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DECAY OF MOLYBDENUM-90

J. A. Cooper, J. M. Hollander, M. I. Kalkstein, and J. O. Rasmussen

March 1965

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ABSTRACT

High resolution beta and gamma-ray spectroscopic studies of the decay of 5.7 hour Mo⁹⁰ have been carried out, with use of the Berkeley 50-cm $\pi\sqrt{2}$ iron-free spectrometer and lithium-drifted germanium gamma-ray detectors. The gamma-ray spectrum revealed transitions of 42.8, 122.5, 163.0, 202.9, 257.5, 323.0, ~420, 445.2, 472.5, ~490, 945, 993, 1272, 1389, 1455, and 1463 keV. The internal-conversion-electron spectrum was studied in the energy range 0 to 400 keV. From measurements of L-subshell internal-conversion ratios, the multipolarities of the 122- and 257-keV transitions were determined to be E2 and E3, respectively. A partial decay scheme for Mo⁹⁰ is proposed that utilizes the data on the 122- and 257-keV transitions together with considerations of the available shell-model states for Nb⁹⁰. This scheme requires the presence: of an undetected low-energy (<3keV) transition in Nb⁹⁰.

I. INTRODUCTION

Molybdenum-90 was first identified by Diamond.¹ This activity, which was produced by the p,4n reaction on Nb^{93} , was shown to decay with a half-life of 5.7 hours. Subsequently, in a study of Mo⁹⁰ decay, Mathur and Hyde² observed gamma rays of 120 and 250 keV and a positron end point of 1.2 MeV. Mathur and Hyde also noted that the levels in Nb^{90} populated by the decay of Mo⁹⁰ are isomeric, and they reported values of 24 seconds and 10 milliseconds, respectively, for the half-lives of the 120- and 250-keV transitions. They reported no other radiations, and spin assignments to the Nb^{90} levels were not made.

The daughter activity, 14.6 hour Nb⁹⁰, the decay of which populates levels in the closed-subshell nucleus $\int_{10} Zr^{90}$, has been the subject of many investigations. When the Nb⁹⁰ decay data are examined together with the known information on Mo⁹⁰, some interesting inconsistencies appear, as follow: From its decay properties, Nb⁹⁰ has been given a ground-state assignment of 8+ or 9+ by Bjørnholm, Nielsen, and Sheline,³ and this can be considered rather definite. The even-even nucleus Mo^{90} , with spin and parity 0+, decays with a small log ft value (5.3, calculated from the positron end point and half-life with use of the theoretical EC/β^+ ratio from Feenberg and Trigg⁴); this low log ft indicates a spin change in the beta transition of 0 or 1, and implies that Mo^{90} decays to a state in Nb⁹⁰ with spin 0 or 1. Only two transitions have been reported in the decay of Mo^{90} , and the above considerations require that the sum of their multipolarities be at least 7 units. On the basis of its half-life, the 250-keV transition was assigned by Mathur and Hyde as an M3 or E3. The data on the 120-keV transition were ambiguous, with an E3 assignment indicated by the half-life and K/L conversion ratio but an E2 assignment indicated by the total conversion coefficient.

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Even if these transitions were both octupoles, as suggested by their half-lives, angular momentum could not be conserved if the suggested spin difference (7 to 9 units) between the initially-populated state and the ground state is correct. Either a higher multipolarity is required for one or both transitions, or additional transitions are required.

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This work was undertaken in the hope that an investigation of the internal-conversion-electron spectrum with the high-resolution $\pi\sqrt{2}$ iron-free spectrometer⁵ and of the gamma-ray spectrum with lithium-drifted germanium detectors might resolve the above inconsistencies and provide a clearer picture of the levels in the odd-odd nucleus Nb⁹⁰.

II. EXPERIMENTAL PROCEDURE

The internal-conversion-electron spectrum was examined with the Berkeley 50-cm radius $\pi\sqrt{2}$ iron-free spectrometer⁵ and a 180-deg permanent-magnet photographic-recording spectrograph.⁶ The counter used in the iron-free spectrometer was a Geiger counter with a window aperture, 1 mm by 20 mm, covered with a formvar film of surface density \approx 50 µg/cm².

