NEW SHORT-LIVED ISOMERS Nd^{140m}, Pm^{141m}, Eu^{146m}, *AND* Gd^{158m}

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Short-lived γ radiation from PrO_4, Sm_2O_3, and Gd_2O_3 samples irradiated by 20.8-MeV protons was detected. The radiation was found to be due to the decay of isomeric states of nuclei produced in the nuclear reactions $Pr^{141}(p, 2n)Nd^{140m}$, $Sm^{147}(p, 2n)Eu^{146m}$, and $Gd^{160}(p, p2n)Gd^{158m}$. The isomeric activity detected earlier in Nd_2O_3 irradiated by protons is identified. The short-lived γ radiation is due to the decay of Pm^{141m} produced in the reaction $Nd^{142}(p, 2n)Pm^{141m}$.

INTRODUCTION

THE comparatively long lifetime of nuclei in isomeric states facilitates the measurement of the quantum characteristics of these states and also permits the direct measurement of the lifetime. Knowledge of these values makes it possible to determine experimentally the value of the matrix element for a radiative transition. The matrix element is determined by the wave functions of the nucleus in the initial and final states, i.e., it depends on the specific model used to represent the nuclear structure. Hence, comparison of the experimentally obtained probabilities of radiative transitions in a nucleus with the theoretically calculated probabilities is an important means of checking the correctness of our ideas on the structure of the atomic nucleus.

At the present time, a number of properties of long-lived isomeric states have been established and explained. For example, the known grouping of isomers with odd values of A into a kind of "island of isomerism" is in good agreement with the views on the shell structure of the nucleus. Our knowledge concerning the isomers of atomic nuclei or half-lives in the region $10^{-4} < T < 1$ sec has grown considerably in the recent period. Thus far, more than twenty cases of isomers with such lifetimes are known. One can already note that the general empirical laws for the long-lived isomers do not extend to the millisecond region. The frequent isomerism of the even-even nuclei Zr^{90m}, Ce^{138m}, Nd^{140m}, Gd^{158m}, W^{180m} (Nd^{140m} and Gd^{158m} are from the results of the present work) can serve as an illustration.

At high energies the basic transitions in the millisecond region appear to be of the M2, M3, and E3 type. It is natural to expect that the characteristic properties will be exhibited here.

Since isomers with half-lives in the millisecond region are usually products of nuclear reactions, the data on the formation of isomeric states can be used to study the mechanisms of the various reactions.

These considerations indicate that the extension of the search for isomers into the millisecond region is very much worth-while. Of particular importance at the present time is the determination of the quantum characteristics of the observed isomeric transitions.

In the present work, we searched for and studied the isomeric states of nuclei among the rare-earth elements.

1. METHOD OF MEASUREMENT

Metastable states can arise in nuclei produced in reactions when a target containing rare-earth elements is bombarded by fast protons. A linear accelerator served as a source for protons with energy up to $E_p = 20.8 \pm 0.25$ Mev. The operating conditions of the accelerator and the experimental arrangement enabled us to investigate γ activity with a half-life in the interval 10^{-4}–10^{-1} sec.

The method of measurement differed little from that described by Morozov, Yampol'skii and one of the authors.[1] We first carried out preliminary investigations of the sample for the presence of short-lived activity. These preliminary measurements were made visually on a pulsed oscilloscope with a single sweep triggered at the instant the proton bombardment of the target ceased. After the detection of the isomeric activity, we measured the γ-ray spectrum on a scintillation spectrometer. The spectrometer consisted of a

30 × 15 mm NaI(Tl) crystal, a broadband amplifier (non-overloading), and a single-channel amplitude analyzer. The amplitude-analyzer circuit permitted simultaneous measurement, at each point of the spectrum, of the long-lived background coming from the direction of the target. In all the energy spectra presented in this article the long-lived background has been eliminated. The resolving time of the entire measuring apparatus was 1 μsec. We knew how the photoregistration efficiency of the NaI(Tl) crystal varied with the γ-ray energy.

The half-life was measured on a 25-channel time analyzer, which was triggered at a given instant (usually 300—500 μsec) after interruption of the proton bombardment. The pulses were applied to the input of the time analyzer and were preselected by the amplitude discriminator in accordance with the investigated γ-ray energy.

