UCRL-16306

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VERY LOW ENERGY ISOMER IN THE DECAY OF Mo^{90}

Berkeley, California

Talk given by J. M. Hollander at the International Conference on the Internal Conversion Process, Vanderbilt University, Nashville, May 10-13, 1965.

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AEC Contract No. W-7405-eng-48

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A reinvestigation of Mo^{90} was undertaken in order to resolve some inconsistencies in the decay scheme of this 5.7 hour activity. The status of the decay scheme, as it was, is summarized in Fig. 1. From the Mo^{90} log ft values,[1] the spin of the 380-keV level of Nb^{90} had been assigned as 0 or 1, and from the Nb^{90} decay properties, the Nb^{90} ground state was considered to have spin 8 or 9. Only two transitions had been observed in the Mo^{90} decay, and these were found to be isomeric, with half-lives 10 msec(250 keV) and 24 sec (120 keV).[2] However, the above spin assignments require that at least 7 units of angular momentum be carried off by the two transitions, and this was not consistent with the half-life and preliminary internal conversion data, which had indicated that the transitions are at most octupoles.[2]

In this work, the multipolarities of the 122- and 258-keV transitions were determined unambiguously by measurements of the L-subshell ratios with the 50-cm iron-free spectrometer. The L-subshell conversion ratios are very sensitive indicators of the multipolarities of transitions, and have usually been definitive whenever the L lines could be resolved. In this particular case the L lines were not completely resolved, but the complex structure of the line could be analyzed so as to characterize the multipolarity of the corresponding transition.

The experimental L-subshell ratios were compared with the theoretical ratios by comparing the experimental L-line shape (plotted on semi-log paper) with a "theoretical line" constructed by using the theoretical L-subshell conversion coefficients of Rose[3] and Sliv.[4] (The individual L-subshell conversion coefficients for the 122-keV transition were interpolated from log-log plots of Rose's conversion coefficients as a function of gamma energy.) The theoretical composite L-line shape was constructed as follows: First the position of the L_ith line was determined from the position of the K line and the difference between the K and L_i electron binding energies. (The assumption was made that the L-line shapes are the same as that of the closest K line. In the 122-keV L group, this was the K line of the 142-keV transition in Zr^{90}, Fig. 2, and for the 257-keV L group it was the 257-keV K line.) The relative intensities of the L lines in the composite line were adjusted to agree with the theoretical L-sub-shell ratios. For the comparison with experiment, this theoretical composite L line was adjusted along the ordinate axis until the best fit of the experimental data points was obtained. The validity of this method of analysis is shown in Fig. 3, 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.

The experimental points for the 122- and 257-keV transitions are compared in a similar manner (Figs. 4 through 7) to the theoretical composite lines constructed for different multipolarities. This analysis determines unambiguously the multipolarities of the 122- and 257-keV transitions to be E2 and E3, respectively. In addition, the K/L ratios obtained for the 122-keV transition, 5.76, and for the 257-keV transition, 5.64, agree very well with this assignment:

But the reported half-life of 24 seconds for the 122-keV transition is longer by a factor of 10^7 than that predicted by the single-particle model for an E2 transition of this energy, and thus a gross inconsistency exists. In addition, the two multipolarity assignments are still inconsistent with the expected large spin difference (7 to 9 units) between the state initially populated by the decay of Mo^{90} and the ground state of Nb^{90}.

To explain the very long half-life measured for the 122-keV E2 transition and to conserve angular momentum, we propose a decay scheme (Fig. 8) in which a very-low-energy (less than 2 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. The most likely multipolarity of the missing low-energy transition is M2, although E3 is also possible.

A search for new transitions in the gamma-ray spectrom revealed many weak lines up to 1500 keV energy but none in sufficient intensity to correspond to the missing 24-second isomeric transition. A search of the internal conversion spectrum also failed to reveal strong conversion lines, above 2 keV. We conclude that the energy of the 24-second isomer is very low, $\lesssim 3$ keV.

It is thus of interest to try to obtain indirect evidence for the isomeric transition. If the transition energy is sufficiently low, the possibility exists of altering the decay constant by changing the chemical environment of the nucleus, as was demonstrated in the case of the 2-keV isomeric transition in Tc^{99m} by Bainbridge, Goldhaber, and Wilson.[5] In that case, a change in decay constant of 0.3% was noted in the comparison of the decay rate of Tc^{99m} in the compounds $KTcO_4$ and Tc_2S_7. In the present case the chemical bonding electrons are in the N and O shells of Nb, with binding energies

$\lesssim$ 50 eV. If the decay energy should be less than 200 eV (binding energy of the M_V shell), internal conversion could take place only in the N and O shells, and in this case appreciable chemical effects might be expected.

Such a chemical effect was demonstrated by an experiment in which an intensity-change of the 122-keV E2 gamma ray (in cascade with the low-energy isomeric transition) was observed following a rapid chemical dissolution with hot HNO_3-HF of Nb metal foils containing Mo^{90} activity. This reaction alters the chemical environment of the 24 sec isomeric state from metallic niobium to a fluoride complex. A four-point growth curve was observed, by recording gamma-ray spectra from a Ge(Li) detector for 18 seconds in each of the four 100 channel blocks of a 400-channel pulse-height analyzer. This experiment was repeated about 200 times to insure the statistical reliability o of the data. The resulting gamma spectrum from one of the 100 channel blocks is shown in Fig. 9.

A plot of the total number of counts under the 122-keV photopeak (Fig.10) shows an exponential growth while the background (133- and 142-keV photopeaks from daughter Nb^{90} activity also present) remains constant. From a difference plot, shown in Fig. 11, a half-life of ~20 seconds was obtained for the isomeric state. By extrapolation of the difference plot back to the time of separation, it was found that the half-life of the isomer had been increased 3.2% by the change of chemical state from metal to fluoride complex. This is believed to be the largest alteration in half-life yet noted for a radioactive isotope.

On the basis of these experiments the decay scheme shown in Fig. 8 is postulated for Mo^{90}.

FOOTNOTES AND REFERENCES

*This work done under the auspices of the U. S. Atomic Energy Commission.

1. J. A. Cooper, J. M. Hollander, M. I. Kalkstein, and J. O. Rasmussen, Nucl. Phys. (in press).

2. H. Mathur and E. K. Hyde, Phys. Rev. 98, 79 (1955).

3. M. E. Rose, Internal Conversion Coefficients, (Interscience Publishers, Inc., New York, 1958).

4. L. A. Sliv and I. M. Band, Coefficients of Internal Conversion of Gamma Radiation, (USSR Academy of Sciences, Moscow-Leningrad, 1956) Part I: K Shell; Part II: L Shell.

