Isomeric transitions in ^{100}Tc

M. Bini, A. M. Bizzeti-Sona, P. Blasi, M. Lorini, and N. Taccetti
Istituto di Fisica dell'Università, Firenze, Italy
and Istituto Nazionale di Fisica Nucleare, Sezione di Firenze, Italy
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Low-lying levels of ^{100}Tc have been investigated with the reaction ^{100}Mo($p,n\gamma$)^{100}Tc in the energy range 4 MeV $\leq E_p \leq$ 6.8 MeV. Gamma-ray cascades have been studied by means of γ-γ coincidences. Two isomeric states have been identified, namely the 200.6 keV (4^+) state with $\tau_m = 11.8 \pm 0.1$ μs and the 243.9 keV (6^+) state with $\tau_m = 4.7 \pm 0.3$ μs. An upper limit $\tau_m \leq 4$ ns has been assigned for the mean life of seven other levels.

[NUCLEAR REACTIONS ^{100}Mo($p,n\gamma$), E_p = 4–6.8 MeV, pulsed beam; measured E_γ, γ-γ coincidences, delayed γ. Two ^{100}Tc isomers observed, deduced lifetimes. Enriched target.]

I. INTRODUCTION

Among the odd-odd isotopes of technetium the one of mass 100 possesses singular properties, at variance with the systematics of the three isotopes from ^{94}Tc to ^{98}Tc. This fact appears to be related to the closure of the $\nu d_{5/2}$ subshell expected at $N = 56$. If the next available shell for the valence neutron is $g_{7/2}$, the ground state configuration $(\pi g_{9/2})^3 \otimes (\nu g_{7/2})$ is expected to include states (of the lowest seniority) with J^π from 1^+ to 8^+, with a ground state 1^+ according to the Nordheim rule. In fact, from the results[1,2] obtained with the ^{99}Tc(d,p)^{100}Tc reaction and with the ^{99}Tc(n,γ)^{100}Tc reaction, the spin sequence $1^+, 2^+, 4^+, (2^-), 6^+, 3^+, 5^+$ has been found for the lowest states. As a consequence no isomeric transitions with high multipolarity are likely to exist in ^{100}Tc, while they have been observed in the lower mass Tc isotopes.[3] On the other hand $E2$ transitions within states belonging to the ground state configuration are expected to exhibit relatively long lifetimes due to the close level spacing.

This paper concerns the measurement of the mean lives of ^{100}Tc low-lying levels, by means of the reaction ^{100}Mo(p,n)^{100}Tc and with a pulsed proton beam at energies ranging from 4 to 6.8 MeV. The pulse duration and repetition rate was varied from ~1 ns at 1 MHz to 20 μs at 10 kHz to cover a wide interval of possible lifetimes. Two isomeric levels have been observed at 200.6 keV and 243.9 keV with mean lives $\tau_m = 11.8 \pm 0.1$ and $\tau_m = 4.7 \pm 0.3$ μs, respectively. The values of the mean lives have been derived from the time spectra of the primary decay products (the 43 keV γ rays for the 244 keV level and the 25.7 keV L-conversion electrons for the 200.6 keV level) and also from the time spectra of the γ transition deexciting the daughter level at 172 keV.

Moreover, an upper limit $\tau_m < 4$ ns has been established for the mean life of seven more levels.

A preliminary result concerning the mean life of the 200.6 keV state, obtained in the early stage of this work, has been published elsewhere.[4] This mean life has also been measured by Bartsch *et al.*[5] and by Martin and Wender[6] whose results are in good agreement with the present one.

In addition to the lifetime measurements, direct spectra of prompt γ rays and of γ-γ coincidences have been studied at E_p = 4 MeV. The results fit well with the level scheme obtained from neutron capture[2] and from ^{99}Tc(d,p)^{100}Tc reaction.[1]

II. EXPERIMENTAL PROCEDURE

Measurements were performed at the 7.5 MV Van De Graaff accelerator of the Laboratori Nazionali di Legnaro. The target consisted of a ~100 μg/cm^2 molybdenum oxide layer, 97.42% enriched in ^{100}Mo, evaporated either onto a graphite backing for γ measurements, or onto a thin carbon foil, for electron measurements. A self-supporting molybdenum foil, 88.7% enriched in ^{100}Mo, was also used for the time measurements performed at the highest beam energy E_p = 6.8 MeV.

An intrinsic germanium detector (IG) of 0.5 cm^3 volume and energy resolution 0.5 keV at 100 keV was used to investigate the low energy gamma-ray spectra. In addition, a 60 cm^3 Ge(Li) counter of energy resolution 2.5 keV at 1333 keV was used for measuring γ-γ coincidences. Radioactive sources of ^{133}Ba and ^{152}Eu and the x rays of molybdenum, lead, and tantalum present in the γ spectrum, together with ^{19}F and ^{100}Mo Coulomb excitation γ rays, were used as reference lines for energy calibration.