Isotopes corresponding to the production of an isomer in the reaction were determined by comparison of the yield of the isomeric radiation from a target of natural isotopic composition and the yield of this radiation from a target enriched in the given isotope. The type of reaction was determined from the threshold and shape of the excitation-function curve of the isomeric activity. It was assumed that the reaction threshold differed from the production threshold for the isomeric activity by the excitation energy of the isomeric state. The proton energy was determined by absorption in aluminum filters and was verified by the threshold for the reaction $C^{12}(p, n)N^{12}$ (E_{thr} = 19.91 MeV). The proton energy was measured with aluminum filters. The target thickness used in the measurements was ~50 mg/cm^2.

2. RESULTS OF THE MEASUREMENTS

We investigated oxide samples of the following elements: Ce, Pr, Nd, Sm, Eu, Gd, Tb, Yb. In the PrO_4, Sm_2O_3, and Gd_2O_3 targets we observed short-lived isomeric activity after bombardment by protons. The detection of isomeric activity with a half-life of 2.2 msec after bombardment of a Nd_2O_3 target by protons has been discussed by Morozov et al.[1]

Praseodymium. The γ spectrum in the case of isomeric activity resulting from the proton bombardment of a praseodymium oxide target is shown in Fig. 1. The three photopeaks indicated by arrows correspond to γ transitions with energies of 1.00, 0.77, 0.435 MeV (the errors here are all 0.02 MeV). The relative intensity of these γ transitions estimated from the area of the photopeak does not

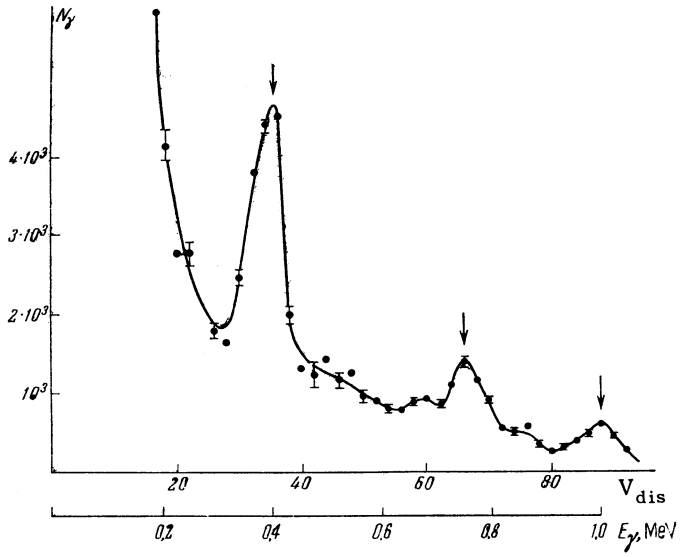

FIG. 1. Nd^{140m} γ-ray spectrum. The arrows indicate photopeaks corresponding to transitions of 0.435, 0.77, and 1.0 MeV. Here and in other figures V_{dis} is the discrimination level in volts.

contradict the assumption that the transitions occur in a single cascade. The decrease in the isomeric activity with time is shown in Fig. 2. The half-life obtained is 0.6 ± 0.05 msec.

In order to determine the reaction responsible for the given isomeric state, we measured the excitation function of the isomeric activity (see Fig. 3). The production threshold for the isomer is $E_{\gamma thr}$ = 12 ± 1 MeV. The excitation energy in the isomeric state is ~2 MeV. Consequently, the threshold for the reaction responsible for the iso-

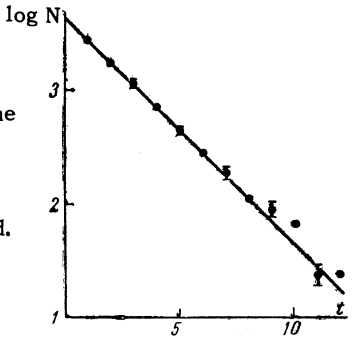

FIG. 2. Decay of Nd^{140m}. The channel number of the time analyzer is given on the time axis. The channel width is 0.39 msec. The long-lived background has been deducted.

