Resonance Capture Reactions with a Total Energy Detector*

CONF-780926--4

R. L. Macklin

Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830

I. Total Energy: What's That?

Nuclear reaction rates can be determined in two basic types of experiment; depending respectively on counting and energy release. A simple example would be to measure the alpha disintegration rate of a sample. The number of alpha particles emitted in a given time could be counted with a charged particle counter or by measuring the helium released. Alternatively, the energy released could be measured with a calorimeter and divided by the (known) energy per alpha particle to arrive at the number of alpha disintegrations per unit time. This second approach has been exploited to measure (n,γ) cross sections of isotopic targets for which the neutron binding energy (BE) is generally well known, ranging from roughly 4 to 11 MeV. The total excitation energy of each compound nucleus formed is computed to good accuracy as:

$$E_x = E_n(A/(A + 1)) + BE$$

where E_n is the incident neutron energy and A is the atomic mass of the stationary target nucleus.

Most if not all of the excitation energy appears as a cascade of prompt ($<10^{-7}$ seconds) gamma rays and it is these that are detected. The efficiency of detection of the gamma rays is typically low and strongly energy dependent for equipment dimensions of a few centimeters.

*Invited paper for the Third International Symposium on Neutron Capture Gamma-Ray Spectroscopy and Related Topics, Brookhaven National Laboratory, September 1978

The Moxon-Rae detector is designed to make its gamma ray detection efficiency a linearly increasing function of energy, thus making its counting rate proportional to the total energy for application in the scheme above. In its simplest form, this result is approximated by counting the electrons emerging from the rear face of a converter plate. Gamma rays produce electrons nearly uniformly throughout the converter, with energies distributed below the gamma ray energy. The finite range of the electrons, however, ensures that those reaching the back face to be counted come only from a thickness proportional to the energy. Only the highest energy electrons (∿10 MeV) produced near the front face of the converter penetrate all the way through it to be counted. Various improvements to the Moxon-Rae detector's linearity have been achieved through multilayered arrangements and adjustment of the converter plate composition.

A similar proportionality to total energy can be achieved for gamma ray detectors sensitive throughout their volume, with, correspondingly, a much higher total efficiency. This is done by assigning a weight (or quantitative importance) to the fraction of the gamma ray energy deposited in the detector. This fraction is commonly measured by scintillation counting, leading via a photomultiplier to a digitized pulse height, thus the process is called pulse height weighting. If the pulse height always corresponded to the full energy of the detected gamma ray, the appropriate weight would just be proportional to the pulse height and the reciprocal of the detector's gamma ray efficiency at that energy. As gamma detectors give a full range of pulse heights up to the incident gamma ray energy (smeared out by a resolution function) the problem appears at first sight insoluble. Maier-Leibnitz has shown, however, that a weighting function exists even in this situation and techniques have been found that allow it to be calculated stepwise from the pulse height response matrix of a detector system. The essential feature seems to be a monotonic increase in maximum pulse height with gamma ray energy. The basic equation is

$$\Sigma G(I)P(I,E_\gamma) = E_\gamma$$

where G(I) is the weighting function for pulse height channel I and $P(I,E_\gamma)$ is the probability of detecting a pulse in channel I induced by a gamma ray of energy E . As there are, in principle, no restrictions on the pulse height channel width, the weighting function is continouous and can be well represented as a power series in the pulse height E_p

$$G(E_p) = \sum_{i=1}^{n} a_i \, E_p{}^i$$

Using such a weight function, the gamma rays from neutron capture cascades are repeatedly sampled to get a statistically significant measure of the total gamma ray energy. For one detector the statistics for an average cascade were about 1.2 times the Poisson counting statistics. The ratio rose to about 1.6 for a single ground state transition.

Dividing the total energy estimate by the excitation energy in the prompt cascade gives the total number of (n,γ) reactions and that, combined with the neutron fluence and isotopic sample size gives the capture probability per nucleus. Many small effects need to be calculated as with any other system* to arrive at the corrected (n,γ) cross section.

*These include electronic deadtime corrections, three measured background effects, resonance selfprotection, Doppler broadening and elastic scattering in the sample (also inelastic scattering at higher energies), scattered neutron sensitivity of the detector and sample impurity corrections.

II. Neutron Capture Cross Sections

A. Measuring Prompt Gamma Ray Yields

The pulse height weighting method has been especially useful in measuring neutron capture gamma ray yields where the often complicated details of the gamma ray cascades are to be disregarded. Where the interest is in individual gamma rays, other techniques including spectrum stripping are usually appropriate although in a few cases high bias pulse height weighted results have offered advantages.