5. K. T. Bainbridge, M. Goldhaber, and E. Wilson, Phys. Rev. 90, 430 (1953).

FIGURE CAPTIONS

Fig. 1. Mo^{90} decay scheme as known at the start of this investigation.

Fig. 2. K line of 142-keV transition from Nb^{90} decay, measured with 50-cm iron-free $\pi\sqrt{2}$ spectrometer.

Fig. 3. Comparison of experimental and theoretical L-group line shapes for 132-keV E3 transition from Nb^{90} decay. Normalization procedure is described in text.

Fig. 4. Comparison of experimental L-group line shape of 122-keV transition of Mo^{90} decay with theoretical composite L-group line for M2 (---), M3 (——), and M4 (···) multipolarities.

Fig. 5. Comparison of experimental L-group line shape of 122-keV transition of Mo^{90} decay with theoretical composite L-group line for E1 (– –), E2 (——), and E3 (---) multipolarities.

Fig. 6. Comparison of experimental L-group line shape of 257-keV transition of Mo^{90} decay with theoretical composite L-group line for M2 (– –), M3 (——), and M4 (---) multipolarities.

Fig. 7. Comparison of experimental L-group line shape of 257-keV transition of Mo^{90} decay with theoretical composite L-group line for E2 (– –), E3 (——), and E4 (---) multipolarities.

Fig. 8. Postulated decay scheme of Mo^{90}.

Fig. 9. One of the 4 gamma-ray spectra resulting from about 200 experiments.

Fig. 10. Growth curve of 24-second Nb^{90} isomer back into equilibrium with Mo^{90} parent after dissolution of niobium target foils in HNO_3-HF.

Fig. 11. Difference plot made from growth curve of Fig. 9, showing approximately correct value of isomeric state half-life.

