citation functions. Inelastic angular distributions for the particle-hole states at the $f_{7/2}$, $p_{3/2}$, and first $f_{5/2}$ analog resonances were fit with a single-level R-matrix formula expressed in a j-j coupling scheme in order to determine hole configurations, assuming that only $s_{1/2}$ and $d_{3/2}$ holes are involved. Proton inelastic partial widths determined from the fits were used to calculate $s_{1/2}$ and $d_{3/2}$ spectroscopic factors for the particle-hole states, the degree of admixture of the hole configuration in the structure of a given state being determined by the relative magnitude of the respective $s_{1/2}$ and $d_{3/2}$ spectroscopic factors. Using the results of this analysis in conjunction with the resonant behavior of the corresponding inelastic excitation functions, it was possible to determine the particle-hole configurations and probable final spin assignments for 17 particle-hole excited states in ^{136}Xe.

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Sensitivity of the Half-Life of Nb^{90m} to Chemical Environment*

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Changes in the decay constant of the isomer Nb^{90m} were measured. We found that $(\lambda_{\mathrm{Nb\ (fluoride\ complex)}}-\lambda_{\mathrm{Nb}})/\lambda_{\mathrm{Nb}}=(-3.9\pm0.8)\times10^{-2}$; $(\lambda_{\mathrm{Nb_2O_5}}-\lambda_{\mathrm{Nb}})/\lambda_{\mathrm{Nb}}=(-1.9\pm0.5)\times10^{-2}$. The Nb (fluoride complex) result confirms the value previously obtained by Cooper *et al.*, which had been questioned because of the negative result obtained by Weirauch *et al.* in a similar measurement. Some possible causes of this high sensitivity to chemical environment are discussed.

INTRODUCTION

There has been recent interest in the sensitivity of the half-life of the 20-sec isomer Nb^{90m} to different chemical environments. This isomer is believed to decay through a highly converted 2.4-keV $M2$ transition, although evidence for this is indirect.[1] This transition is followed by a prompt 122.4-keV $E2$ γ ray (see Fig. 1). The isomeric state is populated by the decay of 5.7-h Mo^{90}, and may be produced directly by the reaction $Zr^{90}(d, 2n)Nb^{90m}$.

Cooper *et al.*[2] have reported a 3.6% increase in the half-life when a niobium foil containing Nb^{90m} is dissolved in a 2:1 mixture of concentrated HF and HNO_3. This is by an order of magnitude the largest change in half-life observed in any isomer. Weirauch *et al.*[3] have measured directly the half-life of Nb^{90m} in Zr metal, and in a fluoride complex produced by dissolving a Zr foil containing Nb^{90m} in a mixture of HF and HNO_3 containing dissolved Nb powder. They were unable to find any difference in the half-life with an experimental error of 1%. One might attribute this discrepancy to the slightly different chemical environment used by each group. However, there was also a difference in the techniques of the various workers.

Motivated by the large environmental sensitivity reported by Cooper *et al.*, we recently measured a small change in the Nb^{90m} half-life induced by superconductivity.[4] In view of Weirauch's measurements, we decided to make an independent check of Cooper's result, taking particular account of source geometry changes due to the source diffusing through the sample volume, and also the possibility of volatile Nb compounds evolving after the reaction. We have also observed a difference in half-life between Nb metal and Nb_2O_5.

METHOD

The change in half-life of the 20-sec isomeric state was measured with the Nb^{90m} in equilibrium with its parent Mo^{90}. This is the method first used by Cooper *et al.*[2] A sudden increase in the Nb^{90m} decay rate will increase the intensity of the 122-keV line; this will decay back with a 20-sec half-life to reestablish radioactive equilibrium with the parent Mo^{90}. If the decay