Liquid scintillators based on C_6D_6 or C_6F_6 have been used for fast timing (1-2 ns) and insensitivity to scattered neutrons at several Van de Graaff accelerators (including ones at Cadarache, France and Oak Ridge) and at electron linac pulsed neutron sources (including those at Livermore and Oak Ridge, Geel in Belgium and Kyoto in Japan).

B. Resonance Shape Fitting to Capture Yields

At the 40 meter installation in Oak Ridge that I am most familiar with, a resolution of 0.1 ns/m gives an energy resolution $\Delta E/E$

(FWHM) down to 1/700, the limit set by neutron moderation time. This is adequate to clearly show up hundreds of narrow p-wave and d-wave resonances never seen before. To cope with such a flood of data, a non-linear least squares fitting program was developed, capable of fitting single-level resonance parameters for 15 resonances in 500 channels of capture yield data in less than 10 seconds. Additions to the code to deal with broad resonances (multiple scattering, scattered neutron sensitivity of the detector) continue to be made.

C. Strength Functions and Average Capture

At high enough neutron energies, ranging from 5 keV to nearly 1 MeV for different isotopes, individual resonances can not be adequately resolved, yet useful average properties of the resonances can still be extracted from the data. Theory provides an over abundance of free parameters in this case and one is left with the unhappy choice of lopping off some theoretician's pets. The shape and magnitude of average capture yields below about 120 keV have been fitted by non-linear least squares in many cases with just four free parameters, S^0, S^1, S^2 and Γ_γ/D_0. Even these few often show very high (up to 0.99) correlation (or anti-correlation) coefficients among themselves. Neutron transmission data and systematic trends with target mass often help to constrain S^0 and S^1. Additional parameters such as ℓ dependent radiative widths, doorway states and other J^π dependent intermediate structure may well be present but indistinguishable in the capture yield data. The onset of inelastic scattering is usually evident as a decrease in capture yield above the inelastic threshold, but a theoretically adequate formulation of this effect involves several more parameters.

D. Fluctuation Analysis

One further parameter can be squeezed out of the average capture yield data based on the fluctuations above and below the fitted strength function average. After averaging over some tens of resonances the remaining fluctuations are most strongly dependent on the average level spacing. In the simplified model where $D_J = (2J + 1)\ D_0$ varies only slowly with energy, D_0 is proportional to the relative mean squared deviation of the cross section and the chosen energy interval. The proportionality constant is near 1/2 and weakly dependent on energy through the relative contributions of each spin and parity to the capture cross section. For instance, below 10 keV or so the Porter-Thomas (χ^2 with $\nu = 1$ degree of freedom) s-wave neutron width distribution is dominant, giving a coefficient near $\nu/2 = 1/2$. At higher energies the p-wave contribution becomes important with the coefficient depending on the number of channel spins contributing to each total spin J as well as (slightly) on the widths of the spacing and Γ_γ distributions. This behavior has been used to show that substantial changes in spacing are present over the 2.5-100 keV neutron energy range for some targets but not for others. What sort of intermediate structure or alternative explanation this behavior corresponds to has not been determined although matching the averaged data by Monte-Carlo simulations drawn from prescribed distributions has been suggested.

III. Neutron Capture Data in the Larger Scheme of Things

A. Astrophysics and Nucleosynthesis

Current understanding of our universe starts from a brief high density, high temperature phase dubbed the Big Bang. Most of the matter proceeding from it consisted of hydrogen (mass 1) and a small fraction of helium (mass 4). The observed abundances of heavier elements (up to mass 240 or so) are viewed as products of stellar nucleosynthesis. Stars, particularly ones much larger than our sun in the course of their evolution got hot enough to build up the elements up to iron (mass 56 or 58) by charged particle reactions such as (p,γ) and (α,γ). Dispersed into space when the star blew up such heavier material was later incorporated in other stars where, at certain stages an appreciable neutron flux was produced by (p,n) and (α,n) reactions on isotopes such as ^{13}C and ^{22}Ne. Through sequences of successive neutron captures (and beta decays) all the heavier elements up to lead by a slow (10-150 years per neutron captured) process and past plutonium by a rapid (seconds per neutron captured) process could be produced. The slow process follows the path of stability on a chart of the isotopes and predicts quantitative relations between observed (isotopic) abundances and average neutron capture cross sections at neutron energies (15-40 keV typically) produced at thermal equilibrium inside a star.

