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Continuous Electron Spectrum Accompanying K Capture*

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The continuous electron spectrum accompanying K capture has been observed, in the case of Cs^{131}, in three different ways: by measuring coincidences between one K x-ray and an electron, by measuring coincidences between two K x-rays and an electron, and by measuring the single electron spectrum. Furthermore, coincidences between two K x-rays have been measured. The coincidence data were taken with scintillation counters while the single electron spectrum was taken with a magnetic spectrometer. The electron spectrum, including the absolute intensity, and the probability for a double hole in the K shell are both found to be in agreement with the theory of Primakoff and Porter.

I. INTRODUCTION

As is well known, beta decay is accompanied by a continuous gamma-ray spectrum. Theory and experiment are in good agreement.[1] The emission of atomic electrons has been observed with β^- decay as well as β^+ decay. X-rays following ionization of inner electron shells have also been found.[2] In the case of orbital electron capture, there have been some observations on the radiation resulting from the filling of two holes in the K shell.[3,4] The spectral distribution of the ejected electrons, however, has not been established experimentally.[5]

The theory of the atomic excitation and ionization accompanying orbital electron capture has been given by Primakoff and Porter[6]; for a discussion of the accuracy the reader is referred to the original paper. According to these authors the probability per K capture for the production of a double hole in the K shell is

$$P_{KK}\cong 3/(16Z_i^2). \tag{1}$$

Z_i is the atomic number of the initial nucleus. The probability per K capture of ejecting a K electron is

$$P_{\mathrm{ejec}}\cong 0.08/Z_i^2. \tag{2}$$

The difference between Eqs. (1) and (2) is due to processes where the electron removed from the K shell occupies a bound state. The momentum distribution of the ejected K electrons is given by

$$D_{\mathrm{ejec}}(p_e)=D_{\beta^-}(p_e)\exp\left[\frac{-4(Z_i-1)}{137\beta_e}\tan^{-1}\left(\frac{137p_e}{Z_i-\frac{1}{2}}\right)\right]\times\left[1+\left(\frac{137}{Z_i-\frac{1}{2}}\right)^2 p_e^2\right]^{-4}, \tag{3}$$

with

$$D_{\beta^-}(p_e)=A\times p_e^2[E_0-p_e^2/2]^2\times\left[\frac{2\pi(Z_i-1)}{137\beta_e}\times\left(1-\exp\left[\frac{-2\pi(Z_i-1)}{137\beta_e}\right]\right)^{-1}\right]. \tag{4}$$

p_e is the electron momentum in units of mc, β_e is the electron velocity in units of c, and E_0 is the maximum energy of the continuous electron spectrum in units of mc^2. The constant A in Eq. (4) can be normalized by integrating Eq. (3) graphically and setting the result equal to Eq. (2). After this normalization, $D_{\mathrm{ejec}}(p_e)dp_e$ is the probability per K-capture decay that an electron is ejected with a momentum between p_e and p_e+dp_e. The electrons whose momentum distribution is represented by Eq. (3) are all from the K shell; the contribution from the L shell, in the approximation of Primakoff and Porter, vanishes.

The two practically simultaneous radiations resulting from the filling of the two holes in the K shell will not have the same energy. An electron jumping into a completely empty K shell will liberate a larger amount of energy than an electron with the same initial state but jumping into a K shell already occupied by one electron. The energy difference is of the order of

$$\Delta E\cong 2s_K E_K/Z_f, \tag{5}$$

where $s_K\leqq 1$ is the screening constant[7] for one K electron, Z_f is the atomic number of the final nucleus, and E_K is the K binding energy for $Z=Z_f$.

Both atomic excitation and ionization as described

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[1] C. S. Wu in, *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (Interscience Publishers, Inc., New York, 1955), Chap. XX.

[2] For a survey see: G. A. Renard, J. phys. radium **16**, 575 (1955); A. Michalowicz and R. Bouchez, *ibid.* **16**, 578 (1955); J. S. Levinger, *ibid.* **16**, 556 (1955); G. Charpak, *ibid.* **16**, 567 (1955).

[3] G. Charpak, Compt. rend. **237**, 243 (1953); J. phys. radium **16**, 567 (1955).

[4] J. A. Miskel and M. L. Perlman, Phys. Rev. **94**, 1683 (1954); **95**, 612 (1954).

[5] Miskel and Perlman[4] give a comparison between the theoretical distribution, modified for experimental resolution width, etc., and the measured curve but this covers only the range 0–4 kev out of 800 kev. The distributions are not in good agreement.

[6] H. Primakoff and F. T. Porter, Phys. Rev. **89**, 930 (1953).

