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COMPARISON OF GAMMA RAY SPECTRA FROM CAPTURE OF THERMAL AND FAST NEUTRONS

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Gamma ray spectra of radiative capture of neutrons from a uranium-water reactor are presented. Measurements of the γ ray spectra were made above 3—4 MeV, using a single crystal scintillation spectrometer, for Mn, Co, Fe, Ni and Cu. The results are compared with those under the same geometrical conditions for radiative capture of thermal neutrons.

THERE is a large amount of experimental material on spectra of radiative capture of thermal neutrons by many different materials.[1] Investigations with resonance neutrons (cf., for example,[2,3]) show that the partial γ-ray widths may change markedly from one resonance to another. This is especially noticeable for high energy γ transitions, since the separation of levels near the ground state is large.

To understand the mechanism of capture, it is meaningful to extend such experiments to higher energies, where the effect of individual resonances will not be a factor. Recently a paper by Bergqvist and Starfelt[4] appeared, which gives spectra from radiative capture of neutrons with $E_n = 0.1$ MeV for the three elements Ni, Au, and Ag.

In comparing the capture γ-ray spectra from thermal neutrons with those for neutrons with $E_n = 0.1$ MeV, it appeared that the latter are much softer for the case of nickel and gold. The authors gave the following possible causes of the difference in the spectra.

1. The neutrons may be captured either via the compound nucleus or by a direct capture mechanism. Naturally the capture γ-ray spectrum will change when the relative contributions to the total cross section of these two components change.

2. For thermal neutrons, only S capture is possible, but with increasing neutron energy the probability for P capture increases. This also may affect the radiative capture spectrum.

In the present work we have studied the capture γ ray spectra from five elements: manganese, cobalt, the normal mixture of iron isotopes, nickel, and copper, using a broad spectrum of neutron energies. The neutron source was a zero-power uranium-water reactor. The elements chosen have, on the one hand, relatively high cross sections for fast neutrons and, on the other hand, in the γ ray spectrum from thermal capture they have a large fraction of high energy γ quanta.

EXPERIMENTAL ARRANGEMENT AND RESULTS OF MEASUREMENTS

Figure 1 shows the arrangement of the experimental equipment for studying spectra from radiative capture of neutrons in samples of manganese, cobalt, iron, nickel, and copper. The computed neutron spectrum at the sample position is shown in Fig. 2.[1)] An experimental check of the computation was made by total activations of various

[1)]The spectrum was computed by V. P. Kochergin et al.

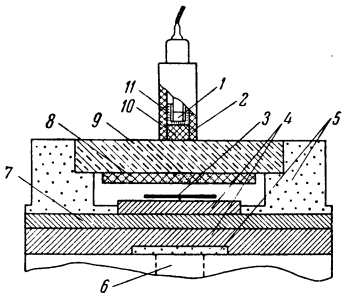

FIG. 1. Schematic of arrangement of equipment for measuring γ rays from radiative capture of fast neutrons: 1 – NaI crystal, 2 – bismuth shield for detector, 3 – sample, 4 – bismuth shield of reactor, 5 – boron carbide shield, 6 – active zone of the reactor, 7 – lead shield, 8-11 – neutron shield (8 – paraffin and B_4C, 9 – lucite and B_4C, 10 – paraffin, 11 – B_4C).

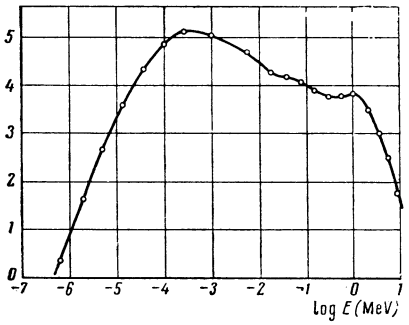

FIG. 2. Neutron spectrum from reactor (computed). Ordinate is log N(E) (neuts/MeV).

neutron detectors whose relative efficiencies were measured in a thermal neutron flux.

The γ-ray detector was a single crystal scintillation spectrometer using a NaI(Tl) crystal 40 mm in diameter and 40 mm high. The pulses were analyzed with a 128-channel pulse analyzer [5].

