action of Christian and Hart (containing the "halfexchange" dependence proposed by Serber) is used. In Fig. 4, $Q(\theta)$ is plotted for energies of 40, 90, 200, 285 Mev. A similar plot of the polarization $Q(\theta)/I(\theta)$ was given in an earlier report.⁷ A comparison of $Q(\theta)$ with Q/I illustrates the point that $I(\theta)$ alone carries almost the entire energy dependence of the polarization.

If odd-state forces were introduced into the triplet n-p interaction (by changing the $1+P_x$ dependence), the polarization could be considerably larger because of the contribution from S-P interference:

$$Q_{SP} = (1/8k^2) Im \{ B_0^{1} [B_1^2 - B_1^0 + \frac{3}{2} (A_1^2 - A_1^1)]^* \} \sin\theta.$$
(12)

⁷ Don R. Swanson, Phys. Rev. 84, 1068 (1951).

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Isomeric Transitions in Tc⁹³ and Tc⁹⁶[†]

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The 390-kev γ -line reported to be associated with Tc⁹² is found to be Tc⁹³ and is shown to be a M4 isomeric transition. Mass assignment to Tc⁹⁶ is made for the isomeric transition previously known to have 34.4-kev energy and 51.5-minute half-life, and a weak positron branching was found.

ASS assignment of the many technetium activities which are usually produced by irradiation of molybdenum is guite difficult because the latter element has seven stable isotopes of roughly equal natural abundance. From the standpoint of nuclear shell structure, knowledge of these activities is of particular interest since according to this model, technetium with 43 protons lies in a region of isomerism. Until now characteristics of isomeric transitions were known for the odd-even isotopes 95, 97, and 99. This report concerns isomeric states in the nuclei 93 and 96.

The three strongest activities produced when enriched Mo⁹² is bombarded with protons of energies between 5 and 10 Mev decay with half-lives of 4.5 minutes, 43.5 minutes, and 2.7 hours.¹⁻³ At the proton energies used (p, n) and (p, γ) reactions are the most probable, so the activities are expected to be associated with Tc⁹² or Tc⁹³. In order to attempt to fix the mass of the 390-kev γ -transition of 43.5-minute half-life as being Tc⁹³, the activity was produced by two additional separate reactions. Thresholds calculated from

the empirical mass formula⁴ for (d, n) and (d, 2n) reactions on Mo^{92} are -2 Mev and +10 Mev. The 390kev line was observed in 1-mil molybdenum foils throughout the full range of 20-Mev deuterons; however, it could not be produced with neutron bombardment on the same type of foil. Thresholds for $(\alpha, 4n)$ and $(\alpha, 5n)$ on Nb⁹³ are calculated to be 34 and 45 Mev, respectively. The activity was found to be produced in slight amount with α 's between 39 and 40 Mev. Although this evidence cannot be considered conclusive, it appears to indicate that the 390-kev line is Tc⁹³ instead of Tc⁹² as previously reported.

To obtain some idea of the magnitude of this term,

suppose the same amount of triplet odd-state interac-

tion were introduced into the n-p Hamiltonian as was

used for the p-p interaction in the preceding paper.¹

Interpolating the p-p phase shifts to obtain rough

values at 200 Mev, the result is $Q_{SP} \approx 0.5 \sin\theta$ milli-

barns leading to $R(\pi/2) \approx 1.03$. Hence, although the

asymmetry is appreciably influenced by the presence

of odd states, the quoted uncertainty in the experi-

mental results of Wouters⁶ is too great to permit any

sharp conclusions to be drawn on the question of the exchange dependence of the n-p interaction. The

desirability of further experiments on n-p double

scattering is, however, indicated.

The multipole order of the 390-kev line was determined by measurement of the K/L ratio and the conversion coefficient. The activity for the multipole order measurements was produced by bombardment of enriched Mo⁹² with 9.5-Mev protons from the 60-inch cyclotron and the activity observed in a β -spectrometer. Chemical separation of the Tc activities was made by heating the bombarded molybdenum oxide in a glass tube open at one end. By controlling the temperature, the technetium oxide can be made to condense in the cooler part of the tube whereas the less volatile molvbdenum oxide is not affected. The activity is removed with a drop of dilute ammonium hydroxide and mounted on a thin Tygon foil. Figure 1 shows the K- and L-

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¹ E. E. Motta and G. E. Boyd, Phys. Rev. 73, 1470 (1948).
² D. N. Kundu and M. L. Pool, Phys. Rev. 74, 1775 (1948).

³ Medicus, Preiswerk, and Scherrer, Helv. Phys. Acta. 23, 299 (1950).

