pp → $d\pi^+$ reaction becomes probable in view of instrumental limitations.

In conclusion, the authors would like to thank V. P. Dzhelepov for his interest in their work and for making the synchrocyclotron available, and to K. A. Ter-Martirosian and I. M. Kobzarev for a discussion of the results.

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THE DECAY SCHEME OF Mo^{99}

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We have measured the angular correlation of the 742 — 180 kev cascade γ-rays emitted in the decay of Mo^{99}. By chemical separation of Tc^{99m} it was shown that the 1.23-Mev β-transition in Mo^{99} goes to the isomeric state in Tc^{99}, while $(7 \pm 1)\%$ of the 140 kev γ-quanta are not associated with the isomeric transition. In the discussion of the data, arguments are presented for assigning the quantum numbers $3/2^+$ to the ground state of Mo^{99}, $3/2^+$ or $5/2^+$ to the excited state of Tc^{99} at 922 kev and $7/2^+$ to the 180 kev state in Tc^{99}.

1. INTRODUCTION

THE radiation accompanying the decay of Mo^{99} has been investigated in many papers, in which the decay scheme of the isotope is also discussed. Study of the isomeric state of Tc^{99} which is produced in the decay of Mo^{99} has made it possible to assign with certainty the quantum numbers of the first two excited states of Tc^{99}, which have excitation energies of 140 and 142 kev.[1-3] A direct measurement gave $I = 9/2$ for the total angular momentum of the ground state.[4] The location of the levels in Tc^{99} at 180 and 922 kev has also been definitely established.[5-7] Different authors are in agreement on the branching ratio[1,5] for the two most intense β-transitions in Mo^{99} (with end points 0.445 and 1.23 Mev). The data of reference 8, in which a direct β-transition from Mo^{99} to the ground state of Tc^{99} was detected, are of interest. All the data enumerated above still do not enable us to make reliable assignments for various excited levels in Tc^{99} and for the ground state of Mo^{99}.

The excited state in Tc^{99} at 922 kev, which is produced in the β-decay with end point $E_\beta = 0.445$ Mev (Fig. 1), is the starting point of two γ-cascades: 742 — 180 kev and 742 — 40 — 140 kev.[7] We have measured the angular correlation for one of these cascades in order to make quantum assignments for the 180 and 922 kev levels in Tc^{99}. The results of these measurements will be presented later in the paper. We also shall consider the

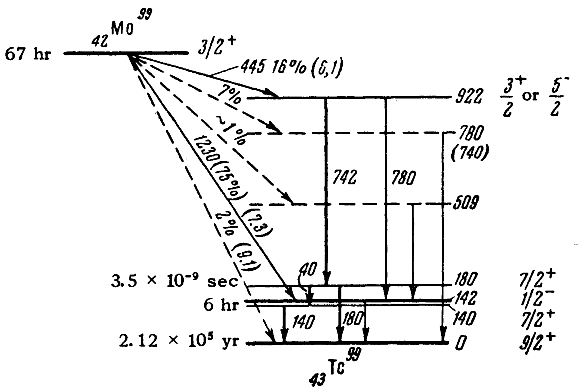

FIG. 1. Decay Scheme of Mo^{99}.

problem of determining which level in Tc^{99} is fed by the 1.23 Mev β-transition in Mo^{99}.

2. MEASUREMENT OF ANGULAR CORRELATION OF THE 742 — 180 kev CASCADE

The measurements of the angular correlation of the cascade γ-rays from Mo^{99} were done in an apparatus consisting of two scintillation counters (Fig. 2), using 20 mm thick stilbene crystals

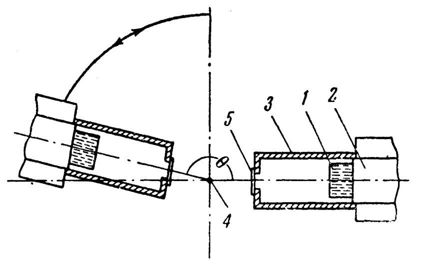

FIG. 2. Arrangement of apparatus for measurement of (γ, γ) angular correlation. 1 — stilbene crystal, 2 — FEU photomultiplier, 3 — lead case, 4 — source, 5 — lead filters in front of entrance window in the shielding.