The main difficulty in measuring the γ-ray spectra from radiative capture comes from the γ-ray background emerging from the reactor and the background from neutron capture in the iodine of the detector itself. To reduce the background, a layer of bismuth 9 cm thick was placed over the boron carbide filter (Fig. 1), and then a layer of lead 8 cm thick, and finally a bismuth layer 8.5 cm in thickness. The γ ray detector was shielded by a neutron shield consisting of a mixture of lucite and boron carbide; in addition the detector was surrounded by a case of boron carbide and a bismuth shield 4 cm thick.

The samples, which were 40 × 40 cm and had a thickness equal to (or less than) a free path for γ rays in the 5–7 MeV range, were placed on top of the bismuth. For backgrounds the sample was replaced by a thickness of lead which produced the same attenuation of the γ rays in the 5–7 MeV

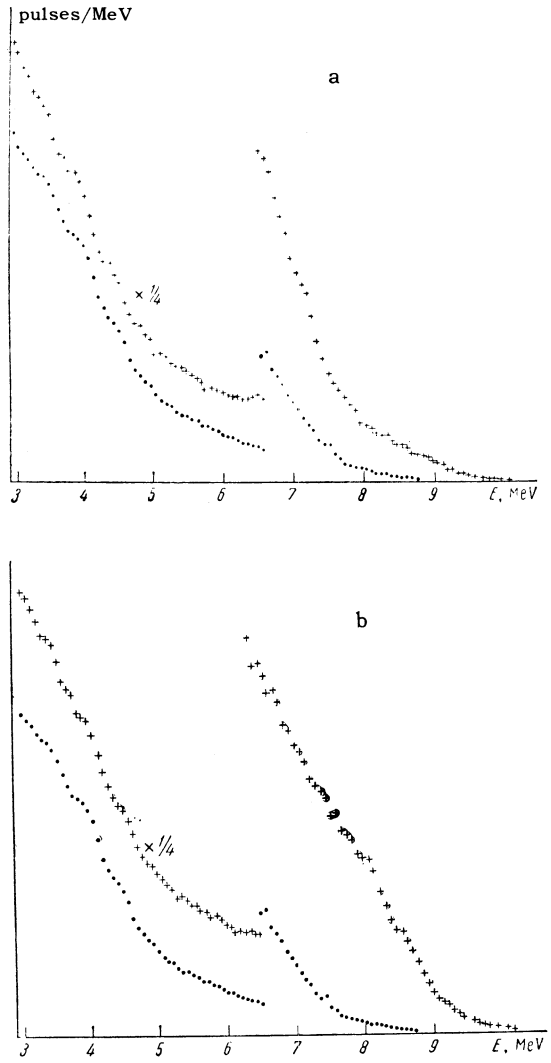

FIG. 3. Pulse height spectra: a) iron, b) nickel; + – effect plus background, ● – background.

range. It should be noted that the pulse distributions measured with and without the lead samples coincide to within 5% for γ-ray energies above 4 MeV.

The measured spectra were compared with spectra from thermal neutron capture under the same geometrical conditions. For this purpose, the samples were placed between two layers of paraffin 4–5 cm in thickness; the background was determined by shielding the samples with boron carbide. The cadmium ratio, measured from activation of Cu^{65}, was ~25. The energy scale of the spectrometer was calibrated and checked periodically with the γ rays from a Po-α-Be source (4.47 MeV).

The ratio of background and effect and the statistical spread of the points from measurement of the γ ray spectrum from capture of neutrons from the reactor are shown in Fig. 3. In the measure-

ments of γ ray spectra from thermal neutron capture, the background was 10—30%.

Figure 4 shows the γ-ray spectra from radiative capture of thermal neutrons and those obtained from capture of neutrons from a broad spectrum of neutron energies. The curves are normalized at a γ-ray energy of 6—7 MeV. Naturally such a normalization permits one only to judge relative changes in the parts of the spectra for $E_\gamma > 3$ MeV. The spectra shown are the average of four to six measurements. The graphs show the mean square errors of the measurements. The mean square errors of the spectra from thermal neutron capture are ~3—5%.