An electrostatic preaccelerator system was used for the study of the very low energy region in the iron-free spectrometer. An exploded view of the elements of the preaccelerator system is shown in Fig. 1. This assembly was designed to replace the standard spectrometer source holder which fits into the source chamber from the top. All source elements were electrically insulated and connected to "feed through" electrodes in the top plate of the machined lucite mounting block. The accelerating electrodes were made from 30/cm mesh nickel grid which has > 90% open area.⁷

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The Mo^{90} activity used for electron spectroscopy was produced by bombarding natural zirconium foils (99.9+% Zr) with 65-MeV alpha particles in the Berkeley 88-inch (224-cm) cyclotron. The Zr foils were dissolved in concentrated hydrofluoric acid, the resulting solution evaporated to dryness, and the activity taken up in 5N HCL-0.06N HF. This solution was placed on a Dowex-1 anion exchange column of dimensions 7- to 8-cm long by 0.4-cm in diameter. Molybdenum forms anionic complexes that stick tightly to the column while niobium and zirconium pass on through. The Nb and Zr are quantitatively separated by eluting with several column volumes of 5N HCL-0.06N HF. The Mo activity was stripped from the column with 1N HCl and further purified and concentrated into 2 drops by repeating the above column procedure with a column 8-mm long by 1-mm in diameter.

The Mo⁹⁰ source for the iron-free spectrometer was prepared by vacuum sublimation of the dried chloride solution from a tungsten boat at $\geq 2000^{\circ}$ C through a collimator 1 mm by 10 mm onto an aluminum foil backing of surface density $\approx 7 \text{ mg/cm}^2$. The source for the permanent-magnet spectrograph was prepared in a similar manner, except that the activity was sublimed onto a 1-cm length of 0.25-mm platinum wire.

The Mo^{90} source for the gamma-ray studies was also produced and separated by the procedure described above. In addition, Mo^{90} gamma sources were produced by bombarding natural NaBr with 80-MeV N¹⁴ ions in the Berkeley Heavy-Ion Linear Accelerator (Hilac), and by bombarding Nb metal foils with 47-MeV protons in the Berkeley 88-inch cyclotron.

When NaBr was used as the target, the separation of Mo⁹⁰ from the NaBr . and other activities was made by dissolving the NaBr in 5N HCL-0.06N HF. This solution was passed through a Dowex-1 column and eluted with 5N HCL-0.06N HF. All impurities pass through the column. The purified Mo was stripped from the column with 1N HCL.

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When Nb metal foils were used as the target, they were dissolved in a mixture of concentrated HF and HNO_3 , and the solution evaporated to dryness. The Mo activity was then taken up in 5N HCl-0.06N HF and purified further with a Dowex-l anion column as described above.

The gamma-ray spectrum was studied with a lithium-drifted germanium detector with an active volume of 6 cm² by 9-mm deep. It was maintained at "liquid-nitrogen temperature" (-196°C) with use of a 10-liter gravity-feed liquid-nitrogen reservoir of commercial manufacture.⁸ The associated electronics consisted of a low-noise, low-capacity pre-amplifier and biased-amplifier system designed by Goulding and Landis^{9,10} and constructed at this Laboratorý. Pulse-height analysis of the spectrum was made with a 400-channel analyzer.¹¹

III. EXPERIMENTAL RESULTS

A. Gamma-Ray Spectrum

The gamma-ray spectrum observed with the Ge(Li) detector clearly shows the two prominent gamma rays previously reported by Mathur and Hyde,² but in addition a number of low-intensity gamma rays are found. (The gamma rays were shown to have originated from the decay of Mo^{90} by their half-lives and their relative intensities, which were independent of the method used to produce the activity.) Examples of our recorded spectra are shown in Figs.2a-e, and the energy and intensity information on the Mo^{90} photons is summarized in Table I. (The photon relative intensities were determined by making measurements of the areas under the peaks and correcting for the variation of the Ge(Li) photopeak efficiency with energy. The Ge(Li) photopeak efficiency function was experimentally determined with use of a number of isotopes with well-known photon intensities, and this efficiency curve is reproduced in Fig. 3.)

