Thermal-Neutron Activation Cross Sections of Ge and the Isomeric Ratio Rule

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The thermal-neutron activation cross sections of Ge^{74} and Ge^{76} were determined. Contrary to indications obtained from previously published neutron cross-section compilations, our results are in agreement with the isomeric ratio rule, which states that the ratio of cross sections for the production of two isomeric states by slow neutron capture is such that the isomeric state with spin close to that of the compound nucleus is favored.

SOME of the past investigations into the formation of isomeric states have revealed an interesting empirical rule concerning a quantity called the "isomeric ratio."[1] When isomeric states are formed through some process, such as radiative neutron capture, the ratio of the cross sections for the production of each state is known as the isomeric ratio. The rule was discovered empirically, that in slow neutron capture the two isomeric states of a nucleus having large spin differences are formed in unequal amounts and the isomeric state with spin closer in value to that of the compound state is favored over the other. Although exceptions to this rule were known, it was nevertheless established well enough to be invoked in arguments in favor of proposed spin assignments for isomeric levels.[2]

Under the Bohr assumption of compound nucleus formation, the capture of a neutron by a nucleus A leads to a state of high excitation in nucleus $A+1$, which decays promptly through the emission of electromagnetic radiations. The new nucleus eventually ends up in the ground state of nucleus $A+1$, or in some low-lying metastable level. The details of the decay process will determine in which of these two states the nucleus will find itself most frequently; the spins and parities of the initial and final states available for the nuclear transitions as well as the energy differences and the density of these states bear upon the nature of the decay. Dipole transitions are favored over transitions of high multipolarity, the energy dependence favors the emission of high-energy gamma rays and the density of levels is such that the emission of lower energy γ rays is favored. Experimental evidence indicates that for the intermediate and heavy elements (except for shell effects) the gamma-ray spectrum is very complex, with an average multiplicity[3] of 2 to 4. If a cascade of this nature were to consist mainly of dipole radiations, then clearly only those states with spins close to that of the compound nucleus will be populated with any appreciable frequency. If isomeric states exist in the nucleus, implying a large spin value difference in the low-lying levels, then the state with spin closer in value to that of the compound nucleus will be populated in preference to the other.

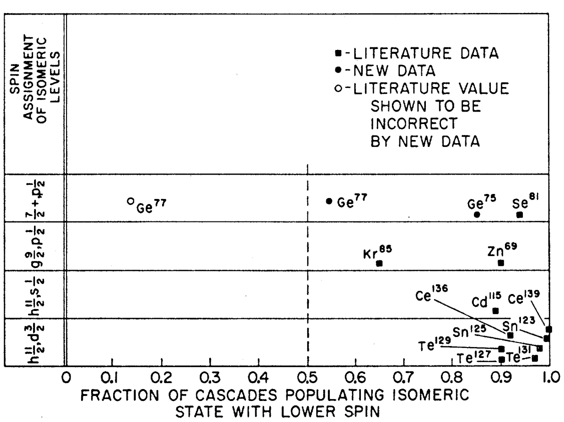

FIG. 1. Experimental evidence for rule concerning isomeric ratios: neutron capture in even-even nuclei only is included in the figure, so that the resulting compound nucleus states all have a spin $\frac{1}{2}$ and even parity. The fraction of cascades populating the member of an isomeric pair with low spin is plotted along the x axis while the spins and parities of the isomeric states are given to the left.

It appeared to us to be of sufficient interest to investigate the rule in the light of new data on neutron activation cross sections, spin assignments, and decay schemes of isomeric states. In Fig. 1 we have summarized the data on isomeric ratios for isomers formed by neutron capture in even-even nuclei. In these cases, the neutron capture leads to a compound nucleus with a definite spin and parity assignment, $\frac{1}{2}+$. Along the x axis there is plotted the fraction of the cascades, following compound nucleus formation, which populate the isomeric state with a low spin; that is, with a spin close to that of the compound nucleus. The data are divided into groups associated with the spins and parities of the isomeric levels. These spins are given at the left of each group of data. Inspection of the data reveals that only one point is plotted to the left of the 0.5 line, namely Ge^{77}. This point (the open circle plotted at $\sim$0.15) was obtained from the neutron cross-section compilation by Hughes and Harvey[4] and represents a clear violation of the rule. Reference to

* Under the auspices of the U. S. Atomic Energy Commission.
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[4] *Neutron Cross Sections*, compiled by D. J. Hughes and J. A. Harvey, Brookhaven National Laboratory Report BNL-325 (Superintendent of Documents, U. S. Government Printing Office, Washington, D. C., 1955).

