Ionization of Inert Gases by Positive Potassium Ions*

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Ionization of the inert gases by relatively low-energy potassium ions has been investigated. The use of relatively intense ion beams and phase-sensitive detection of ionization signals yields a large increase in sensitivity over that obtained in former investigations. The enhanced sensitivity permits detection of ionization at lower ion energies than heretofore reported. Evidence is presented for a real ionization threshold potential for K^+ ions in Kr.

Both the absolute ionization cross section and its derivative with respect to ion energy are reported. The latter shows a fine structure which is characteristic of the bombarded gas.

The electron energy distribution of the ionization electrons is determined from stopping potential curves and is found to be characteristic of the bombarded gas. In general, the energy distribution of the ionization electrons is independent of the incident ion energy.

The results of this investigation support the qualitative theory of ionization by ions proposed by Weizel.

I. INTRODUCTION

DIRECT evidence for ionization of gases by relatively low-energy positive ions was first obtained by Sutton[1] in 1929. During the following decade the ionization of inert gases by positive ions of the alkali metals was studied systematically by a number of investigators.[2–5] In two instances[2,5] the apparatus sensitivity was great enough to allow detection of ionization when the energy of the incident ions was less than 100 ev. In each of these two instances definite ionization onset potentials were reported. The published values of the ionization thresholds were in considerable disagreement, however, and even the existence of definite ionization thresholds for positive ions has since been questioned.[6]

The object of the present research is to explain the discrepancies in published threshold values and to investigate the reality of ionization thresholds for positive ions.

II. EXPERIMENTAL APPARATUS

The experimental arrangement ultimately employed in this investigation is seen schematically in Fig. 1. The experimental tube was evacuated by two mercury diffusion pumps. Pyrex glass construction permitted prolonged baking at 420°C, and a liquid nitrogen trap was used to remove Hg and other condensable vapors. The system was evacuated to at least 10^{-8} mm Hg before the experimental gas was introduced. A large McLeod gauge was used to measure the experimental gas pressure of about 10^{-2} mm Hg. The inert gases were purchased in Pyrex flasks. The impurity was almost entirely nitrogen which was present to about 2 parts in 10^4. Since nitrogen is less readily ionized by alkali ions than are the inert gases, this amount of nitrogen impurity was considered negligible.

The source of K^+ ions was a Kunsman[7] catalyst deposited upon a hollow nickel cylinder S. About one microampere of K^+ ions could be drawn from this source when it was heated with the internal heater. Ions from S in Fig. 1 were accelerated to G_1, traversed the equipotential ionization chamber, and were collected on P. The tube was operated in a uniform magnetic field of about 200 gauss along the axis of the tube. While this field had little effect upon the positive ions, it constrained the electrons to move in tight helices coaxial with the tube. Thus the magnetic field tended to guide ionization electrons originating below S_3 onto C, but secondary electrons originating on P were prevented from reaching C.

The elimination of extraneous charges produced by ion bombardment of metal surfaces has been perhaps the most difficult problem faced by the investigator of ionization by positive ions. It was necessary to make C 45 volts positive with respect to S_3 in order to prevent multiply reflected positive ions from reaching C. While the magnetic field proved to be effective in eliminating most secondary electrons, it was found desirable to operate with a 6-volt electron stopping potential between S_3 and S_2. Under these conditions the secondary electron current was reduced and held to less than one electron per 10^8 alkali ions, a figure which actually exceeds the over-all sensitivity of several of the earlier experiments. Of the secondary electrons that still persisted in reaching the collector, less than one percent had energies above 12 volts, as determined by stopping potential measurements. Those having such high energies could potentially ionize Xe and lead to spurious currents; since only one secondary for each 10^{10} primary ions could reach the region between S_3 and S_2 with this energy under the existing

