electron spectra for different molecules. This similarity allows one to scale the cross sections measured for one target molecule for use in determining the continuum-electron distribution in the region of the Auger-electron peak in the spectrum of another molecule. The K-shell ionization cross sections reported in this paper were determined from Auger yields obtained by integrating the area under Auger peaks such as the one shown in Fig. 1, after the contribution due to the continuum had been subtracted. It should be pointed out that although the shape of the continuum-electron distribution from hydrogen and ethane look similar, subtle differences made it more reliable to use the oxygen continuum in determining the baseline under the carbon K-Auger peak. The oxygen K-Auger transitions occur at sufficiently high energy (450 to 550 eV) that their influence on the continuum in the energy region of the carbon Auger lines is negligible. In Fig. 2 are shown the carbon K-Auger-electron distributions superimposed on the continuum for 1.0-MeV protons on ethane and acetylene. The dashed line indicating the contribution due to "knock-on" electrons was obtained by normalization of the oxygen spectra in the electron energy region between 150 and 170 eV. The oxygen continuum, when normalized, was found to fit the carbon continuum very well at electron energies both above and below the carbon Auger lines. The total area under the Auger-electron peaks was measured by means of a planimeter and the cross section for K-Auger emission (Auger-electron yield) was obtained from this area measurement. The cross sections shown in Fig. 2 are differential in electron energy only, i. e., after integration of the measured double-differential cross sections with respect to electron emission angle. Since K-Auger-electron emission is isotropic the total cross sections for Auger-electron emission can also be obtained from the double-differential cross sections by integrating the area under the Auger peak measured for a given angle and then multiplying this result by 4π to account for the solid angle. The advantage of using double-differential cross sections is that a backward angle can be chosen where the contribution from continuum electrons is small. K-shell ionization cross sections reported in this work were determined from an average of Auger-electron yields obtained at 125°, 90°, and from the integral over angles. In

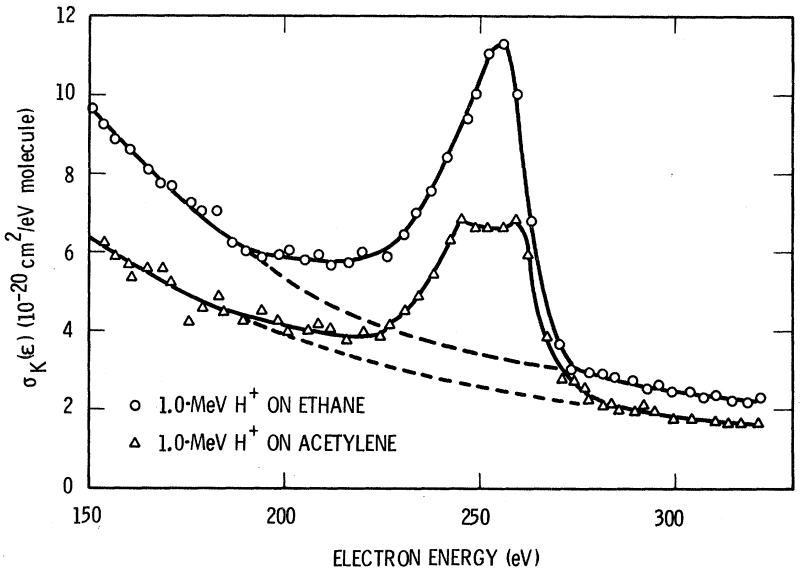

FIG. 2. Auger-electron spectra of carbon obtained for 1.0-MeV protons on ethane and acetylene.

general the cross sections obtained from each of these three yield determinations were in agreement to within ± 5%. The cross sections reported here are an average of values obtained from data acquired with two different electrostatic analyzers and three different arrangements for the collimators which define the solid angle into which electrons are accepted. These various conditions used for measuring emission cross sections served to cross check our ability to evaluate the experimental parameters such as solid angle, electron absorption, and analyzer transmission. (An analysis of the uncertainties in the parameters used to determine absolute values for the cross sections from the raw data can be found in Ref. 7.) The relative uncertainty in the cross sections based on electron yield measurements under the variety of conditions discussed above is estimated as ± 10%. When cross sections for various molecules were measured using the same electrostatic analyzer and collimator system the relative uncertainties in the cross sections were reduced to ± 5%. In obtaining K-shell ionization cross sections one must also consider the uncertainties associated with the area under the Auger lines and with the determination of the shape of the electron continuum used. By use of the electron emission spectra of oxygen normalized to the carbon continuum and by carefully measuring the peak areas, additional uncertainties of less than 10% are introduced. The uncertainty in the absolute value of the K-shell ionization cross sections is, therefore, estimated as ± 20%, due primarily to the over-all uncertainty in the measurement of the emission cross sections. The electron energy scale was calibrated to ± 1.0% using the measured Auger-line energies of Moddeman[12] and the proton energy has been calibrated to 0.5% using the Li^7 (p, n) Be^7 threshold reaction.

