An Upper Limit for Atomic Orbital Electron Ejection Accompanying K-Capture in Fe^{55}†

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An upper limit has been determined for the number of atomic orbital electrons ejected during the decay of Fe^{55} by orbital capture. From cloud-chamber observations it is concluded that less than 0.6×10^{-6} electron are ejected per disintegration with energies in the range 30 kev to 205 kev. The theoretical prediction gives the order of 10^{-6} electron per disintegration for the frequency of the process in the energy range studied.

I. INTRODUCTION

THE fact that a continuous photon spectrum accompanies nuclear beta-decay and orbital electron capture has been verified[1–5] and is in good agreement with the theory.[6–8] In addition, Bruner[9] has observed electrons in a magnetic spectrometer accompanying the decay of Sc^{44}, a positron emitter. Novey[10] has observed weak x-radiation associated with the decay of the supposedly pure negatron emitter RaE. These last two observations suggest the possibility that atomic orbital electrons may be excited and even ejected from an atom during the processes of nuclear beta-decay and orbital electron capture. A theoretical treatment of the problem of atomic electron excitation and ejection accompanying orbital electron capture appears in this issue of *The Physical Review*.[11] In the present paper experiments will be described which place an upper limit on the number of atomic orbital electrons ejected with energies greater than 30 kev accompanying orbital electron capture in $_{26}Fe^{55}$.

II. SOURCE MATERIAL

It is well established[12] that Fe^{55} decays only by orbital electron capture. The source material used in the present investigations was produced by deuteron bombardment of 100 percent abundant Mn^{55} as the oxide. The target material of high chemical purity was obtained through the courtesy of the Mallinckrodt Chemical Company. In particular, it was specified that iron was present in less than one part in 10^5 by weight. Our own chemical analyses verified this figure. The cyclotron target backing was aluminum. Iron content of the backing was barely detectable chemically and was much lower than other copper and brass backings available. The aluminum backing was washed repeatedly in HCl after machining to remove all traces of iron. These precautions were taken to assure a minimum of Fe^{59} (a negatron emitter) from an $Fe^{58}(d,p)Fe^{59}$ reaction. If only Mn^{55} were present in the target, the reactions $Mn^{55}(d,p)Mn^{56}$ and $Mn^{55}(d,2n)Fe^{55}$ would be the only reactions to be considered.

The Mn^{55} was bombarded for 10 hours at 180 microamperes with the 10-Mev deuterons from the Washington University Cyclotron. The target was aged for three weeks to allow the Mn^{56} (2.6 hr) to decay. The MnO_2 target material was dissolved in warm 8N HCl; a few milligrams of cobalt hold-back carrier were added, and an extraction in di-isopropal ether carried out. The ether fraction was washed twice with warm 8N HCl and finally the iron fraction was back-extracted into H_2O.

Aluminum absorption curves of this carrier free iron fraction were obtained using a 3.5 mg/cm²-mica end-window counter. A straight line was obtained on the usual semilog plot out to 50 mg/cm² of Al; the slope or half-thickness in Al corresponded to the characteristic K_α x-ray of Mn following the K-capture in Fe. In order to have still a better verification of the activity, a small fraction of the material was studied in a proportional counter and compared to a known low specific activity source of Fe^{55}. With a resolution of 19 percent for the 5.9-kev Mn x-ray, the identification was established without doubt that the radiation from the sample was that of the Mn K_α x-ray.

Most of the sample was deposited on a source for the magnetic spectrometer[13] in order to observe any gross number of charged particle radiations. The spectrometer source strength was (measured in a way similar to that described below) approximately 10^7 disintegrations/sec. The spectrometer counter window had a 10-kev low energy cutoff for beta-particles. No negatrons or positrons could be detected above background.

III. CLOUD-CHAMBER OBSERVATIONS

A more sensitive means of determining the upper limit for orbital electron ejection was sought in the cloud chamber. A portion of the Fe^{55} source solution was deposited on a 2 mg/cm² mica backing, supported

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[4] D. Maeder and P. Preiswerk, Phys. Rev. **84**, 595 (1951).
[5] Anderson, Wheeler, and Watson, Phys. Rev. **87**, 668 (1952).
[6] J. K. Knipp and G. E. Uhlenbeck, Physica **3**, 425 (1936).
[7] F. Bloch, Phys. Rev. **50**, 472 (1936).
[8] P. Morrison and L. I. Schiff, Phys. Rev. **58**, 24 (1940).
[9] J. A. Bruner, Phys. Rev. **84**, 282 (1951).
[10] T. B. Novey, Phys. Rev. **86**, 619 (1952).
[11] H. Primakoff and F. T. Porter, Phys. Rev. **89**, 930 (1953). This article also gives reference to and discussion of the related problem in $\beta^{\mp}$ decay.
[12] National Bureau of Standards Circular No. 499 (1950) and supplements.
[13] F. T. Porter and C. S. Cook [Phys. Rev. **87**, 464 (1952)] mention recent modifications of the spectrometer.

by a thin brass frame, and inserted in a 25 cm diameter, automatically cycled, cloud chamber[14] of conventional design. With a field of 200 gauss, 1400 frames were taken and scanned by reprojection. The criteria for an acceptable event were that the track appear to originate in the source and that it have the curvature appropriate to an electron in the field. The apparent radii of curvature of all tracks were measured in addition to counting them. In the first 1400 pictures 15 electron tracks were observed. A second sequence of 1500 pictures produced 28 acceptable tracks. Of the 43 tracks, only 4 had curvatures indicating energies greater than 205 kev (the transition energy[3,4] of $Fe^{55} \rightarrow Mn^{55}$). No positrons were observed to originate in the source in all 2900 pictures.