Conversion electrons have been studied by means of a small magnetic spectrometer[7] with a momentum resolution $\Delta p/p = 0.07$. The electrons coming from the target were focused on a 3 mm thick Si(Li) detector, cooled at $-70°$C, with an energy resolution of ~4 keV at 26 keV.

A data acquisition system consisting of an HP 2116 B computer and up to three analog to digital converters (ADC) working in a multiparameter mode, were used for γ-γ coincidences and for lifetime measurements. The relevant information from the different ADC's was stored sequentially in the memory and transferred to magnetic tape for further analysis.

The γ-γ coincidences were performed by using the IG detector and the Ge(Li) counter placed at $\vartheta_1 = +100°$ and $\vartheta_2 = +20°$ with respect to the beam direction. The electronic chain was a conventional fast-slow system.

For the mean-life measurements two different techniques have been used:

(1) Delay spectra of signals from the IG counter, with respect to the bursts (FWHM ~ 1 ns) of the 1 MHz-pulsed proton beam, have been collected at 4 MeV proton energy. The usual fast-slow coincidence system was used.

(2) The time spectra of the 172 keV and 43 keV γ rays and of the L-conversion electrons of the 28 keV transition have been measured with the continuous proton beam chopped by means of a fast electrostatic deflector[4] placed halfway between the output slits of the analyzing magnet and the target. In the different series of measurements the "beam-on" period was chosen in the range from 6 μs to 20 μs while the corresponding "beam-off" period ranged from 35 μs to 80 μs.

The electrostatic deflector synchronization signal, correlated by means of an adjustable delay to the beginning of the beam-off period, was used to start a time-to-amplitude-converter (TAC). In some measurements the stop signals for the TAC were derived from one (or more, working in OR) timing single channel analyzer which performed the desired energy selection. The "valid stop" signals of the TAC were used to validate the corresponding energy signals.

Other measurements were performed with the standard fast-slow system; in this case the TAC stop signals were obtained by means of a constant fraction discriminator. The time scale was determined by means of an ORTEC 462 time calibrator. Energy signals were shaped at 1 μs with a

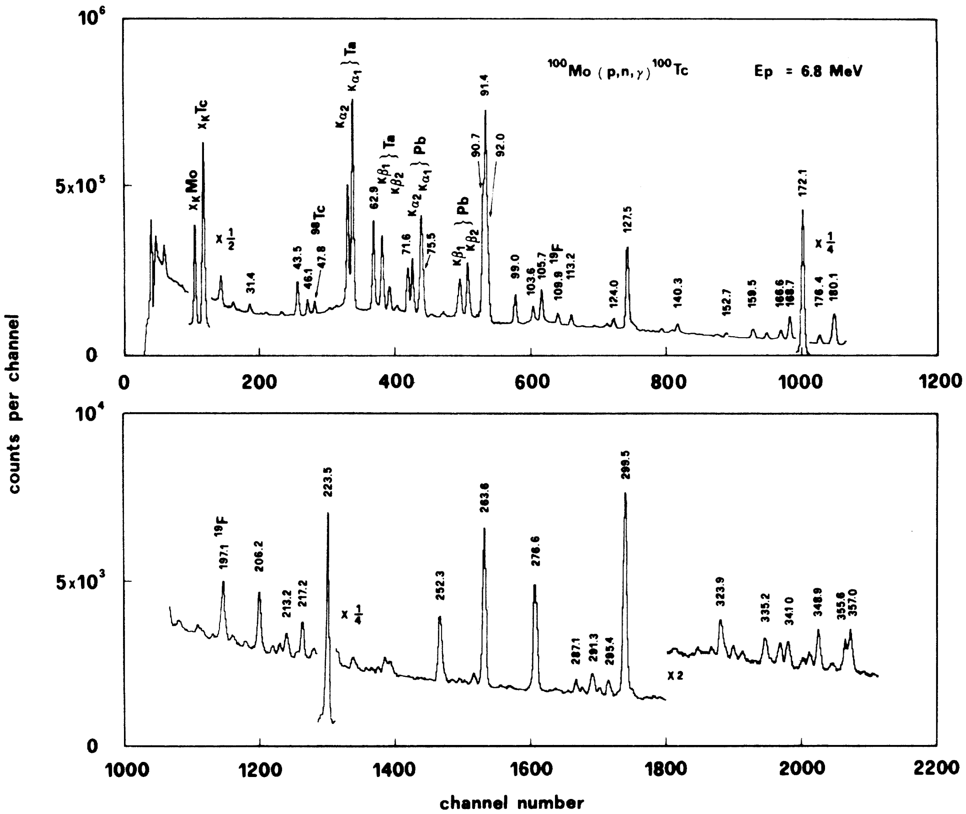

FIG. 1. Typical γ-ray spectrum from the reaction ^{100}Mo $(p, n\gamma)^{100}$Tc, obtained with a 0.5 cm^3 intrinsic germanium detector. The γ-ray energies are given in keV.

