The Disintegration Scheme of I^{131}*

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The radiations of I^{131} have been studied with a magnetic lens spectrometer and coincidence techniques using "Kallmann" counters. Four gamma-rays of 80, 283, 363, and 638 kev were detected both by their internal conversion electrons and by photoelectrons emitted from different converters. A beta-ray spectrum of 600-kev maximum energy constitutes about 85 percent of the disintegrations and leads to a state of Xe^{131} with 363-kev excitation. In about 6 percent of the disintegrations, the transitions from this level to the ground state take place by the successive emission of an 80-kev and a 283-kev gamma-ray. Fifteen percent of the disintegrations proceed by the emission of a 315-kev beta-spectrum followed by a 638-kev gamma-ray.

INTRODUCTION

THE nuclear radiations of I^{131} were studied by Downing, Deutsch, and Roberts in 1942.[1] The beta-ray spectrum was found to be simple, and each beta-ray to be followed by an 80-kev and a 367-kev gamma-ray in cascade.

The development of nuclear reactors made possible the preparation of large quantities of I^{131} of high specific activity. The therapeutic application of radioactive iodine called for careful dosage measurements which revealed inconsistencies when the simple disintegration scheme was used. Absorption measurements[2] showed the presence of a 650-kev gamma-ray, the intensity of which is 15 percent of the 367-kev gamma-ray. It was therefore thought desirable to reinvestigate the radiations of I^{131} using the much better sources now available.

THE BETA-RAY SPECTRUM

"Carrier free" NaI** was deposited on a 0.04-mg/cm² "Formvar" film and covered with a second film of about the same thickness in order to prevent contamination of the spectrometer. The thickness of the source was less than one mg/cm². The momentum distribution of the beta-rays from I^{131} obtained with a magnetic lens spectrometer,[3] using a mica counter window of 0.5-mg/cm² thickness, is shown in Fig. 1. In addition to the conversion peaks resulting from the 80-kev and 367-kev gamma-rays already found by DDR, further conversion lines at 248 kev and, very weak, at about 603 kev, give evidence of the existence of two more gamma-rays of 283 kev and 638 kev, respectively. The Fermi plot (Fig. 2) of the spectrum looks very similar to the one given by DDR where at 250 kev a faint peak is visible, but was, together with the deviation from the straight line, ascribed to experimental uncertainty. Assuming the Fermi plot of a simple spectrum to be a straight line, one can split the actual spectrum into two groups, the maximum energies of which are 315 ± 20 kev and 600 ± 5 kev. An experimental justification of this procedure will be given in connection with the beta-gamma-coincidences.

Integration of the two partial spectra leads to an intensity ratio of $I_{315}:I_{600}=1:6$.

THE GAMMA-RAY SPECTRUM

Figure 3 shows the spectrum of secondary electrons ejected by the gamma-rays of I^{131} from a uranium converter of 25-mg/cm² thickness. The source was contained in an aluminum capsule thick enough to stop all beta-rays. The gamma-ray energies corresponding to the different photoelectron peaks are 283 ± 3, 363 ± 3, and 638 ± 5 kev. The relative abundances of these gamma-rays can be calculated from the heights

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[1] I. R. Downing, M. Deutsch, and A. Roberts, Phys. Rev. **61**, 686 (1942), referred to as DDR in this paper.

[2] C. M. Davisson and R. D. Evans, Bull. Am. Phys. Soc. **23**, No. 3, 45 (1948).

