First Excited States in Zr^{92} and Zr^{94}†

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(Received April 23, 1956)

Fast neutron inelastic scattering by separated isotopes of Zr was studied by observing the de-excitation gamma-ray spectra. The (926±9)-kev photopeak observed with normal Zr scatterers was shown to be produced by two gamma rays differing in energy by less than 2.5% and originating in the isotopes Zr^{92} and Zr^{94}. This is interpreted as evidence for a first excited state at about 926 kev in both isotopes. Relative excitation curves for gamma-ray production by neutron inelastic scattering have been measured from threshold to $E_n=5.5$ Mev for the 850-kev gamma ray from Fe^{56} and the 926-kev gamma rays from Zr^{92+94}.

I. INTRODUCTION

THE presence of a (920±20)-kev gamma ray resulting from fast neutron bombardment of zirconium has been reported,[1] although the origin of this gamma ray was uncertain at that time. This gamma ray has been reported by others,[2] but no positive isotopic assignment could be made. The purpose of this work was to determine the isotopic origin of this gamma ray as well as to identify the nuclear process involved.

II. APPARATUS

The scatterers were bombarded with neutrons produced by the Westinghouse electrostatic generator by means of the D(d,n) or the T(p,n) reaction. The single-crystal NaI(Tl) spectrometer method described by Sinclair[3] was used to observe the resultant gamma-ray spectra at 90°. Samples of electromagnetically enriched (~95%) zirconium oxide were available[4] for the zirconium isotopes 90, 91, 92, and 94 in the amounts of 13.5, 4.1, 7.9, and 8.3 grams, respectively, of the oxides. A current integrator measured the charge delivered to the neutron-producing target and a flat-response, shielded long counter[5] monitored the neutron flux.

III. ISOTOPIC ASSIGNMENT

Enriched zirconium oxides contained in thin, paper pill-boxes were bombarded by 4.1-Mev neutrons produced by the D(d,n) reaction. Under these experimental conditions, the peak-to-background ratio was such that the 926-kev photopeak could be detected unambiguously from a 26-gram normal zirconium oxide scatterer. Only the 926-kev photopeak could be seen with these samples since only the more intense photopeaks were discernible. It was impossible to obtain a threshold curve for the production of the 926-kev gamma ray from these enriched samples because of the unfavorable peak-to-background ratio existing when using the T(p,n) reaction with the Westinghouse generator.

While precise energy measurements could not be made from these small samples, unmistakable photopeaks were observed with nominal energies of 926 kev for both the Zr^{92} and the Zr^{94} samples. Typical pulse-height spectra for each of the four isotopes as well as a comparison spectrum taken with a 22-gram normal metallic zirconium scatterer are shown in Fig. 1.

The photopeak shapes and half-widths of the 850-kev gamma ray from Fe^{56} and the 926-kev gamma rays from zirconium were compared by alternate bombardments of metallic scatterers of iron and zirconium. The 926-kev photopeak showed no evidence of structure and, within experimental error, the half-widths observed for the two photopeaks differed by the amount expected for two gamma rays of 850 and 926 kev.

Emmerich and Sinclair[6] have compared the 926-kev

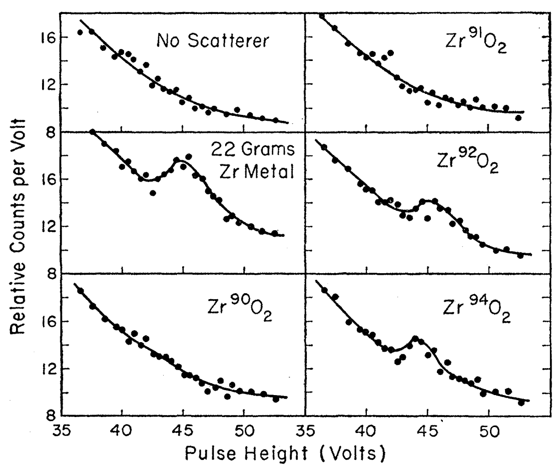

FIG. 1. Typical pulse-height spectra obtained with a single NaI(Tl) spectrometer for inelastic scattering of 4.1-Mev neutrons by separated isotopes of zirconium and by normal metallic zirconium. The oxide scatterers were enriched to approximately 95% in the indicated isotope of zirconium. Each experimental point has a statistical accuracy of about 2.5%.