(1 in the figure), an FEU-19 photomultiplier (2), and a coincidence circuit with a resolving time of $\tau = 3 \times 10^{-8}$ sec. The angle between the emerging γ-rays could be varied by shifting one of the counters. The crystals of the scintillation counters were covered by lead cases (3) which were 4 mm thick and had entrance windows which did not limit the solid angle subtended by the counters at the source (4). The lead shield protected the counters from the effect of scattered γ-radiation. In a control experiment with a Cs^{137} source, it was shown that there are no coincidences due to scattering from one crystal into the other for angles in the range $90° \leq \theta \leq 165°$. Such coincidences were observed for angles of 170 to 180°. We therefore limited ourselves to $\theta < 165°$ in the measurements on Mo^{99}.

The electronic equipment used in the experiment did not have amplitude discrimination, since the stilbene crystals did not permit us to separate the 140 and 180 kev γ-rays which belong to the different cascades 742-180 and 742-40-140 kev in Mo^{99}.[7] To separate the cascades we used lead filters (5 in Fig. 2) covering the entrance windows in the shielding 3. A one mm filter gave a satisfactory ratio of intensities of the 140 and 180 kev γ-lines transmitted through the filter. By measuring the number $N_{\gamma\gamma}$ of twofold coincidences as a function of filter thickness x, we determined the ratio of the contributions of the different cascades. Without filters over the entrance windows, the contribution to $N_{\gamma\gamma}$ from the 742 — 140 cascade was ~70% of that from the 742 — 180 kev cascade. With the 1 mm Pb filter used in the experiment, the contribution of the 742 — 140 kev cascade was $(10 \pm 5)\%$ of that of the 742 — 180 kev cascade (the 40 kev quanta were cut out because of biasing in the amplifiers of the electronic circuit). The effect of the 742 — 140 kev cascade on the correlation function will be considered later on. The method used enabled us to take account of contributions from the different cascades without bringing in additional data on intensities I_γ and efficiencies of γ-counting ϵ_γ ($N_{\gamma\gamma}$ is proportional to $I_\gamma\epsilon_\gamma$).

The difficulty in measuring the angular correlation of the cascade γ-rays from Mo^{99} was that it constituted a small fraction of the total γ-radiation. Under such conditions it is impossible to achieve a favorable ratio of true coincidences N_t to accidentals N_a. The measurements were done with counting rates of $\sim 10^4$ pulses/sec in the channels. N_t was ~0.3 pulses/sec while $N_a \sim 1.2$ pulses/sec, i.e. $N_t/N_a \approx 1/4$.

The Mo^{99} source in the form of the metal or oxide was obtained by slow neutron irradiation of molybdenum. No differences were observed in experiments with different sources.

The coincidence measurements with the Mo^{99} source were done at angles θ equal to 90, 140 and 165°. We collected $\sim 9 \times 10^4$ true coincidences at each angle, which assured an accuracy of 1.4% in the measurements. Setting $W(\pi/2) = 1.00$, we find for θ = 140 and 165 values of the correlation function $[W(\theta)/W(\pi/2)]_{exp} = 0.93 \pm 0.02$ and 0.92 ± 0.02, respectively. This result contradicts the data of Ref. 9, in which measurements of the angular correlation of the Mo^{99} cascade γ-rays gave $W(\pi)/W(\pi/2) = 1.18 \pm 0.04$. However the separate cascades were not resolved in that work. Also their coincidence circuit had a resolving time of $\tau = 2 \times 10^{-7}$ sec, which led to additional trouble in measurement of the low intensity cascade. Apparently the data of reference 9 are incorrect.