** Obtained from Oak Ridge National Laboratory.

[3] M. Deutsch, L. G. Elliott, and R. D. Evans, Rev. Sci. Inst. **15**, 178 (1944).

of the photoelectron lines if the effect of finite converter thickness and the variation of the photoelectric cross section with energy are taken into account. Using the formula given by Deutsch, Elliott, and Evans[3] and combining the results obtained with different converters (Sn, Au, Pb, U), we found for the relative abundance of the gamma-rays: $I_{283}:I_{363}:I_{638}=8:100:20$. The measurements with the uranium converter gave no evidence of the existence of the 80-kev gamma-ray. In order to get information concerning the intensity of this low energy gamma-ray, we performed some experiments with a tin converter of 10-mg/cm² thickness and could detect a weak K-photo-line caused by the 80-kev gamma-ray. The comparison with the other photoelectric lines showed that the 80-kev gamma-ray is about forty times less intense than the 363-kev gamma-ray. We should like to point out that for the 80-kev line the correction resulting from the absorption of the photoelectrons in the converter becomes very large and the estimate of the intensity, therefore, rather uncertain. In spite of this, the absence of an appreciable photoelectron line for the 80-kev gamma-ray, combined with our knowledge of the number of conversion electrons, enables us to exclude the disintegration scheme hitherto used. If the two gamma-rays of 80 and 363 kev were in cascade, the low intensity of the photoelectric line caused by the 80-kev gamma-ray could only be explained by assuming that more than 90 percent of the 80-kev transitions are internally converted. The numbers of internal conversion electrons of the two lines, however, are the same within a factor of two and, as we shall see later on, the conversion coefficient of the 363-kev gamma-ray is less than 2 percent. The conversion of the 80-kev transition could, therefore, not be higher than 4 percent, which is in strong disagreement with the 90 percent deduced above. We therefore conclude that the 80-kev gamma-ray is either not in cascade with the 363-kev ray or that there are other gamma-rays competing with the 80-kev line. The latter possibility, however, is excluded by the fact that the sum of the intensities of the 80-, 283-, and 638-kev gamma-rays is decidedly smaller than the intensity of the 363-kev gamma-ray alone. The 363-kev gamma-ray, therefore, leads to the ground state of the product nucleus Xe^{131}

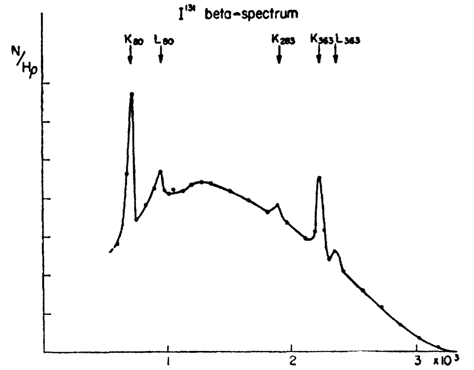

FIG. 1. The beta-ray spectrum of I^{131}. Arrows mark internal conversion lines.

and is connected with the main beta-spectrum ($E_{max}=600$ kev). Moreover, it follows that the beta-ray spectrum has to be complex.

In order to determine the correlation of the other lines, we performed the following beta-gamma- and beta-electron coincidence measurements:

BETA-GAMMA-COINCIDENCES

DDR showed that, within the uncertainty of their experiments, the number of beta-gamma-coincidences per beta-ray is independent of the thickness of the absorber inserted between source and beta-ray counter. Their measurements, however, do not exclude the possibility of the spectrum being complex because the effi-

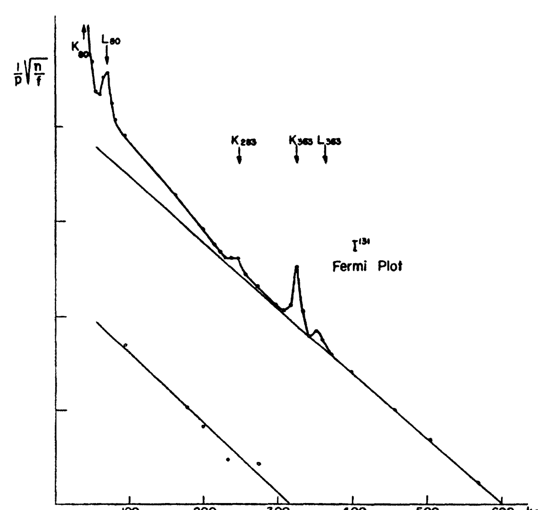

FIG. 2. Fermi plot of the beta-ray spectrum of I^{131}.

