GAMMA RADIATION FROM RADIATIVE CAPTURE OF THERMAL NEUTRONS BY Mo^{95}, Ag^{107}, Te^{123}, *AND* Cs^{133} *NUCLEI*

L. F. KALINKIN, A. S. MELIORANSKII, and I. V. ESTULIN

Scientific Research Institute for Nuclear Physics, Moscow State University

Submitted to JETP editor April 5, 1958

J. Exptl. Theoret. Phys. (U.S.S.R.) **35**, 592-598 (September, 1958)

A single-crystal scintillation spectrometer adjusted for operation at high counting rates was used to measure the energies and absolute intensities of γ rays in the region of 20 to 1000 kev, emitted after radiative capture of neutrons in samples enriched in isotopes of Mo^{95}, Ag^{107}, Te^{123}, and Cs^{133}. The observed γ rays come from transitions between low lying states of these nuclei.

MEASURING TECHNIQUE

THE present paper is a continuation of previous work of the authors[1] on the investigation of the γ radiation emitted by nuclei after capture of thermal neutrons. The measurements were carried out in the range of γ-ray energies from 20 to 1000 kev. The neutron source was the modernized heavy-water reactor of the Academy of Sciences, U.S.S.R.[2] The increased power of the reactor and the use of more complete neutron collimation than in the previous experiments enabled us to obtain a flux of the external beam in the region of the target of $\sim 10^8$ neutrons/sec cm^2.

In measurements of neutron-capture gamma rays using magnetic spectrometers,[3-5] the shielding of the neutron beam from pile γ rays was accomplished by means of thick layers (~ 10 cm) of lead or bismuth. In the present work it turned out that it was sufficient to cover the neutron beam inside the collimator with a thin combined filter (1.7 mm of lead, 1.5 mm of tin, and 0.5 mm of zinc), which strongly absorbed the soft γ rays and produced a negligible weakening of the neutron flux.

The γ-ray detector was a single crystal scintillation spectrometer with a NaI (Tl) crystal 30 mm in diameter and 19.7 mm high. The use of photomultiplier FEU-11, a non-saturating pulse amplifier,[6] and a fast acting non-saturating single-channel analyzer enabled us to work with high integral counting rates (up to $\sim 10^5$ pulses/sec) without any change in the resolution and the energy calibration of the spectrometer. The window width of the analyzer corresponded to energy intervals of 2.4, 6, and 20 kev for measurements in the energy ranges $E_\gamma \leq 50$, $E_\gamma \leq 400$, and $E_\gamma \geq 400$ kev, respectively. The line shape, the photopeak efficiency, and the resolution of the spectrometer were determined experimentally, under the geometrical conditions existing in the capture gamma-ray measurements, for γ rays of 22, 25, 27, 31, 59, 72 kev (the K radiations of Ag, Sn, Te, Cs, W, and T, respectively), 159 kev (Te^{123M}), 279 kev (Hg^{203}), 323 kev (Cr^{51}), 480 kev (γ-rays from the B^{10} (n, α) Li^{7*} reaction), 662 kev (Cs^{137}), 762 kev (Nb^{95}), and 1120 kev (Zn^{65}). The resolving power of the spectrometer in the energy region E_γ = 279 to 1120 kev can be written as

$$\eta = (240/\sqrt{E_\gamma}) + 0.2. \quad (1)$$

where the resolution η is in percent and E_γ is in kev. For $E_\gamma < 279$ kev, the values of $\eta(E_\gamma)$ are somewhat smaller than those given by formula (1). Figure 1 shows the dependence of the photopeak efficiency ϵ_Φ of the spectrometer on E_γ:[1] (1) for a NaI (Tl) crystal with h = 10 mm, d = 28 mm; (2) for the crystal with h = 19.7 mm,

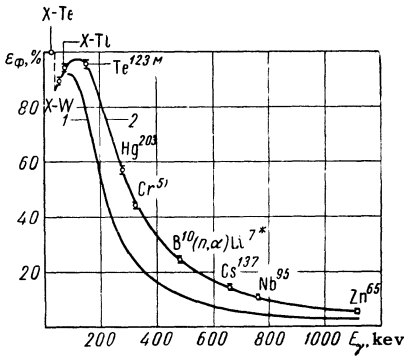

FIG. 1. Photopeak efficiency of spectrometer with NaI (Tl) crystal: 1) crystal with h = 10 mm, d = 28 mm; 2) crystal with h = 19.7 mm, d = 30 mm.

