

Inelastic Scattering of 0.3, 0.77 and 1.0 mev Photoneutrons*

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(Submitted to JETP editor December 2, 1955)

J. Exptl. Theoret. Phys. (U.S.S.R.) 30, 1017-1024 (June, 1956)

We have measured the cross section for inelastic scattering of neutrons of three energies: 0.3, 0.77 and 1.0 mev by the nuclei of 13 elements: uranium, bismuth, lead, mercury, wolfram, antimony, tin, cadmium, copper, nickel, iron, aluminum and sodium. For heavy nonmagic nuclei (uranium, mercury and wolfram), the cross section for inelastic scattering of neutrons with energies 0.8 and 1 mev was found to be 1-2 barns. For light and magic nuclei the cross section for these same neutrons was found to be small, of order 0.1-0.2 barns. In addition, for incident neutrons with $E_0 = 1$ mev, an estimate was made of the average energy of the inelastically scattered neutrons, and the energy levels excited were determined for some nuclei.

1. INTRODUCTION

THE investigation of the inelastic scattering of neutrons in the 0.3-1 mev energy region from nuclei of various elements has great theoretical and practical interest. Measurements of the inelastic effect are especially important for those elements which are structural materials for reactors. In addition, data from such measurements are a test of existing theoretical descriptions of nuclear structure. Unfortunately, the great experimental difficulties associated with such measurements are an obstacle to any complete investigation of the effect. Thus, up to now, too little effort has been devoted to the study of inelastic scattering, while the published results are usually very crude and often insufficiently reliable. The investigation of inelastic scattering of neutrons of energy 1 mev and below is especially difficult. For these energies there are no published data at present. The present work deals with just this energy region.

2. METHOD AND MEASURING TECHNIQUE

As sources, we used photoneutrons from Na-Re, La-Re and Na-D₂O (see Fig. 1). The inelastic effect was measured in experiments using spherical geometry, from the change in the initial neutron spectrum in the materials under study; the materials were in the form of spheres (for convenience, they consisted of two hemispheres) with diameter $D = 230$ mm, having a small cavity at the center, of diameter 50 mm for the neutron sources.

The neutrons were recorded by using nuclear recoils in a spherical ionization chamber filled with hydrogen or helium. The type of chamber used depended on the neutron energy: for La-Be and Na-Be neutrons the measurements were made with a helium chamber (volume 4 liters, pressure

* This work was completed in 1953-1954. The results were reported at the Geneva International Conference on Peaceful Uses of Atomic Energy.

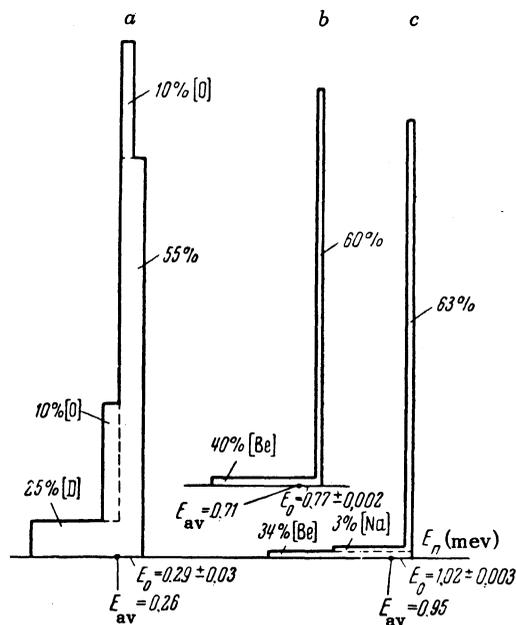


FIG. 1. Neutron spectrum from spherical sources: a--Na-D₂O, b--La-Be, c--Na-Be, used in the present work. The last two sources are especially monochromatic. We give, in percent, the fraction of the neutrons which suffer a single elastic scattering in the source itself, for each of the source materials.

3 atm), for the Na-D₂O neutrons they were done with a hydrogen chamber (volume 4 liters, pressure 1 atm). The experiments consisted in measuring the initial spectrum (from the source without scatterer) and the secondary spectrum (from the source placed at the center of the scatterer), reduced to the same time and source distance from the ionization chamber. Individual pulses from the ionization chamber were amplified in a type ULZD proportional amplifier and recorded by a twenty channel

analyzer. The duration of each measurement was 30-60 minutes, during which time we obtained 900-1000 pulses per channel.