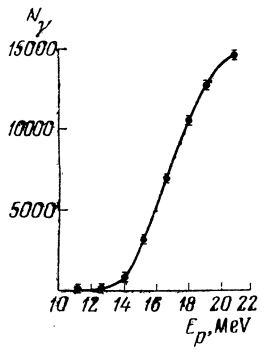

FIG. 3. Excitation function for the isomeric activity of the Pr^{141} (p, 2n) Nd^{140m} reaction. E_p is the proton energy; N_γ is the isomeric activity yield.

meric state should be ~10 MeV. Praseodymium has only one isotope (A = 141). The probable reactions are $Pr^{141}(p, pn)Pr^{140}$, Q = −9.57 MeV and $Pr^{141}(p, 2n)Nd^{140}$, Q = −10.5 MeV. The values of Q for both reactions lie within the measurement error of the threshold value. As a rule, however, the yield for the (p, pn) reaction is considerably less at these proton energies than for the (p, 2n) reaction. In our case, the reactions was marked by a relatively greater yield of isomeric activity.

Moreover, as experiment shows, reactions on heavy nuclei which yield charged particles always give values of the measured threshold enhanced by several MeV. This is connected with the fact that the Coulomb barrier for the emitted charged particles greatly lowers the probability of the (p, pn) reaction close to the threshold when the energy of the outgoing proton is considerably less than the barrier potential. When the excitation-function curve is extrapolated to the threshold value, this effect leads to an increased threshold energy.

Comparing the calculated values of the threshold energy −9.57 MeV for the (p, pn) reaction and −10.5 MeV for the (p, 2n) reaction, with the measured value 10 MeV and taking into account the foregoing remarks, we should conclude that the isomeric state arises in the reaction $Pr^{141}(p, 2n)Nd^{140m}$. A confirmation of this conclusion is provided by the fact that when a Ce^{140} target (its concentration in a natural target is 88.5%) is bombarded by protons, the Pr^{140} isotope produced in the $Ce^{140}(p, n)Pr^{140}$ reaction is not observed to have an isomeric state.

The $_{60}Nd^{140}$ nucleus is even-even. The ground state should be assigned a spin 0 and positive parity. It is natural to assume that the first excited state is 2^+. For the second excited state we should take the spin and parity as 4^+, since no transition is observed from this level to the ground state. It is known that for the first two levels of the even-even nuclei the rule $E_{4+}/E_{2+} \approx 2.2$ must be satisfied. Here, E_{4+} and E_{2+} are the excitation energies of the second (4^+) and the first (2^+) levels. This rule for even-even nuclei in the rare-earth region has been confirmed recently by Funk et al.[2] They concluded that for the rare-earth elements with 84, 86, and 88 neutrons a sequence of excited states 2^+ and 4^+ apparently occurs, where $E_{4+}/E_{2+} = 1.85-2.35$.

We obtained good agreement with this rule for the succession of levels if we assume that the 0.77-MeV level corresponds to the first excited state of the Nd^{140} nucleus and the 1.77-MeV level corresponds to the second excited state; in this case the ratio E_{4+}/E_{2+} is 2.3. Poorer agreement is given by the sequence of levels 0.43 and 1.2 MeV. Moreover, in this case the isomeric transition will have an energy of 1.0 MeV, which contradicts the assumption that the Nd^{140m} nucleus has a very large spin.

Our proposed scheme for the Nd^{140} levels is shown in Fig. 4. The value of spin 7 is assigned to the isomeric state on the basis of the assumption that the transition from this level is accompanied by electromagnetic radiation of the E3 type. The E3 transitions give good agreement between the measured lifetime and the theoretical estimate of the probability of this transition.[3,4]

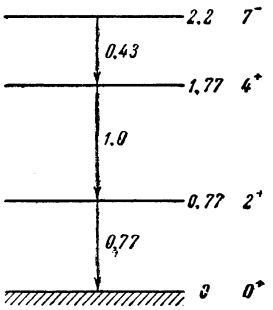

FIG. 4. Nd^{140m} decay scheme.