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[1] R. M. Sutton, Phys. Rev. **33**, 364 (1929).
[2] O. Beeck and J. C. Mouzon, Ann. Physik **11**, 737, 858 (1931).
[3] Carl Frische, Phys. Rev. **43**, 160 (1933).
[4] M. Nordmeyer, Ann. Physik **16**, 706 (1933).
[5] R. N. Varney, Phys. Rev. **46**, 235 (1934); **47**, 483 (1935).
[6] H. Massey and E. Burhop, *Electronic and Ionic Impact Phenomena* (Oxford University Press, New York, 1952), p. 530.
[7] C. H. Kunsman, Science **62**, 269 (1925).

experimental conditions, the potential error is negligible by contrast with the far larger effects observed.

The tube and its associated electronic apparatus were designed to give either the ionization cross section curve or its derivative with respect to ion energy, as desired. The ionization cross section curves were obtained by sending the ionization current through a resistor of 10^{12} ohms. The resulting signal, after dc amplification, was displayed on a cathode ray oscilloscope and photographed. A voltage increasing linearly with time applied to the ion source provided the accelerating potential.

The derivative of the ionization curve was obtained by superimposing a small 30-cps signal on the ion accelerating potential. Landale[8] has shown that the corresponding ac component of the ionization current is proportional to the derivative of the ionization curve. The source and its adjacent grid were both pulsed so that the beam current was independent of the phase of the modulating voltage. After ac amplification, the ionization signal was fed into a phase-sensitive detector of the type developed by Schuster.[9] A long output time constant was obtained by feeding the output of the detector to a pair of cathode followers through appropriate $R-C$ elements. The output from the cathode followers was fed into an oscilloscope for photographic recording of data.

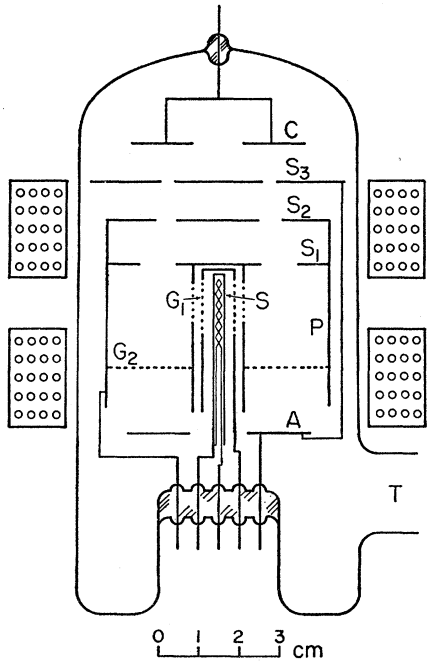

FIG. 1. The positive ion tube. Ions emitted by the Kunsman source S are accelerated through grid G_1, traverse the equipotential ionization chamber, and are collected on P. The slit system S_1, S_2, and S_3 prevents reflected ions and secondary electrons originating at P from reaching the ionization collector C. The Helmholtz coils shown in cross section produce an axial magnetic field which constrains electrons to move in tight spirals along the tube axis. The tube is usually operated with a 45-volt ion stopping potential between C and S_3, and a 6-volt electron stopping potential between S_3 and S_2.

[8] S. E. A. Landale, Proc. Cambridge Phil. Soc. **25**, 355 (1929).
[9] N. Schuster, Rev. Sci. Instr. **22**, 254 (1951).

The ac mode of detection is well adapted to an investigation of onset potentials. Any discontinuity in the ionization curve at onset can be readily detected in the derivative curve, while the minute "vacuum" current, which increases nearly linearly with ion energy, gives a practically constant and therefore easily subtractable signal.

The present apparatus was designed primarily to detect minute ionization signals rather than to measure accurate ionization cross sections. It became apparent during the process of taking readings that some sort of measure of the sensitivity of the equipment was essential for numerous reasons. Thus in particular, a failure to detect ionization would necessitate assurance that the equipment was potentially capable of the detection if it exceeded some minimum figure. Such a calibration of instrumental sensitivity may be expressed as a collision cross section provided that the characteristics of the apparatus are reasonably well known. It would be unrealistic to claim that ionization cross sections have been determined in the present work more precisely than by a factor of two, although the results are nonetheless interesting and significant because they indicate improvements in sensitivity of several orders of magnitude over previously reported works.

