Metastable State of Ge^{73}

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A new short-period isomeric activity with a gamma-ray energy of 54 kev and a half-life of 0.53±0.03 sec has been observed following thermal neutron absorption in Ge^{72}. The gamma-ray transition is shown to arise from a metastable state of Ge^{73} at 67.4-kev excitation energy, known from the decay of As^{73}, but not previously characterized as to half-life. Chemical separation of Ge^{73m} from As^{73} by a gas-sweeping technique was sufficiently rapid to allow decay measurements of the separated gas. The half-life so determined is within experimental error the same as that found in the neutron-produced activity.

INTRODUCTION

RAPID techniques of neutron irradiation and of chemical separation have been used to characterize the metastable state of Ge^{73} at 67.4-kev excitation energy. These techniques were developed since decay of Ge^{73m} ($t_{\frac{1}{2}}=0.53$ sec) was too slow to permit successful use of the standard delayed-coincidence methods, and too rapid for application of standard chemical-separation procedures. The present work is part of a program to investigate isomeric states with half-lives in the region 1 to 10^{-3} second. Some of these investigations have previously been published.[1]

The half-life of Ge^{73m} has already been noted in various compilations[2,3] from data supplied by the present authors.

The existence of metastable states in Ge^{73} was first suggested by Johansson.[4] In an investigation of the decay of As^{73} he found transitions of 13.5 and 53.9 kev following electron-capture decay of As^{73}. From the observation of fewer prompt coincidences than expected between conversion electrons and x-rays or gamma rays, he concluded that probably both transitions proceeded from metastable states of Ge^{73}. The limits for the expected half-lives of states giving rise to the 13.5-kev transition and the 53.9-kev transition were calculated by Johansson to lie between 1 and 10 microseconds, and between 1 and 10^{-3} second, respectively. Welker *et al.*[5] showed that the highly converted 13.5-kev transition followed the 53.9-kev transition and measured the half-life of the state giving rise to the 13.5-kev transition to be 4.6 microseconds. By using an extraction method for separating Ge from As, a procedure which required about 15 seconds, they were able to show that the half-life of the state giving rise to the 53.9-kev transition was shorter than 10 seconds, and by a delayed-coincidence method, that it exceeded 900 microseconds. They quote a preliminary value communicated to them by the present authors. More recent experiments have revealed that the earlier measurements contained systematic errors. The value of the half-life reported here supersedes that previously quoted.

1. NEUTRON IRRADIATION OF Ge^{72}

The short-period activity in Ge was first found[6] in the course of a systematic search for new neutron-induced short-period nuclides by means of the fast pneumatic tube[7] installed in the Oak Ridge Graphite Reactor. With this device a small sample carried by a rectangular nylon "rabbit" can be propelled in and out of the reactor at high speed by CO_2 gas. The transit time of a sample from the center of the reactor to a shielded NaI scintillation spectrometer outside the reactor can be made as short as 0.1 sec.

Irradiation of a few milligrams of high-purity germanium in the fast pneumatic tube for a few seconds immediately revealed the existence of a short-period decay with a half-life in the fractional-second range. However, the presence of other intense germanium activities, mainly 49-sec Ge^{75m} and 52-sec Ge^{77m}, complicated measurement of the gamma energy and half-life. A preliminary value of 60 kev was estimated for the gamma energy from short time-exposure photographs of the NaI pulse-height spectrum displayed on an oscilloscope. The half-life, based on a decay which could be followed only over a factor of 3, was estimated to be 0.35 sec.

The use of isotopically separated germanium[8] increased the yield of the short-period activity and reduced the background of interfering activities. The

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[1] P. H. Stelson and E. C. Campbell, Phys. Rev. **97**, 1222 (1955); Campbell, Peele, Maienschein, and Stelson, Phys. Rev. **98**, 1172(A) (1955); E. C. Campbell and P. H. Stelson, Bull. Am. Phys. Soc. Ser. II, **1**, 263 (1956); E. C. Campbell and F. Nelson, J. Inorg. Nuclear Chem. **3**, 233 (1956).

[2] Way, King, McGinnis, and van Lieshout, *Nuclear Level Schemes*, $A=40-A=92$, Atomic Energy Commission Report TID-5300 (U. S. Government Printing Office, Washington, D. C., 1955).

[3] Wm. H. Sullivan, *Trilinear Chart of Nuclides*, U. S. Atomic Energy Commission Report (Superintendent of Documents, Government Printing Office, Washington, D. C., 1957).

[4] S. Johansson, Arkiv. Fysik. **4**, 273 (1952).

[5] Welker, Schardt, Friedlander, and Howland, Phys. Rev. **92**, 401 (1953).

[6] E. C. Campbell, Oak Ridge National Laboratory Report ORNL-1365, 1952 (unpublished), and Oak Ridge National Laboratory Report ORNL-1798, 1954 (unpublished).

[7] E. C. Campbell (unpublished).