Identification of the energy region in which the neutron capture occurred was done from the computed neutron spectrum and from radiative capture cross sections found by V. I. Golubev and M. L. Nikolaev (private communication).

DISCUSSION OF RESULTS OF MEASUREMENTS

1. Manganese. The majority of the captures (~ 70%) in manganese for measurements with the reactor spectrum occur in the region of the 377 eV resonance. A comparison of the spectra (Fig. 4a) of γ rays from radiative capture of thermal neutrons and of neutrons mainly in the region around 337 eV shows that

1) within the resolution of the spectrometer and the errors in the measurements, the shapes of the spectra are the same above 4.5 MeV;

2) in the energy range of 3—4.5 MeV, there is a slight difference between the spectra, which shows that the intensity of the γ-ray group near 5 MeV is comparatively weaker in the case of capture of resonance neutrons.[2)]

The difference in the intensity of γ transitions with energy ~ 5 MeV and those to levels close to the ground state (~ 7.2 MeV) is difficult to explain in terms of a change in partial γ-ray widths, since in the capture of thermal neutrons excited states with spin and parity 2^- are formed with a probability ~ 0.8 and those with 3^- are formed with probability ~ 0.2, while in the capture of resonance neutrons the 2^- states have probability ~ 0.7 and the 3^- states have probability ~ 0.3.[3)]

2. Cobalt. In the case of Co^{59}, most of the radiative capture of reactor neutrons occurs in the region of the 132 eV resonance. The γ ray spectra from capture of resonance and thermal neutrons coincide within the limits of error of the measurements (cf. Fig. 4b). The agreement in the shape of the spectra is understandable if we consider that the capture in the thermal region is due mainly to the 132 eV resonance (for this it is sufficient to have $\Gamma_\gamma \sim 0.5$ eV).

3. Iron. In this case more than half (~ 60%) of the neutrons are captured with energy greater than 100 keV. Changes in the γ-ray spectrum were mainly of two kinds:

a) the intensity of transitions with energies above 7.6 MeV (transition to the ground state of Fe^{57}) increased for capture of fast neutrons, compared with the 7.6 MeV transition;

b) the intensity of transitions with energy < 6 MeV rose relative to the 7.6 MeV transition.

The increase in intensity of γ rays with energies above 7.6 MeV is related to at least two causes. As has been shown in many papers (cf., for example,[7] and especially [8]), for energies above a few tens of keV the capture cross section of even-even isotopes is 5—15 times less than the capture cross section of even-odd isotopes. The normal mixture of isotopes of iron contains 2.2% of Fe^{57}; after radiative capture, Fe^{58} is formed with a binding energy of 10.0 MeV. Analysis of the experimental data shows that the intensity of γ rays with energies above 8 MeV increases by a factor of 6—10 relative to the 7.6 MeV transition. In addition the probability for transition to the ground state in Fe^{58} after P capture may be greater if we assume that in the capture of thermal neutrons there is a sizable contribution from formation of an excited state with spin-parity 0^-.

The reduction in intensity of γ transitions with energy 7.62 MeV compared to the lower energy transitions in the capture of fast neutrons can be explained either by a large contribution from capture of P neutrons (estimates have shown [4)] that the radiative capture cross section for P neutrons is three times as large as for S neutrons, for neutron energies above 100 keV), or by the possibly important role of direct neutron capture in the thermal region.

4. Nickel. Most of the neutron captures (~ 70%) for the reactor spectrum occur in the energy range

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[2)]The shape of the spectrum with this spectrometer is such that the most intense peak corresponds to $E_\gamma - 2m_0c^2$.

[3)]The contributions of the 2^- and 3^- excited states to the capture of thermal neutrons and neutrons in the reactor spectrum were estimated using $\Gamma_\gamma = 0.5$ eV. The values of Γ_n were taken from [6].

