

a sufficient cause is to be found in the symmetry properties of the ground-state wave function. The permutation symmetry corresponding to a pairing of spins is not enough by itself, however, and has to be extended to the more thorough-going type of pairing described by Racah's notion of lowest seniority.

The results presented in Table II are based on  $jj$  coupling, but  $LS$  coupling appears to give an equally good account of the present experimental data. In fact, the larger odd-even effect predicted by  $LS$  coupling has the advantage of producing better accord between the assumption of charge independence and the observed energy of the  $0+$  state in  $N^{14}$ . Further measurements are needed to establish whether a discontinuity in Coulomb energy and radius occurs after  $Z=14$ .

The oscillator model gives appreciably smaller values for the rms radius of the nuclear charge distribution than does the uniform density model in which  $E_c = (3/5)Z(Z-1)e^2/R_{eq}$  and  $\Delta_1(Z) = (6/5)(Z-1)e^2/R_{eq}$ . Part of the difference is due to the absence of exchange terms<sup>20</sup> in the latter case, but it is not clear that ex-

<sup>20</sup> The separation between direct and exchange terms is sometimes defined in a different way. When the wave function is taken to be a single determinant of one-particle functions, it is often formally convenient to add a sum of "self-energy" integrals,  $\frac{1}{2}\sum_i \int \int |\psi_i(\mathbf{r}_1)\psi_i(\mathbf{r}_2)|^2 (e^2/r_{12}) d\mathbf{r}_1 d\mathbf{r}_2$ , to the direct terms and

change effects are sufficient to account for the whole difference. For instance, if the Slater exchange integrals are completely omitted in calculating  $\Delta_1$  for  $A=17$ , the result obtained is  $10.967e^2(\nu/\pi)^{\frac{1}{2}}$ , only 6 percent larger than the value with exchange. The corresponding  $r_0$  for  $O^{17}$  would be 1.34 without exchange, compared with 1.52 from the uniform density model. This remaining discrepancy is caused by the change in shape of the shell-model charge distribution when an outer proton turns into a neutron.

The values of  $r_0$  listed in Table II show a steady decrease from  $C^{13}$  to  $Al^{27}$ . It is to be hoped that further measurements of Coulomb energies will soon determine whether this decrease continues without interruption in the heavier mirror nuclei, and that electron scattering experiments will provide an independent set of results for comparison.

It is a pleasure to express our gratitude to Professor E. P. Wigner for valuable discussions.

subtract the same sum in the form of exchange contributions. We do not include such integrals in the definitions of "direct" and "exchange"; it seems superfluous to attribute direct and exchange Coulomb energies of equal magnitude and opposite sign to a system containing only one charged particle, and undesirable to attribute exchange energy to an unsymmetrized many-particle wave function with no positional correlations.

## Radiations from $Ir^{192m}\dagger$

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The photon and electron spectra of  $Ir^{192m}$  (1.42 min) have been thoroughly investigated. Study of the photon spectrum with scintillation spectrometers and an aluminum-lined proportional counter spectrometer revealed the presence of a quantum continuum, iridium  $L$  x-rays, and a 58-keV gamma ray. The quantum continuum was shown to be bremsstrahlung engendered by conversion electrons from the isomeric transition. Previous to this study, the quantum continuum had been interpreted as possibly caused by "two-quantum" emission. A high-resolution,  $180^\circ$  permanent magnet spectrograph was employed for the examination of the conversion electron spectrum. The energy of the isomeric transition was found to be  $58.0 \pm 0.4$  keV from the conversion electron studies. Lifetime-energy considerations and the observed  $L$ -subshell conversion ratios show that the 58.0-keV gamma ray is  $E3$ . The  $L$ -shell conversion coefficient was determined from scintillation spectrometer data to be  $\geq 870$ .

### I. INTRODUCTION

PREVIOUS investigations<sup>1,2</sup> have shown that the decay of  $Ir^{192m}$  (1.42 min) is accompanied by iridium  $L$  x-rays,  $L$ -shell conversion electrons from an  $\sim 60$ -keV gamma ray, and a quantum continuum with peak intensity at  $\sim 30$  keV and upper energy end point at  $\sim 60$  keV. Conversion electron analyses have been

made by Hole,<sup>3</sup> Caldwell,<sup>4</sup> and Weber<sup>5</sup> yielding energy values for the isomeric transition of 55.5, 57.4, and 56.0 keV, respectively. The reported quantum continuum has been interpreted as possibly caused by "two-quantum" transitions.<sup>1,2</sup> Sachs<sup>6</sup> has suggested that nuclear decay through transitions of the type  $0 \rightarrow 0$  (yes) can take place by simultaneous emission of two quanta, one quantum and one electron or two elec-

<sup>†</sup> Work performed under the auspices of the U. S. Atomic Energy Commission.