[7] For the effect of screening see: A. Sommerfeld, *Atombau und Spektrallinien* (Friedrich Vieweg und Sohn, Braunschweig, 1951), Vol. 1.

above can be observed by detecting the filling of the two holes in the K shell, either with summing in one counter or with coincidences between two counters. In principle, one can also use the energy shift as given by Eq. (5). These methods, however, do not give any information about the electron spectrum nor whether the K electron not captured by the nucleus is kept in a bound state or has been ejected into an unbound state. There are essentially three methods of observing the electron spectrum: (a) measuring the single electron spectrum, (b) measuring the electron spectrum in coincidence with the radiation resulting from the filling of one K hole, and (c) measuring the electron spectrum in coincidence with the radiations resulting from the filling of both K holes. The radiation resulting from the filling of a K-shell hole may be either an x-ray quantum or an Auger electron.

Charpak[3] observed, with two x-ray proportional counters in coincidence, the double vacancy in the K shell in the Fe^{55} decay. He obtained a value of $P_{KK}=(3.9\pm0.9)\times10^{-4}$. Miskel and Perlman,[4] using a proportional counter and the summing method, obtained a value of $P_{KK}=(5.3\pm1.0)\times10^{-4}$ in the case of Ar^{37}. The theoretical values according to Eq. (1) are 2.8×10^{-4} and 5.8×10^{-4}, respectively, indicating good agreement between experiment and theory.

In order to observe the electron spectrum from the decay of Fe^{55} Porter and Hotz[8] tried two different methods. With a magnetic spectrometer they did not observe any counts above background. With a cloud chamber they observed some electrons in the correct energy range. From these data they concluded that there are less than 6×10^{-7} ejected electrons per decay with energies greater than 30 kev. The theoretical value for the same energy range is 36×10^{-7}. For a detailed discussion the reader is referred to the original paper.

According to Eqs. (1) and (2) it seems to be advantageous to work with a low-Z isotope. This is, however, not completely correct. It is simpler, and therefore gives more accurate results, to observe higher energy x-rays (or Auger-electrons). Furthermore, when observing x-rays it is advantageous to have an element with a high fluorescent yield. Both effects shift the optimum Z value to higher figures. Moreover, the intensity of electrons of a given energy, Eq. (3), does not show the $1/Z_i^2$-dependence but is less disadvantageous for higher Z_i values.

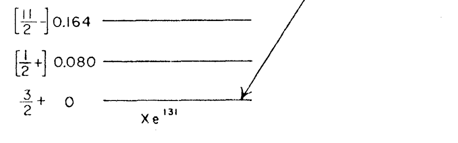

FIG. 1. Decay scheme of Cs^{131} and low-lying levels of Xe^{131}. Energies in Mev.

Cs^{131} was chosen for the present experiments. Figure 1 shows the decay scheme and the low-lying levels[9] in Xe^{131}. No transitions to either the 80-kev level or the 164-kev level are observed; the upper limits are 10^{-7}/decay and 10^{-10}/decay, respectively.[10] The maximum energy of the continuous bremsstrahlung spectrum is 0.32 Mev.[9] Since the K binding energy of the daughter is 35 kev, the maximum energy of the continuous electron spectrum is 0.29 Mev. The half-life of Cs^{131}, 9.9 days, allows one to follow the decay of the observed radiation, thus providing a further check.

Because of the uncertainty involved in experiments on small order effects, it seemed worthwhile to use as many independent experimental methods as available. The single electron spectrum was investigated with a magnetic spectrometer. Coincidences between one K x-ray and an electron were taken with two scintillation counters in coincidence. Coincidences between two K x-rays and an electron were taken with the same arrangement but with the gamma channel set for the summing peak of the two K x-rays. Finally, coincidences were measured between both K x-rays, each x-ray being detected by one scintillation counter. In all four experiments the effect searched for was observed and, within experimental error, was found to be in agreement with the theory.[6]

II. EXPERIMENTAL TECHNIQUES

For the production of Cs^{131}, pure $BaCO_3$ was irradiated in the Brookhaven reactor. From the Ba^{131}, the daughter Cs^{131} was separated carrier-free.[11] From the resulting solution one liquid-drop source was prepared for the magnetic spectrometer, with an intensity of 6.5×10^7 K-capture decays per sec at the time the beta-spectrometer work started. Because of the large amount of solution and reagents involved in the chemical separation, the resulting source had a thickness of 9 mg/cm^2. Later, much weaker sources of negligible thickness and various strengths were made for the scintillation spectrometer work.