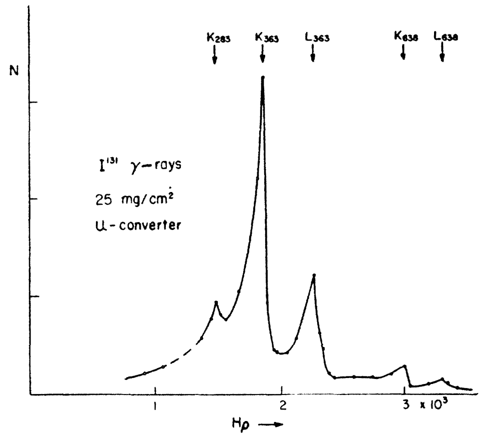

FIG. 3. Secondary electron spectrum caused by the gamma-rays of I^{131}.

ciency of platinum screen cathode gamma-ray counters varies only slightly between 350- and 650-kev gamma-ray energy[4] and, therefore, the probability of causing a coincidence is about the same for all beta-rays. On the basis of our disintegration scheme (Fig. 6), we would expect a change of less than 4 percent in the coincidence rate per beta-ray if the entire soft beta-spectrum is absorbed. Such a small variation is compatible with the experimental evidence of DDR. Similar considerations can be applied to the other coincidence experiment of DDR (reference 1, Fig. 5). Both earlier coincidence measurements are, therefore, reconcilable with the existence of a low energy beta-ray spectrum of the abundance given by our analysis of the Fermi plot (Fig. 2).

In order to give positive evidence of the reality of the second beta-ray spectrum and of its connection with the 638-kev gamma-ray, the number of beta-gamma-coincidences between beta-rays, focused in the spectrometer, and gamma-rays, detected by a "Kallmann" counter,[5] was measured for different beta-ray energies with and without 4 grams per cm² of lead in front of the gamma-ray counter.

The "Kallmann" counter consisted of a reasonably clear piece of naphthalene of about $1\times1.5\times3$ cm³ attached to a RCA 931-A photo-multiplier tube which was cooled with dry ice and operated at about 70 volts per stage. The pulses were amplified by a Los Alamos Model 501 amplifier (0.1-μsec. rise time) and fed into a coincidence circuit with a 0.1-μsec. gate for the G-M tube pulses and a 0.3-μsec. gate for the photomultiplier pulses.

At first the magnetic field of the spectrometer interfered with the proper operation of the multiplier tube, the counting rate being strongly dependent on the coil current of the spectrometer. Since the magnetic field of a thin lens spectrometer falls off quite fast along the axis, it was not difficult, at least for the energy range used, to provide sufficient shielding for the multiplier tube by using an iron—instead of a brass—end plate for the spectrometer. For the same reason, the calibration of the spectrometer was only slightly altered by this change. Throughout all the work described here, the spectrometer was used with a focal length of 12.5 cm.

In Fig. 4 the two beta-ray spectra are shown; some typical settings of the spectrometer for the beta-gamma coincidence measurements are indicated by vertical lines. In positions II and III the hard component alone contributes to the beta-gamma-coincidences, whereas in position I the soft spectrum participates appreciably. Figure 5 summarizes the results of these experiments. For beta-energies above 300 kev, the attenuation of the beta-gamma-coincidences corresponds to the one which is to be expected for a mixture of a 363- and a much less abundant 283-kev gamma-ray. For low beta-energies the attenuation of the coincidences is less strong than the one measured for the total gamma-ray beam, which demonstrates that part of these electrons are connected with a high energy gamma-ray (638 kev). The shape of the curve is consistent with an upper limit of about 300 kev for the low energy spectrum.

BETA-ELECTRON COINCIDENCES

For the purpose of determining the internal conversion coefficient of the 363-kev gamma-ray, DDR had measured coincidences between beta-rays and conversion electrons focused in the spectrometer. In addition to the conversion line of the 363-kev gamma-ray, a second peak at about 250 kev appeared, which can now be identified as the conversion line of the 283-kev gamma-ray.