To reduce the results to the initial activity, additional time was added to the duration of each successive measurement to correct for the decay of the source, using the fact that the half life for Na^{24} is 14.8 hr, for La^{140} , 42 hr. The difficulties in measurement arose mainly from the strong γ -radiation accompanying the neutrons, since the number of γ -quanta was approximately 10^5 times the number of neutrons. Therefore, the copiously formed secondary electrons which enter the gas from the walls of the chamber give rise to a large background, whose superposition on the neutron pulses causes a statistical spread in their amplitudes.

To shield the chamber from γ -radiation we placed over the scatterer and auxiliary lead shield of spherical shape and wall thickness 100 mm (which produced 100-fold attenuation).

It was shown by separate experiments that there is no inelastic scattering in lead in the energy region from 0.3 to 0.9 mev (cf. below). Therefore, the use of lead as a shield does not introduce errors into the measurements, and the spectrum of the source surrounded by only the lead shield may be taken to be the primary spectrum. Another method which was used to separate the neutron pulses from the background of secondary electrons was the introduction into the amplifier circuit of a differentiating network with low RC, equal to 3 microseconds.

3. CONSTRUCTION AND OPERATION OF THE SPHERICAL IONIZATION CHAMBER

A spherical chamber filled with pure hydrogen or helium is most effective for recording photon neutrons. Especially simple in construction is the glass spherical chamber, silvered on the inside and coated with a layer of electrolytic copper on the outside for withstanding high pressures. The advantages of the spherical chamber over other types are:

- 1) small capacity of the collecting electrode, of order 5-10 cm;
- 2) efficiency independent of the direction of incidence of neutrons;
- 3) optimal ratio of chamber wall surface to working volume, and consequent lower background from γ -radiation producing secondary electrons in the walls of the chamber;
- 4) small inductive effect, of the order of 5%, from positive ions;

5) constant effective time for electron collection, independent of place of formation of charges in the chamber.

These advantages as well as proper operation of the spherical chamber are possible only when it is filled with pure gases with 100% electron collection. Even small impurities of oxygen and water vapor lead to electron attachment which in general distorts the spectrum to be measured. For this reason, commercial hydrogen and helium were purified by passing over copper turnings heated to 300° , and tested for purity by filling a chamber and observing pulses from polonium α -particles. Only in pure gases are the amplitude and the leading edge of pulses determined by electron collection. In addition, the effective electron collection time is the same, about 10 microseconds, independent of the amount or place of formation of charge; pulses of different amplitude then differ from one another only in the slope of their leading edge. This fact enabled us to introduce into the amplifier a differentiating circuit with $RC = 3$ microseconds, a time constant which is smaller than the effective collection time for electrons. Such a network produces almost complete discrimination of pulses from background. The pulse shape is altered and the amplitude decreased, but pulses at the output of the circuit are still proportional to the input pulses.

The shape of the overall spectrum (spectrum of recoil nuclei) depends on the chamber filling. We know that the spectrum of recoils in hydrogen is uniform in energy for monochromatic incident neutrons. Unlike the case of hydrogen, the scattering of neutrons in helium is anisotropic^{1,2} and preferentially fore and aft, so that the recoil spectrum rises toward the low and high energy sides. From an analysis of the spectrum of recoils in a helium chamber for various neutron energies, we were able to show that in the 0.3-1.0 mev region the distributions of recoil nuclei have a common, approximately straight portion independent of the energy of the incident neutrons. Thus, for incident neutrons with energy E_0 and scattered neutrons with energy $E < E_0$, the recoil spectra can be described in general as:

$$N_1(E) \approx (A + BE)n(E_0), \quad (1)$$

$$N_2(E) \approx (A + BE) \int_E^{E_0} n(E) dE, \quad (2)$$

where A and B are parameters which can be eliminated by dividing:

$$\frac{N_2}{N_1} = \int_E^{E_0} n(E) dE. \quad (3)$$

The spectrum of scattered neutrons is obtained by differentiating Eq. (3):

$$n(E) = -\frac{d}{dE}(N_2/N_1). \quad (4)$$

One can see from Figs. 2 and 3 that the anisotropy of scattering in helium is especially large for energy $E_0 \approx 1$ mev, for which the ratio of maximum to minimum in the recoil spectrum amounts to 2:1 and the effect to be measured is, as a result, more strongly emphasized. On the other hand, in the low energy region $E_0 = 0.2-0.3$ mev the helium chamber does not have these advantages, and it is therefore more convenient to use a hydrogen chamber as neutron detector.