While the crucial experiments to check these predictions were done many years ago, similar data for many more isotopes continue to be needed in unscrambling the contributions of the rapid (supernova) process and the time scales of nucleosynthesis. Currently interest is returning to the question of the age of our galaxy, as derived from the extent of radioactive decay of ^{187}Re to ^{187}Os.

B. Capture Reaction Mechanisms

Data on partial radiative widths, their energy dependence and correlations with neutron widths provide most of the evidence for mechanisms such as hard sphere external capture, valence or channel capture and doorway state mechanisms as detailed by S. F. Mughabghab's talk at this conference. The pulse height weighting method lends itself most readily however to determinations of total radiative widths. In special cases, where the level spacing is particularly wide some partial width information can also be derived. S. Raman is to speak on such results applied to giant electromagnetic multipole influences in excited levels and resonances of the closed shell ^{208}Pb system.

Even in the data on total radiative widths significant correlations have been found, particularly near mass 90, indicating that valence capture is operating in addition to the statistical compound nucleus mechanism. For very broad resonances, neutron scattering by a sample followed by capture in the detector contributes to the measured yield and must be carefully corrected for to avoid a spurious Γ_γ vs Γ_n correlation.

IV. Summary

The total gamma energy detection method has been applied successfully to radiative neutron capture cross section measurements. While many of the isotopic samples studied derive from needs in fission reactor design, the results have contributed importantly to stellar nucleosynthesis studies and the elucidation of nuclear reaction mechanisms. A bibliography is appended, including most of the published papers reporting neutron capture cross sections measured by the pulse height weighting technique.

Allen, B. J., A. R. de L. Musgrove, J. W. Boldeman and R. L. Macklin, "Valence Neutron Capture in ^{54}Fe", Nucl. Phys. A283, 37 (1977).

Allen, B. J., C. Y. Fu, R. R. Winters and R. L. Macklin, "Comments on the Doorway State in ^{206}Pb", PHys. Rev. C 7, 2598 (1973).

Allen, B. J., R. R. Winters, C. Y. Fu and R. L. Macklin, "Neutron-Capture Cross Sections of the Stable Lead Isotopes", Phys. Rev. C 8, 1504 (1973).

Allen, B. J. and R. L. Macklin, "The Neutron Capture Cross Section of ^{207}Pb", Neutron Cross Section and Technology, Proc. 3rd Conf., U.T., Knoxville, Tenn., March 1971, CONF-710301, 2, 764 (1971).

Allen, B. J. and R. L. Macklin, "Neutron Capture Cross Sections of ^{13}C and ^{16}O", Phys. Rev. C 3, 1737 (1971).

Allen, B. J. and R. L. Macklin, "Fast Neutron Capture Cross Section Facility", Nucl. Instrum. Methods 91, 565 (1971).

Allen, B. J. and R. L. Macklin, "Non-Resonant Neutron Capture in ^{207}Pb", Phys. Rev. Lett. 25, 1675 (1970).

Auchampaugh, G. F., J. Halperin, W. M. Howard and R. L. Macklin, "keV $^{33}S(n,\alpha)$ and $^{33}S(n,\gamma)$ Cross Sections: Importance in the Nucleosynthesis of the Rare Nucleus ^{36}S", Phys. Rev. C 12, 1126 (1975).

Boldeman, J. W., B. J. Allen, A. R. de L. Musgrove and R. L. Macklin, "The Neutron Capture Cross Section of ^{89}Y", Nucl. Eng. 64, 744 (1977).

Boldeman, J. W., B. J. Allen, A. R. de L. Musgrove and R. L. Macklin, "Valence Neutron Capture in ^{88}Sr", Nucl. Phys. A269, 397 (1976).

Boldeman, J. W., A. R. de L. Musgrove, B. J. Allen, J. A. Harvey and R. L. Macklin, "The Neutron Total and Capture Cross Sections of $^{92,94}Zr$", Nucl. Phys. A269, 31 (1976).

Boldeman, J. W., B. J. Allen, A. R. de L. Musgrove and R. L. Macklin, "The Neutron Capture Cross Section of Natural Silicon", Nucl. Phys. A252, 62 (1975).