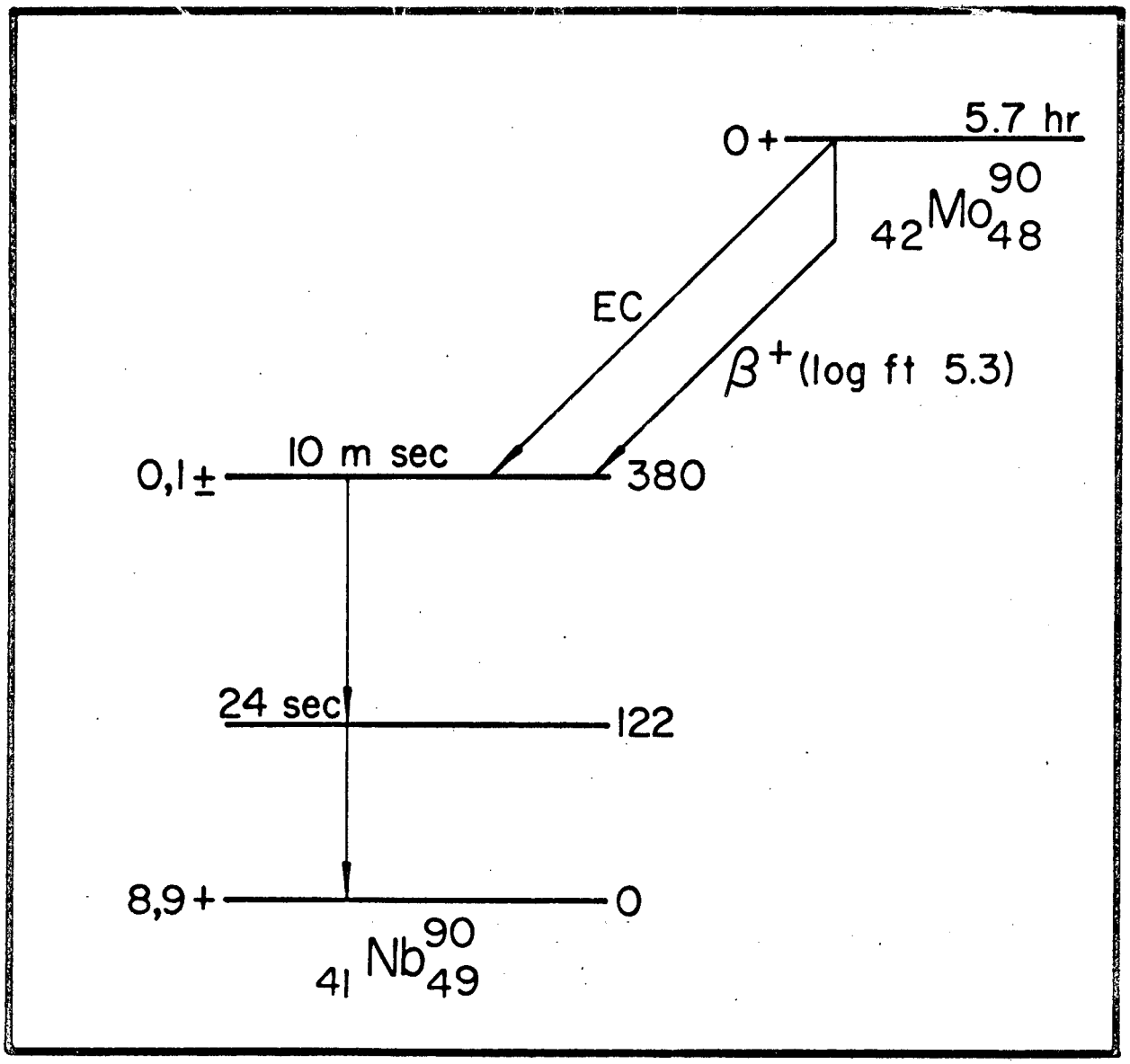


Figure 1

MUB-6593

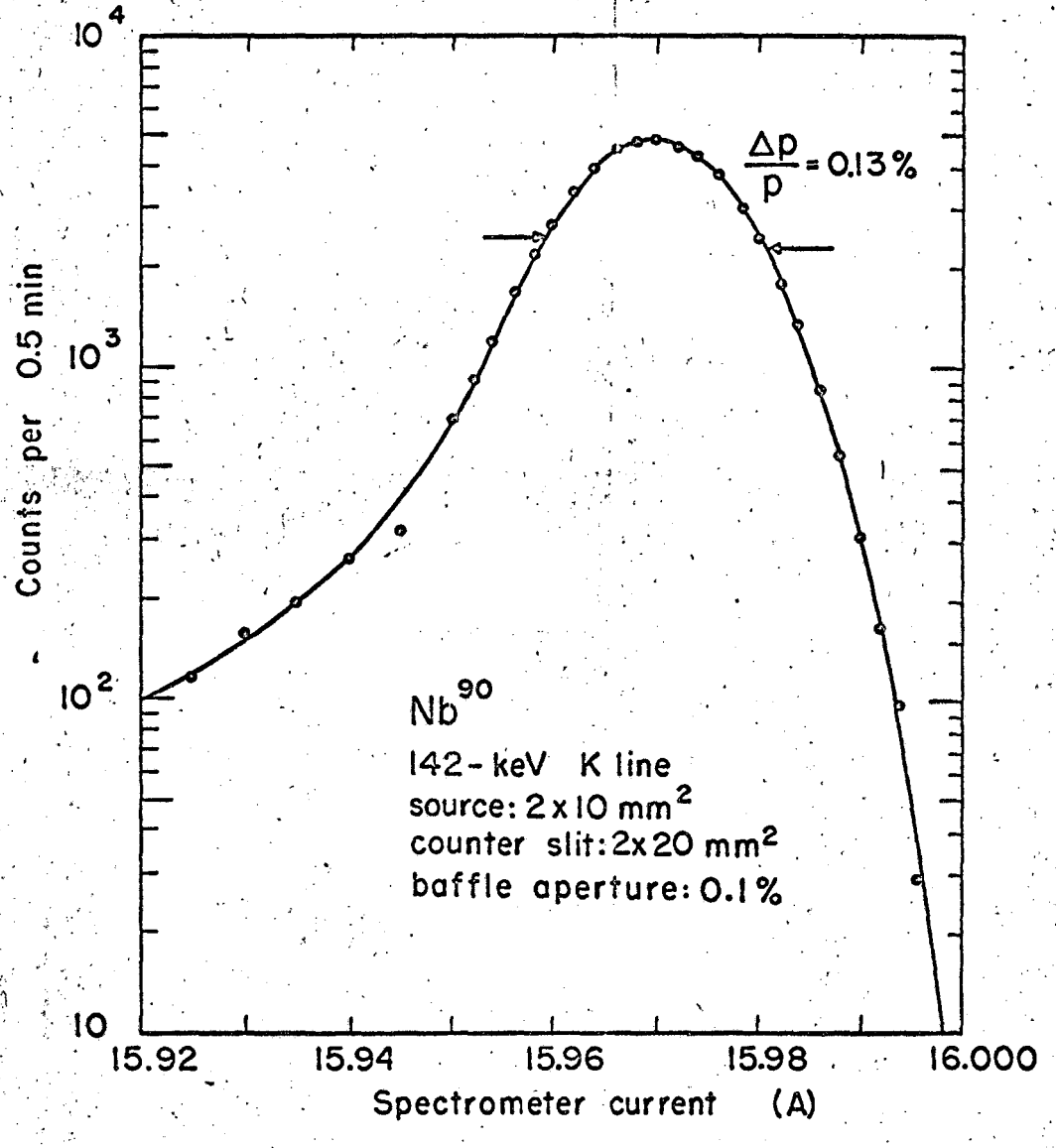

10^4
10^3
10^2
10
Counts per 0.5 min
$\frac{\Delta p}{p} = 0.13\%$
Nb^{90}
142-keV K line
source: 2x10 mm^2
counter slit: 2x20 mm^2
baffle aperture: 0.1%
15.92
15.94
15.96
15.98
16.000
Spectrometer current (A)
MUB-5030

Figure 2

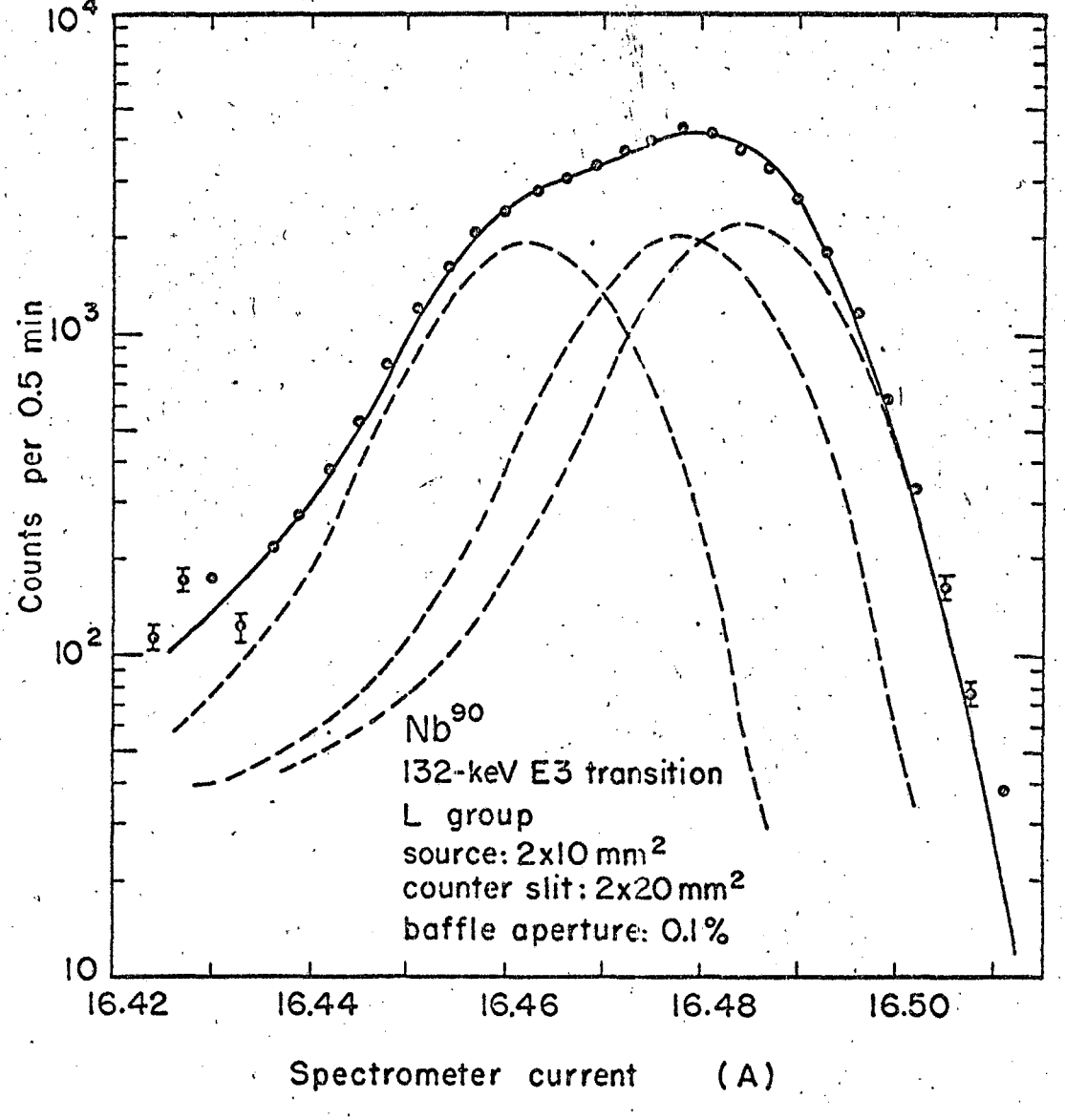

Counts per 0.5 min
10^4
10^3
10^2
10
16.42
16.44
16.46
16.48
16.50
Spectrometer current (A)
Nb^{90}
132-keV E3 transition
L group
source: 2x10 mm²
counter slit: 2x20 mm²
baffle aperture: 0.1%
MUB-5031