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It is interesting to note that although many other gamma rays were observed in addition to the prominent 122.5- and 257.5-keV radiations, none has an intensity greater than 12% of the 257-keV gamma intensity, and therefore unless one or more of the transitions is very highly converted, none can be in 100% cascade with the 122- and 257-keV transitions.

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B. Internal-Conversion-Electron Spectrum

The internal-conversion spectrum of Mo^{90} (including Mo^{93m} and Nb^{90}), taken with the 180° permanent-magnet spectrograph, is reproduced in Fig. 4. The energy range of this spectrum is from 10 to 400 keV, and it shows clearly the K and L lines from the 122- and 257-keV transitions from Mo^{90} decay, the 132- and 142-keV transitions from Nb^{90} decay, and the 262-keV transition from Mo^{93m} . In addition, M lines from the 132-, 142-, 257-, and 262-keV transitions and the very weak K lines from the 163- and 203-keV transitions of Mo^{90} are also visible. From this spectrum it is obvious that there are no strong, highly converted transitions within the energy range 10-400 keV that have been previously unreported.

A scan, with the iron-free spectrometer, of the energy range from 10 keV to 100 keV revealed only the low intensity K and L lines from the 42.76-keV transition (Figs. 5 and 6) and a large number of lines from 13 to 18 keV of about the same low intensity as the K line from the 42.76-keV transition. This region was not scanned in detail but it is noted that the energies of these lines correspond to the energies expected for the KXY Auger electrons from Zr, Nb and Mo due to the decay of Nb⁹⁰, Mo⁹⁰, and Mo^{93m}.

.C. Search for Low-Energy Transition

The region from about 0 keV to 10 keV was studied with the preaccelerator, using an accelerating potential of -4.69 kV. The spectrum observed in the initial scan of this region is shown in Fig. 7. The region below 0.235 keV was not studied until several half-lives later because of its high intensity. The spectrum observed at that time is shown in Fig. 8. These lines could not be identified unambiguously, but from their energies and the fact that they decayed with a complex half-life, it is thought that they are associated with the LXY Auger and $L_{iL_{j}X}$ Coster-Krönig transitions. We cannot, however, rule out the presence of a very low energy conversion line, mixed in with the Auger lines. From this experiment, we can say that there is no <u>intense</u> transition in Mo⁹⁰ decay with conversion lines in the region 2.5-10 keV; below 2.5 keV the results are not definitive.

D. Multipolarities of the 122- and 257-keV Transitions

As pointed out in Sec. I, the multipolarities of the 122- and 257-keV transitions are crucial to the interpretation of the Nb⁹⁰ level scheme. In an effort to determine these multipolarities unambiguously we have made careful measurements of the L-subshell conversion intensity ratios with the iron-free spectrometer. The L-subshell conversion intensity ratios are a very sensitive indicator of the multipolarity of a transition and have always been definitive whenever the L lines could be resolved. However, the L-subshell-ratios method has not in the past been used for low-Z nuclei because of the difficulty in resolving the lines. In this particular case the L lines were not completely resolved, but with the accumulation of good counting statistics the complex structure of the line could be analyzed so as to characterize the multipolarity of the corresponding transitions.

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The following method was used to make comparisons between the experimental L-subshell ratios and the theoretical ratios for various multipolarities: The experimental composite line was plotted on semi-log paper after background subtraction, and compared with a "theoretical line" constructed by using the theoretical L-subshell conversion coefficients of Sliv.¹² (The individual theoretical L-subshell conversion coefficients for the 122-keV transitions were interpolated from log-log plots of Sliv's conversion coefficients as a function of gamma energy; for the 257-keV transition the theoretical values for k=0.5 were used.) The theoretical composite L-line was constructed as follows: First the current position of the L,th line was determined from the current position of the K line and the known K-L, electron binding energy difference. (Since the difference in the K and L natural line widths is less than 5 eV, 13 it was assumed that the L-line shapes are the same as that of the closest K line. In the case of the 122-keV L group, this was the K line of the 142-keV transition in Zr⁹⁰ (Fig. 9), and for the 257-keV L group it was the 257-keV K line.) With use of the experimental line shape, the theoretical composite line was constructed with relative intensities of the individual L-lines in the composite taken from the theoretical L-subshell ratios. For the comparison with experiment, this theoretical composite L line was adjusted along the ordinate axis until the best fit of this line with the experimental data points was obtained. The validity of this method of analysis is shown in Fig. 10, where a comparison is made between the theoretically-constructed L group for the known 132-keV E3 transition in Zr^{90} and the experimental L-group points. The agreement is seen to be excellent.