⁴ E. Fermi, Nuclear Physics (University of Chicago Press, Chicago, 1950), p. 7.

conversion lines measured with a double focusing spectrometer of 1 percent resolution. It was not possible to separate the L and M lines. The result of the K/(L+M)ratio is 5.8 ± 0.3 . Determination of the conversion coefficient was made with a lens spectrometer by measuring the intensities of the conversion electron line and of the photoelectrons from a uranium converter. The converter efficiency was determined using the 390-kev K-line of Sr⁸⁷ which Mann and Axel⁵ have shown to be an M4 transition. Taking the theoretical K-conversion coefficient of 0.194 from the new tables of Rose et al.⁶ for the Sr^{87} isomer, the K-conversion coefficient for Tc^{93} is 0.31 ± 0.07 . This is in good agreement with the theoretical value of 0.295 given by Rose for magnetic 2^4 -pole emission at this energy and Z value. The measured K/L ratio is within experimental error on the curves given by Goldhaber and Sunvar.⁷ The half-life of 43.5 minutes is too small by a factor of at least three to agree with the empirical formula of Goldhaber and Sunvar even if one assumes that the initial state has spin $\frac{1}{2}$. Since the decay scheme of Tc⁹³ is not known at present, it is possible that the apparent half-life is shortened by competing processes, K-capture and positron emission.

An isomer of 51.5 ± 1.0 minutes half-life and 34.4-kev transition energy is produced among the other activities formed by the irradiation of isotopes of natural molybdenum.³ Until now it has not been possible to assign this isomer to a given Tc isotope. The obvious solution of assigning this activity to the positron emitter Tc⁹⁴,

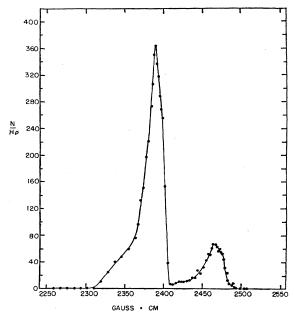


FIG. 1. K- and L-conversion electrons from the 390-kev transition in Tc⁹³

TABLE I. Relative counting rates for different Tc isotopes produced by proton bombardment of enriched Mo isotopes.

Enriched isotope	Isomer 51.5-min 34.4-kev <i>L</i> -line	Tc ⁹³ 43.5-min 390-kev <i>K</i> -line	Tc ⁹⁴ 52.5-min β ⁺ -spectrum at 835 kev	Tc ⁹⁵ 20-hour 762-kev line	Tc% 104-hour 842-kev line	Tc ⁹⁷ 90-day 96-kev <i>K</i> -line
Mo ⁹²	1000	1080	170	0.32	0.19	0.32
Mo^{94}	1000	3.6	5400	0.69	0.31	
Mo ⁹⁵	1000	58	23	4.4	0.22	0.08
Mo^{96}	1000		0.80	0.050	0.18	0.017
Mo ⁹⁷	1000		67		0.28	20.0
Mo^{98}	1000	6.3	79	0.80	0.28	
Mo^{100}	1000	3.4	43	0.19	0.16	0.30
Natural Mo	1000	9.9	91	0.35	0.22	1.1

which has a half-life $(52.5 \pm 1.0 \text{ minutes})$ within the experimental error of that of the isomer, has been proved incorrect.3

By bombarding enriched molvbdenum oxides⁸ with 9.5-Mev protons, it has been possible to fix the mass number of the isomer. Only activities produced by (p, n)processes are important at this energy. The $F^{18}\beta^+$ activity of 0.64-Mev maximum energy produced by O¹⁸ did not interfere with the measurements since a helical baffle could be used in the spectrometer to block all positrons. The intensity of the L-line of the isomer was compared in each sample with intensities of known lines of Tc isotopes 93, 95, 96, and 97 and the positron spectrum of Tc⁹⁴ at 835 kev. No comparison of Tc⁹⁸ and Tc¹⁰⁰ could be made, since the energy and half-life of these activities are not known at present. Table I gives the relative counting rates with that of the isomer normalized to 1000. All values are corrected in time to the end of bombardment (as a rule, $1\frac{1}{4}$ hour in duration). It will be noticed that among the ratios for the various isotopes, only those for Tc⁹⁶ give a reasonably constant value

Tc⁹⁶ alone was produced by irradiating Nb⁹³ foils with α -particles of 13.5 and 15 Mev. These energies were chosen in order to suppress the $(\alpha, 2n)$ reaction which at 17.5 Mev was found to have a small cross section compared to (α, n) . In both cases the L-line of the isomer was observed. As a further proof that the activity is not associated with the positron emitter Tc⁹⁴, the isomer was produced with 3.5-4.0 Mev protons from the Radiation Laboratory electrostatic accelerator on natural molybdenum. According to the decay scheme,³ the (p, n) threshold energy required to produce the Tc⁹⁴ activity is at least 5.1 Mev.

It appears that the ground state of Tc⁹⁶ and the isomer are formed by bombardment in amounts of comparable magnitude. This results from knowing the number of L-conversion electrons for the isomeric transition along with the number of K+L electrons for the three 0.8-Mev transitions in cascade. By considering the K/L ratio of the isomeric transition, the ab-

⁶ L. G. Mann and P. Axel, Phys. Rev. 80, 759 (1950). ⁶ Rose, Goertzel, and Perry, Oak Ridge National Laboratory Report 1023 (unpublished).