Figure 3

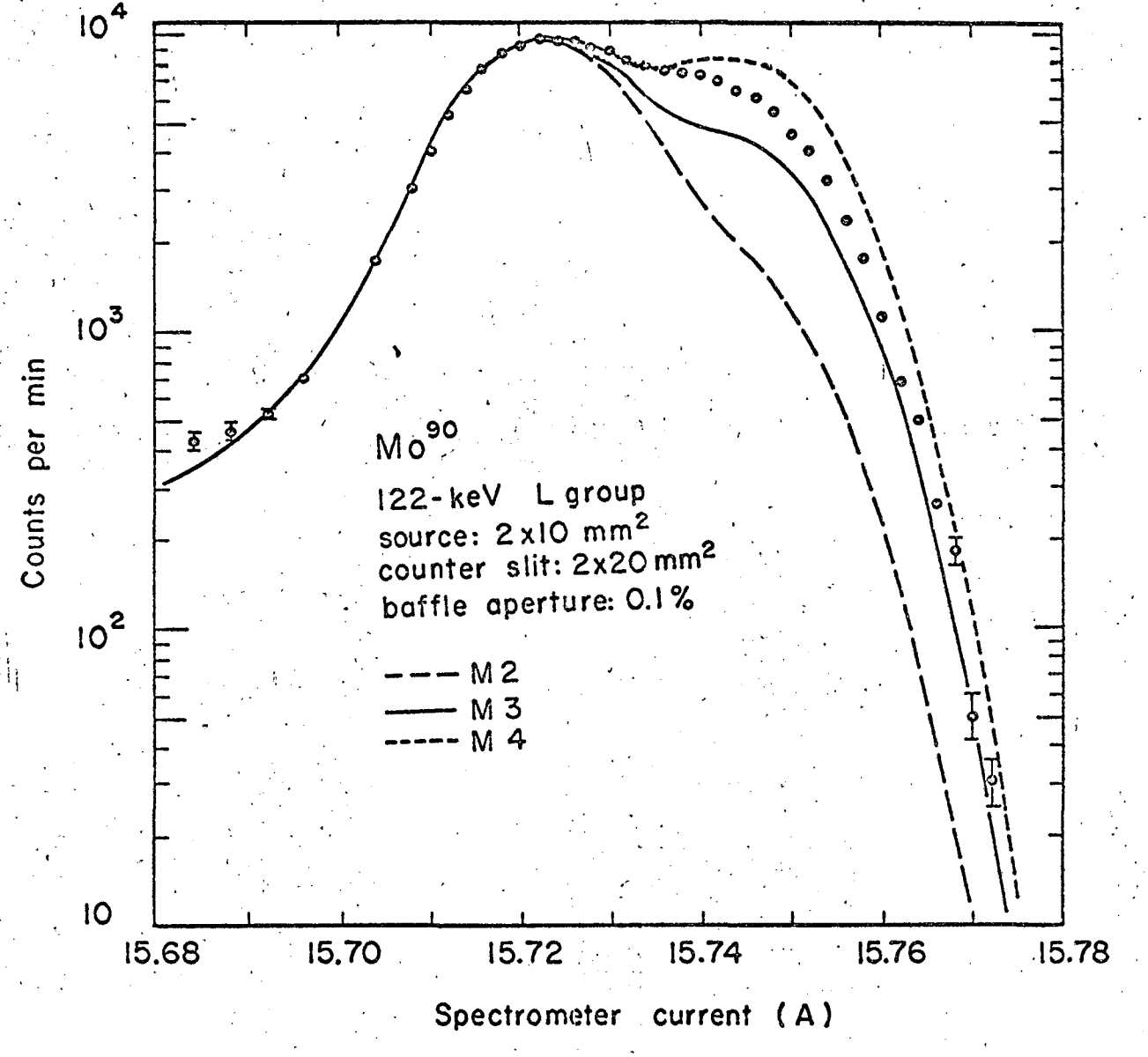

Counts per min
10^4
10^3
10^2
10
Mo^{90}
122-keV L group
source: 2x10 mm²
counter slit: 2x20 mm²
baffle aperture: 0.1%
M 2
M 3
M 4
15.68
15.70
15.72
15.74
15.76
15.78
Spectrometer current (A)
MUB-5033-A