[4)]In computing the cross sections we used the expression for the radiative capture cross section in a region of nonoverlapping resonances ($\overline{\Gamma}_n/\overline{D} \leq 1$), averaged over many levels of the compound nucleus. The values of the penetration factors were taken from [9], $\Gamma_\gamma = 0.5$ eV and $D = 10^3$ eV.

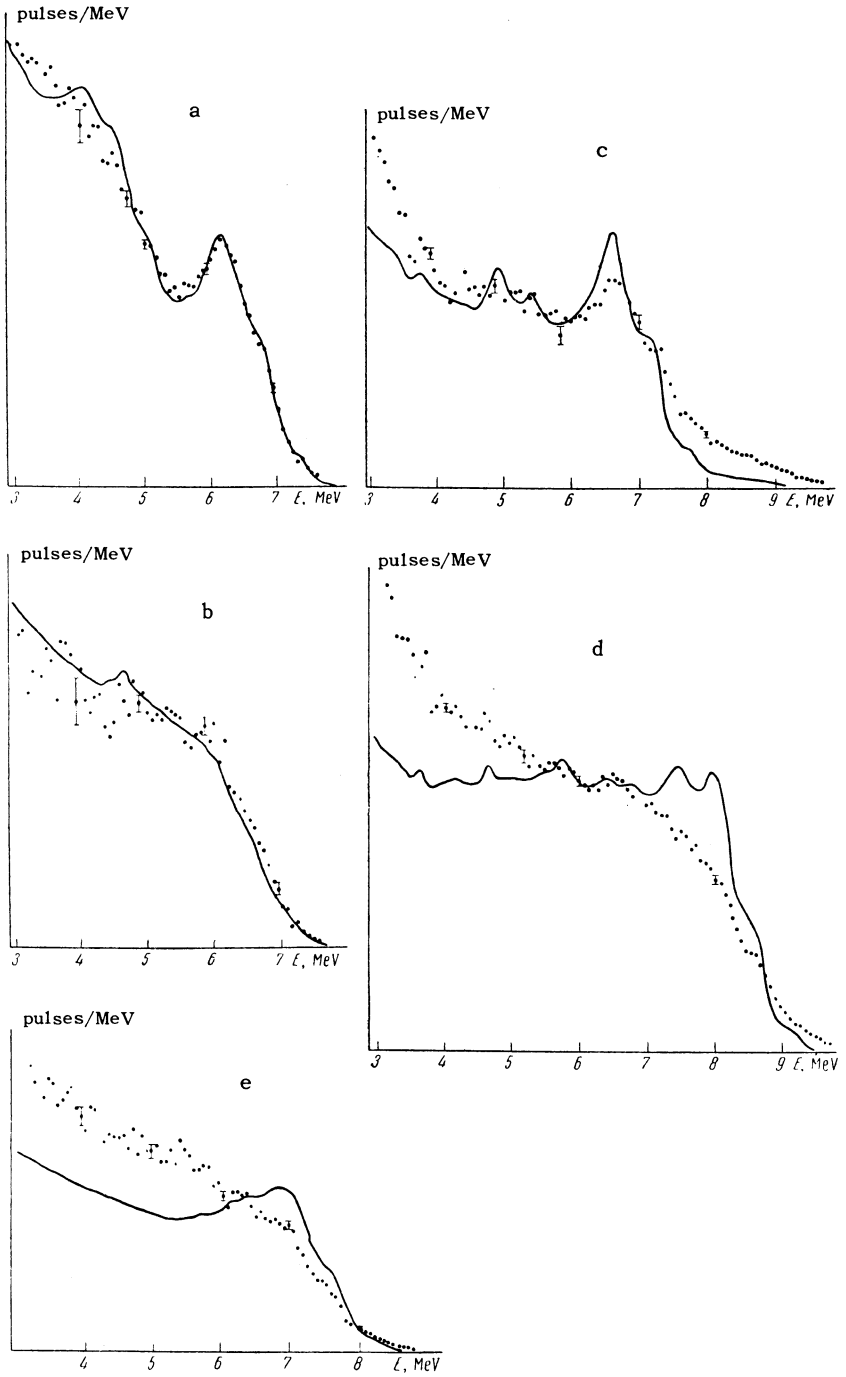

FIG. 4. Gamma spectra from radiative capture of thermal neutrons (solid curve) and neutrons from the whole reactor spectrum (dotted curve): a – Mn, b – Co, c – Fe, d – Ni, e – Cu.

above 100 keV. It is completely clear that the γ rays from capture of fast neutrons are much softer than those for thermal neutron capture (Fig. 4). High energy quanta with energies above 9 MeV appear. The simplest explanation for the change in the spectrum is a change in the relative contributions of the various isotopes to the total capture cross section when we change from fast to thermal neutrons.