3. DISCUSSION OF RESULTS OF ANGULAR CORRELATION MEASUREMENTS

In discussing the results of the angular correlation measurements on the 742 — 180 kev cascade, we first mention some of the facts about the 180 and 922 kev levels of Tc^{99} (Fig. 1). From the 180-kev level, which has a lifetime of 3.5×10^{-9} sec,[10] radiative transitions with energies of 40 and 180 kev can occur and have approximately equal probability.[7] Data on the lifetimes of the radiative transitions, K-shell internal conversion coefficients α_K, and K-to-L ratios α_K/α_L[5-7] allow one to assert that the 40 and 180 kev γ-transitions are either M1 or E2 or a mixture M1 + E2. Consequently the 180-kev level can have the assignments $5/2^+$, $7/2^+$, $9/2^+$ or $11/2^+$. All of these values of the spin I of the 180-kev level were tried in calculating $W(\theta)$.

The 922-kev level in Tc^{99} decays mainly to the 180-kev level. Since $\beta\gamma$-coincidences are observed with the electrons from the 0.445 Mev β-transition,[7,11] the lifetime of the 922-kev level is less than 10^{-7} sec, i.e., the 742-kev transition has a multipolarity no higher than quadrupole. If the transition were octupole, the lifetime would be considerably longer, and in addition transitions with lower multipolarity should occur to the lower levels. In calculating $W(\theta)$ we therefore chose the value of I′ for the 922-kev level to satisfy the condition $|I - I'| \leq 2$, for each choice of I for the 180-kev level.

We tried 23 different sets of values of I and I′. The correlation function $W(\theta)$ was expanded in Legendre polynomials in accordance with the formulas of reference 12, which were used in the calculations. Three of the sets, sets 1, 2, 3, (cf. the table) fit the results within experimental error. The other sets give values of $W(165°)$ $W(\pi/2)$ which differ from the experimental data by more than two standard errors. The table gives the computed values of the coefficient a_2 of the polynomial P_2, and the ratio $W(\theta)/W(\pi/2)$ corrected for the contribution of the 742 — 140 kev cascade. This correction changes the ratio by no more than 1%. Uncorrected values in the table are indicated by an asterisk.

The comparatively long lifetime of the 180-kev level in Tc^{99} compels us to consider the question of possible distortions of $W(\theta)$ due to effects of atomic magnetic fields or electric quadrupole interactions, which would result in a reduction of the correlation. Only case 4 in the table gives a correlation of the same sign, but larger in magnitude, than that observed experimentally. An internal magnetic field $H \approx 5 \times 10^4$ gausses could bring case 4 into agreement with the experiment. We shall therefore include case 4 among the possible explanations of the experimental data.

So far we have listed cases of pure radiative transitions. But mixed transitions like M1 + E2 are known to occur in nuclei. It is therefore natural to consider the possibility of explaining our experimental result by such a mixed transition. We should note as a probable mixture, giving different values for the spins, case 5: $5/2\,(D)\,7/2\,(M1 + E2)\,9/2$. For the mixture of radiation with this set of spins, a_2 is determined by the equation

$$(1 + \delta^2)\, a_2 = 0.050 + 0.097\delta^2 + 0.486\delta, \qquad (1)$$

where δ^2 is the ratio of intensity of E2 to M1 radiation. Figure 3 shows the variation of a_2 with

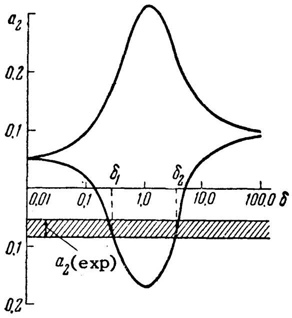

FIG. 3. Dependence of angular correlation coefficient a_2 on mixing parameter δ for the $5/2$ (D) 7/2 (M1 + E2) $9/2$ transition.