Figure 4

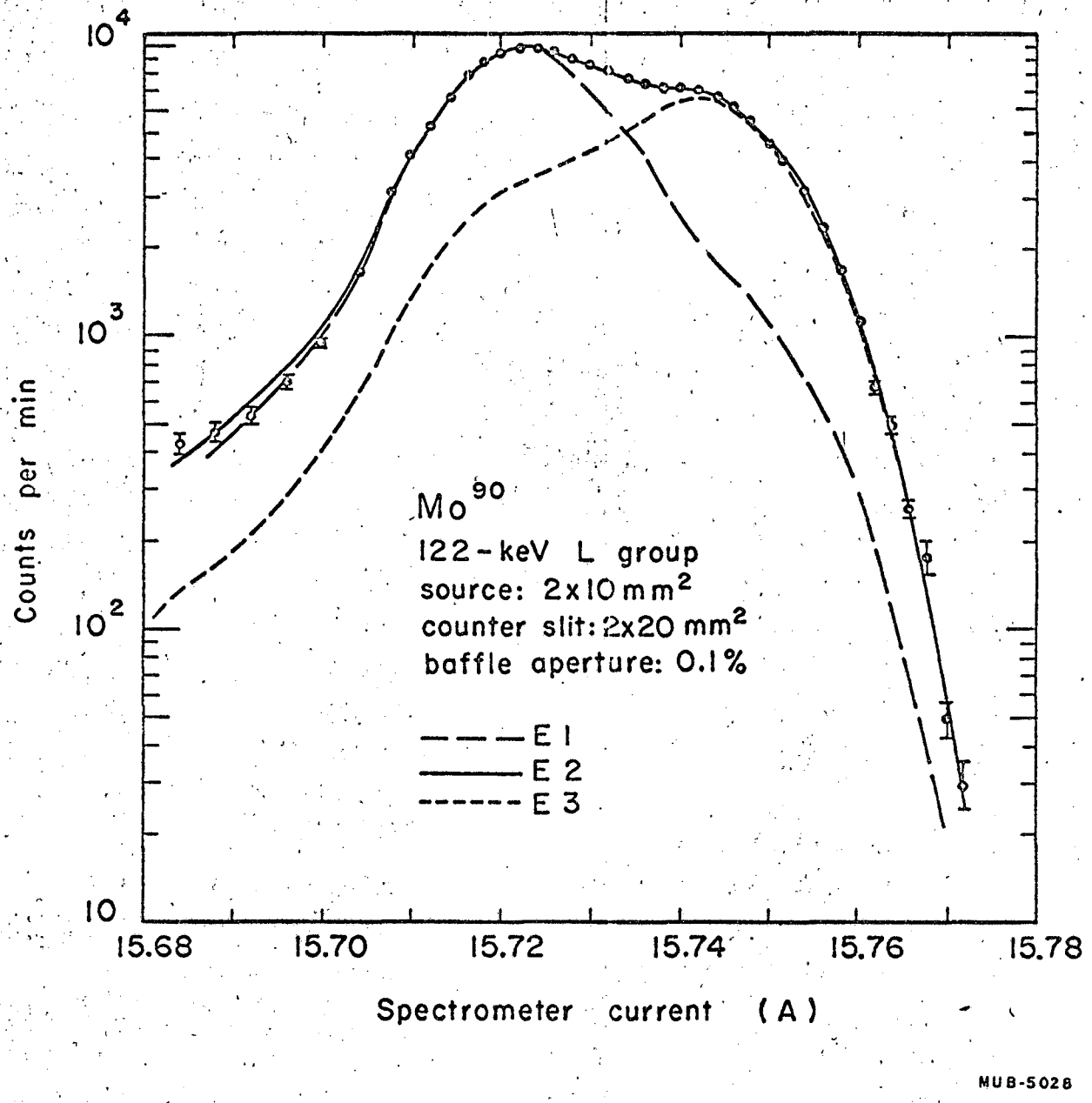

10^4
10^3
10^2
10
Counts per min
Mo^{90}
122-keV L group
source: 2x10 mm²
counter slit: 2x20 mm²
baffle aperture: 0.1%
E 1
E 2
E 3
15.68
15.70
15.72
15.74
15.76
15.78
Spectrometer current (A)
MUB-5028

Figure 5

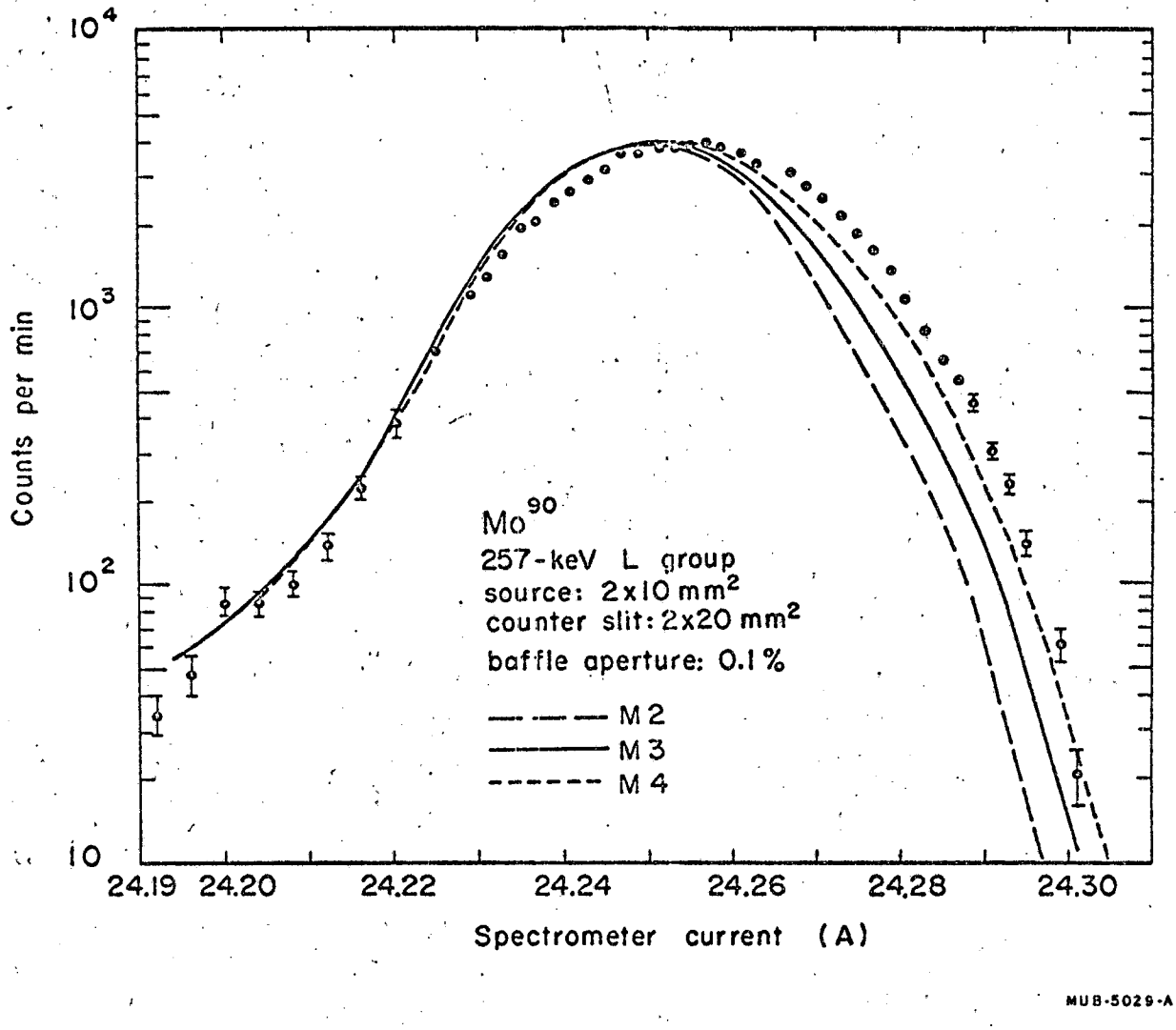

10^4
10^3
10^2
10
Counts per min
Mo^{90}
257-keV L group
source: 2x10 mm²
counter slit: 2x20 mm²
baffle aperture: 0.1 %
M 2
M 3
M 4
24.19
24.20
24.22
24.24
24.26
24.28
24.30
Spectrometer current (A)
MUB-5029-A