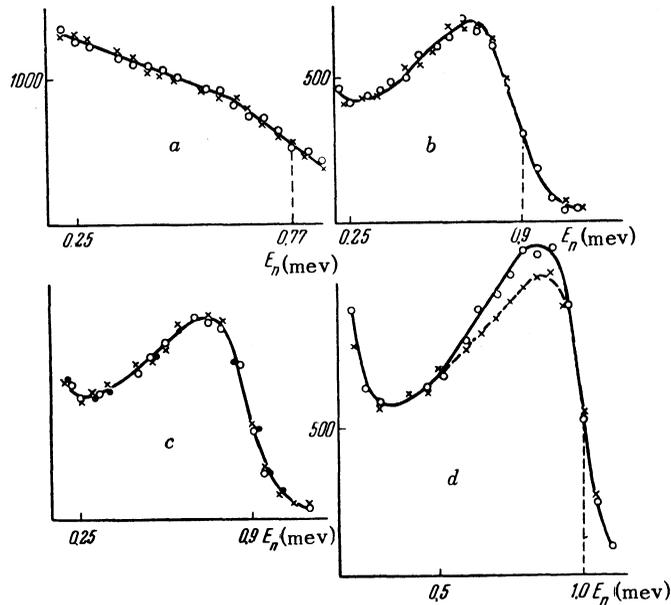


FIG. 2. Spectra of lead and bismuth. *a*--H₂-chamber, *b, c, d*--He-chamber; curves *a, b, d* taken with Pb (crosses) and Bi (circles); curve *c* taken only with Pb for varying thicknesses: 150, 190, 230 mm. In the region above 0.9 mev we observe inelastic scattering in lead corresponding to an energy level $E = 550 \pm 50$ kev.

4. CHARACTERISTICS OF THE PHOTONEUTRON SOURCES

The energy of the photoneutrons is determined by the energy of the γ -ray and by the threshold energy as follows:

$$E = \frac{A-1}{A} (E_\gamma - Q) + \delta. \quad (5)$$

Here δ is the small spread in neutron energies depending on the angle between the directions of the neutron and γ -ray

$$\delta = E_\gamma (\cos \theta) \left[\frac{2(A-1)(E_\gamma - Q)}{931A^3} \right]^{1/2} \approx (0.1-1\%) E_\gamma. \quad (6)$$

In principle, one can obtain neutrons which are almost homogeneous in energy, with a small spread $\delta = \pm(0.1-1\%) E_\gamma$. If we assume $^3 E_\gamma(\text{Na}) = 276$ mev, $E_\gamma(\text{La}) = 2.49$ mev, $Q(\text{Be}) = 1.63$ mev and $Q(\text{D}) = 2.18$ mev, then the maximum energies of photoneutrons as given by Eqs. (5) and (6) for each source are, respectively, equal to the quantities given in Table I. However, because of the slowing down of the neutrons in the sources themselves, the spread in energy, Δ , can reach several percent. For a spherical (Na-Be)-source, made up of two concentric spheres--the inner one 30 mm in diameter containing 13 gm of sodium, and the outer one a hollow sphere of beryllium with wall thickness 10 mm, it can be shown that 34% of all the neutrons are scattered once in the beryllium,

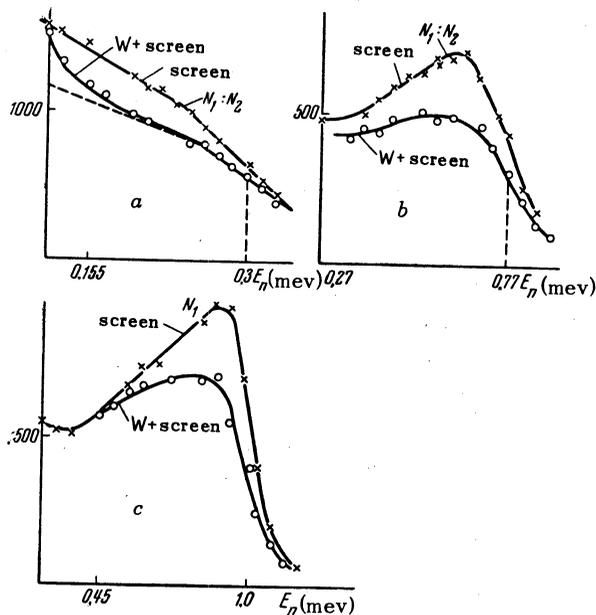


FIG. 3. Wolfram spectrum. *a*--H₂-chamber, *b* and *c*--He-chamber. *a*--Na-D₂O, *b*--La-Be, *c*--Na-Be. Inelastic scattering is observed at all energies. In particular, for $E = 0.3$ mev only the first level at 145 ± 20 kev is excited.