Neodymium. The availability of enriched isotopes of neodymium allowed us to identify an isomer detected when a neodymium target of natural isotopic composition was bombarded by protons. Comparison of the effects from a natural Nd_2O_3 target and a Nd_2O_3 target enriched with the isotope Nd^{142} showed that the isomeric activity is a product of the reaction on the Nd^{142} isotope. Figure 5 shows the spectrum of the isomeric γ radiation from a natural target and from a Nd^{142} target (the Nd^{142} contents in the enriched sample was 98.5%). The γ-ray spectrum consists of two lines: $E_{\gamma 1} = 0.19 \pm 0.01$ Mev and $E_{\gamma 2} = 0.43 \pm 0.01$ MeV.

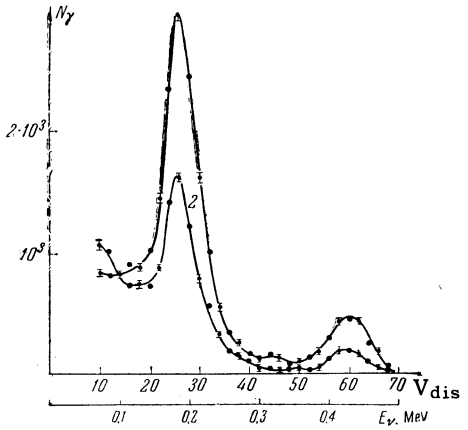

FIG. 5. Pm^{141m} γ-ray spectrum. Curve 1 was taken after bombardment of a target of natural Nd by protons; curve 2 is for the bombardment of a Nd^{142} sample. The thickness of the enriched target is less than that of the natural one.

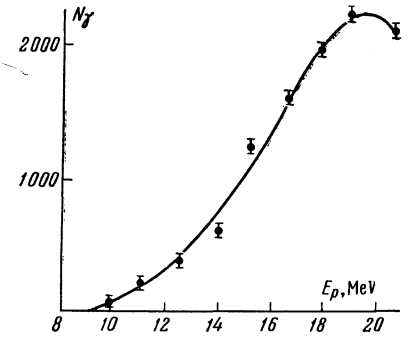

FIG. 6. Excitation function for isomeric activity of the Nd^{142} (p, 2n) Pm^{141m} reaction.

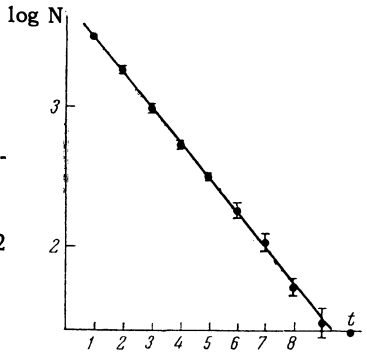

FIG. 7. Eu^{146m} decay. The measurements were made on the 0.36-MeV γ line. The channel number of the time analyzer is given on the abscissa axis. The channel width is 0.2 msec

The measured threshold for the reaction (Fig. 6) leading to the isomeric state is $E_{thr} = 8.5 \pm 1$ MeV. Possible reactions are $Nd^{142}(p, pn)Nd^{141}$ and $Nd^{142}(p, 2n)Pm^{141}$. For the first reaction $Q = -9.7$ MeV. For the second reaction Q was not calculated, since we did not know the mass of Pm^{141}, but, as a rule, the threshold of the (p, pn) and the (p, 2n) reactions differ little. On the basis of considerations concerning the shape of the excitation-function curve close to the threshold for the (p, pn) reaction similar to those in the identification of the Nd^{140m} isomer, we should expect that the isomeric state is produced in the $Nd^{142}(p, 2n)Pm^{141m}$ reaction. This conclusion agrees with the fact that we did not observe the activity after praseodymium was bombarded by protons, where the excited states of the Nd^{141} nucleus should have been obtained from the reaction $Pr^{141}(p, n)Nd^{141}$. According to Kistiakowsky,[5] a 20-minute activity of Pm^{141} results from the bombardment of Nd^{142} by 20-MeV protons.

In the estimate of the relative intensity of the γ transitions $E_{\gamma 1} = 0.19$ MeV and $E_{\gamma 2} = 0.43$ MeV, it turned out that the intensity of the 0.19-MeV transition exceeds the intensity of the other transition beyond the limits of experimental error. To explain this fact, we can assume that the 0.43-MeV transition is a conversion. Our measurements yielded $N_{0.19}/N_{0.43} = 1.6 \pm 0.3$. The total conversion coefficient in this case is $\alpha \approx 60\%$ and the most probable type of transition for $E_\gamma = 0.43$ MeV is M3. The measured[1] half-life of the Pm^{141m} isomer 2.2 msec is in good agreement with the assumption that the isomeric state has an M3 transition.

