Radiative Electron Capture of Fe⁵⁵, A³⁷, and Ni⁵⁹

W. S. Emmerich, S. E. Singer, and J. D. Kurbatov Department of Physics, The Ohio State University, Columbus, Ohio (Received December 18, 1953)

The radiative electron capture spectra of Fe⁵⁵, A³⁷, and Ni⁵⁹ were studied with a scintillation spectrometer. The end-point energies were found to be 226±10 kev, 815±20 kev, and 1065±30 kev, respectively. A method was devised to correct the experimental intensity distributions for distortions due to the Compton effect in the scintillation crystal. This was applied to the spectra of A³⁷ and Ni⁵⁹ above 320 kev. In this energy range, the experimental spectrum of \hat{A}^{37} was found to agree with the calculated spectral shape. In the case of Ni⁵⁹, a dependence of the spectral shape on the type of transition could not be detected within present experimental limits.

I. INTRODUCTION

HE occurrence of radiative orbital electron capture provides a means of determining directly the transition energy associated with the disintegration. The shape of the spectrum may in some cases give information regarding the type of transition involved. Morrison and Schiff¹ have given an expression for the relative probability of photon emission per unit time, dP, with respect to the total K-capture probability per unit time, P_{K} . For allowed transitions, this ratio is given, except at low energies, by

$$dP/P_{K} = (\alpha/\pi) (1 - E/E_{0})^{2} E dE, \qquad (1)$$

where α is the fine structure constant, E is the energy of the photon in units of mc^2 , and E_0 is the energy available for the transition. Jauch² has shown that E_0 is obtained by plotting $(N/E)^{\frac{1}{2}}$ against E, where N is the observed activity per unit energy interval. If the experiments are in accordance with the derived distribution, the plot should be linear and intersect the axis at E_0 , in analogy to a Kurie plot in beta-ray analysis. The total radiation probability per K capture is approximately given by

$$\int dP/P_K = (\alpha/12\pi) (E_0)^2.$$
⁽²⁾

Experimental studies of radiative electron capture have been reported by a number of groups. The earliest measurements were carried out with Fe55 by Bradt et al.,³ using absorption technique. Later results obtained by Maeder and Preiswerk⁴ with scintillation counters showed that the end-point energy of the radiative capture spectrum is within experimental error with the value calculated from the $Mn^{55}(p,n)Fe^{55}$ reaction threshold.⁵ It was also shown that the experimental intensity distribution corresponds to the calculations. Bell, Jauch, and Cassidy⁶ have reinvestigated the spectrum with a scintillation spectrometer and analyzed their data in the form of a linear plot.

The radiative capture spectrum of A³⁷ has been measured by Anderson, Wheeler, and Watson.⁷ These authors applied the linear plot analysis to their scintillation spectrometer results and obtained an end-point energy in agreement with that calculated from the $Cl^{37}(p,n)A^{37}$ reaction threshold.⁸ The data were corrected by calculating the efficiency of the scintillation crystal for detection of electromagnetic radiation, but not for the effect of Compton scattering.

The possibility of studying radiative electron capture in a forbidden transition presents itself in the case of Ni⁵⁹. Up to the present time, no investigation has been reported for radiative electron capture in a forbidden transition except Tl²⁰⁴, for which the end-point energy has been determined.9 Nickel59 decays by orbital electron capture to the ground state of Co⁵⁹ with a half-life of about 7×10^4 years.¹⁰ The threshold energy of the $Co^{59}(p,n)Ni^{59}$ reaction¹¹ indicates that an endpoint energy of 1066 kev is expected for the radiative capture spectrum.

II. PREPARATION OF RADIOACTIVITIES

The activities for the present study were produced by neutron reactions. Natural iron was irradiated in the Oak Ridge reactor and aged for five years before measurements were initiated. This eliminated interference from the activity of Fe⁵⁹ which decays with a half-life of 46 days. The iron, as ferric chloride, was separated from a small amount of Mn⁵⁴ activity present in the sample, by ethyl ether extraction. It was then precipitated repeatedly at low pH as hydrous ferric oxide, in the presence of carriers consisting of potassium, sodium, manganous, cobaltous, nickel, zinc, strontium, and barium chlorides. Lanthanum and ceric ion carriers were then added and removed as the

¹ P. Morrison and L. I. Schiff, Phys. Rev. 58, 24 (1940).
² J. M. Jauch, Oak Ridge National Laboratory Report ORNL-1102, 1951 (unpublished).
⁸ H. Bradt *et al.*, Helv. Phys. Acta 19, 222 (1946).
⁴ D. Maeder and P. Preiswerk, Phys. Rev. 84, 595 (1951).
⁵ P. H. Stelson and W. M. Preston, Phys. Rev. 83, 469 (1951).
⁶ Bell, Jauch, and Cassidy, Science 115, 12 (1952).

