Neutron Activation Cross Sections at 25 kev*

REX BOOTH, WILLIAM P. BALL,† AND MALCOLM H. MACGREGOR
University of California Radiation Laboratory, Livermore, California
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Neutron activation cross sections have been measured at 25 kev for 31 isotopes. An Sb-Be photoneutron source was used, and thermal activations served to calibrate the beta- and gamma-detector efficiencies. The cross sections were measured relative to iodine. A comparison was made between measured cross sections and predictions based on known low-energy resonance parameters.

INTRODUCTION

THE energy region of a few kiloelectron volts has proved to be a difficult one for neutron cross-section measurements. In the case of total cross sections, where only a transmission measurement is required, the upper energy limit of reactor fast-choppers and the lower energy limit of monoenergetic accelerator neutron sources overlap, so that the entire region is covered. However, the measurements of the partial cross sections, which in this region are principally elastic scattering and neutron capture cross sections, require neutron fluxes an order of magnitude greater. In such a situation, antimony-beryllium photoneutron sources compete favorably with reactor and accelerator neutron sources for the measurement of capture cross sections at 25 kev, particularly when a cross section averaged over a range of energies is all that is needed.

Capture cross sections in the kev region are important both for cosmological studies and for reactor calculations. Since current cosmological theories of the formation of elements assume temperatures in the range corresponding to a few kev,[1] a knowledge of the capture cross sections at these energies is essential in carrying out build-up calculations. Some of the fast-reactor designs also require knowledge about this energy region. Measurement of all the required cross sections is a difficult and lengthy task; measurement of a portion of the list, however, is very useful for two reasons: (1) the measured cross sections serve as a test for the computation of capture cross sections by means of low-energy resonance parameters; (2) if a capture cross section is known at one energy, it can be calculated at nearby energies.[2]

Determination of capture cross sections by the activation technique limits measurements to cases where neutron capture leads to a radioactive nucleus having a suitable half-life and suitable beta or gamma activity. Detector calibration is accomplished by measuring known thermal cross sections for the same isotope. Similar measurements have been carried out by Hummel, Hamermesh, and Kimball,[3,4] and by Macklin, Lazar, and Lyon,[5] the latter group using absolute gamma-ray counting to obtain the detector efficiency.

NEUTRON SOURCES

The photoneutron source consisted of an antimony cylinder suspended inside a beryllium annulus. The initial Sb^{124} activity was about 100 curies, with a half-life of 60 days, and the initial total neutron production was 10^8 neutrons/sec, as measured with the manganese bath technique. The geometry of the source was chosen to be similar to that used by Hamermesh,[3,4] since measurements have been made to infer the neutron spectrum from such a source.[6] The "effective" neutron energy is about 25 kev, with an energy spread of about ± 5 kev. A sample holder supported five samples adjacent to the beryllium annulus, and it was rotated by a motor so that all samples would be equally irradiated. Neutron scattering from the floor and shielding walls was calculated to be small.[7]

Thermal-neutron activations were made using a water-moderated Po-Be neutron source. The cadmium ratio was determined for each sample, and corrections were made for epithermal neutrons. The Po^{210} source has a half-life of 138 days. In making measurements, both the Sb-Be and the Po-Be source strengths were referred to a fixed time in order to facilitate comparisons. Irradiation times varied from a few minutes to a week.

COUNTERS

Automatic counting and recording equipment registered beta and gamma activities simultaneously. A counter wheel handled up to 24 samples at a time, and data were recorded photographically. A beta counter, consisting of a methane proportional flow counter with a 0.7-mg/cm² aluminum-coated Mylar window, was placed on one side of the sample, and a 2-in. diameter by 1-in.-thick NaI(Tl) scintillation gamma counter was placed on the other side; the counters were run on plateaus and were periodically checked for constancy

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† Now at the Ramo-Wooldridge Corporation, Los Angeles, California.
[1] See, for example, A. G. W. Cameron, Chalk River Laboratory Report CRL-41, Chalk River, Canada, June, 1957 (unpublished).
[2] H. W. Newson (private communication).
[3] V. Hummel and B. Hamermesh, Phys. Rev. **82**, 67 (1951).
[4] C. Kimball and B. Hamermesh, Phys. Rev. **89**, 1306 (1953).
[5] Macklin, Lazar, and Lyon, Phys. Rev. **107**, 504 (1957).
[6] R. Culp and B. Hamermesh, Phys. Rev. **93**, 1025 (1954); see also discussion of this point in reference 5.
[7] D. W. Glasgow, General Electric Company, Hanford Works Report HW-32086, Richland, Washington, 1954 (unpublished).