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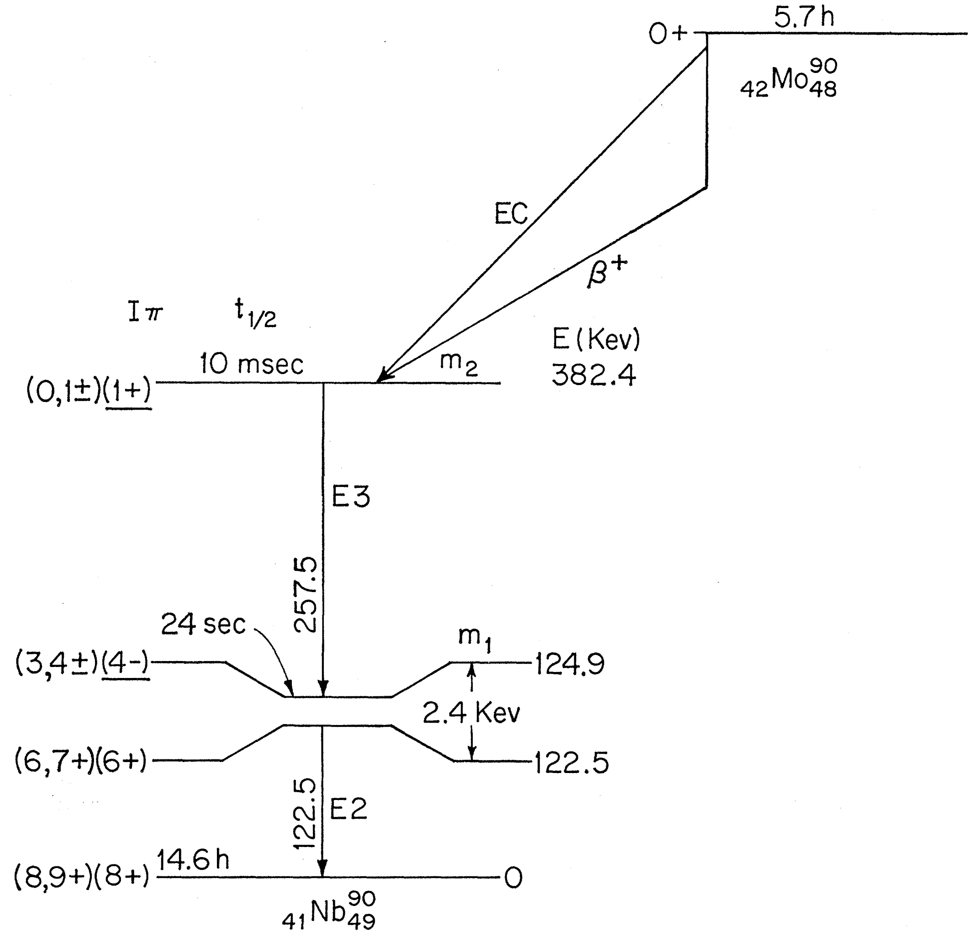

FIG. 1. Partial decay scheme of Mo^{90} as proposed by Cooper *et al.* (Ref. 2). The first metastable state (Nb^{90m}) at 124.9 keV is indicated by m_1 and the second metastable state (Nb^{90m_2}) at 382.4 keV by m_2.

rate changes from λ_2 to $\lambda_2+\Delta\lambda_2$ at $t=0$, we have

$$I=(\lambda_2+\Delta\lambda_2)N_2=A\exp(-\lambda_1 t)+B\exp(-\lambda_2 t), \quad (1)$$

$$A=\lambda_1 N_1(1+\lambda_1/\lambda_2), \quad (2)$$

$$B=(\Delta\lambda_2/\lambda_2)\lambda_1 N_1(1-\Delta\lambda_2 t), \quad (3)$$

Then

$$\Delta\lambda_2/\lambda_2 \approx B/A. \quad (4)$$

Here, I is the intensity of the 122-keV line, λ_1 and λ_2 are the decay rates, and N_1 and N_2 are the number of atoms of Mo^{90} and Nb^{90m}, respectively.

