Disintegration Schemes of Radioactive Substances. VII. Mn^{54} and Co^{58}*

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The radiations emitted by Mn^{54} and by Co^{58} have been studied by means of the magnetic lens spectrometer and coincidence techniques. Mn^{54} decays by orbital electron capture to an excited state of Cr^{54}, followed by the emission of a 0.835-Mev gamma-ray. A study of the x-rays emitted following the capture process indicates that a large—or even an overwhelming—fraction of the captured electrons are K electrons. Few if any capture transitions lead directly to the ground state. Co^{58} decays to a state of Fe^{58} 0.805 Mev above the ground state which in turn decays by the emission of a single gamma-ray. About 90 percent of the disintegrations of Co^{58} occur by K-electron capture. In the remaining 10 percent positrons of maximum energy 0.47 Mev are emitted. This ratio of the two modes of decay is consistent with the idea that the transition takes place with a change of angular momentum of one unit or zero, whether the parity changes in the transition or not, if the tensor theory of beta-decay is the correct one. The lowest known excited states of Cr^{54}, Fe^{56}, and Fe^{58} have excitation energies differing by less than five percent. The difference between the masses of the neutral atoms of Co^{58} and Fe^{58} should be $2.46\pm0.03\times10^{-3}$ a.m.u.

INTRODUCTION

IN contrast to the very complex radiations described in paper VI,[1] this paper is concerned with two nuclear species whose disintegration schemes are quite simple. Inspection of the isotope chart (Fig. 1) discloses that both Mn^{54} and Co^{58} have two stable isobaric neighbors and could thus decay either by increasing or by decreasing the nuclear charge. However, both decay only by the change of a proton into a neutron, i.e., by positron emission or orbital electron capture, with relatively long half-lives. The product nuclei Cr^{54} and Fe^{58}, respectively, both belong to the stable sequence of nuclear type $(4n+2)$ with isotopic number 6 which extends from Ca^{46} to Kr^{78}. Both disintegration schemes were studied by the magnetic spectrometer and coincidence techniques described in the preceding papers of this series.[1]

Mn^{54}

The radioactivity of manganese with about 500-day half-life was definitely assigned to the species of mass number 54 by Livingood and Seaborg.[2] These same authors also reported that this activity emits no charged particles of nuclear origin but does emit gamma-rays of about 0.85-Mev energy as shown by absorption measurements. Alvarez[3] also demonstrated the emission of chromium K x-rays.

Sources of Mn^{54} used in our experiments were prepared from iron bombarded with deuterons. The iron was removed by precipitation with pyridine (paper IV), cobalt by precipitation with KNO_2 (paper VI), and finally the manganese was precipitated as the oxide in nitric acid solution. The sources were allowed to age for about two months because of the presence of the 6.5-day activity of Mn^{52}.

Under the bombarding conditions of the M.I.T. cyclotron at the time when these experiments were performed, i.e., with a deuteron energy of about 11 Mev and thick targets, the great majority of all the gamma-rays emitted by the long-lived activities induced in the iron target are contributed by the soft gamma-rays of Co^{57}. Since the cross section for production of photoelectrons by gamma-rays of these energies (0.119 and 0.131 Mev) is very large, even a slight residual contamination of radiocobalt will be detected when photoelectron spectra are studied in the spectrometer. It will be recalled that such a contamination was found in sources of Fe^{59} prepared from the same targets as the Mn^{54} (paper IV). Deutsch and Roberts[4] reported a 0.850-Mev and a 0.120-Mev gamma-ray from

* Some of these results have been presented at the New York Meeting of the American Physical Society, January 22–23, 1943.

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[1] L. G. Elliott and M. Deutsch, Phys. Rev. **64**, 321 (1943). The previous papers of this series are listed under reference 1 of paper VI, and are referred to here as papers I to VI.

[2] J. J. Livingood and G. T. Seaborg, Phys. Rev. **54**, 391 (1938).

[3] L. Alvarez, Phys. Rev. **54**, 486 (1938).

[4] M. Deutsch and A. Roberts, Phys. Rev. **60**, 362 (1941).