M. Goldhaber and A. W. Sunyar, Phys. Rev. 83, 906 (1951).

⁸ The degree of isotopic enrichment estimated by comparing line intensities with those of natural molybdenum varied between 80 percent and 90 percent.

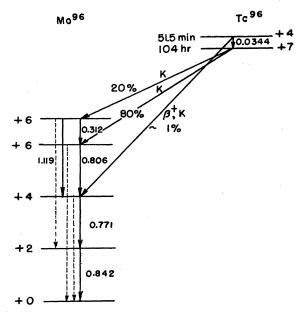


FIG. 2. Decay scheme for Tc⁹⁶.

sorption in the counter window (0.4-mg/cm² Nylon), and the decay, and by assuming that all three 0.8-Mev γ -rays have conversion coefficients corresponding to E2transitions,⁹ one should be able to find the ratio of the number formed in the ground state, to the number formed in the isomeric state. However, this result is subject to a large error due to the difficulty in making counter window and backscattering corrections at the low energy of the isomer.

If one assumes that the isomeric state decays exclusively to the ground state of Tc⁹⁶ with no branching in the form of positron emission or K-capture, an increase in the 104-hour activity should be observed immediately after bombardment. Attempts to observe this increase with one-hour bombardments of Nb foils with 18-Mev α -particles were not successful because the intensity of the sources was too small. It is not possible to observe the increase by bombarding enriched Mo⁹⁶ with protons because the 20-hour Tc⁹⁵ produced from Mo⁹⁵ contamination is strong enough to mask any increase in the 104-hour activity. Branching from the isomeric state was found to occur by observing with a trochoidal analyzer a 51.5-minute positron activity in the separated Tc activity produced by 17-Mev α 's on Nb. At this energy the $(\alpha, 3n)$ reaction leading to the 52.5-minute positron emitter, Tc94, is excluded. By comparing the intensity of these positrons with the 104-hour activity, it can be concluded that there is roughly one positron for each 10⁴ transitions to the

ground state. Although it was not possible to measure the maximum energy of the positrons directly, some estimations can be made. The branching ratio of the K-capture from the Tc^{96} ground state to the two upper states of Mo⁹⁶ is known (Fig. 2). If one assumes that the ft values of these two transitions are equal, one can establish the energy difference between the Tc⁹⁶ and Mo⁹⁶ levels and thus determine the maximum positron energy from the isomeric state as being about 400 kev. The only allowed positron transition from the isomeric state is to the second excited state of Mo⁹⁶. Another estimate can be made by using the (p, n) threshold for Mo⁹⁶ of 3.8 ± 0.3 Mev found by Blaser *et al.*¹⁰ This also leads to a value of 0.4 ± 0.3 Mev for the maximum positron energy. At this energy the K-capture to positron ratio given by Moskowski¹¹ is 50 to 1, so that the branching ratio for the isomeric transitions to Kcapture plus positron emission is in the order of 200 to 1. Since the ratio of half-lives of the ground state to isomeric state is 120 to 1, the expected increase in the 104-hour activity is more or less compensated by direct decay.

The modified decay scheme for Tc⁹⁶ is shown in Fig. 2. The dotted lines indicate weak cross-over transitions of 1.65, 1.89, and 2.39-Mev energy reported by Boyd and Ketelle.¹² A spin of at least 7 must be assigned to the ground state in order to be in agreement with the data for the 104-hour activity (i.e., no strong cross-over transitions in the triple γ -cascade, no positron emission, no direct transitions to the ground state). The half-life of 51.5 min and K/L ratio of 1.2 ± 0.3 allow the assignment of a magnetic 2^3 -pole γ -ray to the isomeric transition and a spin of +4 to the excited state. Since Tc⁹⁶ contains an odd number of both neutrons and protons, a high spin value is not unreasonable. A proton in the $g_{9/2}$ state and a neutron in the $g_{7/2}$ state as given by the shell model¹³ for the 43rd proton and 53rd neutron might add to a spin of 7. One interpretation of the spin of 4 for the excited state would be that the proton remains in the $g_{9/2}$ state while the neutron goes over into a $s_{1/2}$ state.

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⁹ P. Preiswerk and P. Stähelin, Helv. Phys. Acta 24, 300 (1951).

¹⁰ Blaser, Boehm, Marmier, and Scherrer, Helv. Phys. Acta 24, 440 (1951).
¹¹ S. A. Moskowski, Research Program with the Chicago Cyclo-

 ¹¹ S. A. Moskowski, Research Program with the Chicago Cyclotron, Report NP-1493 (unpublished).
 ¹² G. E. Boyd and B. H. Ketelle, Phys. Rev. 83, 216 (1951).

 ¹² G. E. Boyd and B. H. Ketelle, Phys. Rev. 83, 216 (1951)
 ¹³ M. G. Mayer, Phys. Rev. 78, 16 (1950).