The intensity of the 122-keV line was counted with a thin Ge(Li) crystal by setting a 6-keV window on the peak and multiscaling at 1 channel/sec for 100 sec. Corrections must be made for the presence of Compton background in the window and for the effect of pile-up due to the high count rates used. A second multiscalar scan was taken 160 sec after the chemical reaction was completed. A straight line was fitted to the data from this second scan to obtain the effective decay rate of the peak, including the effect of background decay and pile-up. This calculated straight line was subtracted from the first scan, and then a fit was made to (1). This procedure is valid because the half-lives involved in the Mo^{90} decay and Compton background decay are long compared with 200 sec. From the fit to (1), B is determined. The constant A is, to a good approximation, the average count rate in the window from the 122-keV photopeak. It is calculated from the average count rate of the first multiscalar scan and from the peak-to-Compton-background ratio determined from a pulse-height analysis scan. Then $\Delta\lambda_2/\lambda_2$ is calculated from (4).

The channels during which the chemical reaction occurs could be easily seen by the discontinuity in the counting rate due to changing source geometry. This is used to determine the channel for which $t=0$ in (1). Calculations of the least-squares fit include only the data obtained after source conditions are stabilized.

The intensity of the 140-keV Zr^{90} γ peak was scanned on a second multichannel analyzer simultaneously with the 122-keV peak, and the scans received the same method of analysis. This γ ray arises from the decay of ground-state Nb^{90}, so that transients due to changing

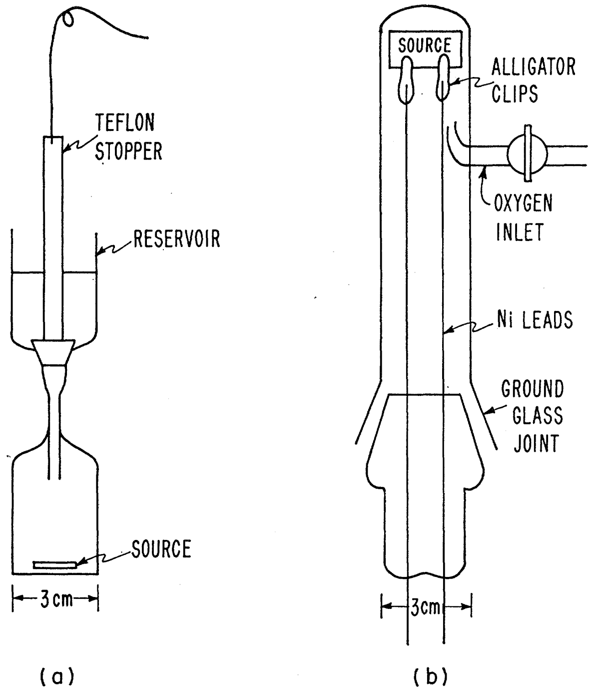

FIG. 2. (a) Apparatus for preparation of Nb (fluoride complex). (b) Apparatus for preparation of Nb_2O_5.

source geometry and escape of volatile Nb compounds would alter the intensity of this line in the same way that the 122-keV intensity would be altered. These effects were detected in the formation of the fluoride complex, but not in the oxide.

SOURCE PREPARATION

An 8-cm×1-cm×0.00125-cm strip of Nb foil was irradiated with 50-MeV protons for 2 h in the internal beam of the Harvard Cyclotron. After bombardment it was cut into eight 1-cm squares.

PREPARATION OF Nb (FLUORIDE COMPLEX)

The irradiated Nb foil was placed in a 35-ml polyethylene bottle and etched with 1 ml of HNO_3. [See Fig. 2(a) for apparatus diagram.] HF and HNO_3 were mixed in the proportion 2:1 in a polyethylene reservoir above the sample bottle to make a total volume of 10 ml. After multiscaling commenced the teflon stopper was drawn from the reservoir, allowing the acid to flow into the sample bottle.

A violent reaction ensued for 2 to 5 sec (as judged by the eye) in which the Nb foil dissolved and an orange gas was evolved. During this period one could observe large fluctuations in the count rate attributable to changes in the source geometry. At least 80% of the Nb foil was dissolved after 2 sec in all cases.

The practice of etching the foil in HNO_3 was adopted to make the reaction rates more uniform. When the etch was omitted, the foil sometimes sat inert for 1 to 2 sec after the HF–HNO_3 was added, and then reacted slowly. This probably means that the foil had a surface coat which was removed by etching in HNO_3.