The electron spectrum from the strong source was measured with a modified Slätis-Siegbahn beta-ray sepctrometer[12] the baffle setting of which could be changed from outside. The detector was an anthracene scintillation counter. The crystal, 14 mm in diameter and 1.2 mm thick, was covered with 150 μg/cm^2 of Al. The cathode of the phototube was cooled to $-55°$C by means of a refrigerator unit. In order to further

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[8] F. T. Porter and H. P. Hotz, Phys. Rev. **89**, 938 (1953).

[9] Strominger, Hollander, and Seaborg, Revs. Modern Phys. **30**, 585 (1958); *Nuclear Data Sheets*, edited by C. L. McGinnis, (National Academy of Science, Washington, D. C., 1958).

[10] B. Saraf, Phys. Rev. **94**, 642 (1954).

[11] B. Finkle and W. E. Cohn, in *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Company, Inc., New York, 1951), Paper No. 287, National Nuclear Energy Series, Plutonium Project Record, Vol. 9, Book III, p. 1654.

[12] To be described in a later publication.

reduce the background the scintillation pulses were selected by a sliding channel that was set to correspond to the energy of the electrons focused by the magnetic spectrometer. The spectrometer was operated at 7% transmission and 7% resolution (these figures are not representative for the spectrometer because all baffles were opened as far as possible). Runs were taken with the intermediate-image baffle open and closed. This technique has proven to be useful for the detection of small electron intensities and has been described in more detail elsewhere.[13] The decay of the electron spectrum was followed for 24 days.

For the e^--x-ray coincidence work the same scintillation beta-counter was used. Here the lower background, because of the cooled cathode, reduced the rate of accidental coincidences. The resolution of the counter at 25 kev was 58%. The *K* x-rays were detected with a 3-in. ×3-in. Harshaw NaI crystal. The pulses in the x-ray branch had to pass a single channel analyzer. The electron spectrum in coincidence with the x-rays was obtained with a RCL 256-channel analyzer. The resolving time of the fast coincidence circuit was $2\tau=2.5\times10^{-7}$ sec. The resolution of the x-ray counter was 31% at 30 kev. The source was placed between both counters with the angle between the counters being 180°. The solid angle was almost 50% of 4π for the x-rays and 28% of 4π for the electrons. An Al absorber, of 70 mg/cm^2, could be placed between the source and the beta crystal to absorb all electrons below about 300 kev. The whole counter arrangement was shielded with copper against low-energy gamma radiation from outside.

The detection probability of the NaI crystal for the *K* x-rays was measured with a Cs^{137} source. The detection probability of such a crystal for Cs^{137} gamma rays is known,[14] as are the number of *K* holes in Ba^{137} per Cs^{137} decay,[9] and the fluorescent yields.[15]

In order to measure coincidences between one *K* x-ray and an electron the single channel analyzer in the x-ray branch was set on the *K* x-ray photopeak while for coincidences between the two *K* x-rays and an electron it was set on the region of the summing photopeak. In each case four different runs were taken: prompt coincidences without and with the Al absorber between the source and the beta crystal and delayed coincidences without and with the Al absorber. The delayed coincidence runs gave the spectra of the accidental coincidences while the runs with the Al absorber in place gave the contribution from processes where a photon was detected by the anthracene crystal. The spectrum of the true coincidences obtained with the Al absorber in place was subtracted from the spectrum of the true coincidences obtained without the Al absorber. An estimate showed that the Al absorber did not noticeably affect the *K* x-ray intensity and, at least at lower energies, there was only a negligible increase in the detection probability of the beta counter for gamma quanta. The source strength for these measurements was centered around 5×10^3 *K*-capture decays/sec.

For measuring coincidences between the two *K* x-rays, the same setup was used except that the anthracene beta-counter was replaced by a second NaI x-ray counter identical to the first one. The source was held in the hole of a copper shield that was placed between the two crystals. Here again, the spectrum of the accidental coincidences was measured with a time delay introduced into one branch. The source strength was of the order of 200 *K*-capture decays/sec.

III. RESULTS

The decay curves taken with the magnetic spectrometer at different energies showed the presence of a beta-active contamination with a half-life of 25 days. This, of course, made the determination of the Cs^{131} electron spectrum more uncertain. With the spectrometer set at $p_e=0.42$ *mc* (43 kev) the decay curve showed a component having a half-life of about 10 days, while at higher momenta ($p_e=0.57$ *mc* and up) one could not deduce the presence of such a component. After correction for self-absorption in the source and counter efficiency a value of $D_{\text{ejec}}=9\times10^{-6}$ was obtained for $p_e=0.42$ *mc*. For the accuracy of this result see Sec. IV.