The determination of cross sections for ionization requires observation of the electron current yield i resulting from a known primary ion current I, the path length L of the ion beam in the sensitive ionization region, and the number of atoms of gas per cm,[3] N. Then the cross section of the atoms for ionizing collisions σ is

$$\sigma=i/(ILN).$$

Since the atomic density and the gas pressure are proportional, the cross section is often written

$$\sigma=i/(IL\rho N_0),$$

where N_0 is the number of atoms/cm^3 at 1 mm Hg pressure, a universal constant, and ρ is the pressure in mm Hg. From the experimental point of view, a critical problem is whether the electron current i truly represents the current of all the electrons created in the path length L and no more. Lastly, by providing a large primary ion current I and a sensitive detector for the electron current i, very small values of σ may be noted.

The complex slit system and magnetic field arrangement, designed primarily to remove false or secondary electrons from the picture, also served to make the cross-section measurements more reliable. Thus the ejected ionization electrons, regardless of initial velocity, was constrained to a tight vertical spiral by the magnetic field so that the slit width S_3 was taken to be equal to

the ionizing path length L, the action of the magnetic field to hold false electrons out of this range aiding to hold true electrons into it. It would appear still that only half of the electrons would reach S_3, as the half with initial downward components of velocity would spirial away from S_3. A factor of two was accordingly introduced in the calculations of σ.

All the upwardly spiraling electrons did not necessarily pass through S_3 because of the 6-volt retarding potential on S_2. Those which failed could be divided into two classes: (a) those with less than 6 ev of kinetic energy, and (b) those with velocity in the axial direction amounting to less than 6 ev of kinetic energy for this degree of freedom. The electrons of type (a) were not recorded, but at least a reasonable correction was made from a study of the electron energy spectrum. The electrons of type (b) going either upward or downward were reflected by the retarding potential into the opposite direction and were essentially in a trap. These electrons would continue to oscillate up and down unless an elastic gas collision redirected them so that they could now penetrate to S_3 at the top or A at the bottom respectively. Since all of the present experiments have been conducted in rare gases, advantage has been taken of the low elastic energy loss per collision and the complete absence of inelastic collisions except at the highest energies encountered in this work. The grid wires in front of electrode A presented a cross section of only 0.4 percent of the total area so presumably could only account for minor errors. Thus ultimately the meter reading i could be regarded as giving very nearly one-half of the total electron current.

The qualitative correctness of the discussion above was tested by reconnecting the tube to stop the action of the electron trap at the bottom. The observed current dropped by a variable factor depending on the stopping potential but averaging nearly ten.

The current i has been set in the ensuing determinations of cross section as the collector current multiplied by two and divided by a factor (one minus fraction of ionization electrons rejected by the 6-volt potential barrier). Despite the various crudities, the suggested possible error of a factor of two in the cross section for ionization seems adequate.

III. EXPERIMENTAL RESULTS

Evidence of Onset Potentials

Figure 2 presents evidence for ionization of Kr by relatively low-energy K^+ ions.

Figure 2A was taken with dc detection and is a plot of the ionization cross section against ion energy. Ionization was first observed at ion energies of about 66 ev. The corresponding ionization onset potentials reported by Beeck[2] and Varney[5] were 80 ev and 69 ev, respectively. In Fig. 2A an ionization cross section of 1.3×10^{-21} cm^2 was taken as the minimum definitely detectable ionization signal. This represents an increase in sensitivity of about 2000 over the apparatus used by Beeck and probably explains the difference in observed onset potentials. Varney's apparatus did not lend itself to absolute measurements, although in the light of the present investigation it seems likely that his apparatus was considerably more sensitive than Beeck's.