[8] Obtained from the Stable Isotopes Division, Oak Ridge National Laboratory.

short period was found in the sample enriched to 89.2% in Ge^{72} and was present but weaker in the sample containing 68.9% Ge^{73} and 11.8% Ge^{72}. With a NaI scintillation spectrometer the gamma energy was determined[9] to be 54 kev. No 13.5-kev gamma rays were observed; this is to be expected since the conversion coefficient of the 13.5-kev transition exceeds 1400.[5] The decay rate was measured by means of a single-channel analyzer with its window set at 54 kev and a recording pen-galvanometer operating from a scale of 800. The initial counting rate was 2×10^4 counts per sec. Analysis of the decay curve gave a half-life of 0.53 ± 0.03 sec.

2. SEPARATION OF Ge^{73m} FROM As^{73}

The similarity between the gamma-ray energy of the 0.53-second activity produced by neutron irradiation of Ge^{72} and that observed in the decay of As^{73} suggested that the same metastable state in Ge is involved in both cases. To determine if a short-lived isomer could be separated from As^{73}, a separation technique which was intrinsically capable of removing the short-lived Ge isomer was devised.

The method used is a modification of conventional techniques for separating Ge(IV) as the volatile tetrachloride from As(V) in concentrated hydrochloric acid solution.[10] On sweeping nitrogen gas through a solution of As^{73} [11] in concentrated hydrochloric acid at 100°C, a rapidly decaying germanium activity, as expected, was found in the exit gas stream. Although the germanium activity could be separated by this technique at 25°C, better yields were obtained for the separation at the higher temperature. The experiments to determine its half-life are described below.

A 5-ml aliquot of solution containing 0.4 millicurie of As^{73} was placed in an 8-ml flask fitted with an inlet for introducing nitrogen gas and an outlet leading to a Lusteroid tube in a shielded well-type NaI scintillation counter. Small-diameter plastic tubing (1.5 mm i.d.) was used between the flask and the tube in the counter to allow rapid transfer of the gas. A small trap was introduced in the line to prevent droplets of active solution from being carried over by the gas stream. The gas collected in the Lusteroid tube could be closed off by a double stopcock system so that losses by diffusion or pressure changes during the decay-rate measurements were avoided. A pen recorder connected to the output of a scaler unit was used to follow the decay.

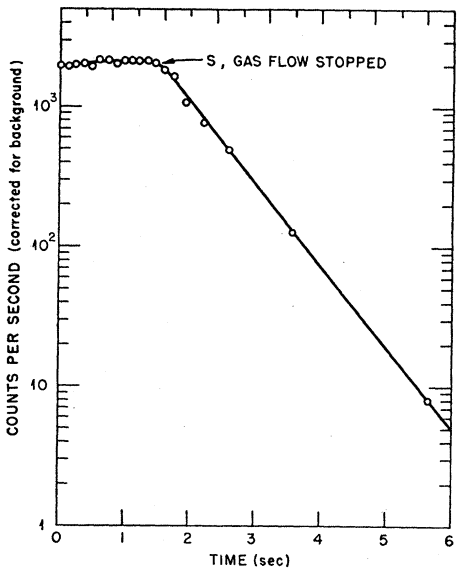

FIG. 1. Counting rate of 0.53-sec Ge^{73m} chemically separated by sweeping active $GeCl_4$ gas from a solution of As^{73} in concentrated HCl.

When nitrogen gas was bubbled through the active solution, the counting rate of the gas flowing through the Lusteroid tube increased rapidly to a few thousand counts/sec, remained at this level until the gas flow was suddenly stopped (indicated by *S* in Fig. 1), and then decayed rapidly. The points plotted in Fig. 1 have been corrected for an observed background counting rate of 29 counts/sec. The average half-life of four separate experiments, each with an initial counting rate of 2500–3000 counts/sec, was 0.52 ± 0.03 second, in agreement with that found for the activity produced by neutron irradiation of Ge^{72}.

3. CONCLUSIONS

By a combination of rapid neutron irradiation and chemical separation techniques, the metastable state of Ge^{73} at 67.4 kev has been shown to decay with a half-life of 0.53 ± 0.03 sec. Partly on the basis of the half-life reported here, the 53.9-kev gamma ray has been identified as an $M2$ transition by the Nuclear Data Group.[2] It is of interest to note that an additional excited state of Ge^{73} at 67.8 ± 0.4 kev has been found in Coulomb-excitation experiments by Temmer and Heydenburg,[12] who show from its excitation and decay characteristics that it is entirely distinct from the metastable level reported here.

[9] The authors wish to thank R. W. Peele and F. C. Maienschein for assistance in the operation of the 40-channel analyzer.

[10] See, e.g., L. M. Dennis and E. B. Johnson, J. Am. Chem. Soc. **45**, 1380 (1923).

[11] Obtained from the Radioisotopes Division, Oak Ridge National Laboratory.

[12] G. M. Temmer and N. P. Heydenburg, Phys. Rev. **96**, 431 (1954).