PREPARATION OF Nb_2O_5

To prepare the Nb_2O_5 [see Fig. 2(b) for apparatus diagram], the foils were irradiated as before. The source was placed in a glass tube filled with pure O_2, then was ignited by passing electric current through it. The foil burned at white heat for about 1 sec, and the Nb_2O_5 product sputtered onto the side of the glass tube.

X-ray diffraction measurements on samples made from nonirradiated foils identify the product as the high-temperature phase of Nb_2O_5. The contamination of unburned Nb and NbO_2 is less than 10%.

RESULTS FOR Nb→Nb (FLUORIDE COMPLEX)

The 14 sources examined showed an apparent $\Delta\lambda/\lambda = (-3.26 \pm 0.80) \times 10^{-2}$ calculated from the 122-keV line data when only the first 5 sec of data were ignored. An analysis of variance shows this data to be statistically inconsistent. The 140-keV line showed an apparent $\Delta\lambda/\lambda = (0.61 \pm 0.31) \times 10^{-2}$. However, the difference values

$$(\Delta\lambda/\lambda)_{122\text{ keV}} - (\Delta\lambda/\lambda)_{140\text{ keV}} = (-3.79 \pm 0.69) \times 10^{-2}$$

were shown to be statistically consistent among runs by the analysis of variance. Similar results were found when the data taken during the first 10 sec following the reaction were ignored. The scatter between runs was smaller here, so that this value is the adopted one.

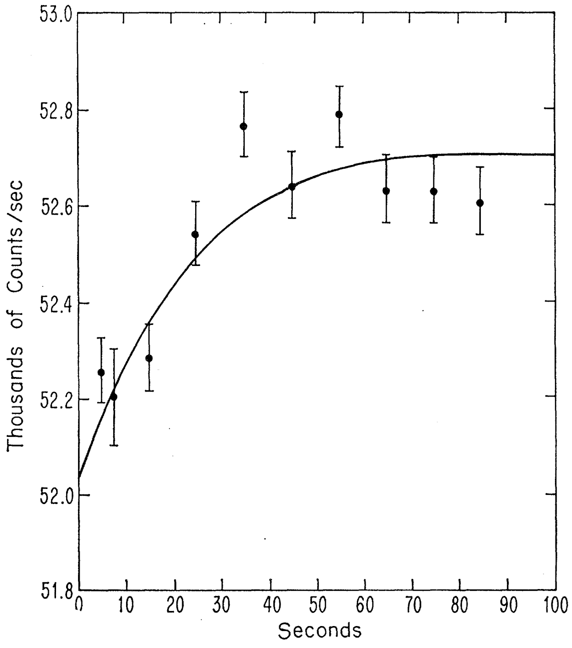

FIG. 3. Total multiscalar scans for Nb→Nb (fluoride complex). Least-squares fit for $T_{1/2} = 20$ sec, $\Delta\lambda/\lambda = (-3.3 \pm 0.4) \times 10^{-2}$, $F = 0.899$.

Account must be taken in the error of the uncertainty in the measurements of the peak-to-background ratio used to determine A. This yields

$$(\lambda_{\mathrm{Nb\ (fluoride\ complex)}}-\lambda_{\mathrm{Nb}})/\lambda_{\mathrm{Nb}}=(-3.9\pm0.8)\times10^{-2},$$

in excellent agreement with Cooper *et al.*

A least-squares fit to the sum of all the 122-keV data is shown in Fig. 3. Each point shown is an average of 10 channels. However, the point shown at $t=7.5$ is an average of five channels only; this is to facilitate comparison with the least-squares curve calculated starting at $t=6$. The effects of changing source geometry are not included, so that the value of $\Delta\lambda/\lambda$ for the Fig. 3 is to be compared to the "apparent" value of $(-3.26\pm0.80)\times10^{-2}$ quoted previously. The standard deviation quoted for Fig. 3 comes from the scatter of points about the least-squares curve, and is the amount expected from counting statistics; the higher value quoted is calculated from the values of $\Delta\lambda/\lambda$ obtained for each source.