The sequence of curves B, C, and D shows the corresponding situation when the phase-sensitive detector was employed with increasing sensitivity of detection. A seventy-fold increase in sensitivity from B to C pushed the observed threshold down from 66 ev to about 63 ev. Figure 2D represents a twenty-fold further increase in sensitivity. The slope of this curve was limited by the long output time constant necessary at high sensitivity. It is noteworthy that the additional increase in sensitivity did not appreciably decrease the apparent ionization threshold.

Since the energy of the ion beam was modulated with a 2-volt square wave in obtaining Fig. 2D, it is reasonable to assume that an appreciable ionization current flowed only during the positive phase of the square wave. Thus the ionization current which flows during the positive phase of the square wave is the observed rms ionization current multiplied by $\sqrt{2}$. An ionization current of 1.0×10^{-15} ampere rms produces $\frac{1}{6}$ of a full scale deflection in Fig. 2D. This corresponds to 1.4×10^{-15} ampere during the positive phase of the square wave which is 1.4×10^{-9} of the primary ion current. The corresponding ionization cross section is 2.0×10^{-23} cm^2. If this cross section is taken as the smallest detectable ionization signal, the present apparatus is 1.2×10^5 times more sensitive than was the apparatus used by Beeck. This figure is either the cross section 2 volts above onset or the increase in cross section for 2 ev increase in accelerating potential. The ionization observed in Fig. 2D cannot be attributed to "above-onset" ionization among dilute contaminants since all investigators have agreed in assigning the lowest apparent onset potential to the ion-atom pair, K^+ in Kr.

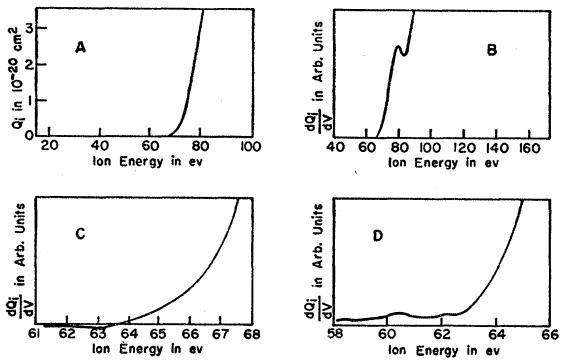

FIG. 2. Ionization curves for K^+ in Kr with increasing apparatus sensitivity. Detection was dc for curve A, and ac for curves B, C, and D. The over-all sensitivities used for curves C and D were respectively 70 and 1400 times greater than those used for curve B.

The fact that an apparent threshold has been reached which is independent of increasing apparatus sensitivity is evidence for a real ionization threshold at 63±0.3 ev. Since ionization by K^+ ions in Kr is energetically possible at all ion energies above 20.5 ev, it cannot be claimed that absolutely no ionization occurs below 63 ev. The abrupt rise in ionization cross section observed at about 63 ev does indicate the onset of a distinctly different and remarkably more efficient mode of ionization. The existence of such an onset potential is predicted by the Weizel-Beeck[10] theory of ionization by ions.

Evidence for a real threshold was less definite for the other gases studied. No abrupt rise in ionization current over vacuum current was observed, and each increase in apparatus sensitivity was accompanied by detection of ionization at lower ion energy. The lowest ion energies at which ionization by K^+ ions was observed in Ne, Ar, and Xe were respectively 115 ev, 67 ev, and 89 ev.

Ionization Cross Section Curves

Figure 3 presents the observed ionization cross section as a function of K^+ ion energy for each of the gases investigated. The observed results for K^+ ions in A and Kr are in good agreement with those reported by Beeck. This agreement is in fact somewhat surprising in view of the large uncertainties inherent in the present method of determining cross sections.