<sup>1</sup> Goldhaber, Muehlhause, and Turkel, *Phys. Rev.* **71**, 372 (1947).

<sup>2</sup> E. der Mateosian and M. Goldhaber, *Phys. Rev.* **82**, 115 (1951).

<sup>3</sup> N. Hole, *Arkiv. Mat. Astron. Fysik* **A36**, No. 9 (1948).

<sup>4</sup> R. L. Caldwell, *Phys. Rev.* **78**, 407 (1950).

<sup>5</sup> C. Weber and A. Flammersfeld, *Z. Naturforsch.* **A8**, 580 (1953); C. Weber, *Z. Naturforsch.* **A9**, 115 (1954).

<sup>6</sup> R. G. Sachs, *Phys. Rev.* **57**, 194 (1940).

trons. Transitions of these types would be characterized by the quantum or electron spectrum having a continuous energy distribution. Goldberger<sup>7</sup> has discussed the reported quantum continuum of  $\text{Ir}^{192m}$  in the light of simultaneous emission of one quantum and one electron according to the mechanism postulated by Sachs.

Since the  $\text{Ir}^{192m}$  quantum continuum seemed to be experimentally unique, it appeared worth while to investigate the continuum in further detail to gain additional insight as to the characteristics of its radiations. In view of the fact that the isomeric transition is believed to be either  $E3$  or  $M3$ ,<sup>8</sup> it also seemed desirable to undertake a new study of the  $L$ -subshell conversion electrons in order to arrive at a unique multipolarity assignment.

## II. EXPERIMENTAL METHODS AND RESULTS

### a. Photon Measurements

The isomeric state of  $\text{Ir}^{192}$  was prepared by irradiating  $\text{IrO}_2$  (of natural isotopic abundance) with thermal neutrons. The  $\text{Ir}^{192m}$  photon spectrum was initially studied with a commercially packaged 2-in.  $\times$  2-in. right circular cylinder of  $\text{NaI(Tl)}$  mounted on a Dumont 6292 photomultiplier tube. A ten-channel differential pulse-height analyzer was utilized for the recording of data. The photon spectrum (Fig. 1) measured with the above apparatus indicates the presence of a 58-keV gamma ray and a broad, low-energy photon distribution (A) which, from resolution considerations, cannot be attributed to a single monoenergetic gamma ray. A study of the 15–40 keV region with a proportional counter spectrometer revealed no additional fine structure. It was thus concluded that the pulse distribution A probably corresponds to the quantum continuum previously reported. The proportional counter data also indicated that the decay of  $\text{Ir}^{192m}$  is accompanied by intense iridium  $L$  x-rays. Since virtually no iridium  $L$  x-rays can be seen in the scintillation spectrum of Fig. 1, it was concluded that the commercially packaged  $\text{NaI(Tl)}$  detector possessed a relatively low quantum detection efficiency at low energies, caused in part by its ambient  $\text{Al}_2\text{O}_3$  reflector.

To facilitate further study of the quantum continuum and iridium  $L$  x-rays, a scintillation spectrometer was designed which would be expected to have essentially constant detection sensitivity in the energy region 10 to 60 keV. The experimental arrangement of the spectrometer is shown in Fig. 2. Since the hygroscopic  $\text{NaI(Tl)}$  detector of Fig. 2 was exposed to the atmosphere throughout the experiment, a continual check was made of its resolution and detection sensitivity. It was established that after the sodium iodide crystal had been polished with alcohol, the resolution

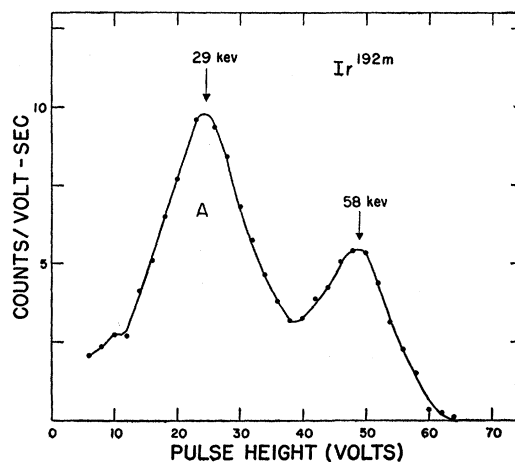


Fig. 1. Photon spectrum of  $\text{Ir}^{192m}$ , obtained with a commercially packaged  $\text{NaI(Tl)}$  crystal mounted on a Dumont type 6292 photomultiplier tube.