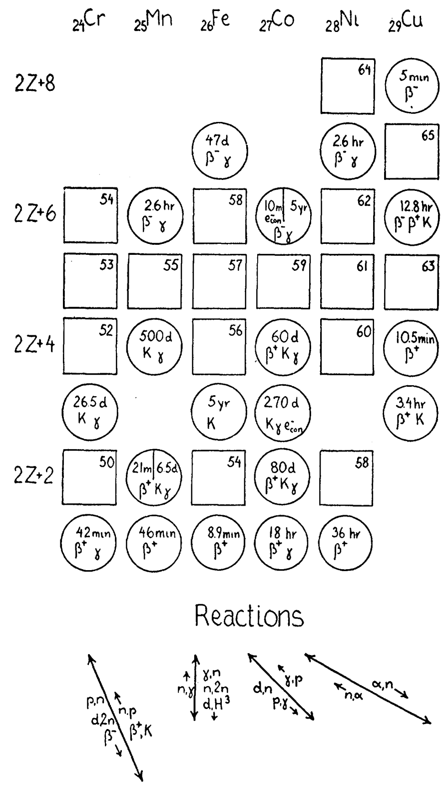

FIG. 1. The known nuclear species of the elements between $Z=24$ and $Z=29$. Squares indicate stable isotopes, circles radioactive isotopes.

Mn^{54}. Because of our experience with Fe^{59}, we re-examined the low energy region, using a source of Mn^{54} which had been freed of cobalt several times, adding inactive carrier material in each purification. No gamma-rays other than the 0.85-Mev radiation were found. Therefore it seems certain that the low energy gamma-ray reported previously[4] was due to a slight contamination by Co^{57}.

The energy of the 0.85-Mev gamma-ray was also redetermined with greater accuracy and is now found to be 0.835 ± 0.015 Mev. (Compare Fig. 9 of paper VI.[1]) In agreement with other observers, we failed to find any charged particles emitted by thin sources of Mn^{54}.

Since Mn^{54} decays only by orbital electron capture, it remained to be shown whether the decay always leaves the Cr^{54} nucleus in an excited state or whether it leads sometimes directly to the ground state. Therefore coincidences between K x-rays and gamma-rays were studied. Our standard "bell" type beta-ray counters, equipped with mica windows and filled with helium at atmospheric pressure (paper II), are quite unsuited for the detection of x-rays of this wave-length. Several of these counters were therefore equipped with thin Cellophane or beryllium windows and filled with pure tank argon at a pressure of about 60 cm Hg. About half of the rays entering these counters should be absorbed in the filling gas and therefore be counted.

Such an x-ray counter was placed in the standard coincidence arrangement (paper V). The observed number of x-gamma-coincidences per recorded x-ray was 1.25 ± 0.04, in good agreement with the known efficiency of the gamma-ray counter for a single gamma-ray of energy 0.835 Mev. (The efficiency of the gamma-ray counter used in this experiment was slightly larger than that of the counter described in paper V). No gamma-gamma coincidences were found. It follows, then, that each K x-ray of chromium emitted following the K-electron capture process is accompanied by a single gamma-ray of energy 0.835 Mev. This scheme is shown in Fig. 2. The disintegration energy is not known, but because of the complete absence of positron emission, it seems reasonable to assume that the energy difference between initial and final states of the disintegration is less than or, at most, very close to mc^2. The particular value chosen in Fig. 2, namely, 0.1 Mev, was arrived at by assuming both Mn^{54} and Co^{58} to perform allowed transitions with the same matrix element.

Marshak[5] has raised the question whether L-electron capture might not occur with a fre-

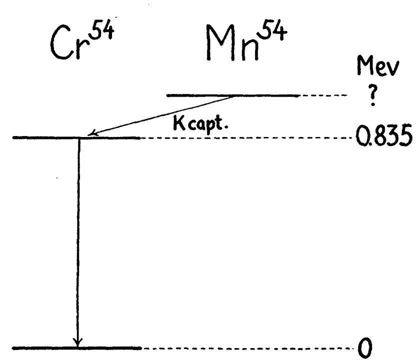

FIG. 2. Disintegration scheme of Mn^{54}.