TABLE I. Coincident γ rays observed in the ^{100}Mo (p, n) ^{100}Tc reaction at $E_p = 4$ MeV. The values in parentheses indicate possible coincidence.

Gate E_γ (keV)	γ rays in coincidence E_γ (keV)
71.6	91, 124, 172, 263
90.7	(172), 180, 299
91.4	(71), 91, 172, (176)
92.0	(63), 104, 263
103.6	92
113.2	169
127.5	172, 176
168.7	172, (180)
172.1	71, 91, 127, 169, 180, 252, 287, 349, 398
180.1[a]	(63), (71), 90.7, 172, 299
223.5	(222), 276, (355), (455), 486
263.6	71, 92
276.6	223
299.5	90.7, (172), 180

[a]This line appears to be a very close doublet which cannot be completely resolved in the coincidence measurements.

simple delay-line differentiator to avoid a pileup effect in the counting periods, starting 1 μs after the end of the beam bursts.

In these measurements a ^{57}Co γ-ray source was placed near the detector to produce pulses uncorrelated with the on-off status of the beam in order to verify the flatness of their delay spectrum.

III. EXPERIMENTAL RESULTS: THE LEVEL SCHEME

A γ-ray spectrum from the proton bombardment of ^{100}Mo at 6.8 MeV is shown in Fig. 1. Most of the prominent γ rays have been attributed to ^{100}Tc. Some of them coincide in energy, within 0.1 keV, with transitions observed in the ^{99}Tc$(n,\gamma)^{100}$Tc reaction.[2] Gamma-gamma coincidence measurements have been performed at proton energy $E_p = 4$ MeV. This measurement has also been valuable in resolving the three γ transitions[8] of energy close to 90 keV (90.7 keV, 91.4 keV, 92.0 keV). The coincidence results are summarized in Table I. The level scheme obtained up to the excitation en-