K-SHELL IONIZATION CROSS SECTIONS

The differences in the shapes of the K-Auger spectra for 1.0-MeV protons on ethane and acetylene shown in Fig. 2 indicate that the magnitude of the carbon K-shell ionization cross sections may depend on the molecular environment of the carbon atom. With the energy resolution available in this work, 3.5%, we are not able to resolve the fine structure in the Auger spectra; however, obvious differences could be observed between the Auger spectra of carbon which were measured from acetylene and ethane. By carefully controlling the experimental conditions, i. e., leaving all instrumental parameters fixed and changing only the target gas, cross sections could be measured with relative uncertainties as small as 5%. Under these conditions the Auger intensities resulting from methane, ethane, ethylene, and acetylene indicated subtle changes in the ionization cross sections as a function of the molecular binding. The results of these measurements are shown in Table I. Also shown in this table are results obtained from Auger-electron emission from carbon monoxide and carbon dioxide; however, these results were not obtained under such carefully controlled conditions, hence the larger uncertainties. The uncertainties quoted in Table I reflect the spread in the measurements corresponding to yield measurements at different angles and provide an indication of the relative uncertainties, i. e., precision rather than the accuracy of the absolute values. These results indicate that only a small

TABLE I. K-shell ionization cross sections for carbon in units of 10^{-18} cm^2 per atom.

Gas molecule	⟨Proton energy⟩ 0.3 MeV	1.0 MeV	2.0 MeV
Acetylene (C_2H_2)	0.94 ± 0.03	1.00 ± 0.06	0.71 ± 0.07
Ethylene (C_2H_4)	0.96 ± 0.03	1.06 ± 0.04	0.74 ± 0.02
Ethane (C_2H_6)	0.99 ± 0.05	1.10 ± 0.07	0.78 ± 0.03
Methane (CH_4)	0.94 ± 0.09	1.08 ± 0.07	
Carbon monoxide (CO)	0.90 ± 0.07	0.98 ± 0.07	0.81 ± 0.08
Carbon dioxide (CO_2)	0.86 ± 0.10	0.96 ± 0.05	0.70 ± 0.06

change in the K-shell ionization cross section is caused by the bonding differences in the molecules studied. There does, however, appear to be a systematic trend toward a decrease in the Auger-electron yield as the carbon-carbon bonding changes from single to triple bonds. Since we have already assumed that the Auger yield was unity in order to obtain K-shell ionization cross sections from the raw data, one cannot conclude whether the K-shell ionization cross sections depend on the chemical surroundings or if the Auger yield, and hence the fluorescent yield, of carbon varies within these molecular configurations. The fluorescent yield will be discussed further in the next section.

Our determination of the K-shell ionization cross sections for carbon from the Auger-electron yields is based on the assumption that each K-shell vacancy is filled by an Auger transition which contributes an electron to the electron peak we observe in the electron spectra between 180 and 300 eV. This, of course, implies no x-ray fluorescence. Although the precise value of the fluorescent yield is uncertain, the largest value which has been proposed ($\omega_K = 0.007$[1]) would influence our measurement by less than 1%. One must also consider the effect of the production of multiple vacancies on the K-shell cross sections measured in this work. Edwards and Rudd[13] have pointed out that the ratio of simultaneous K and L vacancies to total K vacancies produced in neon by 0.3-MeV protons is 0.46. There is, at the present time, no information regarding the dependence of this ratio on proton energy. Auger electrons which follow initial double vacancies, i. e., simultaneous vacancies in both the K and L shell, are shifted in energy but will fall in the energy region between 180 and 300 eV where our yield measurements are made. Since the 3.5% energy resolution of the electrostatic analyzer was not sufficient to resolve satellite lines resulting from multiple ionization, our system did not dicriminate between K-shell ionization and double-vacancy K- and L-shell ionization. The cross sections which we report are for K-shell ionization in which there is a high probability of simultaneous L-shell ionization.