δ, taking into account a possible phase difference. The quantity a_2 varies from -0.17 to $+0.32$. The values δ_1 and δ_2 which are in agreement with the experiment determine two possible mixtures: 93% M1 and 7% E2 or 9% M1 and 91% E2, which

Correlation function $W(\theta)$

Case #	Type of transition for 922-180-0 kev	Coefficient a^2	$W(\theta)/W\left(\frac{\pi}{2}\right)$		
			90°	140°	165°
1	$3/2\,(D)\,5/2\,(Q)\,9/2$	−0.0714	1.00	0.95	0.91
2	$3/2\,(Q)\,7/2\,(D)\,9/2$	−0.0716	1.00	0.94	0.90
3	$7/2\,(D)\,7/2\,(D)\,9/2$	−0.0667	1.00	0.94	0.91
4	$5/2\,(Q)\,9/2\,(D)\,9/2$	−0.119	1.00	0.90*	0.84*
5	$5/2\,(D)\,7/2\,(M1 + E2)\,9/2$	$-0.17 < a_2 < +0.32$			
	Experiment	−0.07 ±0.015	1.00	0.93 ±0.02	0.92 ±0.02

cannot be distinguished by a correlation experiment.

Of the five cases given in the table, we must select the most probable set of spins by invoking other data in addition to those from angular correlation. For this purpose, we consider the problem of assignment of quantum numbers for the Mo^{99} ground state, and the strong 1.23-Mev β-transition in Mo^{99}.

4. THE 1.23-Mev BETA-TRANSITION IN Mo^{99}

In the decay schemes of Mo^{99} which are given in two widely used handbooks,[13,14] the most intense β-transition (constituting 75% of all the β-transitions), which has an end point of 1.23 Mev, leads to the 180 kev level in Tc^{99}. Such a decay scheme for Mo^{99} is in contradiction to the data of various experimental papers.[2,7] In view of the importance of this question for the establishment of the quantum numbers of the Mo^{99} ground state, the question of the end level for the most intense β-transition was subjected to a new experimental check, the results of which are described below.

In the transition of Tc^{99} from the excited state at 180 kev (Fig. 1), 40 and 180 kev γ-quanta are radiated which bypass the isomeric state. Therefore, if 75% of the β-transitions lead to the 180 kev level, we should observe intense γ-radiation at either 40 or 180 kev. Figure 4 shows the spectrum of soft γ-radiation from Mo^{99}, measured

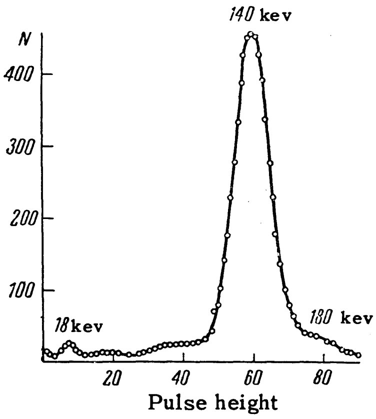

FIG. 4. Soft γ-spectrum from Mo^{99}.

with a scintillation counter having a 10 mm thick NaI (Tl) crystal. As we see from the figure, the intensity of the 40 and 180 kev γ-rays is small compared to that of the 140 kev γ-radiation. The intensity of the characteristic x-rays of Tc ($\sim$18 kev), which are associated with internal conversion of the γ-rays in the K shell of the atom, is also small, so that taking account of possible conversion of the γ-quanta does not lead to equality of the intensities. Consequently the 1.23 Mev β-transition can lead to either the isomeric level at 142-kev or to the 140-kev level.

In a control experiment we measured the β-spectrum of Mo^{99} in coincidence with the 742-kev γ-rays and, separately, with the 140-kev γ-rays. The experiment was carried out on a coincidence apparatus consisting of two scintillation counters (a γ-counter using a NaI (Tl) crystal and a β-counter with a tolane crystal) and having pulse height selection in the channels. The two β-spectra did not differ, and corresponded to the soft β-spectrum of Mo^{99}. No coincidences with the hard β-spectrum of Mo^{99} (E_β = 1.23 Mev) were observed. This experiment confirmed the data of references 2 and 7. In a direct experiment using chemical separation of Tc^{99m} it was shown that the main part of the intensity of 140-kev γ-rays is associated with the isomeric transition in Tc^{99m}.