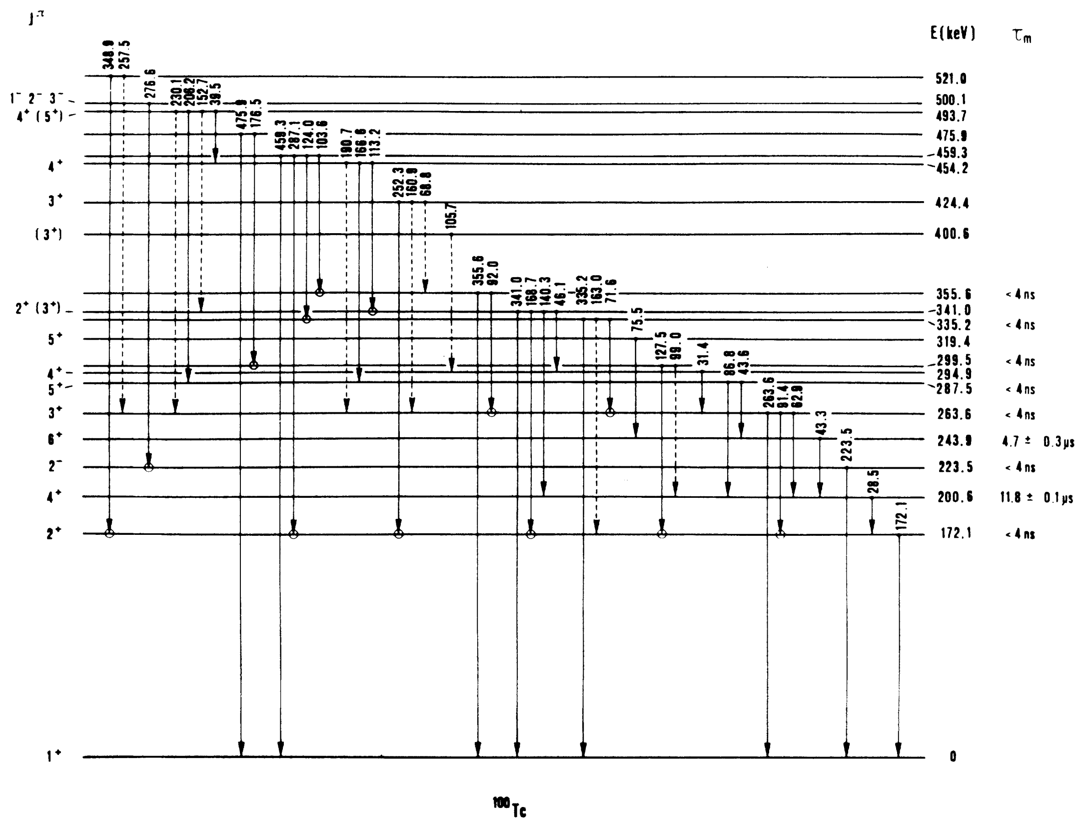

FIG. 2. Level scheme of ^{100}Tc. Dashed lines for the transitions indicate uncertain assignments. The open circles at the tip of a transition arrow indicate observed coincidence with a subsequent γ ray. Spin assignments are from Ref. 2.

ergy of 530 keV is shown in Fig. 2. The level energies have been deduced on the basis of the coincidence measurement and/or energy balance (within 0.2 keV) in the γ-ray cascades.

However, some intense γ transitions do not fit into the level scheme, namely the strong γ cascade involving the 90.7 keV γ, the 299.5 keV γ, and at least one member of the close doublet at 180 keV. In fact the 299.5 keV line could fit (within 0.1 keV) the energy difference either between the 500.1 and the 200.6 keV levels or between the 299.5 keV level and the ground state; however, neither of the above assumptions is in agreement with our coincidence results. In addition one of the components of the 180 keV doublet, being definitely in coincidence with the 172 keV γ, probably corresponds to the decay of a new level at 352 keV.

Most of the levels shown in Fig. 2 have been found also with the ^{99}Tc$(d,p)^{100}$Tc reaction.[1] All the states populated in the (n,γ) reaction up to 455 keV have been observed. Moreover, six levels at 299.5, 335.2, 355.6, 459.3, 475.9, and 521.1 keV have also been found. Owing to their decay properties they appear to have positive parity and spin <3 (<4 for the 521.1 keV state). Some of these levels (namely the 299.5, 335.2, 459.3, and 521.1 keV) have also been recently reported by Martin and Wender.[9]

IV. EXPERIMENTAL RESULTS: LIFETIMES

Lifetime measurements in the nanosecond region have been performed at a proton beam energy $E_p = 4$ MeV. Only the 172 keV γ ray, corresponding to the transition from the first excited to the ground state, shows, in addition to a prompt part, a delayed component with a mean life longer than 1 μs. The other γ lines observed at this beam energy, as well as the prompt part of the 172 keV γ ray, have a mean life $\tau_m < 4$ ns. On this basis, an upper limit has been assigned to the mean life of seven excited levels as shown in Fig. 2.

The delayed part of the 172 keV γ transition must be attributed to the feeding from higher-lying isomer states. The first obvious candidate is the 4^+ level at 200.6 keV (for spin attribution see Ref. 2). This level is expected to decay with an $E2$ transition of 28.5 keV which should be strongly converted ($\alpha = 115$). The corresponding γ ray is weak and lies in a region of the gamma spectrum disturbed by the x ray background; therefore it is difficult to measure directly its mean life. The situation is somewhat better for the conversion electrons. In fact the L-conversion line corresponding to the 28.5 keV γ transition has been clearly observed during the beam-off period. The electrons' energy comes out to be 25.7 ± 0.5 keV.

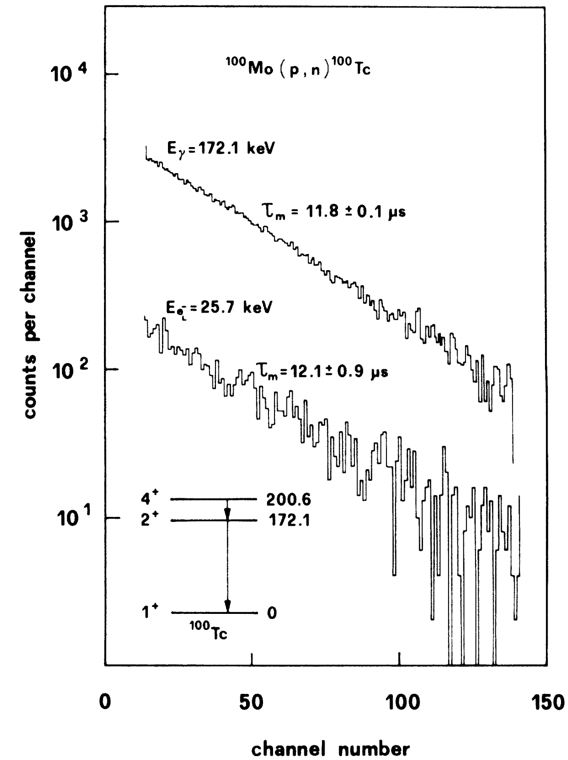

FIG. 3. Time spectra, during the beam-off period, for the 172 keV γ transition and for the L-conversion electrons coming from the 28.5 keV transition, at $E_p = 5.0$ MeV and $E_p = 5.2$ MeV, respectively. The time scale is 0.345 μs/channel.