[5] R. E. Marshak, Phys. Rev. 61, 431 (1942).

quency comparable to that of K-electron capture in the case of Mn^{54}. It seems quite certain, for theoretical reasons, that abundant L-electron capture, if it occurs at all, must also lead to the excited state of Cr^{54}. If the net detection efficiency of the x-ray counter were known as well as the efficiency of the gamma-ray counter, it should be possible to compare directly the number of gamma-rays and the number of K x-rays emitted by noting the corresponding counting rates. A comparison of the observed number of x-rays with the number from a source of Co^{58}, taking into account the variation of the absorption of the two different radiations in the counter gas and in the beryllium window and the different fluorescent yield of iron and chromium, all of which enter into the net detection efficiency, leads to the conclusion that our observations are consistent with the assumption that Mn^{54} decays overwhelmingly by K-electron capture. The experiment is crude, at best, because of the large and uncertain absorption of the x-rays in the source material itself.

Co^{58}

Livingood and Seaborg[6] showed that Co^{58} decays by positron emission to Fe^{58}. They reported the maximum energies of the beta- and gamma-rays, found by absorption methods to be 0.5 Mev and 0.8 Mev, respectively. The assignment of this activity to Co^{58} is definite because it is produced by alpha-particle bombardment of manganese. This distinguishes it from Co^{56} which has a very similar half-life (compare paper VI).[1]

The results of Jensen,[7] which contradict these findings as well as ours, were probably due either to a contamination or to the well-known unreliability of cloud-chamber data.

We have produced Co^{58} by an (n, p) reaction on nickel. About 5 g of nickel metal were fixed to the back of a beryllium target which was bombarded by about 10-milliampere-hours of 14-Mev deuterons. The chemical separations followed the same lines as those described in paper VI[1] for the preparation of sources of Co^{56}. Inspection of Fig. 1 shows that the only known

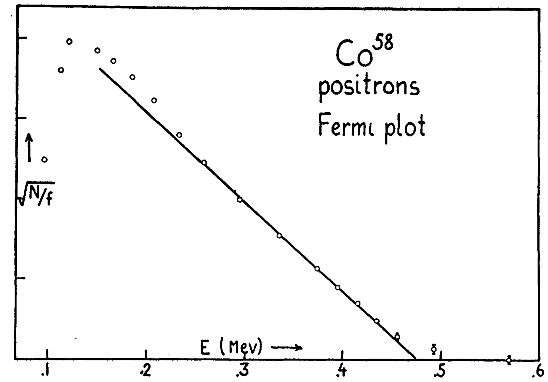

FIG. 3. Fermi plot of the positron spectrum of Co^{58}. The Fermi function was evaluated for $Z=26$.

radioactive species of cobalt expected from fast neutron bombardment of nickel are Co^{58} and Co^{60}. The radiations accompanying the decay of the latter species are well known[8] and consist of soft negatrons and hard gamma-rays. If equal cross sections for the two (n, p) reactions are assumed, a consideration of the relative isotope abundance and half-lives indicates that one should observe about one hundred times as many disintegrations of Co^{58} as of Co^{60}. In fact the number of negatrons was found to be too small to be reliably detected, in the presence of the secondary electron due to the Co^{58} gamma-rays.[9] A small amount of Co^{57} might be found as the decay product of Ni^{57}, produced by an $(n, 2n)$ reaction. However, the intensity of the soft gamma-rays and conversion electrons emitted by this activity, if present at all, was too small to be detected.

Figure 3 shows a Fermi plot of the positron spectrum emitted by Co^{58}. The specific activity of the active material was not sufficient to permit the preparation of a source which could be considered thin for electrons of so low an energy. The shape of the Fermi plot is therefore probably distorted by scattering in the source. Extrapolation of the straight line portion of the plot yields the value 0.470 ± 0.015 Mev for the maximum energy. The apparent tailing off beyond the end point is due to the poor resolution

[6] J. J. Livingood and G. T. Seaborg, Phys. Rev. **60**, 913 (1941).
[7] A. S. Jensen, Phys. Rev. **60**, 430 (1941).
[8] M. Deutsch and L. G. Elliott, Phys. Rev. **62**, 558 (1942).
[9] The magnetic lens spectrometer ordinarily does not distinguish between positrons and negatrons. A special baffle system is provided for this purpose.

necessary because of the low intensity available. The value of the maximum energy is in good agreement with that found with thinner sources containing a mixture of Co^{56} and Co^{58} (paper VI).