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The experimental points for the 122- and 257-keV L lines are compared in a similar manner to the theoretical composite lines constructed for various multipolarities, and the results are shown in Figs. 11 to 14. This analysis

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determines unambiguously that the multipolarities of the 122- and 257-keV transitions are E2 and E3, respectively. In addition, the K/L ratio obtained for the 122-keV transition, 5.76, and that for the 257-keV transition, 5.64, agree very well with the corresponding theoretical K/L ratios. Table II contains a summary of the experimental and theoretical information on these transitions, other than the L-subshell data, which was not obtained in numerical form.

E. EC/β^+ Ratio Determination

The electron capture-to-positron ratio of Mo⁹⁰ decay was determined by measuring the intensity of the 122-keV gamma ray (100% transition abundance) relative to the 511-keV annihilation radiation. The source was located 20 cm from a Ge(Li) detector, 2 cm² × 7-mm deep, and was sandwiched between two lucite absorbers to localize the creation of the annihilation radiation so that the same solid angle would be subtended by the gamma rays and the annihilation radiation. Correction was made for the contribution to the annihilation peak from the daughter Nb⁹⁰ decay. An experimental photopeak relative efficiency curve (Fig. 15) for the Ge(Li) detector used (with a source-to-detector distance of 20 cm) was obtained by using the known equal intensities of the 122-and 257-keV cascade transitions in Mo⁹⁰ decay and the 262- and 685-keV cascade transitions in Mo^{93m} as standards.

The value so obtained for the EC/β^+ is 3.0 ± 0.5. This may be compared to the value of 1.9 ± 0.5 obtained from the curves of Feenberg and Trigg.⁴ The agreement is not unsatisfactory if one considers that there is possibly some pure electron capture to high-lying states. Ten percent pure electron capture to upper levels (which is reasonable in terms of observed gamma-ray relative

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intensities) and subsequent cascade through the 122-keV state would give a value of 2.5 ± 0.5 .

The value for the EC/β^+ , 3.0, together with the half-life, 5.7 hours,¹ and the positron end point, 1.15 MeV,² determine a log ft value of 5.5 ± 0.3 for the Mo⁹⁰ positron decay. This suggests an allowed or first-forbidden (non-unique) decay from the 0+ ground state of Mo⁹⁰ to either a 0± or 1± state in Nb⁹⁰.

IV. DISCUSSION

The definite assignment of E2 for the multipolarity of the 122-keV transition leads to a gross inconsistency because the reported half-life of 24 seconds for this transition is a factor of 10^7 longer than that predicted by the single-particle model for an E2 transition of this energy. In addition, there is a problem with the conservation of angular momentum because the sum of the two multipolarities is at least 2 units less than the spin difference (7 to 9 units) between the state initially populated by the decay of Mo⁹⁰ and the ground state of Nb⁹⁰.

To explain the very long half-life for the 122-keV E2 transition and to conserve angular momentum, we propose a decay scheme (Fig. 16) in which a very low energy (less than 3 keV) transition is postulated as preceding the 122-keV transition. The 24 second half-life is then assigned to this very low energy transition and not to the 122-keV transition.

Let us consider the arguments bearing on spin assignments of the states in the main gamma cascade. The ground state of Nb^{90} decays by more than 90% to the 8+ state in Zr^{90} and less than 0.1% to the 6+ state.³ The log ft value, 6.0,³ indicates either an allowed or first-forbidden transition. According to

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the shell model, the lowest-lying configuration in Nb⁹⁰ (41 protons, 49 neutrons) should be the even parity configuration $\pi(g_{9/2})^{1}\nu(g_{9/2})^{-1}$. Thus the transition from Nb⁹⁰ to the 8+ state in Zr⁹⁰ is probably an allowed transition. The above considerations exclude all spins except 8+ or 9+ for the ground state of Nb⁹⁰.