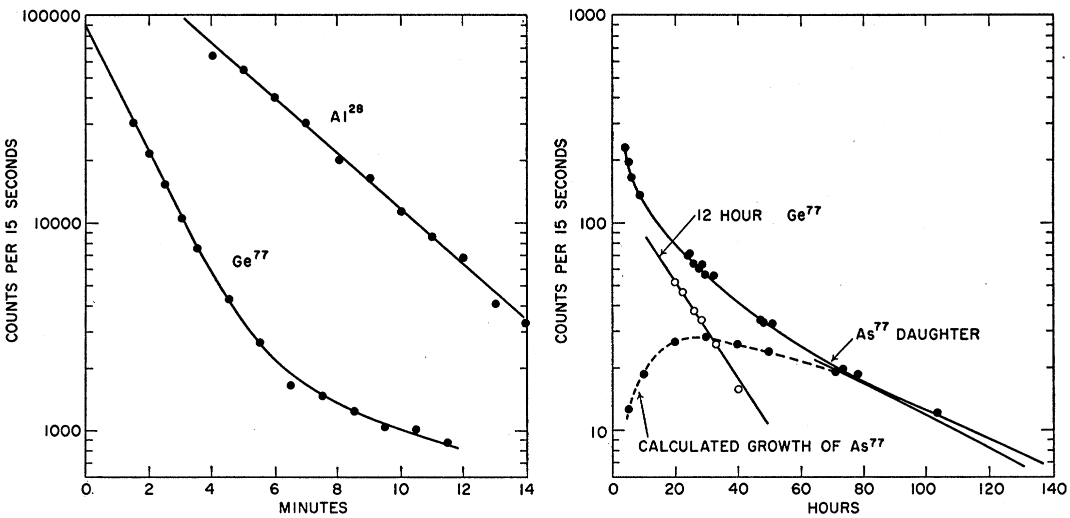

FIG. 2. Decay curve of 59-sec and 12-hr activities of Ge^{77}. These data were taken with an anthracene crystal and indicate the decay of these isomers as detected through their beta rays.

the literature listed in the compilation for the neutron activation cross section of Ge^{76} revealed, however, that a considerable discrepancy existed in the published values of these cross sections, and it was subsequently learned that the values listed in reference 4 were judiciously chosen ones. This suggested the possibility that the cross-section values might be incorrect for Ge^{77} and that further measurements of these cross sections were in order.

CROSS-SECTION MEASUREMENTS

Since there are many similarities in the decays of the Ge isomers, it was found necessary to investigate Ge^{75} in order to avoid errors in the Ge^{77} measurements due to the presence of Ge^{75}. Enriched isotopes[5] were irradiated in the Brookhaven National Laboratory reactor to produce relatively pure samples of these isomers. Cross-section measurements were made relative to the (n,γ) cross sections of Al and Na, leading to 2.3-min Al^{28} and 15-hr Na^{24}, respectively.

The 59-sec isomer of Ge^{77} was compared through its beta decay to the 2.3-min Al^{28} activity by irradiating weighed samples of Ge^{76} and Al simultaneously for short periods of time (irradiations were made for 3 or 5 sec). Al^{28} emits beta rays of 2.865-Mev maximum energy, while 59-sec Ge^{77m} emits two beta-ray spectra of maximum energies 2.7 and 2.9 Mev, respectively. Thus it could be assumed that our detector (anthracene) was equally efficient in detecting these two activities. The irradiation time was sufficiently short to enable us to assume that the production of our activities was linear with time [i.e., λt may be taken as a good approximation for $(1-e^{-\lambda t})$]. Under these conditions the Ge cross section could be calculated from the Al cross section by the following formula:

$$\sigma_{Ge}=\frac{I_{Ge}}{I_{Al}}\frac{t_{Ge}}{t_{Al}}\frac{n_{Al}}{n_{Ge}}\sigma_{Al},$$

where σ is the cross section; I, the initial decay rate; t, the half-life; and n, the number of nuclei.

A typical run is illustrated in Fig. 2. The initial decay rates of Ge^{77m} and Al^{28} were obtained by a straightforward extrapolation to zero time. Corrections had to be made for the internal transition branch of the Ge^{77m} activity and for the isotopic enrichment of the Ge^{76} sample (79.3%). A Cd ratio was obtained for this isomer in order to calculate the thermal cross section. As is seen in Fig. 2, the cross section of the 12-hr activity of Ge^{77} could also be determined relative to Al^{28}. The isomeric ratio of Ge^{77m} and Ge^{77} could also be obtained, independent of the value found for the absolute cross sections. In one run, the 12-hr Ge^{77} activity was compared to the 15-hr Na^{24} activity. The cross sections for production of Ge^{75m} and Ge^{75} were measured in the same manner. The data are tabulated in Table I.

TABLE I. Thermal neutron activation cross sections in germanium.

Isomer	half-life	Cross section (pile neutrons) Final	Initial	Cd Ratio	Initial cross section (thermal neutrons)	Spin and parity
Ge^{75m}	49 sec		0.046	8	0.040±0.008	$\frac{7}{2}+$
Ge^{75}	81 min	0.250	0.204	8	0.180±0.04	$\frac{1}{2}-$
Ge^{77m}	59 sec		0.126	3.25	0.087±0.015	$\frac{1}{2}-$
Ge^{77}	12 hr	0.129	0.113	3.25	0.076±0.015	$\frac{7}{2}+$

5 Obtained from the Stable Isotopes Division, Oak Ridge, Tennessee.

CONCLUSION

With the newly measured values of the cross sections for production of Ge^{77m} and Ge^{77},[6] the one exception to the isomeric ratio rule is removed. Again referring to Fig. 1, we see that the point for Ge^{77} calculated on the basis of our new measurements of the cross sections for production of Ge^{77} and Ge^{77m}, falls just to the right of the 0.5 line.