(~21 percent) and detection efficiency of the spectrometer for the 59.8-keV gamma ray of  $\text{Am}^{241}$  remained constant during each photon spectrum measurement.

Results of the  $\text{Ir}^{192m}$  photon spectrum measurements with the scintillation apparatus of Fig. 2 are presented in Fig. 3. The intense iridium  $L$  x-rays present in the  $\text{Ir}^{192m}$  decay necessitated the placement of a 0.020-inch thick aluminum absorber between source and detector for the studies shown in Fig. 3. The conversion electrons were stopped by either the 0.020-inch thick aluminum foil or by a 5.0-mg/cm<sup>2</sup> polyethylene film. The iridium sources were prepared from  $\text{IrO}_2$  powder and were mounted on the aluminum foil or the polyethylene film. The surface densities of the  $\text{IrO}_2$  sources were estimated from microscopic measurements of aggregate grain size. By this method an average surface density of ~70 mg/cm<sup>2</sup> was obtained for the thick sources, and an average surface density of ~0.7 mg/cm<sup>2</sup> was obtained for the thin sources. Since the  $\text{IrO}_2$  was of questionable chemical purity, the decay of the 58-keV gamma ray was followed through seven half-lives. During that period no deviation was observed from the 1.42 minute half-life assigned to the isomeric transition. Since the quantum continuum and iridium  $L$  x-rays also decayed with a 1.42 minute half-life, the

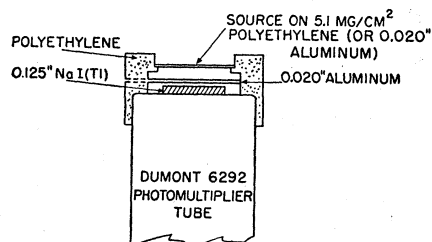


Fig. 2. Schematic diagram of the scintillation counter arrangement.

<sup>7</sup> M. L. Goldberger, Phys. Rev. **73**, 1119 (1948).

<sup>8</sup> M. Goldhaber and A. W. Sunyar, Phys. Rev. **83**, 906 (1951).

photon spectrum shown in Fig. 3 is unambiguously engendered by the decay of  $\text{Ir}^{192m}$ .

The amplitude and shape of the quantum continuum (Fig. 3) were measured as a function of source surface density and the  $Z$  of the conversion electron stopping material. The data have been normalized to the 58-keV photopeak. The quantum continuum is observed to have its greatest intensity when the conversion electrons are stopped in the thick iridium source and aluminum absorber; its weakest intensity is observed when the conversion electrons are stopped in the relatively low  $Z$  polyethylene material (with few conversion electrons stopping in the thin source). If the quantum continuum were of nuclear origin, its relative intensity as a function of source thickness would not behave in the above manner. Instead the intensity of the quantum continuum would exhibit a variation similar to that of the iridium  $L$  x-ray shown in Fig. 3, i.e., the intensity of the continuum (relative to that of the 58-keV gamma ray) would be a minimum for a thick source and a maximum for a thin source. Furthermore, if the continuum were of nuclear origin its magnitude and shape would not be dependent on the  $Z$  of the material in which the conversion electrons are stopped. It can be observed from Fig. 3 that the intensity of the continuum increases with the  $Z$  of the material in which the conversion electrons are stopped.

When the photon spectrum *A* of Fig. 3 is corrected for absorption in the aluminum foil, the peaked distribution of the quantum continuum at  $\sim 30$  keV is removed and the quantum continuum assumes the hyperbolic shape shown in Fig. 4.

Thus the above experimental evidence and evidence to be presented below strongly suggest that the quantum

continuum is not of nuclear origin<sup>9</sup> but is in all probability bremsstrahlung engendered by conversion electrons from the isomeric transition

### b. Coincidence Experiment

To offer further proof that the quantum continuum of  $\text{Ir}^{192m}$  is indeed a bremsstrahlung distribution, a coincidence experiment was performed. Two scintillation spectrometers of the type shown in Fig. 2 were used in the coincidence measurements. Quanta in the energy region 17–57 keV were "gated" by quanta in the same energy region. The singles counting rate in each channel was recorded, and a coincidence rate of only 1 percent of the expected rate for two-quantum

