partial waves of angular momentum of at least two units of $\hbar$.

No attempt has been made to compare the observed cross section with single level resonance theory because a sufficiently high bombarding energy was not achieved for good comparison and also because the level widths and level shifts have not been developed for three-body breakup.[15]

[15] E. P. Wigner and L. Eisenbud, Phys. Rev. 72, 29 (1947).

From Fig. 3 the cross section for the reactions (1) and (2) at 200 kev is at least 2.5 microbarns. Fowler and Lauritsen[16] found that for an assumed cross section of 4 microbarns, the $He^3(He^3,2p)He^4$ reaction is the dominant reaction terminating the $p-p$ cycle in stellar production of energy. The essential correctness of their assumption seems established.

[16] W. A. Fowler (private communication).

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The Disintegration of Mo^{99}†

JAGDISH VARMA* AND C. E. MANDEVILLE
Bartol Research Foundation of the Franklin Institute, Swarthmore, Pennsylvania
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The radiations of Mo^{99} and Tc^{99m} have been reinvestigated by means of scintillation spectroscopy. Employing single-channel pulse-height analyzers in coincidence, it has been established that a triple cascade of gamma rays, 741 kev→41 kev→140 kev occurs. The 741-kev radiation is also coincident with a gamma ray at 181 kev by an alternate branch of de-excitation which is equally probable. Radiation at 780 kev is present but is non-coincident with other gamma rays. From these data, a disintegration scheme can be constructed. A gamma ray of energy 372 kev was also detected, and, although it appeared to have the proper half-period to be associated with Mo^{99}, was found to be not in immediate coincidence with beta rays or other gamma rays, suggesting that it might be related to an impurity.

INTRODUCTION

EARLIER coincidence studies[1] showed the disintegration scheme of Mo^{99} to be rather complex. Subsequent measurements[2–6] have led to the conclusion that Mo^{99} decays with the emission of two, or possibly three, groups of beta rays, and gamma rays having energies of 1.8, 40, 140, 142, 181, 367, 741, and 780 kev. A careful study[5,6] of the disintegration of the 6-hour isomer of Tc^{99} has revealed that it decays with cascade emission of the 1.8-kev and 140-kev quanta, and that the 142-kev gamma ray is the associated cross-over transition. The disintegration scheme of Mo^{99}, as advanced by Medicus *et al.*,[5] is based upon coincidence measurements between spectrometrically selected beta rays and gamma rays detected in an anthracene scintillation counter. However, the problem of the precise location in the scheme of the 181- and 367-kev gamma rays has remained unresolved. Accordingly, with the utilization of two single-channel pulse-height analyzers in coincidence, the gamma-ray spectrum and the various cascade relationships have been reinvestigated.

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* Research Fellow. Permanent address: Morena (M.B.) India.

[1] C. E. Mandeville and M. V. Scherb, Phys. Rev. 73, 848 (1948).

[2] Medicus, Maeder, and Schneider, Helv. Phys. Acta 22, 603 (1949).

[3] Cork, Keller, and Stoddard, Phys. Rev. 76, 986 (1949).

[4] M. E. Bunker and R. Canada, Phys. Rev. 80, 951 (1950).

[5] Medicus, Maeder, and Schneider, Helv. Phys. Acta 24, 72 (1951); Phys. Rev. 81, 652 (1951).

[6] Mihelich, Goldhaber, and Wilson, Phys. Rev. 82, 972 (1951).

THE MEASUREMENTS

For the purposes of the present investigations, a source of Mo^{99} was obtained when a quantity of MoO_3 was irradiated by slow neutrons in the Oak Ridge pile. Because no gamma rays other than those already reported were observed, extensive chemical purification was deemed unnecessary. On occasion, however, Tc^{99m} was separated from its parent element by the method of Coryell and Sugarman.[7]