ACKNOWLEDGMENT

The authors wish to express their appreciation to David Larsen for his assistance in obtaining some of the data in this paper.

[6] Our Ge^{76} thermal neutron activation cross sections are close to older values reported by Seren, Friedlander, and Turkel, U. S. Atomic Energy Commission Report MDDC-408 (unpublished), and J. R. Arnold and N. Sugarman, J. Chem. Phys. **15**, 703 (1947), who give 0.085 barn for production of the 12-hr activity and state that the cross section for the 59-sec activity is 10% higher than that for the 12-hr activity. A new measurement of these cross sections has also been reported by W. S. Lyon and J. S. Eldridge, Phys. Rev. **107**, 1056 (1957) who give 0.14 barn and 0.043 barn for the production of Ge^{77m} and Ge^{77}, respectively, by pile neutrons. Although these values are considerably different from our values, they, too, are in agreement with the isomeric ratio rule.

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Coincidence Studies of the $Ni^{58}(p,2p)$ Reaction

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The $Ni^{58}(p,2p)$ reaction, by far the predominant reaction in that nucleus, was studied by detecting the two outgoing protons in coincidence. Measurements were made of the energy spectra of all protons from the reaction, of the spectrum of the sum of the energies of the two outgoing protons, and of the angular correlations of the outgoing protons with each other and wtih the incident proton. The results indicate quite conclusively that the preponderance of $(p,2p)$ over (p,pn) reactions in Ni^{58} is not due to the relative level densities of the final nuclei, the ineffectiveness of Coulomb barriers, or a high emission energy of the "first" proton leaving emission of a neutron energetically forbidden. Other possible explanations are considered.

There is strong evidence that the $(p,2p)$ reaction mechanism is predominantly a direct one in which the two protons are "knocked-out" simultaneously.

INTRODUCTION

AMONG the strangest anomalies in the field of medium-energy nuclear reactions are the very large (x,pn) and $(x,2p)$ cross sections in medium-weight elements.[1–3] For example, with a 23-Mev bombarding energy, (p,pn) reactions have far larger cross sections than $(p,2n)$ reactions in all elements lighter than zinc,[1] and in many cases, the heaviest example of which is Ni^{58}, the most probable reaction is $(p,2p)$.[2] There has been much speculation[1–4] on the explanation for these effects, but the theoretical analysis of total cross sections is too tenuous to allow positive conclusions to be reached. To obtain a deeper experimental hold on the problem, an investigation of the $Ni^{58}(p,2p)$ reaction was undertaken by coincidence detection of the outgoing protons. Angular correlations between these, and between them and the incident proton were studied. All measurements were made as a function of the energy of each of the two outgoing protons, and as a function of the sum of their energies.

To review the situation, the energetic thresholds for the various reactions in Ni^{58} are listed in Table I. The known excited states of Co^{57} are at 1.38, 1.50, and 1.91 Mev. The observed cross sections are 240 mb for the sum of the $(p,pn)+(p,2n)+(p,np)+(p,d)$ reactions,

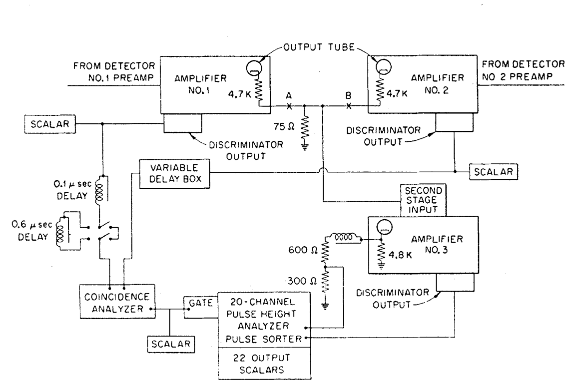

FIG. 1. Electronic circuitry for measuring distribution of sum of pulse heights of coincidence pulses. By breaking the connection at *B* (or *A*), the pulse-height distribution of pulses in detector No. 1 (or No. 2) in coincidence with pulses from the other detector is measured.

* Operated for the U. S. Atomic Energy Commission by Union Carbide Nuclear Company.
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[3] Miller, Friedlander, and Markowitz, Phys. Rev. **98**, 1197(A) (1955); J. M. Miller, and F. S. Houck, Bull. Am. Phys. Soc. Ser. II, **2**, 60 (1957); S. N. Ghoshal, Phys. Rev. **80**, 939 (1950).
[4] J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley and Sons, Inc., New York, 1952), p. 494; also, 1955 Gordon Conference on Nuclear Chemistry (unpublished).