In Fig. 3 are plotted the K-shell ionization cross sections which we have determined from the measured Auger-electron yields along with previous measurements deduced from x-ray yields. A fluorescent yield of 0.0021 was used in obtaining the K-shell ionization cross sections from the x-ray data. This fluorescent yield was chosen since it normalizes the x-ray results of Khan *et al.*[1] to our

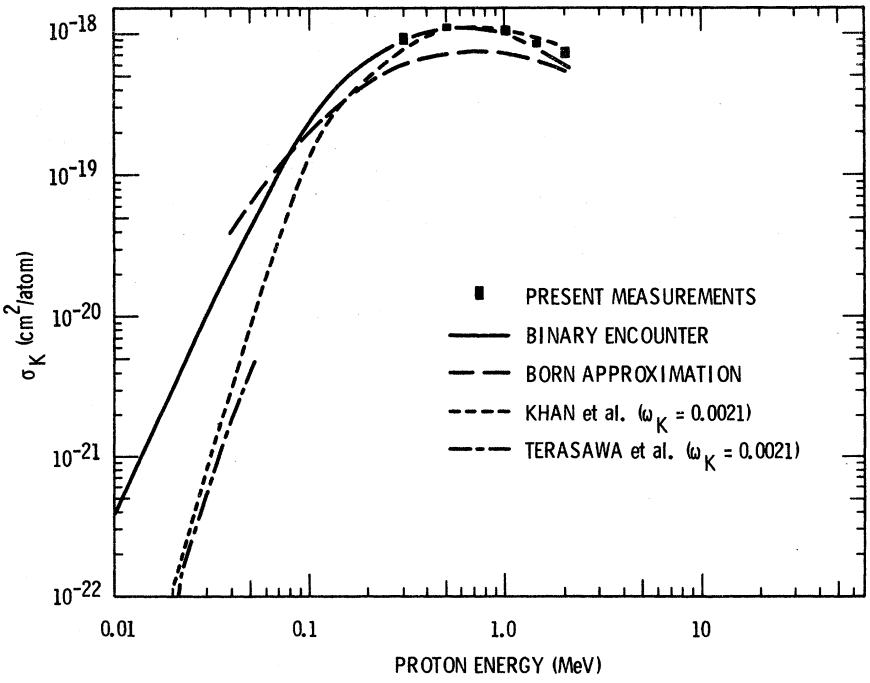

FIG. 3. K-shell ionization cross sections for protons on carbon. Theoretical results: binary-encounter theory (Ref. 14) and Born calculation (Ref. 1). Experimental results: Khan *et al.* (Ref. 1) and Terasawa *et al.* (Ref. 2).

results for proton energies of 0.5 and 1.0 MeV. Our measurements based on Auger-electron yields are shown as a range of values at each proton energy represented by the size of the plotted point. This range of values represents the cross sections obtained for various carbon-containing molecules and does not represent the uncertainty in the absolute value. The agreement between our measurements and the binary-encounter calculation of Garcia[14] is excellent. The results of the Born calculation reported by Khan *et al.*[1] and shown in Fig. 3 are just outside of the ± 20% uncertainty assigned to our values. The shape of the peak in the ionization curve predicted by the Born calculation is in better agreement with the measured shape, however, than is the somewhat narrower peak predicted by the semiclassical calculation. For lower incident proton energies, the measurements of Terasawa *et al.*[2] and of Khan *et al.*[1] are in approximate agreement when the same fluorescent yield is used, but disagree by more than an order of magnitude with the theoretical results of Garcia.[14]