In Fig. 1 is shown the pulse-height distribution generated by the gamma rays of Mo^{99} in a crystal of thallium-activated sodium iodide which is three centimeters thick. In the case of the particular curve of Fig. 1, the radiation incident upon the detecting crystal had been filtered by a lead absorber of thickness about 2 g/cm^2 to reduce in intensity the 140–142 kev radiation relative to the harder gamma rays. The radiation at 78 kev arises from the emission from the absorber of the K line of lead, following photoelectric absorption of the intense 140-kev gamma ray. For the purpose of observing carefully the region of lower

[7] L. E. Glendenin in *Radiochemical Studies: The Fission Products*, edited by C. D. Coryell and N. Sugarman (McGraw-Hill Book Company, New York, 1951), Paper No. 98, National Nuclear Energy Series, Plutonium Project Record, Vol. 9, Div. IV, Part 5, Book 2.

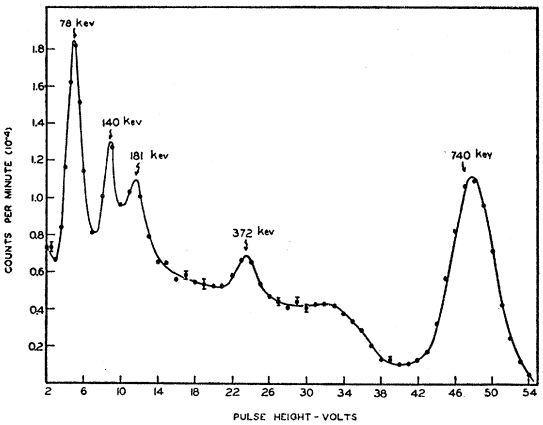

FIG. 1. Pulse-height distribution from NaI(Tl) irradiated by gamma rays emitted in the disintegration of Mo^{99} in equilibrium with Tc^{99m}. The radiations have been filtered by lead (2 g/cm^2) to reduce the intensity of the 140-kev radiation.

energies and to detect the 41-kev radiation, without lead absorber and without interference from the K line, an additional curve, not illustrated in this paper, was obtained. In this case, to reduce the intensity of the 140-kev line, approximately two-thirds of the 6-hour Tc activity was removed from the molybdenum by chemical separation. Unless the intensity of the 140-kev line is reduced somewhat, a source of Mo^{99} strong enough to give a clear picture of the spectral region below 140 kev will at the same time give rise to a piling up of 140-kev pulses at the linear amplifier, distorting the spectral region above 140 kev. From the pulse-height spectrogram of Fig. 1, it is clear that only those gamma-rays which have been previously reported are present in the sample of Mo^{99} under study. These gamma rays were observed many times during twelve half-periods of decay and were found to decay with the same half-period.

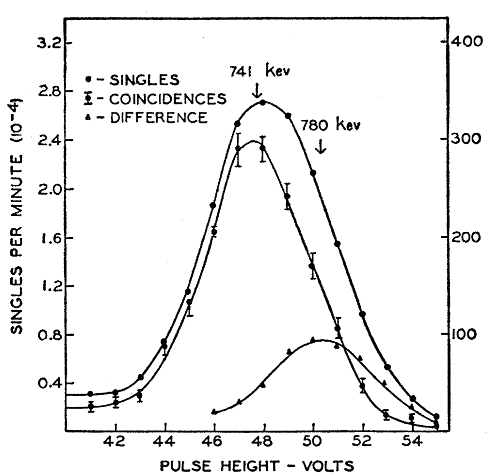

FIG. 2. Coincidence study of the 741-kev–140-kev cascade. The data show that the 780-kev line is noncoincident with any 140-kev radiation.

To study the disintegration scheme of Mo^{99}, gamma-gamma coincidence rates were measured between the two members of all possible pairs of gamma rays.