The cross section observed for K^+ ions in Xe is, however, an order of magnitude lower than that reported by Beeck. It is interesting to note that the author's result would probably remove the one exception to the rule discovered by Beeck which states that of two ions, the one more remote from a given gas in the periodic table will have the lower ionization cross section for that gas. The exception Beeck found was that K^+ ions ionized Xe more efficiently than did Rb^+.

Ionization of Ne by K^+ ions remains in doubtful status. Beeck reported ionization only at ion energies

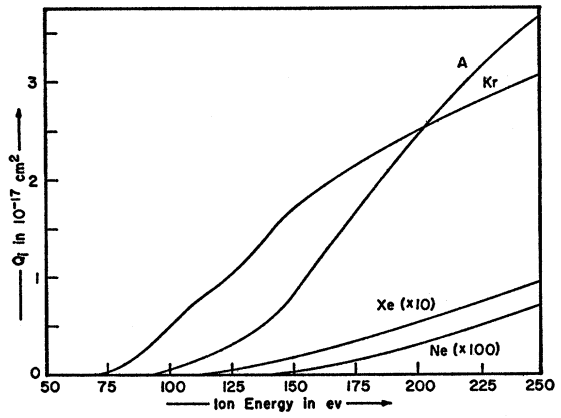

FIG. 3. Observed ionization cross section as a function of ion energy.

[10] W. Weizel and O. Beeck, Z. Physik **76**, 250 (1932).

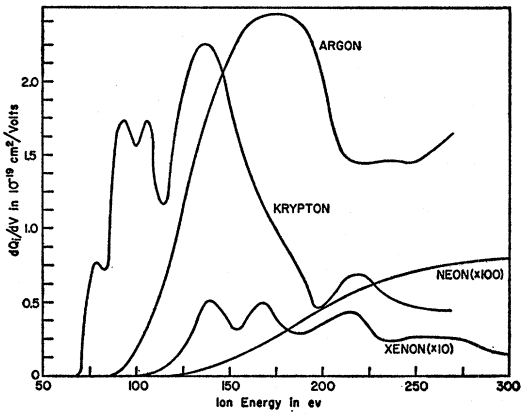

FIG. 4. Observed derivative of the ionization cross section as a function of ion energy.

above 320 ev. The author observed feeble ionization above 115 ev, but at ion energies of 350 ev, his observed ionization cross section was an order of magnitude lower than that reported by Beeck at corresponding ion energies. The electron current observed for K^+ ions in Ne is indeed so small that an indirect effect is not ruled out. Ionization by a Na^+ contaminant or by recoil neutrals were considered but ruled out on the basis of observations of the dependence of ionization current on beam current and gas pressure. The possibility remains that the observed electrons were photoelectrons caused by radiations from excited Ne. Such an origin is suggested by the fact that Varney, whose apparatus was evidently more sensitive than Beeck's but insensitive to photoelectrons, was unable to detect ionization of Ne by K^+ ions.

Derivative of Ionization Cross-Section Curves

The experimental results obtained by using a pulsed ion beam and the phase-sensitive detector at ion energies from 0 ev to 300 ev are presented in Fig. 4. As mentioned above the observed signals were proportional to the derivative of the corresponding ionization curve. The existence of a fine structure in the differentiated curves of Kr and Xe was an unexpected result. The form of the fine structure in general depended only upon the experimental gas; however, for Kr the various peaks were associated with different distributions of kinetic energy of ionization electrons (see next paragraph) as shown by the fact that the peak at 93 ev could be selectively removed by increasing the electron stopping potential of S_3 to 11 volts as compared with 13.5 volts necessary to stop all of the ionization electrons. Ne is unique in the continued rise of the derivative throughout the entire range of ion energies used.