TABLE I

Source	E_0 in mev	Spherical		Cylindrical	
		E_{av} in mev	Δ in %	E_{av} in mev	Δ in %
Na — Be	1.02 ± 0.003	0.95	5	0.80	20
La — Be	0.77 ± 0.002	0.71	8	0.62	20
Na — D ₂ O	0.29 ± 0.03	0.26	10	0.22	33

and 3% in the sodium, while the remaining 63% of the primary neutrons with energy $E_0 = 1$ mev emerge without being scattered. From these data, we can construct the neutron spectrum, and obtain from it the average neutron energy E_{av} (cf. Fig. 1). Table I gives the characteristics of the spherical sources used in the present work, and includes, for comparison, the characteristics of the cylindrical sources with which Wattenberg and Hughes worked^{3,4}. As one sees, the spherical sources are more monochromatic. Moreover, for the first two sources, Na-Be and La-Be, the homogeneity is so great that we can, for practical purposes, take as the effective energy the maximum energy E_0 which is 1.0 mev for the Na-Be source and 0.77 mev for the La-Be source.

5. RESULTS OF MEASUREMENTS AND CALCULATION OF σ_{in}

Assuming that because of inelastic scattering the number of neutrons with the initial energy E_0 decreases exponentially (to be more precise, in cases where the inelastic effect is large as in uranium, mercury and wolfram, we must correct l for the inelastic scattering itself),

$$N_2(E_0) = N_1(E_0) e^{-nl\sigma}, \quad (7)$$

from which we can obtain a formula for computing σ_{in}

$$\sigma_{in} = (1/nl) \ln(N_1/N_2), \quad (8)$$

where N_1/N_2 is the ratio of the number of pulses in the primary and secondary recoil spectra, respectively, with energy E_0 , and l is the mean distance traversed by the neutrons in the scatterer, calculated in the diffusion approximation (where we neglect the volume of the cavity in which the neutron source is located),

$$l = \frac{R_1^2}{2\lambda_1} + \frac{R_1^2}{\lambda_2} - \frac{R_1^3}{\lambda_2(R_2 + 0.7\lambda_2)} \quad (9)$$

R_1 and λ_1 are the scatterer radius and the transport mean free path in the scatterer, R_2 and λ_2 are the radius of the lead shield and the transport free

path in lead.

The errors which are introduced into σ_{in} because of the use of the diffusion approximation for computing the mean distance traversed and because of the assumption of exponential attenuation of the primary spectrum are relatively small. The errors in σ_{in} because of the corrections for neutron absorption in the scatterer⁴ and inelastic scattering in the lead shield are also small.

The significant errors in σ_{in} will depend mainly on the accuracy (5%) with which the primary and secondary spectra are measured, and on the accuracy (10-15%) of the published⁵ values of λ . The combined errors are included in Table II.

TABLE II. Inelastic Scattering Cross Section σ_{in} in Barns

Neutron Energy, E_n	0.3 mev	0.77 mev	1.0 mev
Uranium	0.4 ± 0.1	0.9 ± 0.3	1.6 ± 0.5
Bismuth	0	0	< 0.1
Lead	0	< 0.1	0.2 ± 0.1
Mercury	0.1 ± 0.06	0.8 ± 0.2	1.5 ± 0.4
Wolfram	0.4 ± 0.2	1.3 ± 0.4	2.6 ± 0.8
Antimony	0	0.4 ± 0.2	0.7 ± 0.3
Tin	0	< 0.1	0.4 ± 0.2
Cadmium	< 0.1	0.6 ± 0.2	1.0 ± 0.2
Copper	0	0	0.2 ± 0.1
Nickel	0	0	< 0.1
Iron	0	0	0.3 ± 0.1
Aluminum	0	< 0.1	0.2 ± 0.1
Sodium	0	> 0.2	0.4 ± 0.2

In addition to the cross sections, whenever it was possible, we determined from the spectrum of recoils from the scattered neutrons the energies of the individual levels which were excited or the average energy of the inelastically scattered neutrons (see Table III). The determination of these energies can be done very roughly, in the most favorable case with an accuracy of $1\frac{1}{2}$ channels out of 20, which amounts to 5-7% of the energy of the incident neutrons. The energies of some of the excited levels are given in the summary table, with this resolution. We determined much more poorly the mean energy of the inelastically scattered neutrons, whose spectra are not measured directly in the present experiments, but can be gotten only by graphical differentiation of the recoil spectra [in accordance with Eq. (4)], which gives a qualitative rather than a quantitative result. The low energy end of the spectrum of inelastically scat-

tered neutrons is determined especially poorly, so that we used the convention of setting the average energy equal to the "median", i.e., the energy above and below which half the recoils occur.

