p-Wave Resonances of U^{238}†

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(Received 26 February 1968)

The neutron transmission of a thick sample of U^{238} has been measured over the energy range 0–200 eV with exceptionally good statistical accuracy. The extremely small transmission dips that are detected are interpreted as p-wave resonances, and their resonance parameters are derived. The probability argument used to identify the p-wave resonances is applicable to most nuclides.

In the study of neutron resonances at very low energy (<100 eV), it is usually assumed that almost all of the observed resonances are excited by s-wave neutrons. Although p-wave resonances are surely present in most heavy nuclides, the centrifugal barrier causes the neutron widths to be so small that the resonances are rarely detected. However, since data of exceptionally good statistical accuracy can now be obtained with the large and efficient bank of boron-loaded liquid scintillators[1] used in transmission measurements with the Argonne fast chopper,[2] we have measured the transmission of a thick sample of U^{238} in an attempt to detect and to analyze the expected tiny transmission dips from p-wave interactions. The sample was cooled to liquid-nitrogen temperature to enhance the depth of the dips by reducing the Doppler broadening. The neutron time-of-flight resolution of the experimental system was about 25 nsec/m, which is adequate for the energy range 0–200 eV that was studied. These measurements are similar to those reported[3] earlier for Th^{232}.

Examples of the transmission data obtained are given in Figs. 1 and 2. The sample had a thickness of 0.1696×10^{24} atoms per cm^2. Although this sample was exceptionally pure chemically and rather pure isotopically, the resonance structure produced by a 0.031% impurity of U^{235} is initially a serious source of uncertainty. However, the transmission dips caused by this isotopic impurity were unambiguously identified in a second run in which the transmission sample consisted of 0.020 in. of U^{235} in addition to the original thick sample of U^{238}. The transmission dips due to U^{235} are easily identified because they are much larger for the composite sample than they are for the U^{238} sample alone. For example, in Fig. 1 it is obvious that the resonances at 9.3, 11.7, and 12.4 eV are due to U^{235} whereas those at 10.25 and 11.32 eV are not. Fortunately, the U^{238} sample is so pure chemically that we detect no resonances from possible chemical impurities.

Although the transmission dips that result from the U^{235} impurity are easy to identify and reject in the energy range shown in Fig. 1, at higher energies there are so many of the unwanted dips that they tend to obscure small dips from U^{238}. However, their effect can easily be eliminated by deducing a corrected spectrum in which the intensity is $N_c=N_a-\alpha N_b$, where N_a is the spectrum obtained with the U^{238} sample alone and N_b is that obtained when the 0.020-in.-thick sample of U^{235} is added, and $\alpha=(t_a/t_b)(n_a/n_b)\ll1$ is a constant. Here t is the running time, n is the effective thickness of U^{235}, and the subscripts a and b refer to the spectra N_a and N_b, respectively. The subtraction procedure eliminates the influence of the U^{235} resonances only if the U^{235} impurity is small enough that $n\sigma\ll1$ for both of the measured spectra. This condition is satisfied at energies greater than 20 eV. The effectiveness of the procedure may be seen from the corrected spectrum given in Fig.

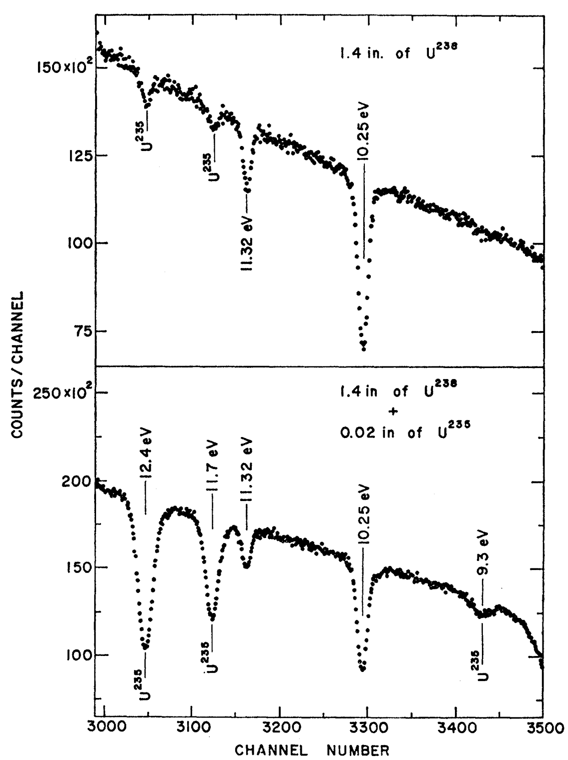

FIG. 1. Measured neutron time-of-flight spectra at very low energies. For the lower curve, 0.020 in. of U^{235} was added to the 1.4-in. sample of U^{238} used for the upper curve. The spectra are almost fully resolved.