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4 Roberts, Elliott, Downing, Peacock, and Deutsch, Phys. Rev. **64**, 268 (1943).

5 See, e.g., M. Deutsch, Nucleonics **2**, No. 3, 58 (1948).

We performed an experiment similar to the experiment of DDR just described, but put 40 mg/cm² of aluminum absorber between the source and the beta-ray counter, cutting out in this way almost the entire soft spectrum. We therefore expect to get beta-electron coincidences only for the gamma-lines accompanying the hard component of the beta-ray spectrum. Focusing the conversion electrons of the 80-, 283-, and 363-kev gamma-rays, we found that, within the experimental error which is about 15 percent for the 80- and 363- and 30 percent for the 283-kev gamma-ray, the number of coincidences per conversion electron is the same for all these three gamma-rays. The 80- and 283-kev gamma-rays are, therefore, connected with the beta-spectrum in the same way as the 363-kev gamma-ray; that is, they accompany the high energy beta-component. The 363-kev gamma-ray leads, as was already pointed out, to the ground state of Xe^{131}. The energies of the two soft gamma-rays add up to 363 kev, and their intensities are comparable. Furthermore, DDR showed that most of the gamma-gamma-coincidences are absorbed with the characteristics of an 80-kev gamma-ray. All these facts make it practically certain that the 80-kev and the 283-kev gamma-rays are in cascade. With this assignment, the coincidence measurements of DDR no longer lead to a much too low efficiency for a platinum screen cathode counter for 80-kev gamma-rays, but fit well with newer measurements of counter efficiencies.[6]

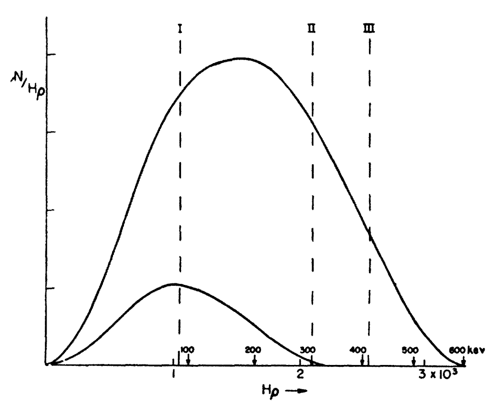

FIG. 4. The two components of the beta-ray spectrum of I^{131} as derived from the analysis of the Fermi plot (Fig. 2). The vertical lines mark typical settings of the spectrometer for the beta-gamma-coincidence experiments.

[6] H. Maier-Leibnitz, Zeits. f. Naturforschung **I**, 243 (1946).

TABLE I. Gamma-rays of I^{131}.

Energy in kev	Intensity rays/100 disintegration	Conversion coefficient $Ne/N\gamma$	N_K/N_L
80 ± 1	6	0.8 ± 0.5	5.5 ± 2.5
283 ± 3	6	0.05 ± 0.02	>2
363 ± 3	79	0.019 ± 0.005	5.2 ± 1.5
638 ± 5	15	<0.005	—

Using the new scheme and the coincidence data of DDR, one finds that the efficiency of a platinum screen cathode counter for 80-kev gamma-rays is about the same or even higher than the one for the 280–650-kev range. The reason for this high efficiency lies in the fact that together with the extremely soft photoelectrons which would not escape, a large percentage of fairly energetic Auger electrons are produced in the absorption of the 80-kev quanta.