K-SHELL FLUORESCENT YIELD

With the Auger-electron yields measured in this work and the published x-ray yields of Khan *et al.*[1] one can, in principle, obtain the fluorescent yield of carbon from the relationship

$$\omega_K = N_X/N_X + N_A \quad , \tag{1}$$

where N_X is the x-ray production cross section and N_A is the Auger-electron production cross section. The results of this calculation are shown in Fig. 4. Although the uncertainties in the yields used in Eq. (1) lead to an uncertainty in the absolute value of the fluorescent yield as large as 30%, the relative uncertainty in each value as a function of proton energy is more nearly 15%. The monotonic increase in the fluorescent yield with increasing proton energy is, therefore, highly indicative of an energy-dependent fluorescent yield. Such an energy dependence could be explained on the basis of multiple vacancies being produced in conjunction with the *K*-shell ionization. Larkins[15] has calculated the *K*-shell fluorescent yield for different atomic configurations with multiple $2p$ vacancies and a filled or empty $3p$ shell. His calculations (based on the argon atom) indicate that a 10% increase in the fluorescent yield can be expected in going from a single $1s$ vacancy to a double vacancy of the type $1s$, $2p$. A further increase is expected as the degree of multiple ionization increases to $1s$, $2p^4$; the fluorescent yield will then decrease with further ionization of the $2p$ shell. An increase in fluorescent yield with proton energy can, therefore, be expected if the degree of multiple ionization increases with proton energy. There is, however, no information available regarding the degree of multiple ionization of carbon as a function of proton energy. If one uses the ratio of the number of double vacancies $(K+L)$ to the total number of K vacancies produced in neon by 0.3-MeV protons as measured by Edwards and Rudd[13] (approximately 0.5) and an estimate of the change in fluorescent yield due to the double vacancy as calculated by Larkins[15] (approximately 10% increase), then our estimate of the single-vacancy *K*-shell fluorescent yield of carbon is $\omega_K = 0.001\,54$. This value is in approximate agreement with the results of the early calculation of Wentzel[16] with parameters reported by Fink *et al.*,[17] which yielded $\omega_K = 0.00144$, but somewhat smaller than the recent calculation

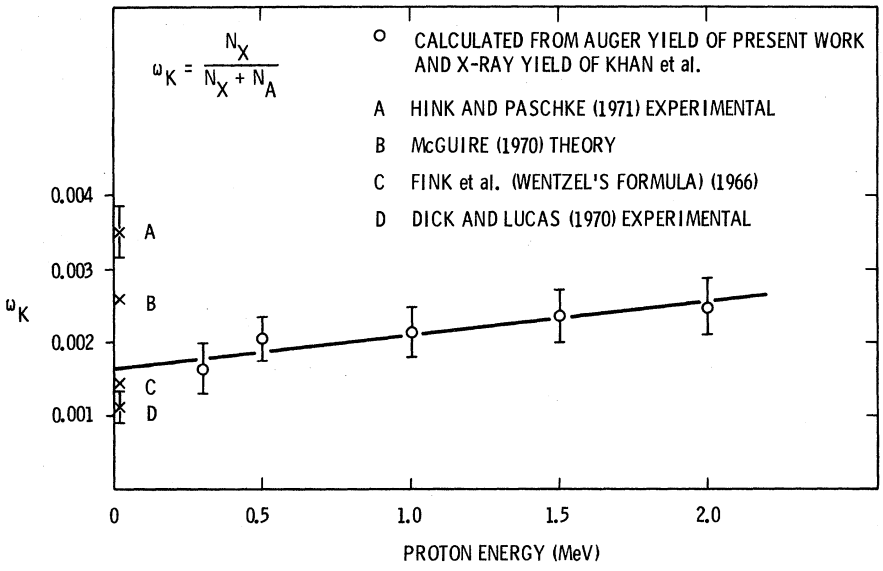

FIG. 4. *K*-shell fluorescent yield for carbon. Experimental results: Hink and Paschke (Ref. 5), Dick and Lucas (Ref. 4), and Khan *et al.* (Ref. 1). Theoretical results: Fink *et al.* (Ref. 17) and McGuire (Ref. 18).

of McGuire which yielded $\omega_K = 0.0026$.[18]