Figure 6

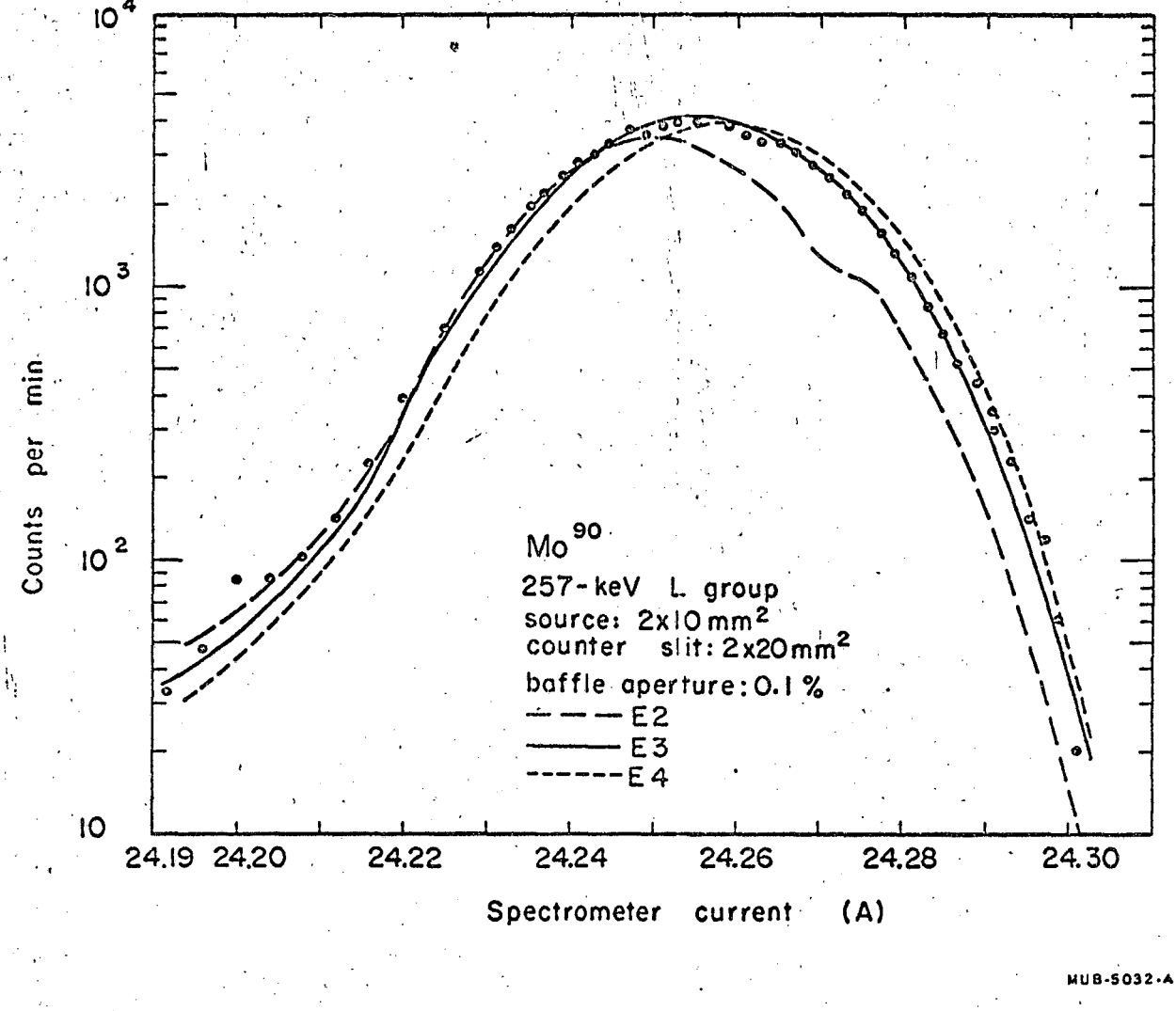
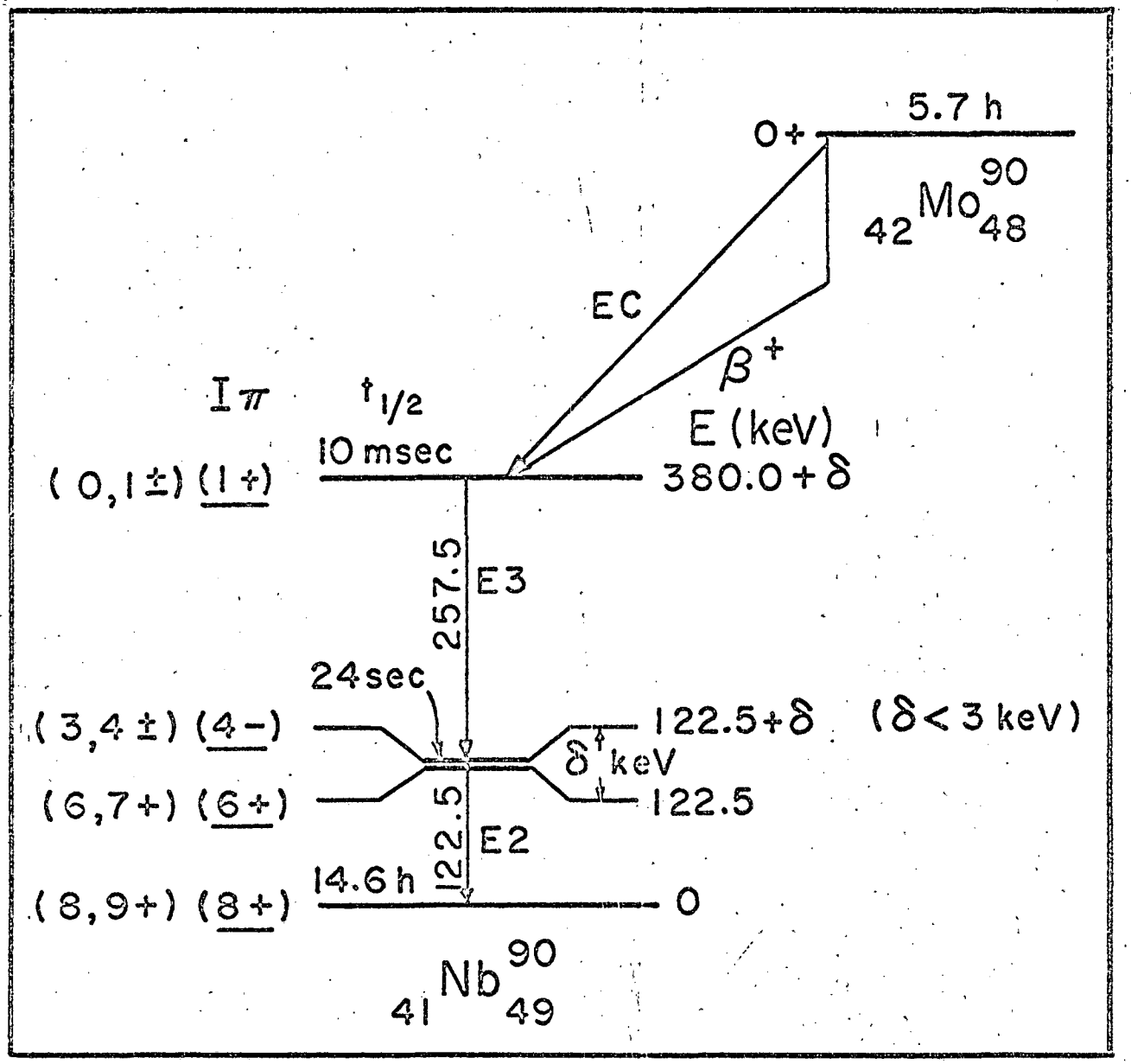