RESULTS FOR $Nb\rightarrow Nb_2O_5$

The data for Nb_2O_5 is all statistically significant, and no effect due to changing source geometry is seen, even when all data after $t=2$ sec were used. However, the difference values $(\Delta\lambda/\lambda)_{122\ \mathrm{keV}}-(\Delta\lambda/\lambda)_{140\ \mathrm{keV}}$ are used in the result to include possible variations in source geometry in the error. No additional account is taken of incomplete combustion of the Nb foil or of the presence of a small amount of NbO_2. This would occur randomly among the sources with the amount of impurity, depending on such things as the nature of the contact between the foil and the alligator clips. Thus, this effect is automatically included in the calculation of the external variance. The adopted value is

$$(\lambda_{\mathrm{Nb_2O_5}}-\lambda_{\mathrm{Nb}})/\lambda_{\mathrm{Nb}}=(1.87\pm0.50)\times10^{-2}.$$

A least-squares fit to the sum of all scans is shown in Fig. 4.

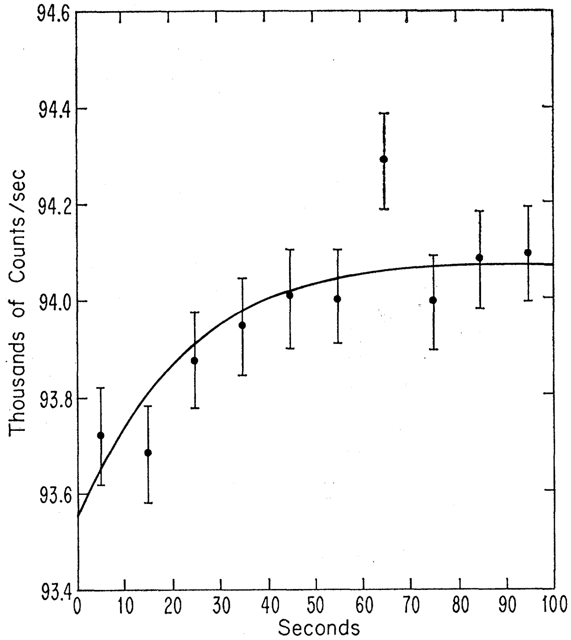

FIG. 4. Total multiscalar scans for $Nb\rightarrow Nb_2O_5$. Least-squares fit for $T_{1/2}=20$ sec, $\Delta\lambda/\lambda=(-1.92\pm0.30)\times10^{-2}$, $F=0.704$.

DISCUSSION

These results confirm the high sensitivity of the Nb conversion coefficient (ICC) to chemical environment. Thus Weirauch's measurement implies that the half-life of Nb^{90m} in Zr metal is a few percent larger than its half-life in Nb metal.

The reason for this high sensitivity to chemical environment is unclear. Calculation of the free-atom ICC's show that conversion is hindered in the d subshells, so that the $4d$ and $5s$ electrons contribute together less than 0.1% to the ICC. However, the atomic d-electron potential exhibits a double-well structure.[5] When one does self-consistent field calculations using such a potential with two wells of equal depth, a metastable situation develops: If the amplitude of the d electron increases at the inner well, then the other electrons are better shielded from the nucleus, and peak further away from it, so that the depth of the inner well increases. Similarly, a small increase in the d-electron amplitude at the outer well leads to an increase in the depth of that well. Under such conditions, small perturbations may cause large changes in the electron density near the nucleus.

The transition energy measured by Cooper *et al.*, namely, 2380±400 eV, is of the order of the L_{III} binding energy of 2371 eV. The width of the L_{III} atomic level[6] is 1.7 eV, and this determines the natural width of the conversion line. Extrapolation of the conversion coefficients from the Hager and Seltzer tables[7] show $\alpha_{L_{\mathrm{III}}}/\alpha_M\simeq1$. Thus if the γ-ray energy happened to be within a few eV of the L_{III} binding energy, then shifts in the chemical potential would have a large effect on the conversion coefficient.

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