For example, in Fig. 2 are given the results of measurements made with lead and bismuth. It is apparent that for three of the energies--0.3, 0.77 and 0.9 mev, the recoil spectra for lead and bismuth coincide completely within the limits of error of the experiments, so that we may assume that absorption and inelastic scattering of neutrons with energy below 0.9 mev does not exceed 0.1 barns. In the region above 0.9 mev (for neutron energy 1 mev) appreciable inelastic scattering is observed in lead: the lead spectrum runs lower over the whole interval from $E_0 = 1$ mev to $E = 0.45$ mev. Thus, we observe a drop in energy equal to 550 ± 50 kev, which agrees well with the known energy⁶ 0.6 mev of the first level in Pb²⁰⁷. The ratio

TABLE III

Neutron Energy, E_n	0.3 mev	0.77 mev	1.0 mev
Uranium	—	$E_{av} = 350$	$E_{av} = 500$
Bismuth	—	—	—
Lead	—	—	$E = 550 \pm 50$
Mercury	$E > 200$	$E_{av} = 300$	$E_{av} = 400$
Wolfram	$E = 145 \pm 20$	$E_{av} = 400$	$E_{av} = 550$
Antimony	—	$E_{av} = 430$	$E_{av} = 450$
Tin	—	—	$E > 800$
Cadmium	—	$E_{av} = 300$	$E_{av} = 400$
Copper	—	—	$E > 800$
Nickel	—	—	—
Iron	—	—	$E = 800$
Aluminum	—	—	$E = 400 \pm 50$
Sodium	—	—	$E = 440 \pm 50$

Note: E is the energy, in kev, of individual levels excited by inelastic scattering of neutrons; E_{av} is the average energy, in kev, of the inelastically scattered neutrons.

$N_1/N_2 = 1.1$ is determined directly from the curves. Then, neglecting the absorption ($\sigma_c = 20$ millibarns), we get

$$\sigma_{in} = \frac{1}{nl} \ln 1.1 = 0.2 \pm 0.1 \text{ barn},$$

where $n = 0.33 \times 10^{24}$ nuclei/cm³ and $\bar{l} = 14.7$ cm.

As another example, Fig. 3 gives the results for wolfram. The scatterer consisted of two hemispheres with $D = 220$ mm, filled with powdered wolfram having a density $\rho = 5.47$ gm/cm³. It is apparent that for incident neutrons with $E_0 = 0.3$ mev the primary and secondary spectra from the hydrogen chamber differ considerably. About 25% of the secondary spectrum is shifted by more than 100 kev, i.e., some of the neutrons have suffered an inelastic energy loss in the wolfram. A rough estimate shows that only one level, the first excited state with energy $E = 145 \pm 20$ kev, has been excited.

The ratio $N_1/N_2 = 1.25$ was found. Then, eliminating the absorption ($\sigma = 0.09$ barns), we get

$$\sigma_{in} = \frac{1}{nl} \ln 1.25 - 0.09 = 0.4 \pm 0.2 \text{ barn},$$

where $n = 0.018 \times 10^{24}$ nuclei/cm³ and $l = 24$ cm.

The measurements for incident neutrons with $E_0 = 0.77$ mev were carried out with a helium chamber. The ratio $N_1/N_2 = 1.45$ was found. Neglecting

absorption ($\sigma_c = 10$ millibarns), and correcting l for inelastic scattering, we find

$$\sigma_{in} = \frac{1}{nl} \ln 1.45 = 1.3 \pm 0.4 \text{ barn}$$

where $n = 0.018 \times 10^{24}$ nuclei/cm³ and $\bar{l} = 16$ cm. An estimate of the average energy of the inelastically scattered neutrons gives $E_{av} \approx 400$ kev. The measurements for incident neutrons with $E_0 = 1$ mev were also done with a helium chamber. The ratio N_1/N_2 was 1.4. A factor of 1.1 must be applied to this ratio to correct for inelastic scattering in the shield ($\sigma_{in} \approx 200$ millibarns). Then, neglecting absorption and correcting l for inelastic scattering we get

$$\sigma_{in} = \frac{1}{nl} \ln 1.4 \cdot 1.1 = 2.6 \pm 0.8 \text{ barn}$$

where $l = 11$ cm. A rough estimate of the average energy gives $E_{av} = 550$ kev.

In conclusion, the authors thank A. I. Leipunskii for general guidance and interest in the work.

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Translated by M. Hamermesh
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