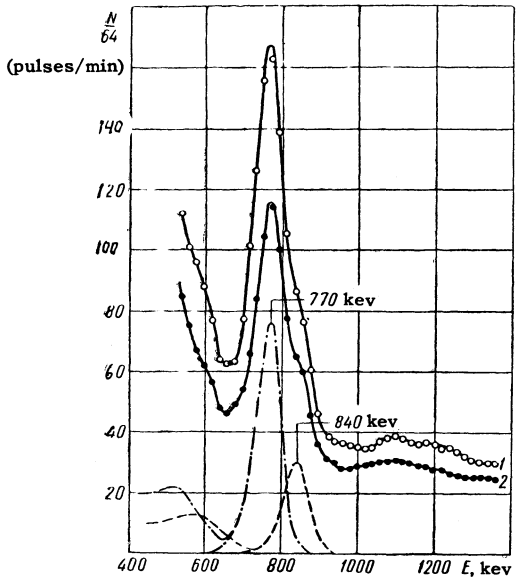

FIG. 2. Spectrum of pulsed from Mo^{96} γ rays: 1) measurements without filter, 2) measurements with 5.490 g/cm² filter.

d = 30 mm, which was used in our work. The reduction in ϵ_Φ for $E_\gamma < 100$ kev, which is shown in curve 2, is caused by the increased probability of escape of iodine K radiation from the crystal and the consequent increase in the "escape" peak at the expense of reducing the area under the peak corresponding to the full energy of the γ rays.

For $E_\gamma < 33$ kev (the binding energy of the electrons in the iodine K-shell), $\epsilon_\Phi \approx 100\%$.

The intensity of the measured γ rays, in numbers of quanta per neutron capture, was determined as previously[1] by comparison with the known γ-ray intensity from the B^{10} (n, α) Li^{7*} reaction, which is 0.935 quanta per capture.[7] The values of the effective neutron capture cross sections were taken from Hughes.[8]

The details of the technique for measurement of neutron capture γ rays (the geometry of the experiment; the extraction of the effect of neutrons from the (nγ) reaction; the measurement of the spectra with thin lead filters to absorb the soft radiation for the purpose of reducing the background from hard γ rays; the resolution of the spectra into components; the determination of the intensity of γ-ray lines, etc) have been described earlier.[1]

RESULTS OF MEASUREMENTS

Molybdenum. A sample of metallic molybdenum weighing 0.666 g was enriched to 88.8% of the Mo^{95} isotope, so that 99.4% of the neutron captures occurred in this isotope. Figure 2 shows the spectrum of pulses from Mo^{96} γ rays in the energy region from 550 to 1350 kev. Curve 1 corresponds

Radiating nucleus	Present work		Results of other authors	
	γ-ray energy E_γ, kev	Number of quanta per neutron capture n_γ, in %	γ-ray energy E_γ, kev	Number of quanta per neutron capture n_γ, in %
Mo^{96}	360±10 770±10 840±10 1100—1240	~2 91±14 43±8 18		
Ag^{108}	22±2* 83±2 117±3 202 {197±3 {215±4 (266±4) ** (302±5)	10±6 18±3 9±2 43 { 21±4 { 22±4 12±3 18±4	82±2 [10] 117±2 [10] 199±3 [10] 187 [11]	20±4 [10] 11±2 [10] 34±6 [10]
Te^{124}	360±10 605±10 725±10	2.5±0.5 58±9 17±4	625±10 [15]; 609 [11]	54±14 [15]
Cs^{184}	31±2* 120±3 184±3 (215±4) (258±4) (310±5)	27±6 20±3 9±2 7±2 5±1 4±1	Spectrum not resolved[11]	

*K x-radiation.