The Brennan-Bernstein rule¹⁴ (which is based on experimental information and delta-force calculations by Schwartz¹⁵) that one less than the maximum spin lies lowest for odd-odd, particle-hole nuclei, suggests that the ground state of Nb⁹⁰ has spin 8 and even parity. Other calculations by Kim and Rasmussen,¹⁶ which include a tensor force, suggest that the 8+ state lies lowest and that the next lowest state is 6+. We choose these assignments for the lowest two states.

The log ft value of 5.5 for the decay of Mo^{90} signifies either an allowed or first-forbidden (non-unique) decay from the 0+ ground state of Mo^{90} (42 protons, 48 neutrons) to either a 0± or 1± state in Nb⁹⁰.

The proposed low-energy gamma transition is in cascade with 122-keV and 257-keV transitions, so its total intensity must be approximately equal to that of the 122- and 257-keV transitions. Since neither a gamma ray nor an electron line of intensity comparable to that of the 122-keV transition was observed above 2.5 keV an upper limit of 3 keV can be set for the energy of this transition. Conservation of angular momentum and parity considerations help us restrict the possible multipolarities of the 24-second isomeric transition. If the state receiving the beta decay is 1+ and Nb⁹⁰ ground state is 8+, an M2 assignment (with possible E3 admixture) is demanded for the 24-second transition in cascade with the E3 and E2 transitions. If the initial state in the cascade is 1-, an E2 assignment is indicated. If the initial state is 0±, an octupole assignment is indicated.

Experimental evidence¹⁷ on the levels in Nb⁹¹ (41 protons, 50 neutrons) shows an energy separation between the $p_{1/2}$ and $g_{9/2}$ proton orbitals of only about 100 keV, whereas the separation between the $p_{3/2}$ and $g_{9/2}$ orbitals is about 1.3 MeV. The experimental evidence¹⁸ on the levels in Zr^{89} (40 protons, 49 neutrons) gives the energy separation between the $p_{1/2}$ and $g_{9/2}$ neutron orbitals as about 600 keV and that for the $p_{3/2}$ somewhat greater. One therefore expects excited states involving the $p_{3/2}$ orbitals to lie higher in the Nb⁹⁰ spectrum than those arising from the $p_{1/2}$ orbitals. The coupling of a $p_{1/2}$ proton (neutron) and $g_{9/2}$ neutron (proton) can lead to 4- or 5- states, and the Nordheim strong rule¹⁹ and that of de-Shalit and Walecka²⁰ puts the 4- level lower. We postulate that the next level above the 6+ in the main cascade of Nb⁹⁰ is of this character with spin 4 and odd parity. Admittedly, the assignment of 4- on shell-model grounds is the weakest link in our chain of reasoning; from the experimental data alone alternative assignments of 3-, 3+, and 4+ cannot be excluded.

The level assigned as 4- is fed by the 257-keV E3 transition from the state that receives primary beta decay. The spin of the latter has already been restricted by ft value considerations to values 0 or 1, but of these only the assignment 1+ is consistent with the 4- assignment. There are several ways to couple the lower-lying $g_{9/2}$, $p_{1/2}$ and $p_{3/2}$ orbitals to form 1+, and it is not obvious which configurations will be predominant. From Table II we see that the E3 transition rate to the 4- state is five times faster than the single-particle rate, according to the graphs of Wapstra et al.²¹ Of the 40 different E3 transitions plotted by Goldhaber and Sunyar,²² only 3 are enhanced; thus the unusual speed of this E3 transition suggests that there is considerable configuration mixing in the 1+ and 4- states of Nb⁹⁰. Descriptions of these states involving collective octupole phonon excitations may be worth exploring.