For the energy calibration the K-conversion electrons corresponding to the 172 keV γ transition and the background peak of photoelectrons produced into the silicon detector by the x rays of the lead shielding have been used.

In Fig. 3 are shown the time spectra for the 172 keV γ rays and for L-conversion electrons coming from the 28.5 keV γ transition, with proper background subtraction. The proton energies were $E_p = 5$ MeV and $E_p = 5.2$ MeV, respectively. The beam was pulsed 20 μs on and 80 μs off. From a minimum χ^2 fit of the time spectra one obtains the mean-life values: $\tau_m = 11.8 \pm 0.1$ μs for the 172 keV γ rays, and $\tau_m = 12.1 \pm 0.9$ μs for the conversion electrons of the 28.5 keV transition.[10] It is therefore reasonable to identify the parent isomer state with the 4^+ level at 200.6 keV and we attribute to it the mean life $\tau_m = 11.8 \pm 0.1$ μs. This result compares well with the values recently reported by Martin and Wender[6] and by Bartsch *et al.*[5]

At higher beam energy, $E_p = 6.5$–6.8 MeV, a new isomeric transition appears. Figure 4 shows the spectra of prompt and delayed γ rays taken at different delay times with respect to the beam burst.

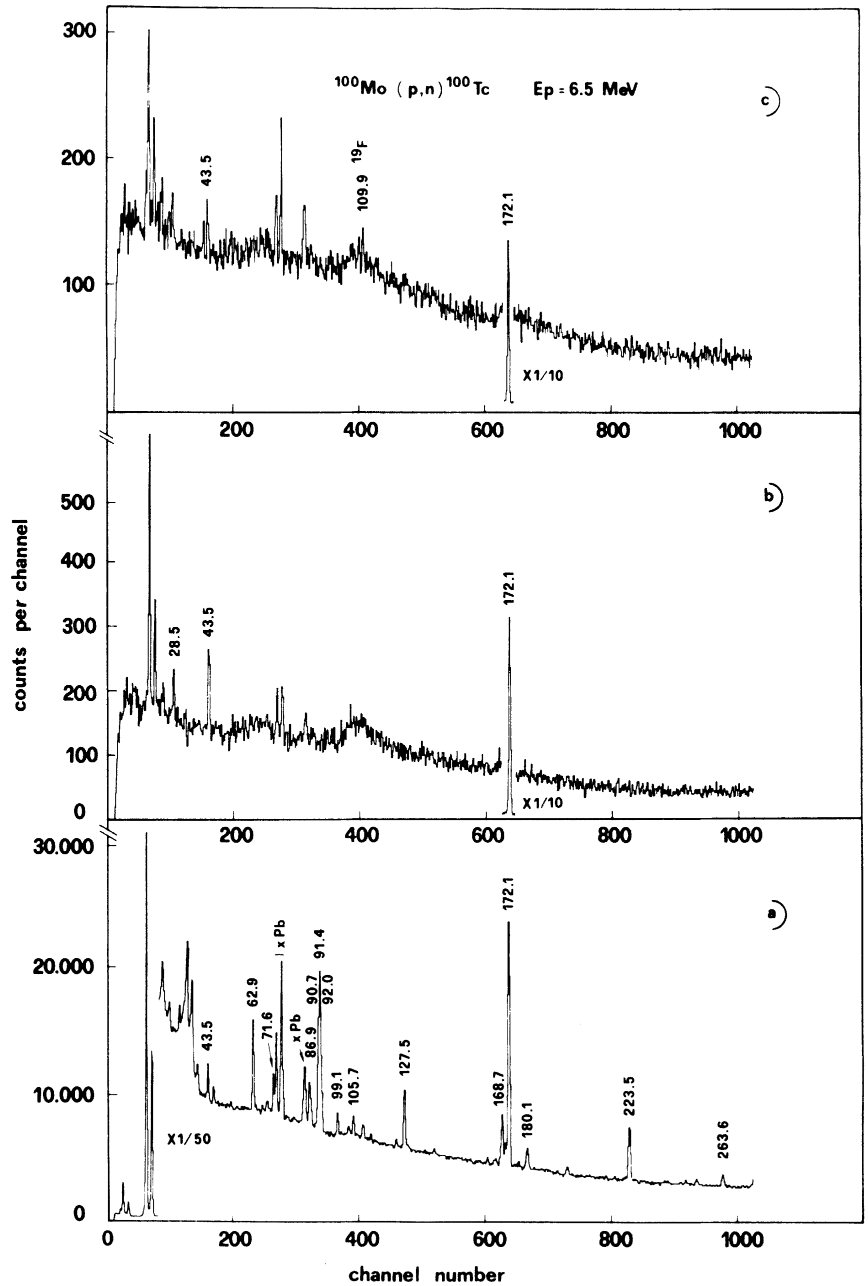

FIG. 4. Energy spectra of γ rays from ^{100}Mo $(p,n\gamma)^{100}$Tc reaction at $E_p = 6.5$ MeV. (a) Prompt γ spectrum; (b) delayed γ spectrum corresponding to the time interval 3.0–10.5 μs; (c) delayed γ spectrum corresponding to the time interval 10.5–26 μs. Delays are measured from the beginning of the beam-off period.