In establishing the lower limit on the electron energy which could be successfully detected, it was noted that the two lowest energy tracks which were observed were apparently between 15–20 kev, but these were badly scattered. In addition, the range of a 30-kev electron in the gas of the chamber (hydrogen at 1.7 atmospheres saturated with ethanol) is calculated to be 5–6 cm. This would seem to give a good chance for detection even though the track would most probably be scattered.

The combination sensitive time solid angle factor for the cloud chamber has been determined by counting the number of electron tracks per picture from a bare source of Co^{60} in the source position of the chamber and with no magnetic field applied. An upper limit for the number of electrons emitted per second by this source has be endetermined by β-counting in known geometries, and the disintegration rate has been checked by comparison γ-counting with a standard Co^{60} source. The solid angle sensitive time factor is not less than 4.9×10^{-3} sec (the most probable value is 5.5×10^{-3} sec).

IV. DISINTEGRATION RATE OF THE SOURCE; RESULTS

The disintegration rate of the Fe^{55} cloud-chamber source was determined by obtaining absorption curves in aluminum of the Mn K_α radiation in a fixed geometry. The straight line semilog plots were extrapolated to zero absorber thickness, considering the 10 mg/cm² of air and 3.5 mg/cm² mica window of the counter to contribute roughly the same absorption as an equivalent thickness of aluminum. Two experimental arrangements were used. The first involved no collimation of the x-rays. The aluminum absorber foils were placed just in front of the counter window, a circular aperture 1.06 inches in diameter. The inside diameter of the counter is 1.12 inches with a sensitive length of approximately 4 cm. This arrangement is the usual one for weak sources of photons.[15]

Extrapolating to zero absorber and applying a solid angle correction involving simply the window diameter and the distance from the source to window (the area of the source is 0.25 cm²), the value 2.5×10^5 disintegrations/sec was obtained, uncorrected for counter efficiency and fluorescence yield. The second arrangement was the same except that a lead sheet $\frac{1}{8}$ inch thick with a $\frac{3}{8}$ inch hole was placed just below the absorbers to act as a collimator. The slopes of the absorption curves in both arrangements were identical (both giving a half-thickness in Al corresponding to the Mn K_α x-ray), but with collimation an extrapolation to zero absorber and correction for solid angle defined by the collimator gave 9.7×10^5 disintegrations/sec uncorrected for counter efficiency and fluorescence yield. The reason for the difference is apparently that in the first case the efficiency is a function of the region in which the x-rays enter the counter. With no collimation it seems probable that x-rays entering near the edge of the counter have less path length in the gas and less probability of causing an ionizing event which can trigger the Geiger tube. In the second case the x-rays all enter the counter relatively close to the central wire and have a chance to traverse the entire sensitive length of the counter. There appears to be no reason to reject the higher figure and some reason to question the lower one, so we take as the disintegration rate of the source 9.7×10^5 disintegrations/sec uncorrected for counter efficiency and fluorescence yield.

The counter efficiency is probably the most uncertain of all the data. The counter gas is predominantly argon; the gas pressure approximately 10 cm of Hg. A consideration of the mass absorption coefficient for 6-kev x-rays in argon at this pressure and of the sensitive length of the counter gives a figure of 25 percent efficiency for detection of these photons. Since we are seeking an upper limit for the number of electrons ejected per disintegration, it seems prudent to double this value in the light of its tenuous basis, so that we may be sure we do not overestimate the disintegration rate of the Fe^{55} source. Further, less than a third[16] of the K-capture events are followed by a K_α x-ray of

TABLE I. Number of electron tracks originating in the Fe^{55} source in the indicated energy range as determined from cloud-chamber observations. The four tracks with energies greater than the available energy ($Fe^{55} \rightarrow Mn^{55}$) are also included.

Electron energy kev	Number of tracks
15–30	6
30–60	13
60–90	8
90–120	6
120–150	4
150–180	3
180–210	3
320	2
500	1
575	1

[14] K. H. Morganstern and K. P. Wolf, Phys. Rev. **76**, 1261 (1949).

[15] See, for example, L. E. Glendenin, Nucleonics **2**, No. 1, 29 (1947).

[16] Steffen, Huber, and Humbel, Helv. Phys. Acta **22**, 167 (1949).