**The energies of γ lines whose existence has not been reliably established are given in parenthesis.

to measurements without a lead filter in the collimator, curve 2 to measurements with a 5.490 g/cm^2 lead filter. The dotted curves are the individual lines resolved from curve 2, whose energy and intensity are given in the table. In the 1100 to 1240 kev energy region there is an unresolved maximum from Mo^{96} γ rays with a total γ-ray intensity of ~18%. In the region $E_\gamma < 770$ kev we observed a small peak from 360-kev γ rays, but this peak is not present in the spectrum of the filtered radiation.

The hard γ rays from radiative capture of neutrons in molybdenum were measured by Kinsey and Bartholomew.[3] From their work, the energy of the first two excited states of Mo^{96} is 760 ± 15 and 1610 ± 20 kev. The 840- and 770-kev γ rays found in the present work are the cascade transition from the second excited state of Mo^{96} at 1610 kev, which is well known from the decay of Nb^{96} and Tc^{96}.[9] Apparently the (360 ± 10)-kev γ rays result from a transition between levels lying higher than the first two excited levels.

In the decay of Nb^{96} and Tc^{96} the Mo^{96} nucleus is excited to an energy ~2.7 Mev. The ground state of Mo^{96} is then reached only by cascade transitions through the first excited level. In neutron capture one produces an excited state of Mo^{96} with an excitation energy of 9.15 Mev.[3] However, in this case also the ground state of the nucleus is reached mainly by transitions through the first excited level (in 91 ± 14% of the cases of neutron capture).

Silver. A metallic silver sample weighing 0.455 g was enriched to 97.8% in the Ag^{107} isotope, so that 94% of the neutron captures took place in this isotope. Figure 3 shows the spectrum of Ag^{108} γ rays absorbed in 2.205 g/cm^2 of lead, and the resolution of the spectrum into components. The energies and intensities of the γ lines are given in the table. It should be mentioned that the half-width of the 202-kev peak exceeds by 30% the half-width for a monochromatic line of the same energy (which is shown by the dashed curve in Fig. 3). This allows us to postulate the presence in the region of the peak of at least two γ lines with energies 197 ± 3 and 215 ± 4 kev, with intensities of 21 ± 4 and 22 ± 4% respectively. This resulution is not unique since there are no reliable arguments for assuming that this maximum consists of two and not more than two lines. However these γ rays with their assigned energies fit well into the γ-ray transition scheme (Fig. 4) discussed below.

According to the data of Skliarevskii, Stepanov, and Obiníakov,[10] the γ rays from radiative capture of neutrons in the Ag^{107} and Ag^{109} isotopes include lines with energies close to one another. The intensities of the Ag^{108} γ lines found in the present work were corrected for the contribution of the radiation following neutron capture in Ag^{109} in the sample (6% of the captures) by using the intensities from reference 10. The values of energy and intensity for those γ lines which occur in both our work and in reference 10 are in good agreement. The 187-kev γ rays found by Hamermesh and Hummel[11] in the radiation accompanying neutron capture in natural silver are apparently due to a superposition of neighboring lines of Ag^{108} and Ag^{110}.[10]

The γ rays at 22 ± 2 kev are naturally interpreted as Ag K radiation, which could result from internal conversion in the K shell and from self-absorption of the γ rays in the target via the photoeffect. Of the observed 22 ± 4 x-ray photons per hundred neutron captures, 12 ± 2 photons are due to the effect of self-absorption. The self-ab-

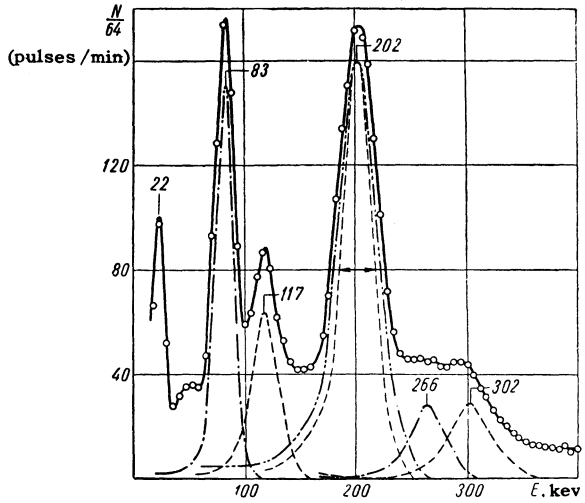

FIG. 3. Spectrum of pulses from Ag^{108} γ rays, absorbed in 2.205 g/cm^2 Pb.