⁷ Anderson, Wheeler, and Watson, Phys. Rev. **90**, 606 (1953). ⁸ Richards, Smith, and Browne, Phys. Rev. **80**, 524 (1950); Schoenfeld, Duborg, Preston, and Goodman, Phys. Rev. **85**, 873 (1952).

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 ⁹ E. der Mateosian and A. Smith, Phys. Rev. 88, 1186 (1952).
 ¹⁰ Brosi, Borkowski, Conn, and Griess, Phys. Rev. 81, 391 (1951); H. W. Wilson (private communication).
 ¹¹ J. J. McCue and W. M. Preston, Phys. Rev. 84, 384 (1951).

oxalates. This was followed by final precipitation of the iron as the hydrous oxide.

The A³⁷ activity was obtained by irradiation of 30 grams of calcium oxide in the Brookhaven reactor for a period of two months. The argon, formed by (n,α) reaction, was extracted by reacting the calcium oxide with water in an atmosphere of common argon and collecting the gaseous products in an evacuated vessel. A half-life measurement of electromagnetic radiation emitted by the sample gave a half-life of 35 days.

Nickel⁵⁹ was obtained by neutron irradiation of 39 grams of nickel metal for a period of 28 days in position A of the NRX reactor. The material was aged for one year before measurements were started. The active impurities present in the nickel sample included millicurie quantities of Co⁵⁸ and Co⁶⁰, and microcurie amounts of Fe⁵⁵ and Mn⁵⁴. The Ni⁵⁹ yield, in terms of ordinary electron capture, was calculated to be about 200 microcuries. Iron was removed by precipitation of the hydrous oxide at low pH. The sample was then treated repeatedly with alpha-nitroso-beta-naphthol,



radiative electron capture spectrum in Fe⁵⁵.

with the addition of cobalt carrier, to remove active cobalt species. The nickel was then separated from Mn⁵⁴ and other contaminants by precipitation of the nickel dimethylglyoxime complex from homogeneous solution using a method devised especially for this investigation.¹² The final sample was obtained by converting the nickel dimethylglyoxime to the oxide. The presence of Ni⁶³ did not interfere with the measurement of the radiative capture because no gamma radiation is emitted in the decay and the beta spectrum has a transition energy of only 67 kev.

III. EXPERIMENTAL PROCEDURE

Measurements were made with a scintillation spectrometer consisting of a thallium activated sodium iodide crystal, one inch long and 1.5 inch in diameter, coupled to a RCA type 5819 photomultiplier tube, followed by a linear amplifier, and a Johnstone-type



FIG. 2. Uncorrected scintillation spectrogram of the radiative electron capture spectrum in A^{37} . Only the energy region above 300 kev was utilized for analysis of the spectral shape.

single-channel pulse-height analyzer.¹³ The experimentally obtained radiative capture spectrum of Fe⁵⁵ is shown in Fig. 1. The spectra for A³⁷ and Ni⁵⁹ above energies of 300 kev are shown in Figs. 2 and 3, respectively. In the energy range from 300 kev to 1300 kev, the detection efficiency of the crystal for electromagnetic radiation and the effect of instrumental change in resolution were determined empirically with the gamma rays of Hg²⁰³, I¹³¹, Au¹⁹⁸, Cs¹³⁷, Zn⁶⁵, and Co⁶⁰. The absolute activity of the Co⁶⁰ sample was known and served as a calibration standard. The other samples were standardized by comparison of their beta-ray spectra with that of Co⁶⁰ using absorption technique.

In order to account for the distortion introduced by the Compton scattering into the intensity distribution of the radiative electron capture spectra, the Compton distributions were measured for a number of gamma rays in the energy region of interest. In addition to some of the above mentioned standards, Be⁷ and Mn⁵⁴ gamma rays were utilized. An empirical correction was attained by dividing the radiative capture spectrum into a number of sections, each corresponding in width to the experimentally observed half-width of a gamma-



FIG. 3. Uncorrected scintillation spectrogram of the radiative electron capture spectrum in Ni⁵⁹. Only the energy region above 300 key was utilized for analysis of the spectral shape.

¹³ C. W. Johnstone (private communication).

¹² Singer, Kurbatov, and Kurbatov (to be published).

ray peak in the instrument. Starting with the highest energy section, the corresponding gamma-ray peak was then normalized to the intensity of that section of the radiative capture spectrum, and the contribution due to the Compton scattering subtracted out at the lower energies. This procedure was repeated always for the highest energy section which remained uncorrected, as shown in Fig. 4 for A³⁷, and in Fig. 5 for Ni⁵⁹. The resultant net intensity of the radiative capture spectrum was then subjected to further correction for crystal efficiency as outlined above.