Mn; the others result in Auger electrons of energies well below the low energy cutoff of the counter window. Consequently, the observed decay rate is increased by a factor 3 to correct for fluorescence yield. Corrections for L capture and for the small "dead space" between the window and the sensitive region of the end-window counter are not made. This also is consistent with the setting of an upper limit for the process. The disintegration rate of the Fe^{55} source is, then, corrected for 50 percent efficiency and for fluorescence yield, 5.8×10^{6} disintegrations/sec.

Finally, taking into account the statistical error in the small number of tracks observed, we conclude there are less than 0.6×10^{-6} ejected electron per disintegration with energies greater than 30 kev.

V. DISCUSSION

The origin of the electrons observed may well be questioned. No doubt some of them are nuclear beta-particles from the decay of impurities in the source. The most likely impurity is Fe^{59}. The currently available information indicates that Fe^{59} has two negatron groups of approximately the same abundance with end points at 260 kev and 460 kev. If a majority of the tracks observed here were to be attributed to an Fe^{59} impurity, the absence of more tracks with energies greater than 210 kev would be difficult to explain. The fact that only 4 tracks corresponded to electrons with energies greater than 210 kev is at least suggestive that a major portion of the observed tracks actually are due to atomic electrons ejected during the decay of Fe^{55} by orbital capture.[17] In any case all tracks below 210 kev are included in setting the upper limit on the frequency of the process.

The theoretical expression[11] for the number of ejected noncaptured K electrons per K-capture was numerically integrated, with $Z_i=26$, from 30 kev to (205–6.5 kev). The result is 3.6×10^{-6} electron/disintegration. The numerical factor (3.6) is sensitive to the value of the lower limit of the integration; thus, if a lower limit of 50 kev is used, the theoretical prediction is reduced to 1.0×10^{-6} electrons/disintegration. We have therefore considered the question as to whether we have actually observed most of the electrons emerging from the source with energies between, say, 30 kev and 50 kev. To what extent multiple scattering and self-absorption in the source may discriminate against observation of the lower energy electrons is a question which does not have a clear quantitative answer. Certainly there is no sharply defined energy below which one cannot observe a track and above which one can observe all the tracks. We seek a lower energy limit above which there is a reasonable chance to observe, say, 90 percent of the tracks in the given solid angle. As was mentioned before, calculations indicate that a 30-kev electron has a mean range of approximately 5 to 6 cm in the cloud chamber and, with a field of 200 gauss, a radius of curvature of approximately 3 cm. Further, it can be said that a majority of the tracks in the 40–60 kev range did not appear to be badly scattered and that the apparent curvature measurements were made with errors of the order of 20 percent. If, on the other hand, one believes that most of the observed events are the result of orbital electron ejection and also believes that the theoretical predictions concerning the momentum distribution are essentially correct, then it would appear that there is considerable discrimination against the observation of the lower energy events.

Actually the point to be emphasized is that the experimental upper limit for the process does not exceed the theoretical prediction. That the upper limit given here could be increased by a factor of 2 seems doubtful; that the theory is refined enough to predict the total number of ejected electrons in the energy range studied within a factor of 2 or 3 is not claimed. Unfortunately, the cloud chamber is not a reliable instrument with which to investigate the momentum distribution of electrons in this energy range even if it had been feasible to obtain enough events to make such a study statistically acceptable, so that nothing can really be said about the the question of the momentum distribution of the ejected electrons from the above observations.

Finally, it is interesting to note that apparently in the case of orbital electron capture in Fe^{55} the number of ejected electrons per disintegration with energies greater than 30 kev is considerably smaller than in the case of positron decay of Sc^{44} (0.04 electron/disintegration).[9] It has been pointed out[11] that even taking into account certain differences in the treatment of K-capture and $\beta^{\mp}$ decay, it is difficult to account theoretically for the large number of electrons observed[9] accompanying the decay of Sc^{44}.

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[17] Other possibilities for the origin of the observed electrons may be considered. First of all there is no evidence available that any orbital electron capture transitions in Fe^{55} go to excited states in Mn^{55}; no nuclear gamma-rays or conversion electrons have been reported. If the electrons detected in the present observations result from Compton or photoelectric interaction in the source and surrounding material of a nuclear gamma-ray, the intensity of this gamma-ray would have to be of such a magnitude as to make its prior detection most likely. On the other hand, if the detected electrons arise from a weak highly converted transition, the branching ratio of this transition cannot be greater than $\approx10^{-6}$, and, of course, the electron spectrum is monoenergetic. Further, since the continuous inner bremsstrahlung photon spectrum has an intensity of the order of 10^{-5} photon/disintegration and since the effective source thickness is certainly less than 5 mg/cm^2, the observed electrons are most probably not the result of Compton or photoelectric interaction of the inner bremsstrahlung photons. *Note added in proof:*—Hausman *et al.*, Phys. Rev. **88**, 1297 (1952) report the existence of 15 excited states in Mn^{55}. Only the lowest of these, 130 kev above the ground state, is energetically attainable by the K-capture of Fe^{55}.