Samarium. When Sm^{147} is bombarded by protons, radiation with an isomeric level half-life 0.24 ± 0.01 msec is produced (Fig. 7). The Sm^{147} content in the enriched target was 93.1%.

The γ-ray spectrum has a very complex form (Fig. 8). This is partly connected with the fact that the amplitude analyzer operated with a considerable overload because of the small half-life. The spectrum has very distinct peaks corresponding to

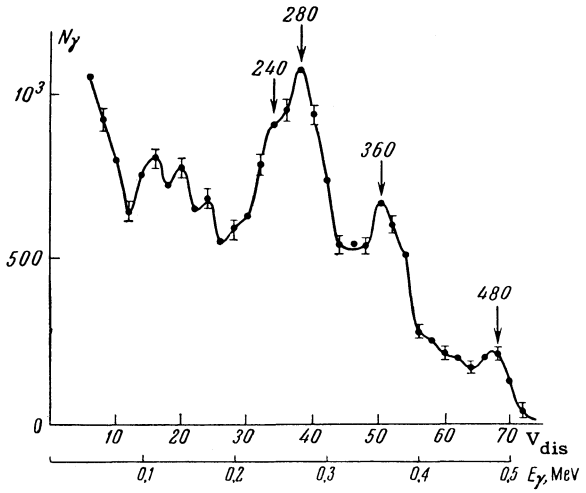

FIG. 8. Eu^{146m} radiation spectrum.

γ-ray energies of 0.28 and 0.36 MeV. The γ-ray spectrum was measured several times. From these measurements we can conclude that the spectrum also has peaks corresponding to γ-ray energies of 0.24, 0.39, and 0.48 MeV.

The value of the reaction threshold obtained from the excitation function of the isomeric activity (Fig. 9) was $E_{thr} \approx 10$ MeV. Since we do not know the decay scheme of the isomeric state, and to determine the reaction threshold we should subtract the excitation energy of the isomeric state from the threshold value for the production of the isomer, we took the excitation energy to be ~1 MeV. The value of Q calculated from the mass for the (p, n) and (p, pn) reactions are −2.6 and −6.5 MeV, respectively. For the (p, 2n) reaction, the mass

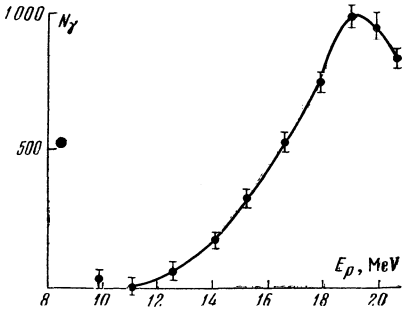

FIG. 9. Excitation function for the isomeric activity of the Sm^{147} (p, 2n) Eu^{146m} reaction.

of Eu^{146} is not known. We used the data of [6] for the decay $Eu^{146} \xrightarrow{e} Sm^{146}$. For the (p, 2n) reaction, we obtained $Q \approx -11$ Mev. On the basis of the reaction threshold, the reaction $Sm^{147}(p, 2n)Eu^{146m}$ is the most probable. However, for the reaction $Sm^{147}(p, pn)Sm^{146}$ the difference between the isomeric threshold and the value of Q for the given reaction can be connected with the Coulomb barrier for the emitted proton. Berlovich et al [6] presented data on the excitation levels of Sm^{146} in the energy region up to 3.5 MeV. Among them are no levels which could give transitions corresponding to the γ rays observed by us. For the excitation region above 3.5 MeV, the reaction threshold leading to an isomeric state should already exceed the value obtained by us. In this connection, the observed γ radiation should refer to the decay of the isomeric nucleus Eu^{146m} produced in the reaction the $Sm^{147m}(p, 2n)Eu^{146m}$.