Yttrium-88 (39 protons, 49 neutrons), which also has an unusually fast E3 transition (factor of 10 over single-proton rate) from a 1+ isomeric state to a 4- ground state, has the same proton and neutron orbitals available as Nb^{90} . This similarity in E3 transition-rate enhancement lends support to our proposed 1+ and 4- level assignments.

The 24-second half-life of the proposed very low-energy transition (< 3 keV) is not unusually short. The gamma-ray transition probability does decrease rapidly with decreasing transition energy but there is also a corresponding increase in the probability of decay by internal conversion. These opposing trends keep the half-life from becoming very long at very low energies. A case in point of this is the 26-min isomeric state in U²³⁵ which is < 1 keV above the ground state.²³

In conclusion, the data reported here have established unambiguously the multipolarities of the two most prominent transitions in Nb⁹⁰ to be E2 and E3. The present experimental and theoretical information strongly suggests a level scheme for Nb⁹⁰ like that of Fig. 16 in which a very low energy transition, less than 3 keV, from a 4- state to a 6+ state has been proposed to conserve angular momentum and parity, and to explain the anomalous half-life of the 122-keV E2 transition.

FOOTNOTES AND REFERENCES

*Thi	s work was supported by the U.S. Atomic Energy Commission.
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Energy (keV)	Relative intensity ^b
42.76 ^a ± 0.08	0.4
122.50 ^a ± 0.1	84.
162.99 ^a ± 0.1	5.8
202.9 ± 0.5	6.6
257.52 ^a ± 0.15	100.
323.0 ± 0.5	8.9
≈420.	≈l. (complex?)
445.2 ± 1.	11.
472.5 ± 1.	2.4
≈490. ± 2.	≈2. (complex?)
945.2 ± 1.5	12.
992.7 ± 2.	2.4
1272.5 ± 2.	9 . 4
1389.0 ± 2.	5.2
1455. ± 3.	3.6
1463. ± 3.	0.6

Table I. Photons of Mo⁹⁰ decay.

^aEnergy values determined from conversion-electron spectrum. Calibration of the iron-free spectrometer was made with K line of the 208.36 ± 0.2-keV transition from Lu¹⁷⁷ decay (P. Marmier, F. Boehm, Phys. Rev. <u>97</u>, 103 (1957)).

^bThe relative accuracy of the quoted intensity figures is thought to vary from 10% for the strongest lines to as much as a factor of two for the weaker lines.

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Fig. 2d. Partial gamma-ray spectrum of Mo^{90} , observed with 6 cm² × 9-mm deep Ge(Li) detector system.

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Fig. 3. Gamma-ray photopeak efficiency curve for the 6 cm² \times 9-mm deep Ge(Li) detector system used for the gamma-ray measurements.

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Fig. 4. Internal-conversion-electron spectrum of Mo⁹⁰ (including Mo⁹³ and Nb⁹⁰), recorded with 180° permanent-magnet spectrograph.



Spectrometer current (A)

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Fig. 5. K line of the 42.76-keV transition from Mo⁹⁰ decay measured with 50-cm iron-free $\pi\sqrt{2}$ spectrometer, with and without 4.69 kV preaccelerator voltage.

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Fig. 10. Comparison of experimental and theoretical L-group line shapes for 132-keV E3 transition from Nb⁹⁰ decay. Normalization procedure is described in text.



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Fig. 11 Comparison of experimental L-group line shape of 122-keV transition of Mo⁹⁰ decay with theoretical composite L-group lines for M2, M3, and M⁴ multipolarities.

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Fig. 12. Comparison of experimental L-group line shape of 122-keV transition of Mo⁹⁰ decay with theoretical composite L-group lines for E1, E2, and E3 multipolarities.



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Fig. 13. Comparison of experimental L-group line shape of 257-keV transition of Mo⁹⁰ decay with theoretical composite L-group lines for M2, M3, and M⁴ multipolarities.

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Fig. 14. Comparison of experimental L-group line shape of 257-keV transition of Mo⁹⁰ decay with theoretical composite L-group lines for E2, E3, and E4 multipolarities.







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Fig. 16. Partial decay scheme of Mo⁹⁰, showing postulated low-energy transition. The preferred state assignments are underlined.

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