† Work performed under the auspices of the U. S. Atomic Energy Commission.

[1] G. E. Thomas, Nucl. Instr. Methods **17**, 137 (1962).

[2] L. M. Bollinger, R. E. Coté, and G. E. Thomas, in *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958* (United Nations, Geneva, 1958), Vol. 14, p. 239.

[3] L. M. Bollinger and G. E. Thomas, Phys. Letters **8**, 45 (1964).

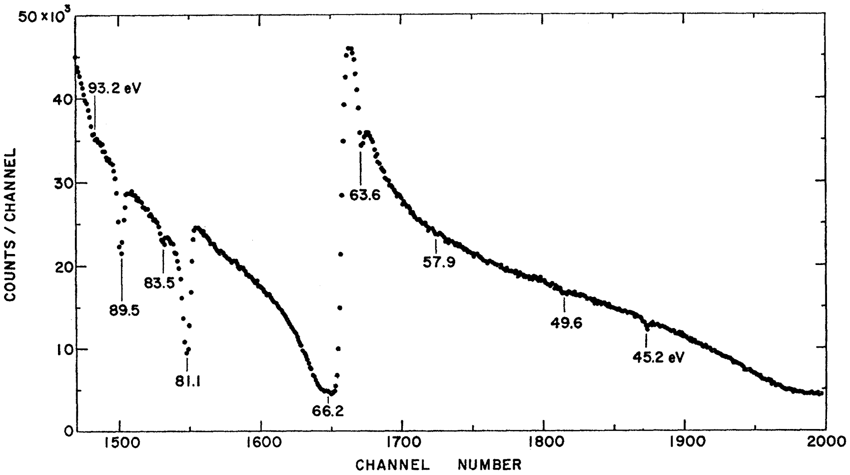

FIG. 2. A time-of-flight spectrum from which the resonance dips caused by the U^{235} impurity in the sample have been removed by the procedure described in the text. The transmission dips to which energy labels are attached are those that are accepted as being real. Other structure that appears significant in the figure (such as near 75 eV) tends to disappear when the data are plotted on a scale that is large enough to preserve their inherent accuracy.

2, where none of the U^{235} resonances[4] can be detected—not even the rather prominent ones in the neighborhood of 56 eV.

As can be seen in Figs. 1 and 2, our spectra contain many small transmission dips that are not caused by U^{235}. The U^{238} sample was so pure chemically that the actual magnitudes of possible chemical impurities could not be determined in a routine chemical analysis. Hence, we depend on the characteristics of the observed resonances themselves to show that they are not caused by chemical impurities. Two kinds of information are used for this purpose. One is the observed width, which for each resonance is small enough to be consistent with the radiation width $\Gamma_\gamma \approx 0.026$ eV and the Doppler width that is expected for U^{238}. On this basis we conclude that the atomic weight of any nuclide responsible for a resonance is fairly high: $A \gtrsim 150$. The second and more useful kind of information consists of the energies and areas of the observed dips. These have been systematically compared with the resonances[4] that would be produced by a small impurity of each element that has a stable isotope. We find that none of the observed transmission dips can be attributed to a stable isotope. Similarly, none of them can be attributed to unstable isotopes of heavy elements (other than U) for which the neutron cross section has been measured. Hence we conclude that all of the observed transmission dips are formed by resonances in uranium.

Our final step in the isotopic assignment of the observed resonances was to compare them with what would be expected from uranium isotopes other than U^{235} and U^{238}. Again this was done by a comparison with resonance parameters determined in previous measurements.[4] For lack of information to the contrary, a small dip observed at 5.19 ± 0.03 eV must be attributed to U^{234}, which is known[4] to have a prominent resonance at 5.19 ± 0.02 eV. Similarly, a barely detectable dip at about 5.49 eV probably results from a known resonance in U^{236}. Using these dips as a measure of the amount of U^{234} and U^{236} in the sample, one can then show that neither isotope could be responsible for any of the clearly observed transmission dips at higher energy. The influence of other possible uranium isotopes is not detected at all.

Since almost all other possibilities have been excluded by the tests outlined above, we conclude that all of the remaining transmission dips are due to U^{238}. All of these resonances that are exceptionally weak, including the previously reported[4,5] ones at 10.25 and 89.5 eV, are listed in the upper part of Table I. For comparison, the well-known large resonances[4] in the same range of energy (usually assumed to be s-wave resonances) are given in the lower part of the table.