† Assisted by a joint program of the Office of Naval Research and the U. S. Atomic Energy Commission.

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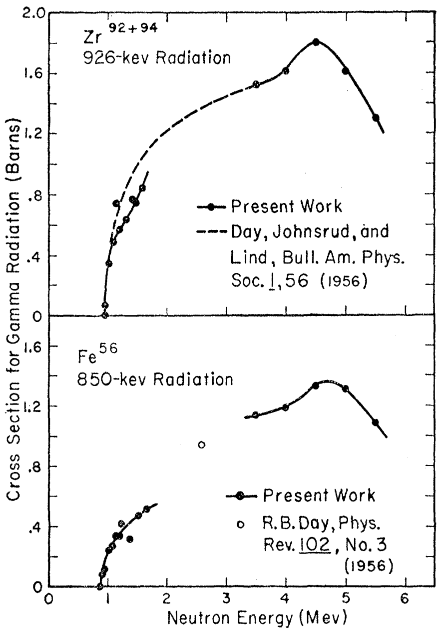

FIG. 2. Cross sections for gamma-ray production by neutron inelastic scattering by the isotopes Fe^{56} and Zr^{92+94}. The cross sections shown are the isotopic differential cross sections at 90° multiplied by 4π.

photopeak observed from fast neutron scattering by zirconium with the known[7] 930-kev gamma ray from Zr^{92} arising from the decay of Nb^{92}. In order to imitate the continuous spectrum due to neutrons scattered into the NaI(Tl) detector during the bombardment of Zr, a similar spectrum produced by neutrons scattered from carbon was added to the gamma-ray spectrum of the Nb^{92} radioactivity by placing the detector and the Nb^{92} source near a carbon scatterer. The energies of the two photopeaks were equal to within one percent. The half-width observed for the photopeak produced by the gamma rays from neutron scattering by zirconium was slightly larger than that from the Nb^{92} decay, although this difference was within the experimental uncertainties.

IV. GAMMA-RAY EXCITATION CURVES

At 4.4-Mev incident neutron energy the time-of-flight technique[8] and the nuclear emulsion method[9] failed to detect any neutron group corresponding to a 926-kev level in zirconium although both methods detected the 850-kev level in Fe^{56}. For this reason, a study was made of the threshold energies and relative cross sections for production of the 850-kev and 926-kev gamma rays from iron and zirconium. Metallic scatterers, $3\frac{1}{8}$ in. $\times 3\frac{1}{2}$ in.$\times\frac{1}{4}$ in., were mounted at 45° to both the incident neutron direction and the central axis of the scintillation detector. Relative gamma-ray yields were measured for the neutron energy-range, threshold to 1.6 Mev and 3.5 to 5.5 Mev.

Since the measured threshold energy for each gamma ray indicated that they were due to direct transitions to the ground state in each case, the ratio of the measured threshold energies was compared to that calculated from the ratio of gamma-ray energies. For inelastic scattering the threshold energy, E_{th}, may be calculated from the relation

$$E_{\text{th}}=(M+1)Q/M,$$

where M is the scatterer mass number and Q is equal to the gamma-ray energy for the case of ground-state transitions. The ratio of the 926-kev and the 850-kev gamma-ray energies was determined by bombarding a laminated, metallic scatterer of Fe and Zr which contained the correct ratio of elements to yield photopeaks of approximately equal size.

The ratio,

$$E_{\text{th}}(926)/E_{\text{th}}(850),$$

of the observed threshold energies was 1.103 ± 0.016, and that ratio calculated from the observed gamma-ray energy-ratio was 1.079 ± 0.010. Thus, these data are consistent with the assumption that the 926-kev gamma rays are due to neutron inelastic scattering through 926-kev levels in zirconium since the 850-kev gamma ray certainly results from inelastic scattering through the 850-kev level in Fe^{56}.

During the above measurements, each gamma-ray energy as well as the energy ratio was determined. The results are as follows: $E(926)/E(850)=1.087\pm0.010$, $E_\gamma(Zr^{92+94})=926\pm9$ kev, and $E_\gamma(Fe^{56})=850\pm8$ kev.