In the delayed spectra a 43 keV gamma is observed in addition to the known 172 keV line. A weak γ transition of 28 keV is also present.

The mean life of the 43 keV transition has been determined with a 6.8 MeV proton beam pulsed 6 μs on and 35 μs off [time spectrum(a) of Fig. 5]. The analysis of this time spectrum was complicated owing to the presence of another 43.5 keV γ ray from ^{98}Tc having a mean life[12] of 21 μs. ^{98}Tc was produced via the ^{98}Mo$(p,n)^{98}$Tc reaction on the ^{98}Mo present in the target at a rate of 3.8%. In order to subtract from the time spectrum of the 43 keV γ line the amount due to the ^{98}Tc contamination, we measured the time spectrum of the ^{98}Tc 43 keV transition, with an enriched ^{98}Mo target, in the same experimental conditions [spec-

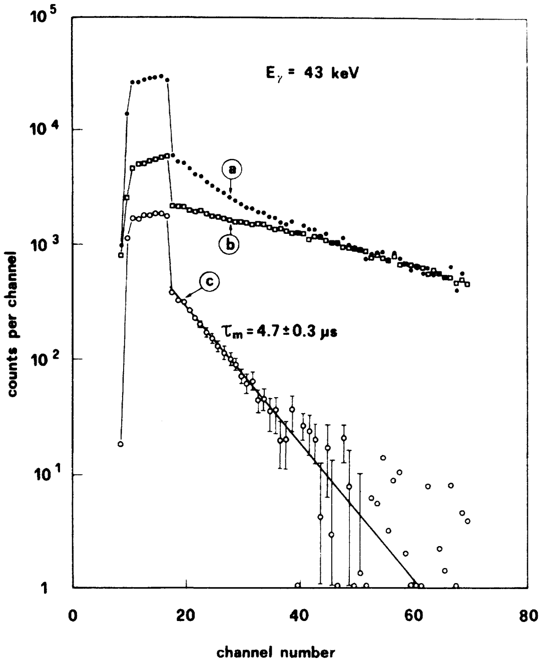

FIG. 5. (a) Time spectrum of the composite 43 keV line obtained with the ^{100}Mo enriched target at $E_p = 6.8$ MeV. (b) Time spectrum of the 43 keV gamma of ^{98}Tc obtained with a ^{98}Mo enriched target in the same experimental conditions as spectrum (a). Spectrum (b) is normalized to spectrum (a) to give the same contribution at the long time side. (c) Spectrum difference between (a) and (b). Spectrum (c) is shifted downward by one decade for the sake of clarity. The prompt part of spectrum (c) is due to the 43.5 keV transition from the (5^+) 287.5 keV level to the (6^+) 243.9 keV level of ^{100}Tc. The time scale is 0.664 μs/channel.