All of the usual methods of making an unambiguous separation of s-wave and p-wave resonances are ineffective for the tiny resonances of interest here, because the neutron widths are so unusually small; the weakest of the resonances (the one at 16.3 eV) has a peak cross section σ_0 of only 0.3 b. However, the probable nature of these resonances can be inferred by comparing the observed neutron widths with the expected widths for s-wave and p-wave resonances. This comparison is made in Table I. First we list values of $g\Gamma_n$ that are obtained from the areas of the transmission dips. From these values we obtain the ratio $g\Gamma_n/\langle g\Gamma_n\rangle$ for s-wave and p-wave excitations by using mean values $\langle g\Gamma_n\rangle$ calculated from the strength function, which is defined[6] as

$$S_l = \frac{1}{(2l+1)\Delta E} \sum g\Gamma_n{}^l, \quad (1)$$

where ΔE is the neutron energy range considered, $\Gamma_n{}^l$ is the reduced neutron width for an orbital angular mo-

[4] Data from many sources are summarized in Brookhaven National Laboratory Report No. BNL-325, Suppl. No. 2, Vols. I–III (unpublished). Information on uranium is given in Vol. III.

[5] L. M. Bollinger, R. E. Coté, D. A. Dahlberg, and G. E. Thomas, Phys. Rev. **105**, 661 (1957).

[6] A. Saplakoglu, L. M. Bollinger, and R. E. Coté, Phys. Rev. **109**, 1258 (1958).

TABLE I. Summary of data on low-energy resonances in U^{238}. The energies and widths of the resonances in the lower part of the table were taken from Ref. 4.

E_0 (eV)	$g\Gamma_n$ (10^{-6} eV)	$g\Gamma_n/\langle g\Gamma_n\rangle$ s wave	p wave	$P(p,g\Gamma_n)$	Asymmetry	l value
4.41±0.01	0.111±0.002	2.94×10^{-5}	0.66×1	0.997	?	1
10.25±0.02	1.56 ±0.01	27	2.62	0.988	?	?
11.32±0.02	0.358±0.006	5.9	0.53	0.995	?	1
16.3 ±0.04	0.053±0.015	0.73	0.045	0.996	?	1
19.6 ±0.04	1.0 ±0.1	12.4	0.64	0.994	?	1
45.2 ±0.2	0.83 ±0.15	6.8	0.15	0.992	?	1
49.6 ±0.2	0.68 ±0.23	5.3	0.11	0.992	?	1
57.9 ±0.3	0.48 ±0.08	3.5	0.06	0.992	?	1
63.6 ±0.3	5.5 ±1.5	37.8	0.59	0.989	?	1
83.5 ±0.4	7.0 ±0.7	42.8	0.51	0.988	?	1
89.5 ±0.4	85.0 ±4	500	5.5	0.908	?	1
93.2 ±0.5	3.0 ±0.6	17.3	0.19	0.989	?	1
125.0 ±0.6	14.2 ±2	70.6	0.56	0.984	?	1
152.6 ±0.8	37.0 ±2	167.0	1.09	0.978	?	1
159.3 ±1	10.4 ±1.3	45.7	0.28	0.985	?	1
173.3 ±1	33.4 ±4	143.0	0.82	0.979	?	1
6.67	1.5×10^3	0.32×1	4.9×10^3	0	yes	0
21.0	8.5	1.03	4.9	0	yes	0
36.7	31	2.8	7.8	0	yes	0
66.2	25	1.7	2.6	0	yes	0
81.1	2.0	0.12	0.15	2.7×10^{-31}	yes	0
102.7	68	3.7	3.7	0	yes	0
116.9	26	1.3	1.2	0	yes	0
145.7	0.70	0.03	0.022	1.3×10^{-3}	yes	0
165.4	3.0	0.13	0.080	3.1×10^{-16}	yes	0
189.6	145	5.8	3.1	0	yes	0

mentum l of the incident neutron, and g is the usual statistical factor $g=\frac{1}{2}(2J+1)/(2I+1)$, J being the spin of the resonance and I the spin of the target nucleus. For the low energies involved in our measurement the reduced width Γ_n^1 for a p-wave resonance is $\Gamma_n^1 \equiv\Gamma_n k^{-2}R^{-2}E_0^{-1/2}$, where E_0 is the resonance energy, k is the neutron wave number, and R is the nuclear radius. Using the value[5] $R=9.1\times10^{-13}$ cm yields $\Gamma_n^1=2.5\times10^5E_0^{-3/2}\Gamma_n$. For s-wave neutrons the reduced width is $\Gamma_n^0\equiv\Gamma_nE_0^{-1/2}$.