Boldeman, J. W., B. J. Allen, A. R. de L. Musgrove and R. L. Macklin, "Valence Component in the Neutron Capture Cross Section of ^{90}Zr", Nucl. Phys. A246, 1 (1975).

Browne, J. C., G. P. Lamaze and I. G. Schroder, "Ratio of Neutron Capture Cross Sections for ^{186}Os and ^{187}Os at 25-keV Neutron Energy", Phys. Rev. C 14, 1287 (1976).

Browne, J. C. and B. L. Berman, "Neutron Capture Cross Sections for ^{186}Os and ^{187}Os and the Age of the Universe", Nature 262, 197 (1976).

Czirr, J. B. and M. L. Stelts, "Measurement of the Neutron Capture Cross Section of Holmium-165 and Gold-197", Nucl. Sci. Eng. 52, 299 (1973).

Czirr, J. B., "^{165}Ho, ^{151}Eu and Natural Europium Capture Cross Section Measurements", (California Univ. Lawrence Livermore Laboratory Report UCRL-50804, Feb. 1970.

Czirr, J. B. and J. S. Lindsey, "A Measurement of the Capture-to-Fission Ratio for Plutonium-239", Nucl. Sci. Eng. 41, 56 (1970).

Czirr, J. B., "A New Technique for Capture and Fission Cross-Section Measurements", Nucl. Instrum. Methods 72, 23 (1969).

Gibbons, J. H. and R. L. Macklin, "Capture-Cross Section Studies for 30-220-keV Neutrons Using a New Technique", Phys. Rev. 159, 1007 (1967).

Kenny, M. J., B. J. Allen and R. L. Macklin, "Resonance Neutron Capture in Scandium Below 100 keV", Aust. J. Phys. (1978)

Kenny, M. J., B. J. Allen, A. R. de L. Musgrove, R. L. Macklin and J. Halperin, "Neutron Capture by the Chromium Isotopes", Australian Atomic Energy Commission Report AAEC/E400.

le Rigoleur, Claude, Andre Arnaud and Jean Taste, "Mesures en Valeur Absolue des Sections Efficaces de Capture Radiative des Neutrons Par le ^{23}Na, Cr, ^{55}Mn, Fe, Ni, ^{103}Rh, Ta ^{197}Au, ^{238}U dans le Domaine de 10 a 600 keV", Centre d'Etudes Nucleaires de Cadarache Report CEA-R-4788 (1976).

Macklin, R. L., J. A. Harvey, J. Halperin and N. W. Hill, "The 292.4 eV Neutron Resonance Parameters of ^{91}Zr", Nucl. Sci. Eng. 62, 174 (1977).

Macklin, R. L. and J. Halperin, "^{232}Th (n,γ) Cross Section from 2.6-800 keV", Nucl. Sci. Eng. 64, 849 (1977).

Macklin, R. L., R. R. Winters and J. Halperin, "Neutron Capture by ^{208}Pb at Stellar Temperatures", Astrophysical Journal 217, 222 (1977).

Macklin, R. L. and J. Halperin, Resonance Neutron Capture by ^{209}Bi", Phys. Rev. C 14, 1389 (1976).

Macklin, R. L. and R. R. Winters, "Stellar Neutron Capture in the Thallium Isotopes", Astrophysical Journal 208, 812 (1976).

Macklin, R. L., "Neutron Capture Cross Section of Nb-93 from 2.6 to 700 keV", Nucl. Sci. Eng. 59, 12 (1976).

Macklin, R. L., "The $^{165}Ho(n,\gamma)$ Standard Cross Section from 3 to 450 keV", Nucl. Sci. Eng. 59, 231 (1976).

Macklin, R. L., R. R. Winters and J. Halperin, "Gold Neutron Capture Cross Section from 3 to 550 keV", Phys. Rev. C 11, 1270 (1975).

Macklin, R. L. and R. R. Winters, "Neutron Capture in Fluorine Below 1500 keV", Phys. Rev. C 7, 1766 (1973).

Mewissen, L., F. Poortmans, E. Cornelis, G. Vanpraet, A. Angeletti, G. Rohr and H. Weigmann, "Neutron Resonance Parameters for ^{237}Np", submitted to Nucl. Sci. Eng. (1978).

Mizumoto, M., R. L. Macklin and J. Halperin, "Neutron Capture Cross Section of Terbium-159 from 2.6 to 700 keV", Phys. Rev. C 17, 522 (1978).