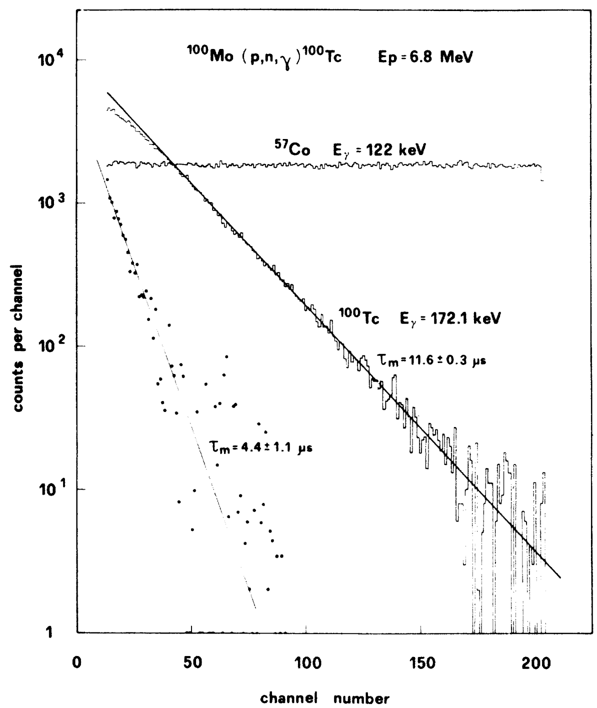

FIG. 6. Time spectrum of the 172 keV γ transition at $E_p = 6.8$ MeV. The thick straight line indicates the exponential decay of the 200.6 keV isomeric state which feeds the 172.1 keV state through the 28.5 keV transition. The full points correspond to the difference between the thick straight line and the experimental spectrum of the 172 keV γ transition. The thin straight line through the points represents the fitted exponential decay of the second isomeric transition feeding the 200.6 keV level. The time spectrum of the 122 keV γ ray from a radioactive source of ^{57}Co is also shown for comparison. The time scale is 0.459 μs/channel.

trum (b) of Fig. 5]. The mean life of the 43 keV transition of ^{100}Tc has been deduced with a minimum χ^2 fit on the difference spectrum [Fig. 5(c)], leaving as a free parameter also the percentage of spectrum (b) to be subtracted from spectrum (a). A value $\tau_m = 4.7 \pm 0.3$ μs has been obtained.

The delayed 43 keV transition can be identified with the decay of the 6^+ level at 243.9 keV to the 4^+, 200.6 keV isomeric level. In fact at the beam energy $E_p = 6.8$ MeV the time spectrum of the 172 keV γ ray is no longer a pure exponential decay, but deviates downwards at the short-time side, as shown in Fig. 6. This is the expected trend if the isomeric level at 200.6 keV is fed from a higher-lying isomer of comparable lifetime. The experimental time spectrum, after proper background subtraction, has been fitted with the four parameter function:

$$f(t) = Ae^{-t/\tau_1} - Be^{-t/\tau_2}.$$

A minimum χ^2 procedure gives the following results for the mean lives of the two isomers:

$$\tau_1 = 11.6 \pm 0.3\ \mu\text{s and } \tau_2 = 4.4 \pm 1.1\ \mu\text{s}.$$

The first value is in good agreement with the one obtained at lower proton energy, while the second is consistent with the observed mean life of the 43 keV transition.

V. DISCUSSION

Available experimental information concerning low-lying levels of ^{100}Tc up to 510 keV are summarized in Fig. 7.

From the results of spectroscopic factors in stripping reactions (Ref. 1) the ground state seems to correspond to a rather pure configuration $(\pi g_{9/2})^3 \nu g_{7/2}$ with the valence protons coupled mainly to seniority 1. The same configuration contains levels with positive parity and spin up to 8: In fact all values of the spin up to 6 are found under