Figure 4 shows the secondary electron spectrum produced in a lead radiator (35 mg/cm²) by the gamma-rays of Co^{58}. Two very pronounced photoelectron lines are found, corresponding to gamma-ray energies of 0.507±0.008 Mev and 0.805±0.012 Mev, respectively. The L-photoelectron group, due to the same gamma-rays, also appears as much smaller peaks. The lower energy line is certainly due to annihilation radiation. The small number of counts at energies above 0.8 Mev is probably due to the Co^{60} gamma-rays of 1.10- and 1.30-Mev energy.[8]

It is apparent from Fig. 4 that the 0.805-Mev radiation is considerably more intense than the 0.51-Mev gamma-ray. When account is taken of the variation of the photoelectric cross section with gamma-ray energy and of the effect of finite radiator thickness on the height of the photoelectron lines, it is found that the 0.805-Mev gamma-ray is 5±2 times as intense as the annihilation radiation. The relatively large uncertainty in this ratio is due to the fact that the effective source of annihilation radiation is slightly larger than the dimensions of the source material because of the finite range of the positrons. Since *two* annihilation quanta are emitted for each positron that is annihilated, we conclude that there are 10±4 times as many 0.805-Mev quanta emitted as positrons.

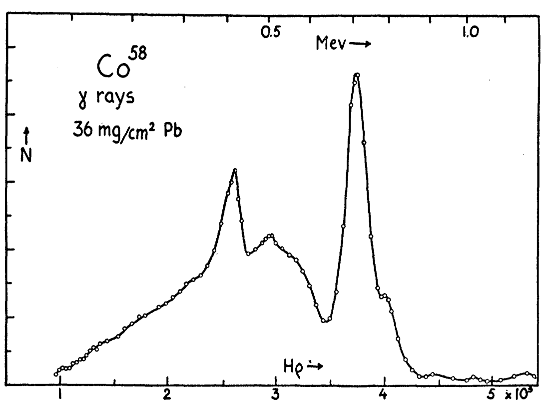

FIG. 4. Secondary electrons due to gamma-rays of Co^{58}. The peaks are photoelectrons ejected from the lead foil. The continuous distribution is due to Compton recoil electrons.

Coincidences between positrons and gamma-rays were observed in the spectrometer since the great abundance of gamma-rays and the presence of negatrons due to Co^{60} interfered with accurate measurements in the standard counter arrangement (paper V). It was found that the coincidence rate per recorded positron was independent of positron energy. One may conclude from this that all disintegrations proceed to the same excited state.

The observed coincidence rate for the positrons of Co^{58} was in good agreement with the efficiency of the gamma-ray for a 0.8-Mev gamma-ray in the geometry used as determined by observing coincidences with the high energy beta-ray spectrum of Mn^{56} (paper VI).

We may conclude that each positron emitted by Co^{58} is accompanied by one 0.805-Mev gamma-ray.

In order to study coincidences between iron K x-rays which are emitted following the K-electron capture process and gamma-rays, the beta-ray counter of the standard arrangement was replaced by an argon-filled x-ray counter as described above in the discussion of Mn^{54}. When all of the positrons were absorbed in beryllium, a true coincidence rate of $1.23 \pm 0.04 \times 10^{-3}$ per recorded x-ray was observed in the standard arrangement. This is in good agreement with the expected efficiency for detecting a 0.8-Mev gamma-ray which is $1.19 \pm 0.05 \times 10^{-3}$. One may conclude that the orbital electron capture process, too, always leaves the Fe^{58} nucleus in the excited state. This is, of course, to be expected since no high energy positrons are observed.

The observations of Co^{58} are summarized in the disintegration scheme shown in Fig. 5.

From this disintegration scheme the mass difference between the neutral atoms of Co^{58} and Fe^{58} is found to be $2.46 \pm 0.03 \times 10^{-3}$ a.m.u. The threshold for the reaction Fe^{58} (p, n) Co^{58} should be 3.10±0.03 Mev.

DISCUSSION

It appears that the determination of the relative probabilities of positron emission and orbital electron capture by Co^{58} represents the first case in which the ratio of the two processes has been found with sufficient accuracy to permit a close quantitative comparison with theory.