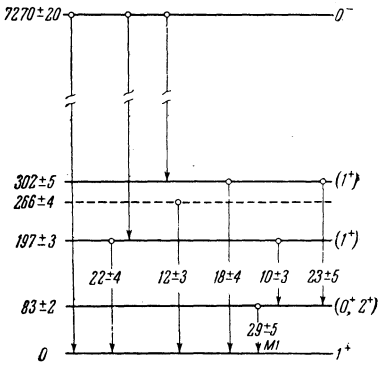

FIG. 4. γ-ray transitions in Ag^{108}. The numbers in the breaks in the arrows give the intensities of the transitions in percent per neutron capture, taking account of conversion ($n_\gamma + k_\gamma$).

sorption was computed for the target thickness 0.283 g/cm^2 employed in the experiment by using the intensities of the Ag^{108} γ lines given in the table. Thus 10 ± 6 x-ray photons are associated with internal conversion. After correcting for the K-fluorescence yield f = 0.82,[12] the number of cases of conversion of all the γ rays will be equal to $K = \Sigma K_{\gamma_i} = (12 \pm 7\%)$ per neutron capture, where K_{γ_i} refers to a single γ line. The "thick" target used in our experiments limited the accuracy in determining K.

The hard γ quanta from the Ag (n, γ) reaction which were found in reference 4 indicate the presence of levels in Ag^{108} with excitation energies of 0.32 ± 0.03 and 0.21 ± 0.03 Mev. The soft γ rays observed in the present work fix more precisely the energies of these levels which serve as the basis for setting up a possible scheme of γ transitions (Fig. 4). In this scheme, which includes the γ lines found by us, excited levels at 83 ± 2 and 266 ± 4 kev are introduced in addition to the levels mentioned above.

The state of Ag^{108} which is produced as a result of capture of a thermal neutron (from now on we call it the initial state) can be either 0^- or 1^- The direct transition to the ground state is an E1 transition. The transitions with comparable intensities to the 197- and 302-kev levels, which were found in reference 4, should also be E1. Thus these levels can be 0^+, 1^+, 2^+.

Using the known intensity of the 83-kev γ rays and theoretical values of the K-shell conversion coefficients taken from the work of Sliv and Band,[13] we can predict the number of cases of conversion K_{83} for radiative transitions of various multiplicities: 133 (M2), 37 (E2), 11.4 (M1), and 43 (E1) (in % per neutron capture). The quadrupole transitions contradict the experimental value of K as determined in the present work from the intensity of the x-rays. We have a choice between the dipole transitions M1 and E1 for the 83-kev γ transition. If we assign this transition to be E1, the state at 83 ± 3 kev must have negative parity. If this is the case, the intensities of the competing transitions of opposite parity from the 302- and 197-kev levels to the 83-kev level and to the ground state (cf. diagram) should be markedly different, whereas the experiments show them to be comparable (cf. table). In addition, in order to make the intensity of the transitions leading to the 83-kev state agree with the intensity of the transitions which deexcite this state, it is preferable to assign the (83 ± 3)-kev transition to M1. Thus this transition should be taken to be magnetic dipole, and the characteristics of the corresponding excited level can be 0^+, 1^+ or 2^+. In reference 4 no γ quanta corresponding to the transition from the initial state to the 83 kev level were found, which can be understood if the initial Ag^{108} state is 0^- and the 83-kev state is 0^+ or 2^+. The 0^- assignment for the initial state in Ag^{108} is directly established by the results of reference 14, in which the first resonance level of Ag^{107} at E_p = 16.6 ev was found to have I = 0. The excited levels at 197 and 302 kev will be 1^+, and the γ transitions shown in Fig. 4 will be either M1 or E2, which does not contradict the experimental value of K.