Tc^{99m} was first separated chemically from neutron-irradiated molybdenum in reference 15. In our work the separation of Tc^{99m} was done using the extraction method proposed in reference 16, with some modifications. After neutron irradiation, MoO_3 was dissolved in a twice-molar solution of K_2CO_3, from which Tc^{99m} was separated by extraction with methylethylketone. In an auxiliary experiment, using Tc^{99m} tracer, it was determined that 99 ± 1% of the Tc^{99m} was extracted. Three samples were prepared for the physical measurements: (1) a sample of the initial Mo^{99}, (2) Mo^{99} after separation of Tc^{99m}, (3) Tc^{99m} extracted from the molybdenum. Using a scintillation counter with a NaI (Tl) crystal, and a multichannel pulse-height analyzer,[17] we measured the time dependence of the intensity of the 140-kev γ-radiation. The results of one of the experiments are shown in Fig. 5. The in-

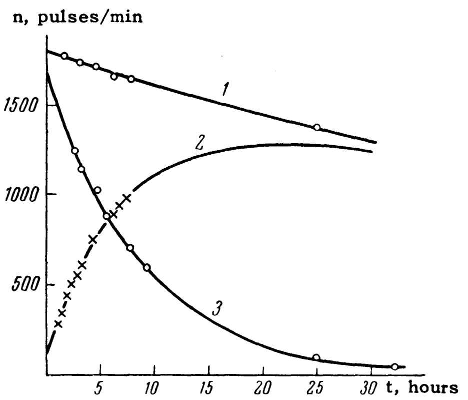

FIG. 5. Variation of intensity of 140-kev γ-rays with time t. 1 – initial Mo^{99}, 2 – Mo^{99} after separation of Tc^{99m} extracted from molybdenum.

tensity of sample 3 varied with the period of the Tc^{99m} isomer (T = 6 hours). In the Mo^{99} sample which had been freed of Tc^{99m} (Curve 2), the intensity increased with time according to the law of radioactive equilibrium (to within 1%), provided we took account of the fact that a portion of the intensity is not associated with the isomeric Tc^{99m} but is formed from Mo^{99} as a result of the 742 — 40 — 140 kev transition (Fig. 1). The magnitude of p was also computed independently from the intensity of sample 3, since the sum of the intensities from samples 2 and 3 should equal the intensity from the initial sample of Mo^{99} (Curve 1).

This experiment using chemical separation showed first that the intense β-transition leads to the isomeric state Tc^{99m}, and second that $(7.5 \pm 1)\%$ of the intensity of 140-kev γ-rays is not associated with the isomeric transition but results from cascade transitions from levels with higher excitation energy. This experiment is a graphic demonstration of how quantitative chemical separation combined with absolute physical measurements can give precise spectroscopic data.

Thus the 1.23-Mev β-transition in Mo^{99} leads to the isomeric level of Tc^{99} at 142 kev (Fig. 1). This β-transition has an $f\tau$ value of 7.3 and is probably first forbidden. The ground state of Mo^{99} can therefore have the quantum numbers $5/2^+$, $3/2^+$ or $1/2^+$. With the $5/2^+$ assignment, we cannot explain the absence of an allowed transition to the 140-kev level. In addition a similar difficulty arises in explaining the absence of the β-transition to the 180-kev level. With spin $5/2$ for the ground state of Mo^{99}, we must use the set of spins of excited states of Tc^{99} which was given by case 4 of our table ($5/2$ (Q) $9/2$ (M1) $9/2$). However, this case is improbable, since it is not clear why the γ-transitions from the 922-kev level to the 140-kev level and to the ground state of Tc^{99} are absent. All of these considerations argue against the assignment of $5/2$ for the ground state of Mo^{99}.

The assignment of $1/2^+$ to the ground state of Mo^{99} does not contradict the results of the correlation measurements, but it is refuted by the data of reference 8 in which a direct β-transtion of Mo^{99} to the ground state of Tc^{99} was observed. The data of the present paper can be brought into accord with the results of reference 8 if the ground state of Mo^{99} has quantum numbers $3/2^+$. The value $3/2^+$ is among the possible values predicted by shell theory, even though it does not agree with the spins of neighboring nuclei having an odd number of neutrons in this same shell.