The various formulations of the Fermi theory[5] all lead to the same ratio of the probabilities in the case of allowed transitions, $\lambda_+/\lambda_K \simeq 0.1$ for Co^{58} in good agreement with the observed value. With the tensor interaction which is most likely to be the correct one, the calculation of the ratio for forbidden transitions is complicated by the occurrence of several matrix elements of unknown ratio. The observed ratio of about 1 : 10 is consistent with a special case of first forbidden transition with a change in nuclear angular momentum of one unit or without change in angular momentum. In particular, the tensor theory predicts the observed value of the ratio if the angular momenta of the initial and final states are 1 and 0 (or vice versa), whether the parity changes or not. For other values of the angular momenta (involving a change of one or zero) the ratio of the transition probabilities may still agree with the observed value if the dominant matrix element is $\int \alpha$ [Eq. (17a), reference 5], which is not unreasonable in this particular case. For larger changes in angular momentum, the tensor theory predicts a smaller relative probability of positron emission at least in some cases. For example, the calculations of Marshak[5] lead to a ratio $\lambda_+/\lambda_K \simeq 0.01$ for an angular momentum change of 2 units and parity change. It may then be said that on the tensor theory the decay of Co^{58} involves an angular momentum change of zero or one, with or without parity change.

If this interpretation of the relative probabilities of the modes of decay is correct, the shape of the positron spectrum of Co^{58} should be that predicted for an allowed transition. It seems difficult, although not impossible, to obtain sources of sufficient specific activity to verify this point.

The similarity between the modes of decay of Mn^{54} and Co^{58} is rather striking: In both cases the transformation of a proton leaves the product nucleus in a state with about 0.8-Mev excitation energy which is then given up in a single gamma-ray. It may be noted that the lowest known excited state of Fe^{56} also has an excitation energy of about 0.8 Mev (paper VI). The excitation energies of the three nuclei are Cr^{54} : 0.835 Mev, Fe^{56} : 0.845 Mev, Fe^{58} : 0.805 Mev (compare Fig. 9 of paper VI). The differences between these energies are known to about ±0.005 Mev even though the absolute values may be in error by three times this amount.[10]

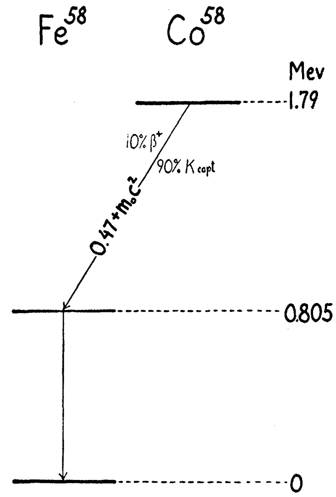

FIG. 5. Disintegration scheme of Co^{58}.

It may be recalled that Rosenblum and collaborators[11] found such similarities of energy level spacings in the case of several natural radioactivities. Another striking similarity exists in the case of Co^{59} (paper IV) and Ni^{60}.[8] The very similar isomeric states of the two stable species of silver,[12] the isomeric states of Br^{78}, of Br^{80}, and of Kr^{83},[12,13] the gamma-ray energies of about 0.05 Mev found in the 19-minute gallium,[13] and the 100-day arsenic[14] activities may be cited as further examples. Sufficient data are not yet available to decide whether these similarities between excitation energies are significant or fortuitous. A study of excited states of isobaric nuclei may shed further light on this question.

The authors wish to express their gratitude to Professor R. D. Evans for his continued interest, to Professor M. S. Livingston and the M.I.T. cyclotron crew for the bombardments, and to Professor J. W. Irvine, Jr., for help with the chemical procedures.

[10] B. R. Curtis, Phys. Rev. **55**, 1136 (1939), reported a gamma-ray of about 0.8 Mev from Co^{55}. A preliminary study of this activity in the spectrometer showed that the gamma-ray energy nearest to this value is 0.915 Mev.

[11] S. Rosenblum and M. Guillot, Comptes rendus **204**, 975 (1937).

[12] A. C. Helmholtz, Phys. Rev. **60**, 415 (1941).

[13] G. E. Valley and R. L. McCreary, Phys. Rev. **56**, 863 (1929).

[14] L. G. Elliott and M. Deutsch, Phys. Rev. **63**, 457 (1943).