Tellurium. The 72.8-mg sample of metallic tellurium used in the experiment was enriched to 28% in Te^{123}, so that 97% of the neutron captures took place in this isotope. In 2.4% of the cases, the neutrons were captured by Te^{124} which constituted 41.5% of the sample. The energies and intensities of the observed γ rays are given in the table. Figure 5 shows the spectrum of γ lines absorbed in a 4.39 g/cm lead filter, in the region of well-resolved lines (shown dotted in the figure). The energy and intensity of the (605 ± 10)-kev γ quanta determined in the present work agrees, within the limits of error, with the results of previous work.[11,15]

The γ quanta with energies of 775 and 605 kev can only come from Te^{124}, because of their high intensity. Similar γ rays are observed in the radioactive decay of Sb^{124} and I^{124},[9] where they correspond to transitions between the first two levels of Te^{124}. The 360-kev γ lines with 2.5% intensity could belong to Te^{125} since the target contains Te^{124}. But in that case the high intensity of this line (~100% per neutron capture in Te^{124})

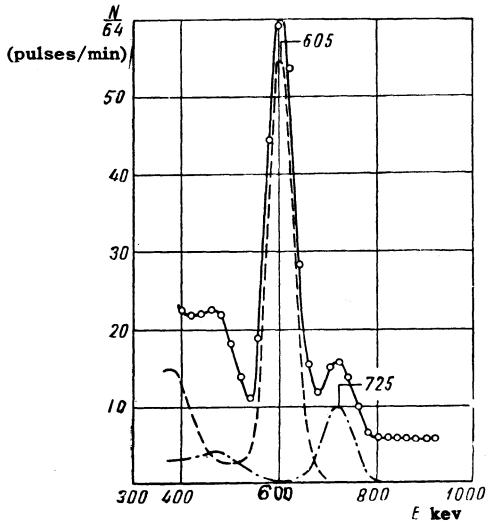

FIG. 5. Spectrum of pulses from Te^{124} γ rays absorbed in 4.386 g/cm^2 Pb.

would require its assignment as a transition from one of the lower lying levels. But it is known from the decay of Sb^{125} (reference 9) that no γ quanta of such energy are emitted in transitions between the low lying states of Te^{125}. Thus the 360-kev γ quanta observed here probably belong to Te^{124} and correspond to transitions between levels with excitation energies greater than 1380 kev.

The intensities of the transitions between the first two excited states of the even-even nuclei Te^{124} and Mo^{96} differ somewhat. In Te^{124} the majority of the cascade transitions from the initial state lead to the ground state, passing over the first two excited states.

Cesium. In the measurements with cesium, we used targets of cesium fluoride weighing 0.540, 2.486, and 8.200 g. The first of these targets was used for the measurements in the region $E_\gamma \leq 50$ kev, where the absorption of the γ rays in the source material is especially important. Figure 6 shows the spectrum of the γ rays, absorbed in 2.205 g/cm^2 of lead, for the 2.486-g target, and its resolution into components (only the photopeaks are shown). The energies and intensities of the γ lines are given in the table. The Cs^{134} nucleus has a level at 10.5 kev whose quantum numbers (5^+) are close to those of the 4^+ ground state.[9] The γ transitions from higher levels to this level and to the ground state can have comparable probabilities. It is possible that the spectrum is poorly resolved in the region $E_\gamma > 180$ kev because of overlapping of γ lines due to such transitions. The neutron capture γ rays of cesium were measured earlier by Hamermesh and Hummel,[11] but the spectrum was not resolved. The γ lines found in the present work are not to be associated with the decay of the three-hour isomer of Cs^{134}, since the cross section for activation of this isomer is small (2×10^{-2} barn). The intense γ radiation with energy 120 ± 3 kev probably corresponds to a transition from a (120 ± 3)-kev state to the ground state. The less intense γ quanta with energy 184 ± 4 kev are possibly due to transitions between levels with excitation energies of 310 ± 5 and 120 ± 3 kev, while the 310-kev γ rays come from the direct transition from the excited state to the ground state.