The relative yields of the 850-kev and the 926-kev gamma rays were calculated from the photopeak areas. Corrections were made for gamma-ray attenuation in the scatterers and for detection efficiency. No corrections were made for attenuation of the incident neutron flux or for multiple scattering. However, these latter two effects do tend to cancel one another, and Day[10] has reported this cancellation to be quite effective for his particular ring geometry. These data are shown as isotopic cross sections in Fig. 2 where both curves have been normalized to an absolute scale using the value of Day *et al.*[2] for the cross section for production of the 926-kev gamma rays in zirconium at $E_n=3.5$ Mev. The cross sections given are the isotopic differential cross sections for gamma-ray emission at 90° multiplied by 4π.

V. DISCUSSION

These data are consistent with the assumption that the 926-kev gamma rays from zirconium are due to

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neutron inelastic scattering. In addition, Cranberg and Levin[11] have observed the inelastically-scattered neutron group for an incident neutron energy of 2.5 Mev, which clearly identifies the process involved as inelastic scattering.

The data obtained using enriched scatterers show conclusively that at $E_n = 4.1$ Mev there are 926-kev gamma rays produced in both Zr^{92} and Zr^{94}. From the relative sizes of the scatterers and observed photopeaks, one can state that the intensities of the two gamma rays are roughly equal, but it is impossible to assign an energy difference to these gamma rays from these data.

From the large-scatterer data, an upper limit of 2.5% is placed on the energy separation of the two 926-kev gamma rays. This figure is based on the observed values of $(8.3\pm0.6)\%$ and $(8.0\pm0.7)\%$ for the half-widths of the 850-kev and 926-kev photopeaks, respectively, and the expected variation in half-width of a composite photopeak as a function of the separation of its two equal components. This variation was determined empirically by a graphical addition of two separated, equal photopeaks.

Without yield curves for these enriched samples one cannot assign unambiguous level schemes for Zr^{92} and Zr^{94}, but the threshold measurements on normal Zr indicate that at least one isotope has a level at 926 kev. From the threshold data of Day *et al.*[2] and the lack of any sudden rise, except at threshold, in the excitation curve for the 926-kev gamma rays, one can exclude the possibility that the 926-kev gamma ray in the other isotope comes from a cascade process. Thus, one can conclude that the first excited state in both Zr^{92} and Zr^{94} lies at 926 kev. This assignment is not unreasonable, based on the nuclear systematics of even-even nuclei,[12] and is in agreement with the known[7] first excited state of Zr^{92}.

ACKNOWLEDGMENT

The assistance of Mr. B. Romeo, who helped set up and run many of these experiments, is gratefully acknowledged.

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PHYSICAL REVIEW VOLUME 103, NUMBER 3 AUGUST 1, 1956

Test of the Statistical Assumption in Nuclear Reactions*

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(Received April 16, 1956)

The angular distribution of protons inelastically scattered from A^{40}, leading to the excitation of the 1.47-Mev level, has been measured at bombarding energies of 9.8, 9.0, and 8.5 Mev. An attempt is made to relate these data to the possibility of correlations in the phases of the levels excited in the compound nucleus.

THE angular distributions of nucleons inelastically scattered from nuclei, leading to the excitation of single levels of the nuclei, will be symmetric about a scattering angle of 90° providing: the reaction goes through the compound nucleus (compound nucleus assumption), many overlapping levels are excited in the compound nucleus (continuum assumption), and the phases of the levels excited by different partial waves are random (statistical assumption).[1,2] The observed departure from symmetry of such angular distributions, in situations where the continuum assumption would probably be satisfied, is usually attributed to a violation of the compound nucleus assumption—i.e., to the presence of direct interactions.[3,4] However the departure from symmetry could be due to a violation of the statistical assumption.

It may be possible to distinguish between these alternatives by measuring angular distributions at several closely spaced bombarding energies.[5] Large changes in the angular distributions for small changes in the bombarding energy would be difficult to explain if the departure from symmetry were due only to a violation of the compound nucleus assumption. This is because the direct interaction angular distributions do not depend in a sensitive manner on the bombarding energy,[3,4] and because the incoherence of the direct interaction and compound nucleus processes (a result of the continuum and statistical assumptions) prevents the existence of interference terms which might have a sensitive dependence on bombarding energy. However, large changes in the angular distributions for small changes in the bombarding energy would be easy to

* Supported in part by the U. S. Atomic Energy Commission.
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