CONVERSION COEFFICIENTS

Table I contains our information concerning the gamma-rays of I^{131}. The number of conversion electrons was determined from the spectrometer data shown in Fig. 1. Owing to absorption in the source and in the counter window, in the case of the low energy line, and to the low conversion of the other lines, our results are rather inaccurate. Our value for the conversion coefficient of the

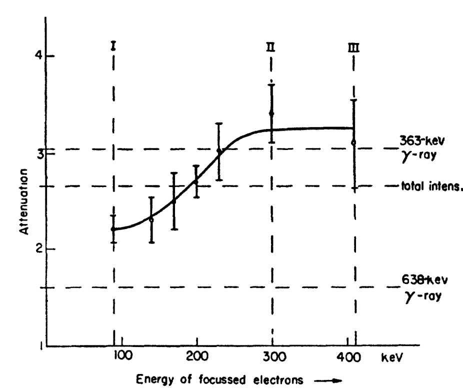

FIG. 5. Attenuation of the number of beta-gamma-coincidences caused by 4 grams per cm² of lead in front of the gamma-ray counter. The vertical lines correspond to the typical settings of the spectrometer indicated in Fig. 4. The horizontal lines mark the attenuation of selected gamma-rays. These attenuations were calculated by using the absorption coefficients given by Glendenin (L. E. Glendenin, Nucleonics **2**, No. 1, 12 (1948)), and an effective absorber thickness was calculated matching the attenuation of the total beam.

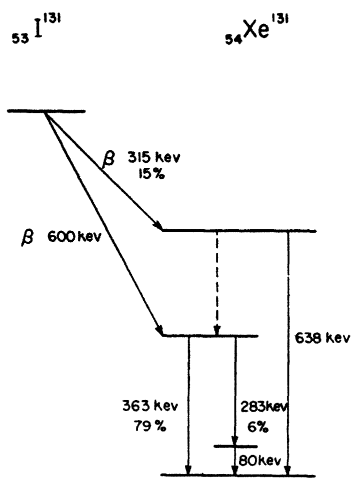

FIG. 6. Disintegration scheme of I^{131}.

363-kev gamma-ray is considerably higher than the value determined by DDR in a more direct way.

The experimental uncertainty and the lack of accurate calculations of internal conversion coefficients for medium atomic numbers make the determination of multipole orders rather ambiguous. The data collected in Table I are compatible with the assumption that all gamma-rays arise from electric quadrupole transitions, the 80-kev line having an appreciable admixture of magnetic dipole radiation. Multipole radiation of higher order is definitely excluded for the 80-kev gamma-ray from lifetime considerations. Electric dipole transition is unlikely for the 283- and 363-kev gamma-rays because of the appreciable conversion coefficients and the relatively low N_K/N_L ratio.

THE DISINTEGRATION SCHEME OF I^{131}

Figure 6 shows the disintegration scheme of I^{131} which we propose, based on our present knowledge. We cannot exclude the existence of further gamma-rays with energies coinciding within a few percent with the known lines and which may be partly connected with a very soft beta-spectrum. A measurement with a spectrometer of high resolving power is therefore desirable, especially as we have some indication of a line of about one percent abundance falling in the low energy tail of the 283-kev line and agreeing in energy with the difference of the two upper levels of Xe^{131} involved in our scheme (dotted line in scheme). On the other hand, in the entire range from 80 kev to 1 Mev, every gamma-ray not coincident with the lines already known would be of less than 3 percent abundance.

The order of the emission of the 80- and 283-kev gamma-rays is not definitely known. The one proposed in this scheme is easier to bring into agreement with lifetime considerations and spin assignments. If the 80-kev line corresponds to an electric quadrupole transition, the lifetime according to the Segrè formula[7] is 6.10^{-8} sec., according to Lowen's[8] calculations, 4.10^{-9} sec.; it, therefore, falls within the range which can be explored by fast coincidence techniques using multiplier tubes.

The $f \cdot t$-values of the two beta-spectra in the notation of Konopinsky[9] are nearly the same and fit very well into the range ascribed in this region of Z to first forbidden spectra.

[7] See A. C. Helmholz, Phys. Rev. **60**, 415 (1941).
[8] I. S. Lowen, Phys. Rev. **59**, 835 (1941).
[9] E. J. Konopinsky, Rev. Mod. Phys. **15**, 209 (1943).