TABLE I. Neutron capture cross sections at 25 kev.

Isotope	$T_{\frac{1}{2}}$	R	ΔR (%)	σ_{th} [a] (barns)	$\Delta\sigma_{th}$ (%)	$\sigma(n,\gamma)$ (millibarns)	$\Delta\sigma(n,\gamma)$ (millibarns)
Na^{23}	15 hr	1.386	12	0.53	4	1.10	0.23
Si^{30}	2.62 hr	0.114	76	0.11	9	1.9	1.5
K^{41}	12.5 hr	0.125	12	1.15	10	22	5
Sc^{45}	85 days	0.0172	50	22	9	56	30
Mn^{55}	2.58 hr	0.0258	12	13.4	22	52	16
Ni^{64}	2.56 hr	0.0365	15	1.6	13	8.7	2.3
Cu^{63}	12.8 hr	0.178	10	4.3	7	114	24
Cu^{65}	5.1 min	0.152	15	2.11	8	48	12
Zn^{68}	13.8 hr	0.388	30	0.097	10	5.6	2.0
Zn^{68}	52 min	0.171	30	1	20	26	10
As^{75}	26.7 hr	1.072	15	4.1	5	650	160
Y^{89}	64 hr	0.150	15	1.26	6	28	7
Mo^{98}	67 hr	5.86	12	0.45	22	390	120
Pd^{108}	13.6 hr	0.329	12	12	25	580	200
Pd^{110}	22 min					>300	
In^{115}	54 min	0.455	10	145	10	980	220
In^{113}	49 days	1.018	15	56	21	8500	2700
Sb^{121}	2.8 days	0.799	10	6.8	22	810	250
Sb^{123}	60 days	0.618	25	2.5	20	230	100
Cs^{133}	3.1 hr	0.243	38	0.017	24	0.6	0.3
Cs^{133}	2.3 yr	0.232	20	26	19	900	300
La^{139}	40.2 hr	0.0390	12	8.4	20	49	15
Pr^{141}	19.1 hr	0.102	10	11.2	5.4	170	40
Dy^{164}	2.32 hr	0.00106	30	2100	15	330	130
Ta^{181}	112 days	0.441	12	21.3	5	1400	310
W^{184}	74 days	1.18	45	2	15	350	180
W^{186}	24.1 hr	0.0532	12	34	9	270	70
Pt^{196}	19 hr	1.77	20	0.8	13	210	60
Pt^{198}	30 min	0.406	20	3.9	20	240	80
Au^{197}	2.7 days	0.596	12	98.8	1	890	190
Bi^{209}	5 days	0.65	30	0.019	10	1.8	0.7

[a] These thermal cross-section values, which were used in Eq. (2), are included so that as better thermal cross sections become available the value for $\sigma(n,\gamma)$ can be adjusted accordingly.

of efficiency by Pm^{147} and Am^{241} standard samples. The gamma counter had an integral bias set at 12 kev. The beta pulse heights were principally proportional to dE/dx, since the beta range in most cases was considerably greater than the stopping power of the beta counter. This yielded nearly flat operating plateaus.