To ascertain the relation between the 140-kev radiation and the harder gamma rays, the data of the curves of Fig. 2 were collected. The single counting rate in the vicinity of 760 kev is plotted along with a coincidence rate which was obtained with two pulse-height analyzers in coincidence by setting one analyzer at the photopeak of the 140-kev radiation and moving the window of the second through the region of 760 kev. On comparing the half-widths and locations in the energy of the two two peaks, it is seen that the 140-kev radiation is coincident only with the 741-kev line. The difference curve, also shown in Fig. 2, is peaked at 780 kev, showing that the 780-kev radiation is noncoincident with the 140-kev line. A similar curve is shown in Fig. 3 in which coincidences have been recorded between the 741-kev gamma ray and the 181-kev line. A third set of curves, not shown as a figure, was obtained

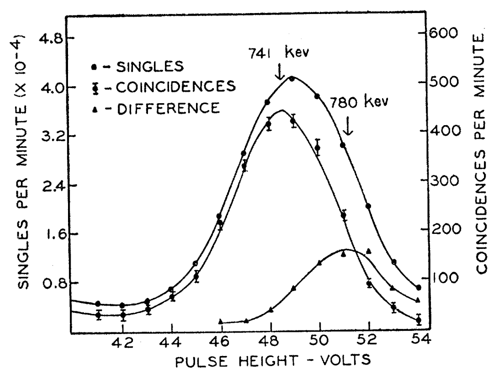

FIG. 3. Coincidence study of the 741-kev–181-kev cascade. The data show that the 780-kev line is noncoincident with any 181-kev radiation.

relative to coincidences between the 41-kev gamma ray and the hard radiations. Again it was found that coincidences were present only between the 741-kev radiation and the softer quantum. To avoid difficulties growing from the presence of the intense 140-kev radiation associated with the decay of the 6-hr metastable level, a partial separation of Tc from Mo was performed before obtaining the data of the curves of Figs. 2 and 3. The soft gamma ray at 41 kev was also found to be coincident with the 140-kev quantum. In performing coincidence measurements between the gamma rays of energies 140 and 41 kev, care was taken that the backscattered quanta from one crystal did not enter the other crystal. A proper choice of absorbers placed between the two crystals eliminated any back scattering effects.

No coincidences were detected between the 372-kev gamma ray and any other gamma rays in the spectrum. A search was also carried out to find two 372-kev quanta in coincidence, but none were found. The resolving time of the coincidence circuit was 10^{-7} sec. Absence of coincidences may, therefore, be interpreted as evidence that the 372-kev transition terminates at a metastable level of lifetime long compared with 10^{-7} sec or that the 372-kev line arises from the presence of a radioactive impurity.‡

The foregoing coincidence data are consistent with the interpretation that the 780-kev gamma ray terminates at the 6-hr metastable level in Tc^{99}, 142 kev above the ground state, and that the 741-kev gamma ray leads to a level 181 kev above the ground state of Tc^{99}. The level at 181-kev decays with the emission of the 181-kev quantum or with cascade emission of the gamma rays of energy 41 and 140 kev. The cascade relation among the various gamma rays is depicted in the disintegration scheme of Fig. 4. To determine the percent of the total beta-ray emission which leads to the level at 922 kev in Tc^{99}, beta-gamma coincidences were measured. The beta-ray counter was an anthracene crystal and phototube biased to count pulses generated

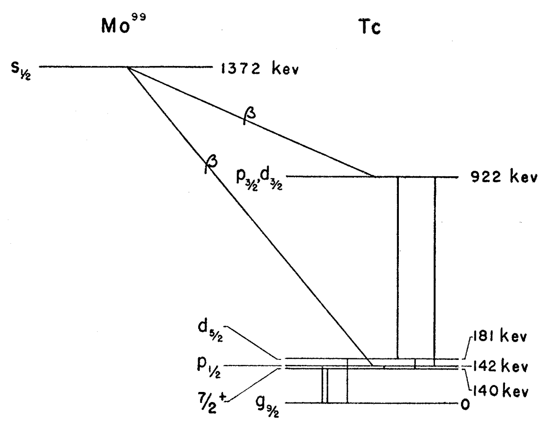

FIG. 4. Term scheme for Mo^{99} and Tc^{99m}.

by beta rays of energy greater than 50 kev. It should also be mentioned that the harder beta-spectrum was found to be non-coincident with gamma rays, in agreement with earlier findings.[1,5] From the gamma-ray counter, all pulses corresponding to a gamma-ray energy greater than 400 kev were accepted. From a carefully calculated solid angle and the counting efficiency of the gamma-ray counter, the 0.5-Mev beta rays were found to constitute 14.5 percent of the total beta emission. This result is in close agreement with the data obtained by Medicus *et al.*[5]