The result of the coincidence experiment with one *K* x-ray in the one branch and the electron in the other is shown in Fig. 2. In order to decrease the statistical

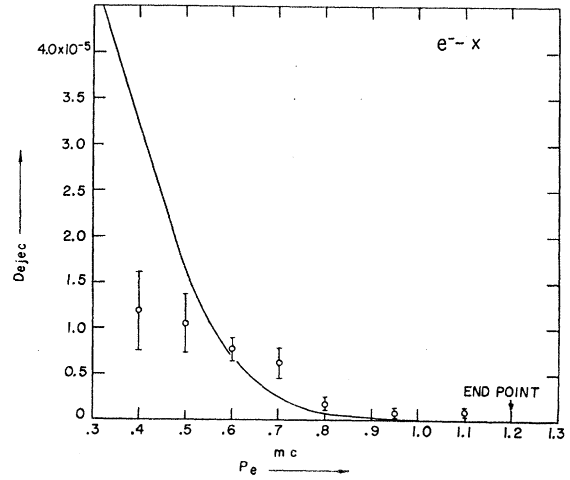

FIG. 2. Electron spectrum in coincidence with one *K* x-ray. The experimental points are shown with their standard deviations and the solid line represents the theoretical curve as given by Eq. (3), normalized with the help of Eq. (2). Both the experimental points and the theoretical curve give the absolute values of D_{ejec}.

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[13] H. Daniel, Z. Physik **150**, 144 (1958).

[14] Wolicki, Jastrow, and Brooks, Naval Research Laboratory, NRL Report 4833, 1956 (unpublished); R. L. Heath, Idaho Operations Office Report IDO-16408, 1957 (unpublished).

[15] Wapstra, Nijgh, and van Lieshout, *Nuclear Spectroscopy Tables* (Interscience Publishers, Inc., New York, 1959).

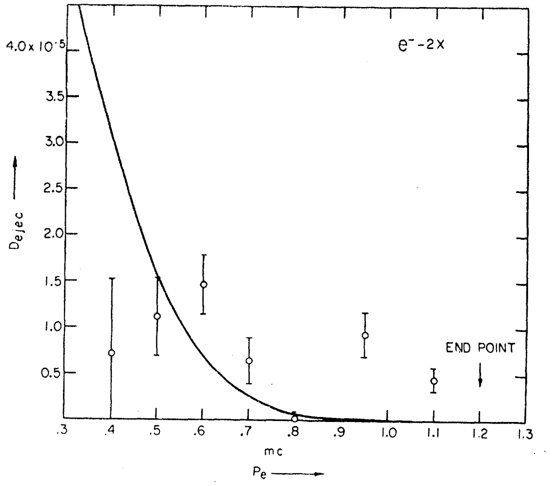

FIG. 3. Electron spectrum in coincidence with two K x-rays. The experimental points are shown with their standard deviations and the solid line represents the theoretical curve as given by Eq. (3), normalized with the help of Eq. (2). Both the experimental points and the theoretical curve give the absolute value of D_{ejec}.

fluctuations, all data of the 256-channel analyzer falling into an energy interval corresponding to p_e between 0.35 and 0.45 *mc* have been treated together. In the same way the other four lower energy points in Fig. 2 correspond to a p_e-range of 0.10 *mc* while the last two points, because of the very low intensity, were chosen to represent a p_e-range of 0.15 and 0.20 *mc*, respectively. The indicated errors are the statistical standard deviations. The solid line represents the theoretical curve as given by Eq. (3), normalized with the help of Eq. (2). Both the theoretical curve and the experimental points give the absolute value of D_{ejec}. The endpoint of the expected electron spectrum is at $p_e=1.20$ *mc*. Data below 0.35 *mc* were distorted by Auger electrons.

Figure 3 shows the electron spectrum obtained in coincidence with the summing peak of the two K x-rays. The ranges in the electron momentum p_e are taken to be the same as in Fig. 2.

The measurement of coincidences between the two K x-rays led to a probability per K-capture decay for the formation of two K holes of

$$P_{KK}=(5.0\pm1.0)\times10^{-5}.$$

The stated error is the standard deviation of counting statistics only. The theoretical value, according to Eq. (1), is $P_{KK}\cong6.2\times10^{-5}$.

The energy of the one K x-ray in coincidence with the other K x-ray was also measured in the x-ray-x-ray coincidence runs. No indication of an energy shift was found when comparing prompt coincidence curves with accidental coincidence curves. The energy shift was found to be less than 1%. Because of the occurrence of accidental coincidences in the prompt runs this does not mean, however, that both K x-ray quanta (for identical initial energy states) have the same energy within 2%.