Musgrove, A. R. de L., B. J. Allen, J. W. Boldeman, D. M. H. Chan and R. L. Macklin, "Odd-Even Effects in Radiative Neutron Capture by Ca-42, 43, 44", Nucl. Phys. A279, 317 (1977).

Musgrove, A. R. de L., J. W. Boldeman, B. J. Allen, J. A. Harvey and R. L. Macklin, ""High Resolution Neutron Transmission and Capture for ^{91}Zr", Aust. J. Phys. 30, 391 (1977).

Musgrove, A. R. de L., B. J. Allen and R. L. Macklin, "Resonant Neutron Capture in ^{139}La", Aust. J. Phys. 30, 605 (1977).

Musgrove, A. R. de L., B. J. Allen, J. W. Boldeman and R. L. Macklin, "Non-Statistical Neutron Capture in ^{140}Ce", Aust. J. Phys. (1978).

Musgrove, A. R. de L., B. J. Allen, J. W. Boldeman and R. L. Macklin, "keV Neutron Resonance Capture In Barium-137", Aust. J. Phys. 29, 157 (1976).

Musgrove, A. R. de L., B. J. Allen, J. W. Boldeman and R. L. Macklin, "keV Neutron Capture Cross Sections in ^{134}Ba and ^{136}Ba", Nucl. Phys. A256, 173 (1976).

Musgrove, A. R. de L., B. J. allen, J. W. Boldeman and R. L. Macklin, "Average Neutron Resonance Parameters and Radiative Capture Cross Sections for the Isotopes of Molybdenum", Nucl. Phys. A270, 108 (1976).

Musgrove, A. R. de L., B. J. Allen, J. W. Boldeman and R. L. Macklin, "keV Neutron Resonance Capture in Ba-138", Aust. J. Phys. 29, 157 (1976).

Musgrove, A. R. de L., B. J. Allen, J. W. Boldeman and R. L. Macklin, "Non-Statistical Effects in the Radiative Capture Cross Sections of the Neodymium Isotopes", Australian Atomic Energy Commission Report AAEC/E401.

Nystrom, G., B. Lundberg and I. Bergqvist, "Neutron Capture Cross Sections in F, Mg, Al, Si, P and S from 10 to 80 keV", Physica Scripta 4, 95 (1971).

Pandey, M. S., J. B. Garg, J. Halperin and R. L. Macklin, "High-Resolution Neutron Capture Cross Sections in ^{63}Cu and ^{65}Cu, II", Phys. Rev. C 15, 615 (1977).

Raman, S., M. Mizumoto, G. G. Slaughter and R. L. Macklin, "Observation of Primary E2 Transitions in the Reaction ^{207}Pb(n,)", Phys. Rev. Lett. 40, 1306 (1978).

Raman, S., M. Mizumoto and R. L. Macklin, "Fine Structure of the Magnetic Dipole Giant Resonance in ^{208}Pb", Phys. Rev. Lett. 39, 598 (1977).

Spencer, R. R. and R. L. Macklin, "Neutron Capture Cross Section of ^{59}Co in the Energy Range 2.5-1000 keV", Nucl. Sci. Eng. 61, 346 (1976).

Wasson, O. A., B. J. Allen, R. R. Winters, J. A. Harvey and R. L. Macklin, "Neutron Resonance Parameters of ^{92}Mo", Phys. Rev. C 7, 1532 (1973).

Weigmann, H., R. L. Macklin and J. A. Harvey, "Isobaric Analogue Impurities from Neutron Capture and Transmission by Magnesium", Phys. Rev. C 14, 1328 (1976).

Weston, L. W. and J. H. Todd, "Neutron Capture and Fission Cross Sections of Plutonium-241", Nucl. Sci. Eng. 65, 454 (1978).

Weston, L. W. and J. H. Todd, "Neutron Capture Cross Section of Plutonium-240", Nucl. Sci. Eng. 63, 143 (1977).

Weston, L. W. and J. H. Todd, "Neutron Absorption Cross Section of Americium-241", Nucl. Sci. Eng. 61, 356 (1976).

Weston, L. W. and J. H. Todd, "Fission-Neutron Multiplicity and Total Prompt gamma-Ray Energy for Resonances in ^{239}Pu", Phys. Rev. C 10, 1402 (1974).

Yamamuro, Nobuhiro, Tamotsu Hayase and Takeshi Doi, "Reliability of the Weighting Function for the Pulse Height Weighting Technique", Nucl. Instrum. Methods 133, 531 (1976).