The level at 181 kev can deexcite by way of either of two modes of gamma-ray emission: by the emission of a single gamma ray of energy 181 kev or the 41- and 140-kev quanta in cascade. To determine the probability of deexcitation of the 180-kev level by either mode, coincidences were measured between the 741-kev gamma ray and the entire low-energy spectrum. The pulse-height distribution of single counts in the region of low energy is plotted along with the coincidences between the low-energy gamma rays and those of energy 741 kev as shown in Fig. 5. In actuality, all pulse heights corresponding to energies greater than 400 kev were accepted in the high energy gamma-ray counter. The low-energy gamma-ray counter was shielded from recoil quanta by a lead absorber of thickness 3 g/cm². The aluminum-magnesium oxide jacket in which the crystal of the low-energy gamma-ray counter was encased was estimated to reduce the intensity of the 41-kev gamma ray by 30 percent. If the conversion coefficients of the 140-kev and 181-kev radiations are neglected, it is evident from a consideration of the areas under the peaks of the coincidence curve that the number of disintegrations proceeding by way of the 181-kev branch is about equal to the number in which deexcitation occurs by way of the 41-kev–140-kev transitions. If the conversion coefficient $[\alpha = N(e^-)/N(\gamma)]$ of the 140-kev gamma ray is taken to be 10 percent,[5] the branching ratio of the 41-kev–140-kev and 181-kev transitions is 1.2/1. A one-to-one ratio should exist for the areas under the 41-kev and 140-kev peaks of the coincidence curve. The actual area of the 41-kev peak recorded in Fig. 5 is considerably smaller, indicating a relatively large

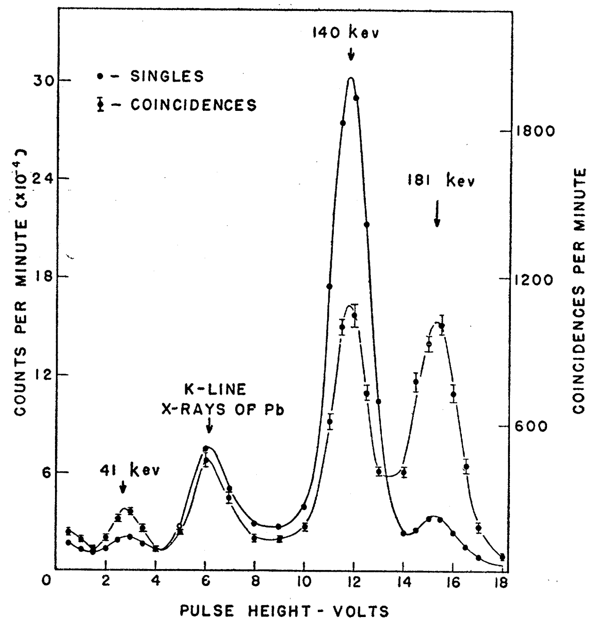

FIG. 5. Gamma-ray spectrum in the region of lower energies of Mo^{99} and Tc^{99m} (Tc^{99m} partially removed by chemical separation). The spectrum of "singles" is to be compared with the spectrum of soft gamma rays in coincidence with the 741-kev radiation.

‡ *Note added in proof.*—Since this paper was submitted for publication, a second source of Mo^{99} has been obtained from Oak Ridge. Coincidences have been observed between the 372-kev gamma ray and a beta-ray spectrum of maximum energy ~0.87 Mev. The 372-kev gamma transition terminates at the 6-hour level of orbital $p_{1/2}$, indicating an additional level in Tc^{99} at 514 kev. The relative abundance of the 0.87-Mev beta-ray group is of the order of one percent.

conversion coefficient for the 41-kev line. From the areas and the conversion coefficient of the 140-kev radiation, the total conversion coefficient for the 41-kev radiation is calculated to be ~ 5. From the determined efficiency for detection of 181-kev radiation, the solid angle of its detector, and the data of Fig. 5, the ratio of intensities of the 741- and 780-kev gamma rays was calculated to be 2.6.