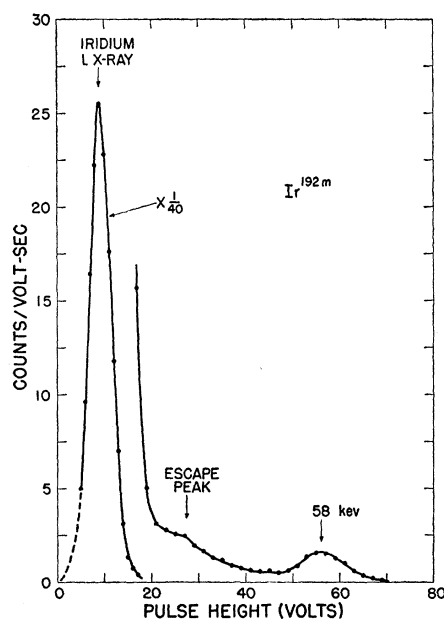


FIG. 4. Photon spectrum *A* of Fig. 3 corrected for absorption in the 0.020-inch thick aluminum absorber.

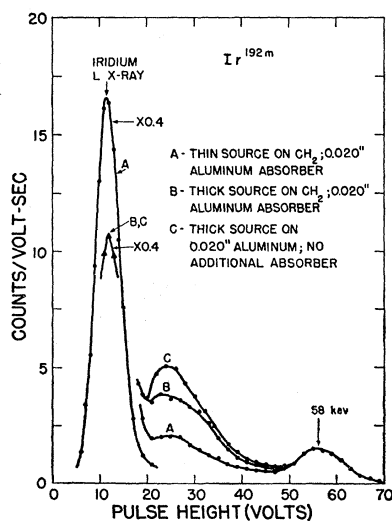


FIG. 3. Photon spectra of  $\text{Ir}^{192m}$ , obtained with the scintillation apparatus shown in Fig. 2.

emission in the above energy region was observed. The observed coincidences could be attributed to Compton scattered events, bremsstrahlung-x-ray coincidences, and chance coincidences.

### c. Internal Conversion Electron Measurements

A 77 gauss, 180° permanent magnet spectrograph of high resolution was employed for the analysis of

<sup>9</sup> Independent research carried out by Weber (reference 5) on  $\text{Ir}^{192m}$  came to our attention after we had reached the above conclusion concerning the nature of the quantum continuum. Weber utilized absorption techniques and found that under the assumption that the entire number of quantum transitions with  $E > 30$  keV are from the gamma-ray continuum:

$$N_{\text{cont.}}/N_{\text{I.T.}} \leq 10^{-3}.$$

Under the assumption that the entire number of quantum transitions with  $E > 30$  keV are unconverted 56.0-keV quanta, the resulting value for the total internal conversion coefficient of the gamma ray has a lower limit of 1000. From these intensity relationships, Weber concluded that he had found no evidence for  $\text{Ir}^{192m}$  decay by "two-quantum" emission.

conversion electrons accompanying the  $\text{Ir}^{192m}$  decay. The spectrograph was calibrated with  $K$  conversion electrons from the  $80.133 \pm 0.005$ -keV transition in  $\text{Xe}^{131}$ <sup>10</sup> and the  $L$  and  $M$  conversion electrons from the  $59.78 \pm 0.02$ -keV transition in  $\text{Np}^{237}$ .<sup>11</sup> The spectra were recorded on Eastman No-Screen X-Ray film. The spectrograph camera was constructed so that radioactive sources could be inserted through a vacuum gate without appreciable loss of the vacuum inside the chamber. At the conclusion of each iridium activation, approximately one minute was required to mount the source and insert it into the spectrograph. A suitable spectrogram of the internal conversion lines was obtained by performing seven successive bombardments and exposures.

Caldwell has observed two  $L$ -subshell conversion electron lines from  $\text{Ir}^{192m}$  by utilizing a high-resolution permanent magnet spectrograph.<sup>4</sup> These lines were interpreted to be  $L_I$  and  $L_{III}$  conversion electrons from a 57.4-keV transition. However, it has been pointed out by Mihelich<sup>12</sup> that the  $L_I$  line in Caldwell's data falls midway between the expected positions for the  $L_I$  and  $L_{II}$  lines on the spectrogram. The 57.4-keV transition has been classified as either  $E3$  or  $M3$  from lifetime-energy considerations.<sup>8</sup> Our measured values of the energies and intensities of the  $L$ -subshell conversion electrons (summarized in Table I) indicate that conversion takes place principally in the  $L_{II}$  and  $L_{III}$  subshells. By using the empirical  $L$ -subshell conversion ratios determined by Mihelich<sup>12</sup> and the recent calculations of  $L$ -subshell conversion coefficients by Rose *et al.*,<sup>13</sup> the radiation is unquestionably electric and must therefore be  $E3$ . An energy value of  $58.0 \pm 0.4$  keV was obtained for the isomeric transition from the data of Table I.