Gadolinium. When a Gd_2O_3 target of natural isotopic composition is bombarded by protons, two isomeric activities are obtained. It has been established that γ radiation with a half-life of 0.46 ± 0.02 msec and energies of 0.08 ± 0.01 and 0.18 ± 0.01 MeV is emitted from the Gd^{160} isotope (enriched to 91.7%). The decay curve and the energy spectrum of this γ radiation are shown in Figs. 10 and 11.

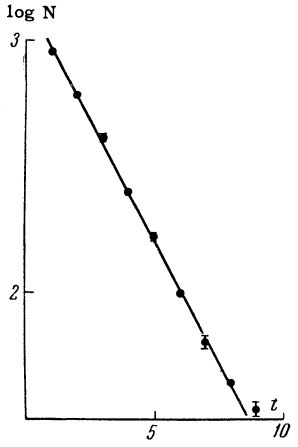

FIG. 10. Gd^{158m} decay. The channel number of the time analyzer is given on the abscissa axis. The channel width is 0.29 msec.

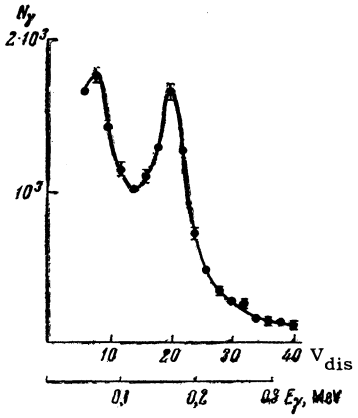

FIG. 11. Gd^{158m} radiation spectrum.

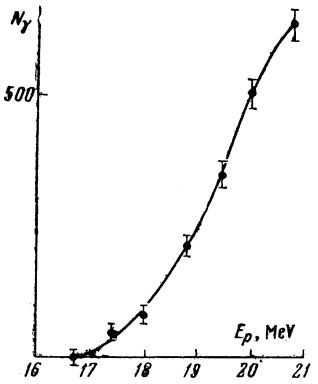

FIG. 12. Excitation function for isomeric activity of the Gd^{160} (p, p2n) Gd^{158m} reaction.

In the case of a natural target, moreover, γ activity with a half-life of ~2 msec and $E_\gamma \approx 0.22$ MeV is observed (other lines may possibly occur in the low-energy part of the spectrum).

The reaction threshold for Gd^{160} leading to a short-lived radiation is 16.5 ± 0.5 MeV (Fig. 12). The reactions close to the value of the threshold are: (p, 3n), Q = −14.2 MeV, and (p, p2n), Q = −13.3 MeV. The (p, 2pn) reaction is unlikely. Since in the case of the two reactions (p, 3n) and (p, p2n) there is no agreement with the measured threshold, it is reasonable to assume that we are dealing with the (p, p2n) reaction. We can then explain the increased value of the measured threshold by taking into account the effect of the Coulomb barrier for the emitted proton. The isomeric nucleus will then be Gd^{158m}.

The γ-transition energies obtained in this work are in good agreement with the known levels of the Gd^{158} nucleus, 79.1 and 260.8 keV. [7] Groshev, Demidov, and Naĭdenov [8] measured the ratio of the conversion coefficients α_K/α_L for the 180-keV transition. They obtained $\alpha_K/\alpha_L = 1.7 \pm 0.3$. The theoretical value of this ratio for transitions of the type E1, E2, E3, M1, and M2 were found by these authors to be ~7, ~2, ~1, ~7, ~5, respectively. Comparing the experimental values of α_K/α_L with the calculated ones, they took the 180-keV transition to be of the E2 type. From a comparison of these values of α_K/α_L, it is seen that the results obtained for the E3 type transition also do not differ very much. In their work, the authors discuss the possibility of an E3 transition for the upper transition (Fig. 13). Taking into account the presently known value of the 260-keV

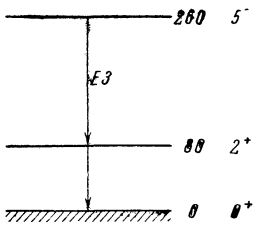

FIG. 13. Gd^{158} level scheme. The 260-keV level is isomeric.

level of Gd^{158} and the results of Groshev et al,[8] we can conclude that the transition from the 260-keV level is of the E3 type, and that the spin and parity of the nucleus in the isomeric state are 5^-.

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