10^4
10^3
10^2
10
Counts per min
Mo^{90}
257-keV L group
source: 2x10 mm²
counter slit: 2x20 mm²
baffle aperture: 0.1 %
E2
E3
E4
24.19
24.20
24.22
24.24
24.26
24.28
24.30
Spectrometer current (A)
MUB-5032-A

Figure 7



MUB-5035

Figure 8

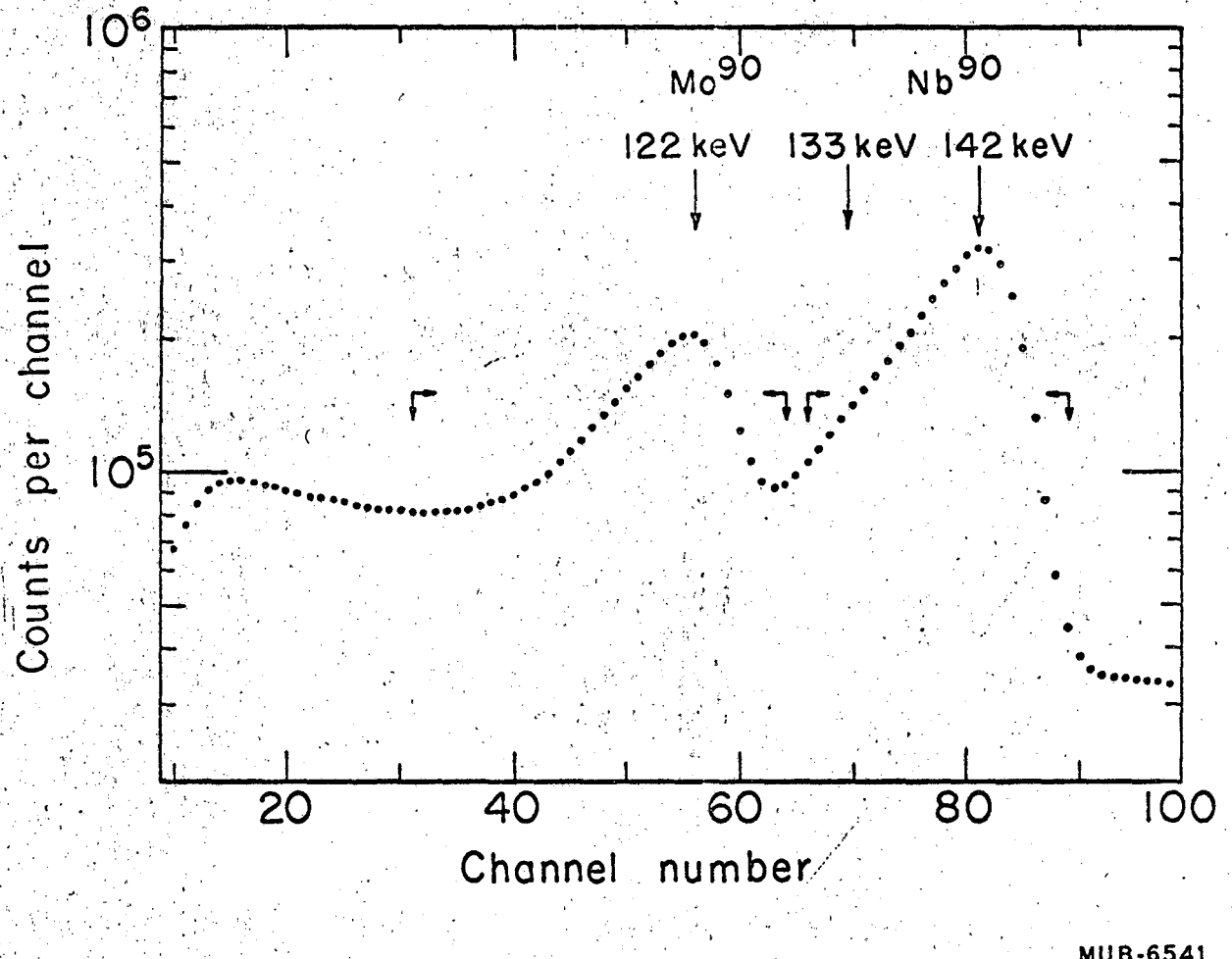

10^6
10^5
Counts per channel
Mo^{90}
Nb^{90}
122 keV
133 keV
142 keV
20
40
60
80
100
Channel number
MUB-6541