DISCUSSION OF RESULTS

The ground state of Tc^{99} has the orbital $g_{9/2}$. The two levels immediately above the ground state at 140 kev and 142 kev are characterized[5,8] by $7/2+$ and $p_{1/2}$. The two beta spectra have values of log ft such that both of them may be considered to fall in the category of either first-forbidden spectra ($\Delta I=0, \pm 1$; yes !) or l-forbidden spectra ($\Delta I=\pm 1$, no ! $\Delta l=\pm 2$). The ground state of Mo^{99} ($N=57$, $Z=42$) may have any one of several orbitals:[9,10] $g_{7/2}$, $d_{5/2}$, $d_{3/2}$, or $s_{1/2}$. It has been previously shown that the harder beta spectrum of Mo^{99} is noncoincident with gamma rays[1] and terminates[5] at the level of orbital $p_{1/2}$. Were the ground state of Mo^{99} a $d_{5/2}$ configuration, an allowed beta transition, $d_{5/2}\rightarrow 7/2+$ would be present rather than the forbidden transition $d_{5/2}\rightarrow p_{1/2}$. Similarly, if the ground state of Mo^{99} be assumed to be a $g_{7/2}$ level, transitions to $7/2+$ or $g_{9/2}$ would be dominant. None of the above-mentioned allowed transitions are observed; therefore, $d_{5/2}$ and $g_{7/2}$ are rejected as possible orbital designations for the ground state of Mo^{99}. The orbital assignment of $d_{3/2}$ can also be eliminated. It is, however, first necessary to establish the characteristics of the 181-kev level in Tc^{99}. The conversion coefficient of the 41-kev gamma ray identifies it as magnetic dipole radiation ($\Delta I=0, \pm 1$; no !). Since this transition terminates at $7/2+$, possible orbital values for the 181-kev level are $d_{5/2}$, $7/2+$, and $9/2+$. Assignment of $7/2+$ or $9/2+$ to the 181-kev level is ruled out by the absence of a gamma-ray transition from the 922-kev level to either the ground state or the level at 140 kev leaving the orbital of the 181-kev level to be $d_{5/2}$. If the ground state of Mo^{99} were of orbital $d_{3/2}$, an allowed beta transition terminating at the 181-kev level would be present. This spectrum is not observed.

The remaining possible orbital assignment for the ground state of Mo^{99} is $s_{1/2}$. If the softer beta spectrum were given a first-forbidden classification, the level at 922 kev in Tc^{99} would have orbitals of $p_{1/2}$ or $p_{3/2}$. If the spectrum is interpreted as l forbidden, the assignment of the 922-kev level would be $d_{3/2}$. The measurements of the present investigation show that the 741-kev and 780-kev gamma-ray transitions to levels of orbitals $d_{5/2}$ and $p_{1/2}$ have the same order of magnitude of probability of occurrence. If the 922-kev level is described as $p_{3/2}$, the multipolarities of the 741- and 780-kev gamma rays are, respectively, $E1$ and $M1$. If, instead of $p_{3/2}$, the level assignment is taken as $d_{3/2}$, the multipole properties of the two hard gamma rays are interchanged. Since the measurements of the present investigation have shown the 741-kev gamma ray to be more intense than the 780-kev line, an orbital assignment of $p_{3/2}$ for the 922-kev level is favored.

The intensity measurements also exclude the possibility that the 922-kev level has the orbital $p_{1/2}$, because were it so, the 741- and 780-kev gamma rays would be classified, respectively, as $M2$ and $M1$, which transitions are known to have greatly differing lifetimes.

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[8] M. Goldhaber and R. D. Hill, Revs. Modern Phys. 24, 179 (1952).
[9] Mayer, Moszkowski, and Nordheim, Revs. Modern Phys. 23, 315 (1951).
[10] M. G. Mayer, Phys. Rev. 78, 16 (1950).