PROCEDURE

Absolute cross sections were not measured directly, but, rather, a double-ratio comparison method was used. Two isotopes were both exposed to the same 25-kev neutron flux, counted, exposed to the same thermal-neutron flux, and counted again. The ratio of the counting rates of the 25-kev and thermal neutrons was independent of counter efficiency, and the ratio of the counting rates of isotopes 1 and 2 in the same flux was independent of flux. Hence if three of the cross sections are known, the fourth can be calculated. The equations are:

$$R=R_{x25}R_{i\text{th}}/R_{x\text{th}}R_{i25}, \tag{1}$$

where

$$R_{x25}=E_xN_x\phi_{25}\sigma_{x25};\quad \text{etc.},$$

and

$$\sigma_{x25}=R(\sigma_{x\text{th}}\sigma_{i25}/\sigma_{i\text{th}}), \tag{2}$$

where x denotes the isotope whose photoneutron cross section is to be measured, i denotes the standard isotope (usually iodine), 25 and th refer to the 25-kev and thermal neutrons, respectively, R is the counting rate extrapolated to saturated activity, E is the counting efficiency, N is the number of atoms of isotope in the target, ϕ is the effective neutron flux, and σ is the activation cross section.

Samples were usually irradiated in the form of mixtures—salts, physical mixtures, or isotopic mixtures. This eliminated any effects of flux perturbation and flux depression. The only assumption then is that the counting efficiency is the same for both thermal and 25-kev activations. This assumption is good for gamma counting; in the case of beta counting, difficulties can arise if the sample is not "thin" to neutrons and if the beta particles from the two isotopes are not of the same energy. The beta-to-gamma counting ratio was a valuable check on the "thinness" of the sample. In some cases metal foils were used instead of mixtures, and then corrections to the data were applied.[8] It should be noted that absolute flux calibrations are not necessary in this method.

Iodine was chosen as the primary standard, and all other cross sections were determined relative to it. In each irradiation iodine, or a secondary standard calibrated against iodine, was included. Isotopes were identified by their half-lives and measurements of most

[8] T. H. R. Skyrme, British Report MS-91, and Appendix (Circa 1943), Atomic Energy Research Establishment, Harwell, Berkshire, England (unpublished).

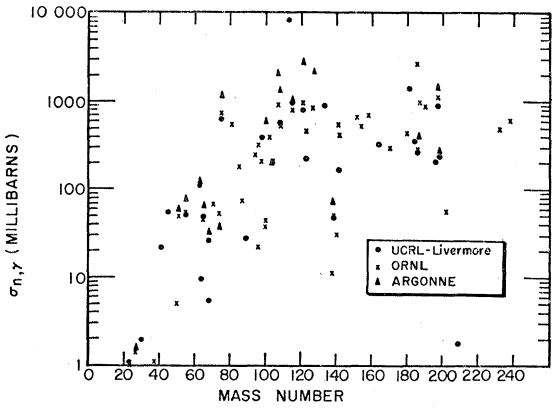

FIG. 1. Neutron capture cross sections at 25 kev *versus* the mass number. The data are from Livermore (present work), Oak Ridge,[5] and Argonne.[3,4]

activities were extended over several half-lives. The experiment was directed principally at the measurement of isotopes with long half-lives, since the long irradiation times required make the use of acclerator neutron sources especially difficult. All counting rates were extrapolated to saturated activities.

RESULTS

Activation cross sections at 25 kev for 31 isotopes are given in Table I. Since the measurements yield R, the R value with its experimental error is quoted separately. Macklin's value of 0.82 ± 0.06 barn[5] for the iodine photoneutron cross section was assumed. An iodine thermal cross section of 5.5 ± 0.5 barns was used,[9] and thermal cross sections used for the other isotopes are listed in Table I. Uncertainties in these latter cross sections make the error in the 25-kev cross section considerably larger than the error in the measurement of R. The uncertainties in the R value originate from (1) statistical uncertainties, (2) uncertainties in the half-lives, and (3) reproducibility.

The results are in general agreement with those of Macklin *et al.*[5] for the activities determined by both groups. An exception is Pr^{141}, where we differ by a factor of 3. The unusually large cross section of 8.5 barns for In^{113} to form In^{114m} warrants further study of this cross section. The Sb^{123} cross section includes activation to the two metastable states as well as the ground state of Sb^{124}. A search for evidence of the 21-minute metastable state was unsuccessful.

Figure 1 is a plot of the 25-kev activation cross section *versus* mass number for data available up to the present time. The characteristic dips at the magic numbers are readily noted.