Energy Distribution of the Ionization Electrons

An indication of the distribution of kinetic energy of the ionization electrons was determined by observing

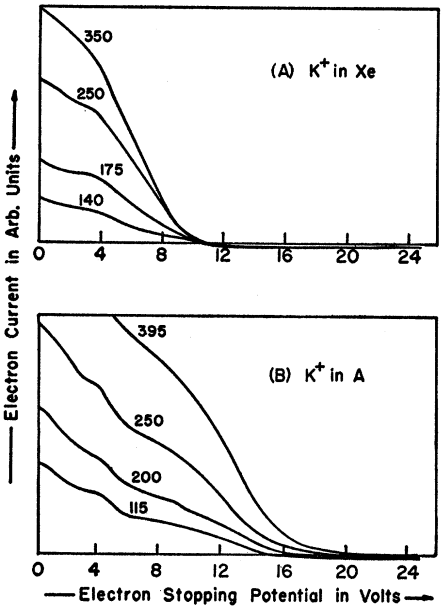

FIG. 5. Electron current reaching the electron collector as a function of the electron stopping potential between electrodes S_3 and S_2 of Fig. 1. Each curve refers to a constant energy of the bombarding ion.

the number of electrons able to overcome given stopping potentials between S_3 and S_2 of Fig. 1. Figure 5(A) shows the number of electrons reaching the collector as a function of stopping potential, with Xe in the tube. Each curve represents a different energy of the bombarding ions, but only ion energies well above the observed threshold potential are used. Thus in all cases the ratio of ionization current to secondary electron current is greater than 10 at a 1-volt electron stopping potential. Little significance can be attached to the curves below a one-volt stopping potential. The maximum electron energy given by Fig. 5(A) was about 12 ev, and it is noteworthy that the value of this maximum energy did not increase with the kinetic energy of the incident ion. While the efficiency of ionization increased rapidly with increasing ion energy, the energy distribution of the emitted electrons remained essentially the same. This observation, in agreement with the Weizel-Beeck theory, implies that the ionization process is not one of direct transfer of kinetic energy from the incident ion to the emitted electron.

Corresponding stopping potential curves for argon are shown in Fig. 5(B). The maximum electron energy in this case was about 20 ev. Similar curves for Kr and Ne gave maximum electron energies of 13.5 ev and 20 ev, respectively.

Rouse,[11] in connection with ion scattering experiments, obtained evidence for ionization electrons of relatively high energy, but in a subsequent investigation he was unable to detect electrons with more than one or two electron volts energy. The evidence for higher energy electrons obtained in the present investigation can perhaps be explained in terms of more efficient elimination of low-energy secondary electrons together with greater apparatus sensitivity to detect the relatively less abundant ionization electrons.

The electron stopping potential curves were differentiated by a point to point calculation of slopes, and the results are plotted in Fig. 6. These curves give the energy distribution of the ionization electrons to the extent that the following argument is valid:

An electron of energy V ev can penetrate a potential barrier between S_2 and S_3 of Fig. 1 up to V volts only if its velocity is directed along the tube axis. An electron of energy V ev for which this condition is not fullfilled will spiral back through the ionization chamber, but will have its axial component of velocity reversed again by the stopping potential between G_2 and A. The electron thus moves in an "electron trap." Each elastic atomic collision will change the axial velocity component, but the energy of the electron will remain essentially constant. Therefore the electron eventually should travel with its velocity directed along the tube axis and thus cross any potential barrier up to V volts.

The shape of the spectra presented in Fig. 6 should be considered as tentative since there are several effects which conceivably could alter the above argument. For example, collisions of trapped electrons with wires of grid G_2 have been ignored. More serious perhaps are inelastic collisions between inert gas atoms and electrons of energy higher than the ionization potential of the inert gas. Each such collision would remove one electron from the high-energy part of the spectrum and yield at least one low-energy electron. The resulting distortion of the spectrum could be considerable if an appreciable fraction of the electrons emitted during an ionizing