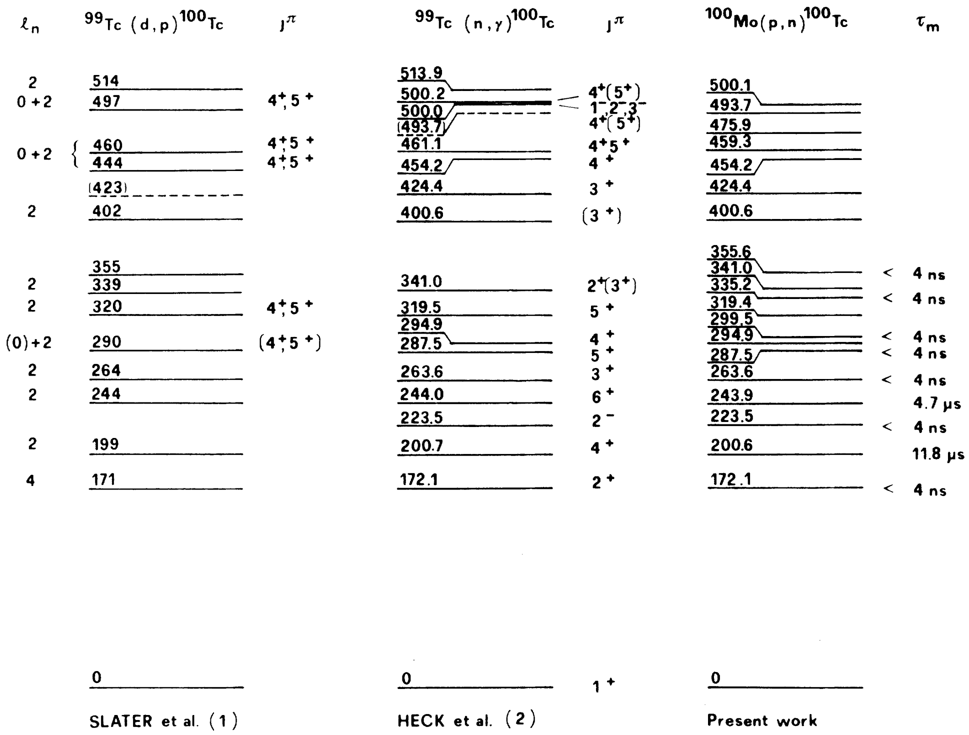

FIG. 7. Low-lying level scheme of ^{100}Tc according to Slater and Booth (Ref. 1), Heck *et al*. (Ref. 2), and to the present work.

320 keV, and levels 7^+ and 8^+ are not expected to be reached with the reactions considered. However, other levels with $J^\pi = 4^+$, 5^+ exist in the same energy range. In addition stripping experiments show a sizable $l_n = 2$ component in the transitions to the lowest 4^+, 6^+, and 5^+ states (although an additional $l_n = 4$ component, as large as in the ground state transition, cannot be excluded). This fact implies either the presence of the $\nu d_{3/2}$ configuration in the wave function of these states, or the presence of $\nu d_{5/2}$ holes in the ground state of ^{99}Tc and possibly in the lowest 4^+, 6^+, and 5^+ states of ^{100}Tc.

Some additional information can be obtained from the measured transition strengths. The $B(E2)$ values for the transitions $6^+ \to 4^+$ and $4^+ \to 2^+$, deduced from the measured mean lives (with conversion coefficients taken from Ref. 13) are: $B(E2)\downarrow = 31.7 \pm 0.2$ e^2 fm^4 (i.e., 1.15 W.u.) for the $4^+ \to 2^+$, 28.5 keV transition, and $B(E2)\downarrow = 44 \pm 3$ e^2 fm^4 (i.e., 1.6 W.u.) for the $6^+ \to 4^+$, 43.3 keV transition. There is therefore no particular indication of enhancement with respect to single particle estimates.

Bartsch *et al.*[5] assume that the 28 keV transition corresponds to the single-neutron transition from the $[(\pi g_{9/2})^3_{v=1} \otimes (\nu 2d_{5/2})^{-1}(\nu 1g_{7/2})^2_0]_4$ to the $[(\pi g_{9/2})^3_{v=1} \otimes (\nu g_{7/2})^1]_2$ configuration. Calculation of the transition strength with these configurations gives, however, very small values for any reasonable assumption on the neutron effective charge (namely 0.28 e^2 fm^4 for $e_n = 1$), due to the neutron spin flip in the transition and to geometrical factors in the coupling to the spectator proton configuration $(\pi g_{9/2})^3_{v=1}$. Instead, the right order of magnitude for $B(E2)$ is obtained if a pure configuration $[(\pi g_{9/2})^3_{v=1} \otimes (\nu g_{7/2})]$ is assumed for both the initial and the final state. In this hypothesis if the effective charges for proton and neutron are taken as $e_p = 1 + \epsilon$, $e_n = \epsilon$ (i.e., the polarization charge is pure isoscalar), the values of the polarization charge ϵ which reproduces the experimental transition strengths are[14] 0.74 for the $4^+ \to 2^+$ transition and 1.18 for the $6^+ \to 4^+$ transition. Similar values ($\epsilon = 1.0$ for the $4^+ \to 2^+$ transition and $\epsilon = 1.36$ for the $6^+ \to 4^+$) would be obtained for transitions between states belonging to the configuration $(\pi g_{9/2})^3_{v=1} \otimes (\nu d_{5/2})^{-1}(\nu g_{9/2})^2_0$.

Although the present information on the wave function of the states is not enough to get definite conclusions, one can note that the values of the polarization charge obtained with the above simple model are comparable with those obtained in the neighboring nuclei.[15,16]

More information on the nuclear wave functions would be highly desirable. We can, however, conclude that at the present status of knowledge, the measured $E2$ strengths appear to be consistent with current shell model descriptions of this region of nuclei.

Note added in proof. We have been informed of new results obtained by J. A. Pinston *et al.*, which are now published.[17] In this paper the authors confirm all the spin and parity attributions they gave in a previous report. In addition, they give values of the mean life of the two isomeric levels at 200.6 and 243.9 keV, which are substantially longer than ours ($\tau_m = 14.7 \pm 0.15$ μs instead of $\tau_m = 11.8 \pm 0.1$ μs and $\tau_m = 6.6 \pm 0.7$ μs instead of $\tau_m = 4.7 \pm 0.3$ μs, respectively). At the moment, the reason for the discrepancy is not clear.

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