CONCLUSION

In the table, we gave the possible sets of total angular momenta of the 922 and 180-kev levels of Tc^{99} which are in agreement with the measured angular correlation of the 742 — 180 kev γ-cascade. As was pointed out in the preceding section, case 4 is improbable. Thus we have confirmed that the values of $[W(\theta)]_{exp}$ are not disturbed by internal fields. Case 3 is also improbable, since the assignment of $I = 7/2$ to the 922-kev level leads to difficulties in explaining the observed radiative transitions from this level (Fig. 1).

Case 1 would be probable if the ground state of Mo^{99} were $1/2^+$. With an assignment of $3/2^+$ for the Mo^{99} ground state, it is difficult in Case 1 to explain the absence of an allowed β-transition to the 180 kev level when a large number of forbidden transitions are present. Consequently the most probable schemes are those of Case 2: $3/2^+$ (E2) $7/2^+$ (M1) $9/2^+$, and Case 5, with a mixed transition ($5/2^-$ (E1) $7/2^+$ (M1 + E2) $9/2^+$), which are difficult to distinguish. In both variants the 180-kev level is definitely $7/2^+$ and the 922-kev level is tentatively assigned (as $3/2^+$ or $5/2^-$). The parity of the 922-kev level was chosen to give optimum agreement with the intensities of radiative transitions from the 922-kev level. To fix more definitely the quantum assignments of these levels of Tc^{99}, it would be desirable to determine the multipolarity of the 180 kev γ-rays by measuring their internal conversion coefficient.

In conclusion the authors thank I. S. Shapiro for discussion of the results of the paper.

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MEASUREMENT OF THE TOTAL CHARGED π MESON PRODUCTION CROSS SECTION IN NEUTRON-PROTON COLLISIONS AT 586 Mev NEUTRON ENERGY

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The total yield of charged π mesons, produced in collisions between neutrons of 586 Mev effective energy and protons, was measured at angles in the range from 15 to 120° (in the laboratory system). Assuming charge independence of nuclear forces, the total cross-section for the production of π^+ and π^- mesons, derived from the experimental data, was found to be $\sigma(np \to \pi^+) = \sigma(np \to \pi^-) = (2.0 \pm 0.5) \times 10^{-27}\ cm^2$.

THE process of production of charged π mesons in neutron-proton collisions has not, so far, been studied extensively. Comprehensive investigations were carried out only for 409 Mev neutrons. One experiment only was carried out at an energy close to 600 Mev.[2] The spectra and yields of π^+ and π^- mesons, emitted at the angle $\Phi = 90°$ in the laboratory system* (l.s.) from a target containing liquid hydrogen bombarded by neutrons originating in the charge exchange of 670-Mev protons, were measured in that work in relative units, using nuclear emulsions. The relatively small cross section for meson production, the fact that three particles are present in the final stage of the reactions studied, and also the fact that the neutron beam used is not monoenergetic, all contribute to the difficulties of experiments on π^+- and π^--meson production in $n-p$ collisions. On the other hand, a detailed study of $n-p$ collisions at energies considerably larger than the meson production threshold necessitates an investigation of these processes. The study of the process of meson production in $n-p$ collisions is also of interest for understanding the character of the interaction between two nucleons with different isotropic spin ($T = 0$ and $T = 1$).

*The angle Φ was measured with respect to the direction of the incident neutron beam.

THE EXPERIMENT

The measurements were carried out with the synchrocyclotron of the Joint Institute for Nuclear Research. The neutron beam used in the experiments was obtained by charge-exchange scattering of 680-Mev protons on a Be target. The energy distribution of the neutrons in the beam had a maximum at 600 Mev and a half-width of 130 Mev.[3]

To determine the differential cross-section for charged π-meson production in $n-p$ collisions,