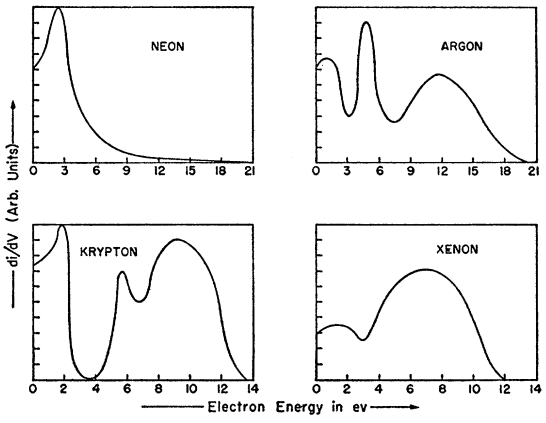

FIG. 6. Derivatives of the electron stopping potential curves for gases bombarded with 250-ev K^+ ions.

[11] A. G. Rouse, Phys. Rev. **52**, 1238 (1937).

event were of high energy. A subsequent investigation employing apparatus primarily designed to measure electron velocity spectra is contemplated. Such spectra are of interest because according to the Weizel-Beeck theory of ionization by ions, a quasi molecule is formed during an ionizing collision, and it seems likely that the fine structure indicated in the spectra of Fig. 6 is related to the energy levels of the quasi molecule.

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Prompt Gamma Rays Accompanying the Spontaneous Fission of Cf^{252}†

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Single- and multiple-crystal scintillation techniques are utilized to measure the energy spectrum of prompt gamma rays emitted in the spontaneous fission of Cf^{252}. It is found that each fission yields an average of 10.3 photons having a total energy of 8.2 Mev. These photons are heterogeneous in energy. An upper limit is placed on the intensity of neutrons and quanta emitted in the order of 10^{-9} second after fission.

INTRODUCTION

A KNOWLEDGE of the prompt gamma rays accompanying fission is fundamental to a complete understanding of the fission process. Furthermore, since prompt emission constitutes a significant portion of the radiation emanating from a critical assembly, it is necessary that the number and energy of the quanta be known in order to design the optimum shielding for such facilities.

Early studies[1,2] of the neutron-induced fission of U^{235} indicated that two prompt quanta were emitted with a total energy of 4—5 Mev. Several recent studies at the Oak Ridge National Laboratory[3,4] show that within 0.3 microsecond after fission more than 7 Mev of energy are emitted as photons. However, the spectral distributions and absolute normalizations of these measurements are not in complete agreement. These discrepancies may well be attributable to the high background associated with the neutron atmosphere required for the experiments.

Recently, intense Cf^{252} spontaneous fission sources have become available. The spontaneous fission of Cf^{252} has been shown theoretically and experimentally[5–7] to be similar to the neutron-induced fission of U^{235}. This offers an opportunity to study the fission phenomenon without complicating backgrounds. By utilizing these clean experimental conditions, the prompt photons and neutrons emitted in the fission of Cf^{252} were measured. The results may be extrapolated to apply to the general fission act.

EXPERIMENTAL TECHNIQUES

All of the measurements carried out in this experiment are of a coincidence type requiring simultaneous response by the fission and neutron-gamma detectors. To obtain the maximum time resolution and minimum background, it is fundamental that the detection system be as fast as possible. The recently developed gas scintillator serves as an excellent fast-fission detector,[8] combining good energy resolution with high speed and an insensitivity to gamma radiation.

Because the prompt gamma-ray spectrum is expected to be of a heterogeneous nature, it is desirable that the technique employed respond to an incident photon in a unique manner. Two methods are utilized. In the first the pulse-height distribution from a single $1\times1\frac{1}{2}$ inch NaI(Tl) crystal is measured in coincidence ($\tau=0.3$ microsecond) with the pulses from the fission dectector. The response of this system to gamma rays is not unique but it is the most sensitive method available. In the second approach, a double-crystal Compton spectrometer is used[9–11] in coincidence ($\tau\geq2.5$ millimicroseconds) with the fission detector. This system does give a unique response to the incident quanta. In most applications of the Compton effect to

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