#### d. The Conversion Coefficient

The  $L$ -shell conversion coefficient for the  $\text{Ir}^{192m}$  transition was calculated from the expression

$$\alpha_L = \frac{A_1 \left[ \frac{F+G}{FD+GE} \right]}{A_2}, \quad (1)$$

where  $A_1$  is the area of the  $L$  x-ray photopeak of Fig. 4,  $A_2$  is the area of the gamma-ray photopeak occurring at  $\sim 57$  volts in Fig. 4 (corrected for the 13-percent loss into the escape peak),  $D$  and  $E$  are the fluorescent yields

<sup>10</sup> Lind, Brown, Klein, Muller, and DuMond, Phys. Rev. **75**, 1544 (1949).

<sup>11</sup> C. I. Browne and I. Perlman, Phys. Rev. **85**, 758 (1952).

<sup>12</sup> J. W. Mihelich, Phys. Rev. **87**, 646 (1952).

<sup>13</sup> Privately circulated tables of  $L$ -shell conversion coefficients by Dr. M. E. Rose *et al.* We wish to thank Dr. Rose for making these values available before publication.

TABLE I. Energies and relative intensities of  $L$ -subshell conversion electron lines of  $\text{Ir}^{192m}$ .

Electron shell	Relative intensity	Electron energy (keV)	Binding energy (keV)	Transition energy (keV)
$L_I$	not observed		13.4	
$L_{II}$	1.1	45.2	12.8	58.0
$L_{III}$	1.0	46.8	11.2	58.0

of the iridium  $L_{II}$  and  $L_{III}$  shells, respectively,<sup>14</sup> and  $F$  and  $G$  are the relative intensities of the  $L_{II}$  and  $L_{III}$  conversion electrons, respectively (Table I). A value of  $\alpha_L \geq 870$  was obtained from Eq. (1).

It must be realized that the above measurement of  $\alpha_L$  is fraught with serious uncertainties which arise from the questionable amount of  $L$  x-ray absorption in the thin iridium source and also in the assumption that the sodium iodide detector of Fig. 2 has a constant photon-detection efficiency from 10 to 60 keV. Therefore, the experimental value of  $\alpha_L$  is only indicative of its proper order of magnitude. When the theoretical  $L$ -subshell conversion coefficient calculations of Rose *et al.* are completed, a more exact value of  $\alpha_L$  can be determined and the above value checked.

### III. CONCLUSIONS

The foregoing study has shown that  $\text{Ir}^{192m}$  is a well-behaved one-step isomer. The isomeric level decays by the emission of a  $58.0 \pm 0.4$ -keV ( $E3$ ) gamma ray. Although the isomer does not decay by "two-quantum" transitions which would engender a continuous gamma spectrum, the observed photon spectrum is somewhat unique in that the 58.0-keV gamma ray is converted to the extent ( $\alpha_L \geq 870$ ) that the remaining nuclear radiation does not obscure the bremsstrahlung spectrum produced by conversion electrons. Furthermore it is clear that unless source thickness, conversion-electron stopping material, and photon-detection efficiency are well known parameters, the resulting bremsstrahlung distribution may not appear to have the characteristic hyperbolic shape. The total intensity of the observed bremsstrahlung distribution can only be crudely estimated because of the presence of the comparatively intense iridium  $L$  x-rays which completely obscure the continuum below  $\sim 20$  keV. If the portion of the bremsstrahlung spectrum above 20 keV represents 10 percent of the total bremsstrahlung intensity, then there are  $< 0.01$  bremsstrahlung quanta emitted per ejected conversion electron. No attempt has been made to estimate what fraction of the observed bremsstrahlung spectrum is caused by continuous gamma radiation accompanying the internal conversion process<sup>15,16</sup> and what fraction is caused by external bremsstrahlung.

<sup>14</sup> B. B. Kinsey, Can. J. Research **A26**, 401 (1948).

<sup>15</sup> H. B. Brown and R. Stump, Phys. Rev. **90**, 1061 (1953).

<sup>16</sup> L. Spruch and G. Goertzel, Phys. Rev. **94**, 1671 (1954).