Figure 9

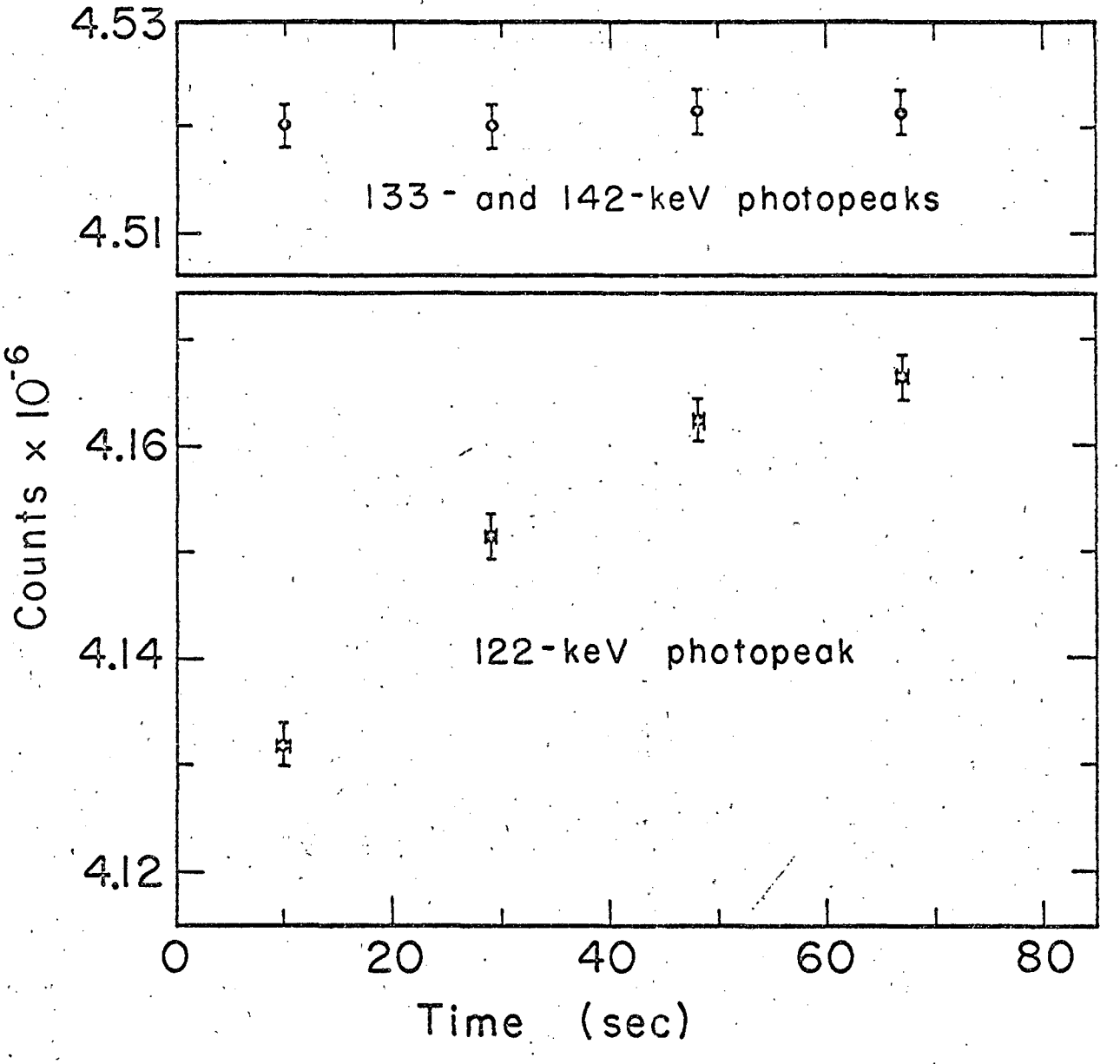

4.53
4.51
133 - and 142-keV photopeaks
4.16
4.14
122-keV photopeak
4.12
Counts x 10^-6
0
20
40
60
80
Time (sec)
MUB-6539

Figure 10

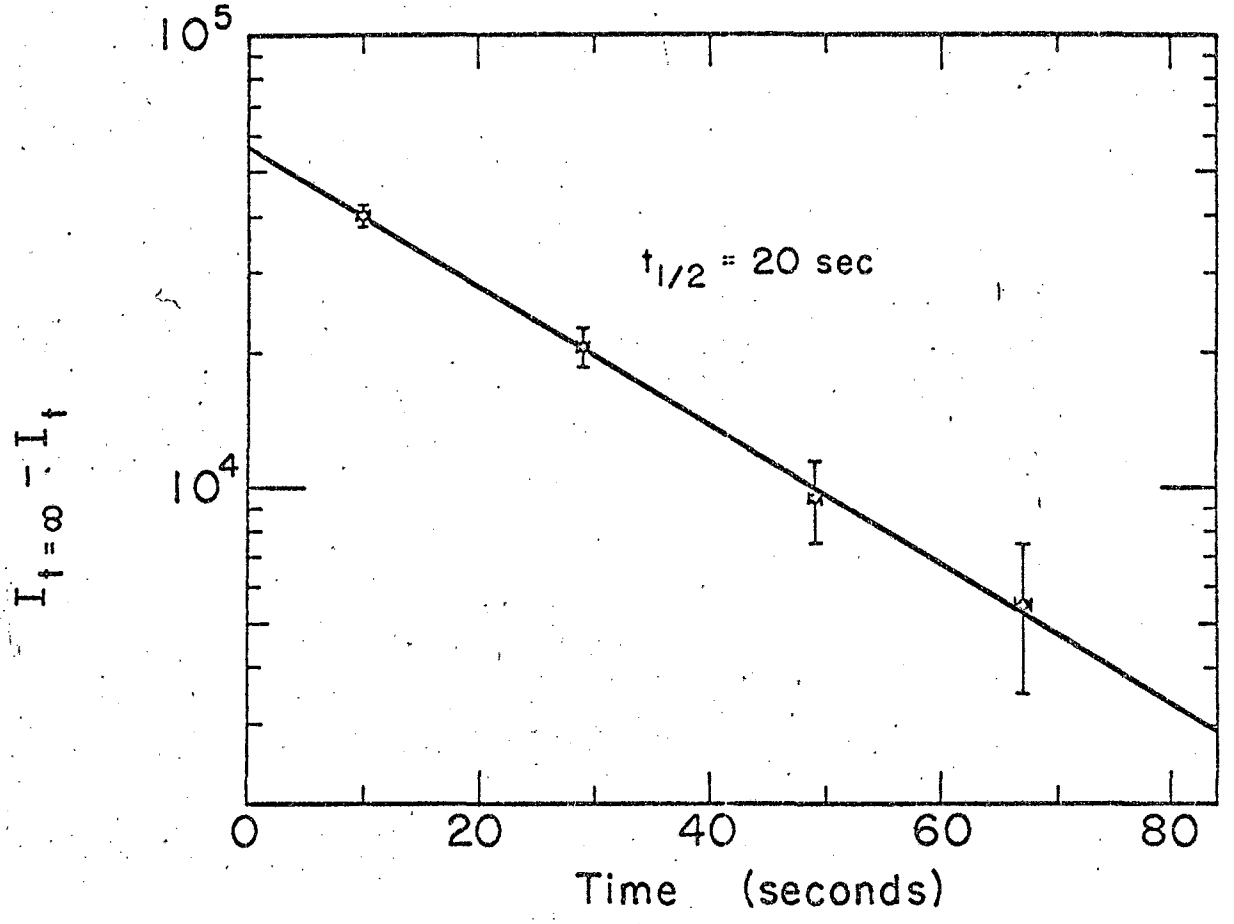


Figure 11