[9] D. J. Hughes and J. A. Harvey, *Neutron Cross Sections*, Brookhaven National Laboratory Report BNL-325 (Superintendent of Documents, U. S. Government Printing Office, Washington, D. C., 1955).

TABLE II. Comparison of experimental 25-kev neutron capture cross sections with values calculated from low-energy s-wave resonance parameters.

Isotope	Mn^{55}	Zn^{68}	In^{113}	In^{115}	Cs^{133}	Ta^{181}	Au^{197}	Bi^{209}
σ_{exp}	0.052	0.032	8.50	0.98	0.90	1.40	0.89	0.002
σ_{calc}	0.115	0.034	1.06	0.50	0.97	1.41	0.57	0.006

COMPARISON WITH THEORY

Neutron capture cross sections in an energy region can be calculated if many resonance levels are contributing and if the average resonance parameters in that region are known. At 25 kev, while a number of capture cross sections have been measured, there is unfortunately little known about the resonance parameters. The question then arises as to whether the resonance parameters which have been measured in the low-energy region (<1 kev) can be assumed to approximate conditions at 25 kev. A definitive answer to this question lies outside the scope of the present report. However, it is instructive to compare experimental and calculated capture cross sections under the very simple assumptions that (1) the low-energy resonance parameters can be used at 25 kev, and (2) only s-wave scattering contributes to the cross section. We use the equation[2,10]

$$\bar{\sigma}_a=\sum_J \frac{2J+1}{2(2I+1)}\frac{\pi}{2}\left(\frac{2.6\times10^6}{E_n}\right)\frac{\Gamma_\gamma}{D}\times\left[1-(b\pi)^{\frac{1}{2}}\left(1-\frac{2}{\sqrt{\pi}}\int_0^{\sqrt{b}}\exp(-t^2)dt\right)\right], \quad (3)$$

where $b=\Gamma_\gamma/(2V_0\Gamma_n^0)$, $D=\bar{D}/(2J+1)$, and $\Gamma_n^0=\bar{\Gamma}_n^0/(2J+1)$. J is the total angular momentum, I is the spin of the target nucleus, E_n is the neutron energy in electron volts, and V_0 is the penetration factor (equal to unity) for zero angular momentum. Γ_γ, $\bar{D}$, and $\bar{\Gamma}_n^0$ are the experimentally measured[9,11–13] low-energy resonance parameters—the radiation width, level spacing, and reduced neutron width, respectively—which are averages over all s-wave resonances. The J-dependent quantities D and Γ_n^0 were used in Eq. (3) to allow for the fact that the strength function $\bar{\Gamma}_n^0/\bar{D}$ is independent of J, but the average level spacing $\bar{D}$ appears to show a $(2J+1)$ dependence.[2] In addition to assumptions (1) and (2) listed above, the validity of Eq. (3) is dependent on the implicit assumptions that many levels are contributing and that the reduced neutron widths follow the Porter-Thomas distribution.

[10] E. G. Bilpuch, thesis, University of North Carolina (unpublished).

[11] Carter, Harvey, Hughes, and Pilcher, Phys. Rev. **96**, 113 (1954).

[12] Harvey, Hughes, Carter, and Pilcher, Phys. Rev. **99**, 10 (1955).

[13] J. S. Levin and D. J. Hughes, Phys. Rev. **101**, 1328 (1956).

The agreement between calculated and experimental values is shown in Table II. Of the eight cases calculated, six were accurate to within a factor of 2. This indicates that assumptions (1) and (2) listed above probably have at least an approximate validity. On the other hand, the agreement for In^{113} is poor. Whether this is due to shortcomings in assumption (1), or in (2), or both, cannot be determined from the present work. P waves and possibly also D waves should be of some importance at 25 kev, and a serious attempt to calculate capture cross sections must include them. This will of course require considerably more experimental data on resonance parameters than are presently available.

The conclusion indicated by the calculations in the present section is that, as far as resonance parameter systematics are concerned, nothing radical occurs in going from the electron-volt to the kiloelectron-volt region.