Owing to the fragmentary nature of the knowledge concerning multiple ionization by protons, the correction to our measurement to provide the fluorescent yield for single K-shell ionization which was discussed in the last paragraph is, at best, a rough estimate. One could argue that owing to the large uncertainty in the absolute values of the individual cross sections, a horizontal line could be drawn through the data points shown in Fig. 4 and that the fluorescent yield is independent of proton energy. Such a line would indicate a fluorescent yield of approximately 0.0021 and, as was shown in Fig. 3, this value normalizes the results of Khan *et al.*[1] to the ionization cross sections reported in our work and to those calculated by Garcia[14] for proton energies between 0.5 and 1.0 MeV. One is, however, not justified in using the uncertainties in the absolute values of each point when determining the shape of a curve drawn through the points plotted in Fig. 4. A line drawn through these points is dependent on the relative uncertainties associated with the measurement, not the uncertainties in the absolute value. It should also be noted that a fixed value of the fluorescent yield will provide agreement between x-ray-determined K-shell ionization cross sections and calculated values for only a limited energy range of the incident protons. This is further evidence that the fluorescent yield, as defined, must be a variable function of proton energy. The straight line drawn through the points in Fig. 4 is a least-squares fit to the data and fits each point to within 10%. An extrapolation of this straight line to zero-energy protons yields a fluorescent yield of 0.00162. From these measurements, coupled with the work of Khan *et al.*,[1] it appears that the fluorescent yield of carbon, extrapolated to single-vacancy production, must fall within the range $0.0011 < \omega_K < 0.0023$. This range includes values obtained from an estimate of the correction for double-vacancy production as well as from straight-line extrapolation and horizontal-line fits to the results shown in Fig. 4. The best estimate of the carbon K-shell fluorescent yield based on estimates of double-vacancy production is $\omega_K = 0.0015^{+0.008}_{-0.004}$. These error limits include uncertainties quoted in the absolute values of both x-ray and Auger-electron production cross sections and in the extrapolation to a single-vacancy fluorescent yield.

SUMMARY

The measurement of K-shell ionization cross sections for elements of low atomic number can readily be obtained from Auger-electron yields. The cross sections for K-shell ionization of carbon reported in this paper have estimated uncertainties of less than 20% whereas previous measurements which were based on x-ray production cross sections and rely on a knowledge of the fluorescent yield of carbon were uncertain by nearly an order of magnitude. By combining the measured Auger-electron yields with published x-ray production cross sections, the fluorescent yield for carbon was shown to vary with proton energy. This adds to the difficulty in interpretation of x-ray yield measurements for heavy-particle impact and emphasizes the need to define the fluorescent yield of an atom in terms of the initial and final states of the atomic or molecular system involved. Cross sections measured in the present work are in good agreement with calculations based on a classical binary-encounter theory[14] and are approximately 30% larger than values obtained using a Born approximation.[1]

ACKNOWLEDGMENTS

The author would like to thank W. E. Wilson and W. A. Glass for many helpful discussions throughout this work and J. E. Choate for his help in the operation of the Van de Graaff and associated instrumentation.

---

*Based on work performed under U. S. Atomic Energy Commission Contract No. AT(45-1)-1830.

[1]J. M. Khan, D. L. Potter, and R. D. Worley, Phys. Rev. 139, A1735 (1965).

[2]M. Terasawa, T. Inouye, and H. Kamei, J. Phys. Soc. Japan 29, 1394 (1970).

[3]W. Crone, Ann. Physik 27, 405 (1936).

[4]C. E. Dick and A. C. Lucas, Phys. Rev. A 2, 580 (1970).

[5]W. Hink and H. Paschke, Phys. Rev. A 4, 507 (1971).

[6]G. Glupe and W. Mehlhorn, Phys. Letters 25A, 274 (1967).

[7]L. H. Toburen, Phys. Rev. A 3, 216 (1971).

[8]M. Inokuti (private communication).

[9]M. E. Rudd, D. Gregoire, and J. B. Crooks, Phys. Rev. A 3, 1635 (1971).

[10]L. H. Toburen and W. E. Wilson, Phys. Rev. A 5, 274 (1972).

[11]L. H. Toburen, *Abstracts, Seventh International Conference on Physics of Electronic and Atomic Collisions* (North-Holland, Amsterdam, 1971), pp. 1120–22.

[12]W. E. Moddeman, Oak Ridge National Laboratory Report No. ORNL-TM-3012, 1970 (unpublished).

[13]A. K. Edwards and M. E. Rudd, Phys. Rev. 170, 140 (1968).

[14]J. D. Garcia, Phys. Rev. A 1, 280 (1970).

[15]F. P. Larkins, J. Phys. B 4, *L*29 (1970).

[16]G. Wentzel, Z. Physik 43, 524 (1927).

[17]R. W. Fink, R. C. Jopson, H. Mark, and C. D. Swift, Rev. Mod. Phys. 38, 513 (1966).

[18]E. J. McGuire, Phys. Rev. A 2, 273 (1970).