IV. RESULTS

After the empirical corrections were applied to the radiative capture spectra, they were plotted in linear



FIG. 4. Correction procedure for the Compton effect applied to the radiative electron capture spectrum of A^{37} . The \times 's indicate experimental points of the spectrum. The dots show instrumental gamma-ray distributions which are superimposed on each other to approximate the continuous energy spectrum. The numbers next to the circles show the relative intensities of the gamma rays used in the analysis.

form according to the method of Jauch. The linear plot of Fe⁵⁵ is shown in Fig. 6. In this energy range, the distortion of the spectrum due to Compton effect is negligible with the crystal used in this work. The spectrum is corrected only for the change in resolution of the instrument. The end-point energy of the plot is 226 ± 10 kev. This is within experimental error of the recent nuclear reaction data of Trail and Johnson,¹⁴ which give an energy value of 230 ± 5 kev for this figures

The end-point energy for the spectrum of A^{37} it 815 ± 20 kev as shown in Fig. 7, which is in agreemen.



FIG. 5. Correction procedure for the Compton effect applied to the radiative electron capture spectrum of Ni⁵⁹. The \times 's indicate experimental points of the spectrum. The dots show instrumental gamma-ray distributions which are superimposed on each other to approximate the continuous energy spectrum. The numbers next to the circles give the relative intensities of the gamma rays used in the analysis.

with the previously published value.⁷ The plot follows a straight line from its end point to an energy of 320 kev, which is the lower limit to which the correction procedure was carried out in this investigation. The Ni⁵⁹ spectrum was analyzed in the same manner as the A³⁷ spectrum and the linear plot is shown in Fig. 8. The experimental end-point energy of this spectrum is 1065 ± 30 kev, which is in agreement with $Co^{59}(p,n)Ni^{59}$ threshold measurement.¹¹ The uncorrected spectrum of Ni⁵⁹, shown in Fig. 3, appears to be different in shape from that of A³⁷. This difference is neutralized almost completely by the application of the Compton correction procedure to both cases. In the corrected linear



FIG. 6. Linear plot of the radiative electron capture in Fe⁵⁵. The extrapolated end-point energy is 226 ± 10 kev.

¹⁴ C. C. Trail and C. H. Johnson, Phys. Rev. 91, 474 (1953).



FIG. 7. Linear plot of the fully corrected radiative electron capture spectrum in A^{37} . The extrapolated end-point energy is 815 ± 20 kev.

graph of Fig. 8, the points follow a straight line within experimental limits down to an energy of 320 kev. An excess counting rate near the end point is believed to be due to the finite energy resolution of the instrument.

v. CONCLUSIONS

The end-point energies of the radiative capture spectra of the three examples studied in this work were found to be in agreement with the energies calculated from the nuclear reaction data. The method can be used, therefore, to determine the transition energy involved in the electron capture disintegration directly by means of nuclear spectroscopy. The experimental spectral shapes agree with the predicted intensity distributions for allowed transitions at least in the region between the end-point energy and 40 percent of that value. For Fe⁵⁵ and A³⁷, the agreement is expected to exist since the transitions are probably allowed. In Ni⁵⁹, it appears from the transition energy and half-life that a second forbidden transition may be involved in the observed decay. A spin change of two is inferred from the measured spin 7/2 of Co⁵⁹, and a



FIG. 8. Linear plot of the fully corrected radiative electron capture spectrum in Ni⁵⁹. The extrapolated end-point energy is 1065 ± 30 kev.

spin 3/2 of the 31st neutron in Ni⁵⁹ as expected from the shell model theory.¹⁵

The radiative capture spectrum for this transition in the range above 320 kev agrees within experimental limits with the shape predicted for allowed transitions. Small variations in the spectral shape may have been lost in the application of the necessary correction technique, but the spectrum does not seem to be fundamentally different in this energy region. Below 320 kev, considerable differences were observed between the uncorrected spectra of A^{37} and Ni⁵⁹. Since no correction could be applied in this energy region, an actual difference, if it exists, remained undetermined.

The transition energy of Ni⁵⁹ exceeds $2mc^2$, so that positron emission is energetically possible. A search for annihilation radiation in the Ni⁵⁹ spectrum yielded negative results. The statistical variations were such that a branching ratio for positron decay greater than 2×10^{-5} could have been detected.

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¹⁵ M. G. Mayer, Phys. Rev. 78, 16 (1950).