IV. DISCUSSION

The agreement between the experimental and theoretical values for P_{KK}, the number of double holes in the K shell per K capture, is excellent. This is in line with previous work on other isotopes.[3,4] The error given above for the experimental value of P_{KK} does not include any systematic error. Systematic errors are expected to be small since the most uncertain quantity, the detection probability of a K x-ray quantum, has been determined experimentally in a straightforward manner. Since P_{KK} was determined with the same arrangement as the K capture rate of the source, the detection probability enters the final result only once.

From the present three measurements of the electron spectrum the data taken in coincidence with only one K x-ray quantum are the most accurate. Not only are the statistics better than in the other two experiments but the method does not involve any uncertainty for the x-ray detection probability in the NaI crystal. The detection probability for the electron in the anthracene counter, however, enters the final result. This value has been obtained from geometrical considerations only. There is, of course, an energy dependent backscattering out of the anthracene crystal. This effect, however, is small, except for very low energies, because of the low average atomic number of anthracene. The aluminum covering on the anthracene crystal is too thin to have an appreciable influence on either detection probability or linearity of the energy response. For the experimental points shown in Fig. 2 it is estimated that the total systematic error does not exceed a factor of 1.5, maybe with the exception of the point at $p_e=0.4$ *mc*.

The electron spectrum taken in coincidence with both K x-ray quanta suffers not only from poorer statistics but there is, as an additional source of systematic error, the dependence on the detection efficiency of the NaI crystal for one K x-ray. Moreover, some single K x-ray pulses may have fallen into the channel set for the summing peak.

The least accurate measurement was that of the single electron spectrum done with the magnetic spectrometer. In this technique any beta-active contamination is counted and makes the determination of the spectrum looked for less precise. The absolute electron intensity was subject to a large error due to the self-absorption of the electrons in the source, an effect which can only be roughly estimated. The intensity measured in this experiment is uncertain within a factor as large as four.

It is easily checked that the internal bremsstrahlung cannot strongly distort the results of experiments like the present one, because of its low over-all intensity and, particularly, because of the low intensity at low energies of the bremsstrahlung connected with *K* capture.[16] Serious distortions would have to be expected if there existed an intense *K*-capture branch to an excited state of Xe^{131}, expecially if the following gamma ray was highly converted. But such a distortion is excluded by the decay scheme of Cs^{131}.

According to the theory,[6] the contribution of *L* electrons to the total spectrum of the ejected electrons is negligible. The measurements of the electron spectrum in coincidence with one *K* x-ray include also *L*-electron ejection while those with the electron spectrum in coincidence with two *K* x-rays do not. The results shown in Figs. 2 and 3 are, of course, not nearly accurate enough to show such a difference. In the same way the contribution from *L*-electron capture to the spectrum of ejected *K* electrons is included in the results shown in Fig. 2 but not in those of Fig. 3. Again the accuracy is not sufficient to show the difference. The ratio of *L* capture to *K* capture in Cs^{131} has been calculated[17] to be $\lambda_L/\lambda_K=0.14$. In principle any contribution from *L* electrons could be observed by measuring coincidences between two *L* x-rays or the corresponding Auger-electrons (with proper corrections for processes involving two holes in the *K* shell). These measurements, however, seem to be difficult.

The authors[6] do not claim that the theory is completely correct. The reader is referred to the original paper[6] and reference 8 for an extended discussion. There is some doubt about a correction factor $\leqq 1$ to be applied to the right hand side of Eq. (1) which could reduce the theoretical value for P_{KK} by as much as 50%.[6] All three experimental values (references 3, 4, and the present paper) for P_{KK} do not suggest such a large reduction. The experiments on the electron spectrum do not suggest that a reduction factor should be applied to the right-hand side of Eq. (3) but, on the other hand, the data are not accurate enough to exclude a small reduction.

For Xe^{131} the energy shift given by Eq. (5) is calculated to be $\Delta E \cong 1.28$ kev, s_K being taken as unity which is the upper limit. In the absence of accidental coincidences in the prompt coincidence runs this would cause a line shift in a *K* x-ray-*K*-x-ray coincidence experiment of about 0.64 kev or 2.17%. With an average ratio of true to accidental coincidences of $\langle n_{\mathrm{true}}/n_{\mathrm{acc}}\rangle_{\mathrm{Av}}=0.175\pm0.034$, as in the present experiment, the upper limit of the expected line shift is too small to exceed the experimental limit of 1%.

It is concluded that, within their respective errors, theory and present experiment are in good agreement on both the shape and absolute intensity of the continuous electron spectrum as well as the probability for a double hole in the *K* shell following *K* capture.

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