In radiative capture of thermal neutrons, the main contribution comes from the two isotopes Ni^{58} and Ni^{60}, having cross sections of 4.2 and 2.5b, while the binding energies of the product nuclei are 8.998 MeV for Ni^{59} and 7.820 MeV for Ni^{61}.

Most of the γ transitions in both isotopes (Ni^{59}, Ni^{61}) go to the ground state or to levels close to it. There is no reason for assuming that the ratio of the capture cross sections for these two isotopes remains the same for fast neutrons as for thermal. Since both isotopes (Ni^{58} and Ni^{60}) are even-even, it is reasonable to assume that they have the same capture cross section for fast neutrons [8]. In this case the contribution

of the two isotopes to the total capture cross section will be proportional to their percentage content in the normal mixture, and one will observe a softening of the spectrum because of the increase of the intensity of the 7.8 MeV transition (Ni^{61}) relative to the 9.0 MeV transition (Ni^{59}). The appearance in the radiative capture spectrum from fast neutrons of γ quanta with $E_\gamma > 9$ MeV is obviously due to Ni^{62} nuclei which have a binding energy of 10.626 MeV. The content of Ni^{61} in the normal mixture is 1.25% and its cross section is 1.9 b. But since this nucleus is even-odd, its capture cross section for fast neutrons may be considerably greater than the cross section for the other, even-even, isotopes of nickel.

5. Copper. As in the case of nickel, the spectrum of capture γ radiation from fast neutrons is considerably softer (Fig. 4e). In the normal isotopic mixture there are only two isotopes, Cu^{63} and Cu^{65}, with thermal neutron capture cross sections of 4.3 and 2.1 b. But for copper one can evaluate the contributions of the individual isotopes to the total cross section, since the product nuclei Cu^{64} and Cu^{66} are unstable and undergo β decay with periods of 12.8 h and 5.1 min. To do this we placed copper foils at the sample position and irradiated with thermal and fast neutron spectra, and compared the ratios of activation of Cu^{64} and Cu^{66}. The ratios agreed to within 7%. In addition, from the known reactor spectrum (Fig. 2) we computed the total activations for the isotopes. Their ratio was compared with the measured ratios of activations of the two isotopes by thermal neutrons. The results again agreed to within 6%. From this it follows that such a large change in the spectra cannot be explained by a change in the relative contribution of the individual isotopes to the total capture cross section.

The observed changes in the spectrum for capture of neutrons from the reactor spectrum (Fig. 2) show that there is a significant decrease (by at least a factor of two) in the intensity of γ rays with energies of 7—8 MeV compared to those in the 4—6 MeV range. This may be explained as an effect of P capture, since the contribution of P neutrons is considerable ($\sim 40\%$ of the neutrons in the reactor spectrum are captured with energies above 100 keV, while the ratio of the cross sections for radiative capture of S and P neutrons in this energy range is $\frac{1}{2}$), if we assume that there are negative parity levels in Cu^{64} and Cu^{66} in the region of excitation energies from 0.5 to 3 MeV. If, however, the negative parity levels have excitations above 2—3 MeV, the change in the capture γ ray spectrum can be explained in terms of a large contribution of "direct" capture to the total cross section for thermal neutron capture. The latter does not contradict theoretical estimates of the upper limit of the cross section for radiative capture of thermal neutrons [10].

In conclusion, the authors take this opportunity to express their gratitude to I. M. Bondarenko, A. V. Malyshev, and V. S. Stavinskiĭ for discussion of this work.

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1