The softest γ radiation with energy 31 ± 2 kev is the x-radiation of cesium, emitted mainly after internal conversion of the γ quanta in the K shell. For the thin target of cesium fluoride, the correction for self-absorption through the photoeffect amounted to 10% of the observed x-ray intensity. Taking into account the K-fluorescence yield f = 0.89,[12] the total number of cases of conversion will be K = (30 ± 6)% per neutron capture. The value of K was compared with the intensities of the γ lines, using theoretical values of the K-shell internal conversion coefficients,[13] in a way similar to that which we described in our discussion of the measurements on silver. This analysis leads to the conclusion that the 120-kev γ transition is E2, while the 184 and 215 kev transitions are M2. In this case we get a value of K = 25 ± 5, in satisfactory agreement with the experimental data. The large number of soft γ rays in the spectrum from the $Cs^{133}(n, \gamma)$ reaction indicates that the odd-odd nucleus Cs^{134} has a high level density even near the ground state.

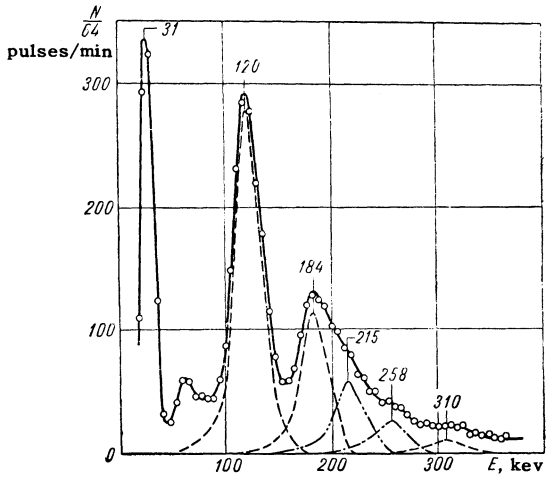

FIG. 6. Spectrum of pulses from Cs^{134} γ rays absorbed in 2.205 g-cm^2 Pb.

The authors thank I. S. Shapiro for his interest in the work and discussion of the results; S. A. Gavrilov, A. P. Shilov, and the entire operating crew of the physics reactor, and also Ia. A. Kleiman, A. M. Safronov and V. F. Tsarakaev for assistance in carrying out the experiment.

[1] Estulin, Kalinkin, and Melioranskii, J. Exptl. Theoret. Phys. (U.S.S.R.) **32**, 979 (1957), Soviet Phys. JETP **5**, 801 (1957).

[2] Short communication in Атомная энергия (Atomic Energy) **3**, 276 (1957).

[3] B. B. Kinsey and G. A. Bartholomew, Can. J. Phys. **31**, 1051 (1953).

[4] G. A. Bartholomew and B. B. Kinsey, Can. J. Phys. **31**, 1025 (1953).

[5] Ad'iasevich, Groshev, and Demidov, Сессия АН СССР по мирному использованию атомной энергии 1—5 июля 1955 г. (Session of the Academy of Sciences of the USSR on peaceful uses of atomic energy, July 1-5, 1955). Session of the Division of Physical and Mathematical Sciences, Acad. Sci. Press, p. 270, 1955.

[6] A. S. Melioranskii and Iu. M. Ostanevich, Приборы и техника эксперимента (Instrum. and Meas. Engg.) (in press).

[7] J. A. DeJuren and H. Rosenwasser, Phys. Rev. **93**, 831 (1954).

[8] D. J. Hughes, Pile Neutron Research, Addison-Wesley, 1953.

[9] V. S. Dzhelepov and L. K. Peker, Схемы распада радиоактивных изотопов (Decay Schemes of Radioactive Isotopes), Acad. Sci. Press, Moscow-Leningrad, 1957.

[10] Skliarevskii, Stepanov, and Obiniakov, Атомная энергия (Atomic Energy) (in press).

[11] B. Hamermesh and V. Hummel, Phys. Rev. **88**, 916 (1952).

[12] C. E. Roos, Phys. Rev. **105**, 931 (1957).

[13] L. A. Sliv and I. M. Band, Таблицы коэффициентов внутренней конверсии гамма-излучения (Tables of Internal Conversion Coefficients of Gamma Radiation), Part 1. Acad. Sci. Press, Moscow-Leningrad, 1956.

[14] Rae, Collins, Kinsey, Lynn, and Wiblin, Nucl. Phys. **5**, 89 (1958).

[15] Fenstermacher, Bennett, Walters, Bockelman, and Schultz, Phys. Rev. **107**, 1650 (1957).

Translated by M. Hamermesh
125