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Directional Correlation of Gamma Rays in Ge^{72}†

R. G. ARNS* AND M. L. WIEDENBECK
Harrison M. Randall Laboratory of Physics, University of Michigan, Ann Arbor, Michigan
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Directional correlation measurements have been made on the 0.63–0.835 Mev, 2.20–0.835 Mev, 2.49 and 2.51–0.835 Mev, and 1.88–1.46 Mev gamma-gamma cascades in Ge^{72} following beta decay of 14.2-hour Ga^{72}. The level at 0.835 Mev has a spin and parity assignment of 2+ and proceeds to the 0+ ground state by pure electric quadrupole radiation. The 0.63-Mev transition proceeds from a 2+ level at 1.46 Mev to the 0.835-Mev level by radiation which is largely electric quadrupole with a small magnetic dipole admixture. The correlation data are consistent with spin assignments of 2 or 3 for the levels at 3.04, 3.32, and 3.34 Mev.

I. INTRODUCTION

THE decay of Ga^{72} has been the subject of many investigations.[1] A portion of the decay scheme proposed by Kraushaar *et al.* is shown in Fig. 1. Important features of the level structure of Ge^{72} were confirmed by studies of the decay of As^{72}.[2]

The first excited state of $_{32}Ge_{40}^{72}$ has spin zero and even parity. This represents a departure from the usual 2+ first excited state found in even-even nuclei. The only other known exceptions (O^{16}, Ca^{40}, Zr^{90}, Pb^{208}) result when both neutrons and protons form closed shells. In view of this peculiarity it was felt that direct measurement of the spins and parities of some of the higher levels might be of value in establishing the relation of Ge^{72} to the systematics of other nuclei in this region.

II. EXPERIMENTAL METHOD

A conventional fast-slow coincidence circuit with a resolving time of 1×10^{-8} second was employed in the angular correlation measurements.[3] A 4-in.×5-in. NaI(Tl) crystal and DuMont 6364 photomultiplier were used to detect the 1.88-Mev gamma ray in the weak 1.88 Mev–1.46 Mev correlation. The detectors in all other cases consisted of 2-in.×2-in. NaI(Tl) crystals mounted on RCA 6342 photomultipliers. Energy selection was provided by differential analyzers.

The Ga^{72} sources were obtained from Oak Ridge National Laboratory as gallium chloride in a dilute HCl solution. No interfering activities were present in the source material.

The half-life of the 0.835-Mev level has been measured to be $(3.2\pm0.8)\times10^{-12}$ sec[4] and the half-life of the 1.46-Mev level is expected to be short. This, together with the fact that the source was in dilute solution, eliminates the possibility of attenuation of the correlation function due to extranuclear fields.

Data were taken in a double quadrant sequence. The real coincidence rate was normalized to correct for source decay and electronic drift. A least-squares fit of the data was made to the function

$$W'(\theta)=\alpha_0+\alpha_2P_2(\cos\theta)+\alpha_4P_4(\cos\theta).$$

The resultant expansion coefficients were then normalized and corrected for finite angular resolution by a collimated beam method.[5] This yielded a correlation function of the form

$$W(\theta)=1+(A_2\pm\sigma_2)P_2(\cos\theta)+(A_4\pm\sigma_4)P_4(\cos\theta).$$

The σ_2 and σ_4 are the root-mean-square errors as defined by Rose, Eq. (30).[6]

† Supported in part by the Michigan Memorial Phoenix Project and the Office of Naval Research.
* Dow Chemical Company Fellow in Physics.
[1] Kraushaar, Brun, and Meyerhof, Phys. Rev. **101**, 139 (1956), and references cited therein.
[2] Brun, Kraushaar, and Meyerhof, Phys. Rev. **102**, 808 (1956).
[3] Stewart, Scharenberg, and Wiedenbeck, Phys. Rev. **99**, 691 (1955).
[4] F. R. Metzger, Phys. Rev. **101**, 286 (1956).
[5] A. M. Feingold and S. Frankel, Phys. Rev. **97**, 1025 (1955).
[6] M. E. Rose, Phys. Rev. **91**, 610 (1953).