Values of $g\Gamma_n/\langle g\Gamma_n\rangle$ were calculated under the assumption that the strength function S is 1.0×10^{-4} for s-wave resonances and 2.5×10^{-4} for p-wave resonances, as reported by Uttley *et al.*[7] In the table one sees that the neutron widths of the newly found resonances are all of about the size expected for p-wave resonances and that they are all much smaller than would be expected for the typical s-wave resonances. This in itself suggests strongly that they are p-wave resonances.

A more quantitative statement about the nature of the tiny resonances may be made by considering Bayes's theorem[8] on conditional probability. Let us apply this theorem to our problem under the assumption that all resonances are either s-wave or p-wave resonances. Then, when a resonance of unknown orbital angular momentum has a known value of $g\Gamma_n$, the probability that the resonance is excited by p-wave neutrons is

$$P(p,g\Gamma_n)=\frac{\pi_p\bar{\rho}_p(g\Gamma_n)}{\pi_p\bar{\rho}_p(g\Gamma_n)+\pi_s\bar{\rho}_s(g\Gamma_n)},\qquad(2)$$

where π_p and π_s are the *a priori* probabilities that the resonance is excited by p and s waves, respectively, and $\bar{\rho}_p(g\Gamma_n)$ and $\bar{\rho}_s(g\Gamma_n)$ are the probabilities of obtaining a value $g\Gamma_n$ within some arbitrary interval when a resonance is known to be p wave or s wave, respectively.

Let us assume that the density of states is proportional[9] to $2J+1$, independent of parity, where J is the spin of the state. Then, since states with $J=\frac{1}{2}$ and $\frac{3}{2}$ are formed by p-wave interactions, whereas only states with $J=\frac{1}{2}$ are formed by s waves, the *a priori* probabilities are $\pi_p=\frac{3}{4}$ and $\pi_s=\frac{1}{4}$. Also, it appears that the appropriate lower limit on the arbitrary interval associated with $\bar{\rho}$ is the uncertainty in $g\Gamma_n$. Thus, when the error is small, we may let $\bar{\rho}_p\to\rho_p$ and $\bar{\rho}_s\to\rho_s$, where ρ_p and ρ_s are the probability density functions of $g\Gamma_n$ for p- and s-wave resonances, respectively. As a result, for the U^{238} resonances Eq. (2) reduces to

$$P(p,g\Gamma_n)\approx(1+\tfrac{1}{3}(\rho_s/\rho_p))^{-1}.\qquad(3)$$

If the neutron strength function is assumed to be independent of J, then for the special case of a target

[7] C. A. Uttley, C. M. Newstead, and K. M. Diment, in *Proceedings of the Conference on Nuclear Data for Reactors, Paris, 1966* (International Atomic Energy Agency, Vienna, 1967), Vol. I, p. 165.

[8] Bayes's theorem is discussed in most books on probability. For example, see H. Cramer, *The Elements of Probability Theory* (John Wiley & Sons, Inc., New York, 1954).

[9] Evidence concerning the J dependence is reviewed, for example, by L. M. Bollinger, in *Nuclear Spectroscopy*, edited by F. Ajzenberg-Selove (Academic Press Inc., New York, 1960), Part A, p. 433.

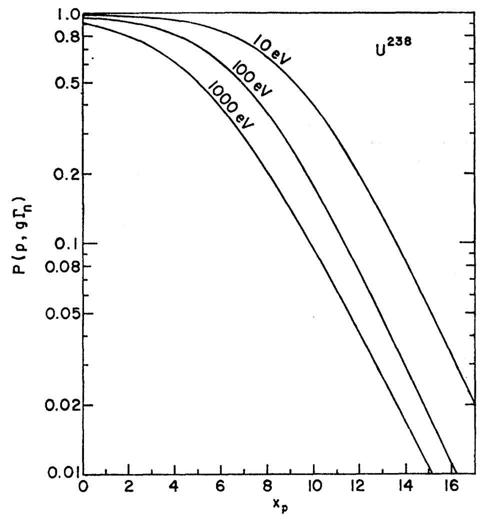

FIG. 3. Plots of $P(p,g\Gamma_n)$ versus x_p for U^{238} at representative values of the neutron energy. The abscissa is $x_p \equiv \Gamma_n/\langle\Gamma_n\rangle_p$, and the probability $P(p,g\Gamma_n)$ was calculated from Eq. (4) with $S_0=1.0\times10^{-4}$ and $S_1=2.5\times10^{-4}$. The ratio $\langle x_s\rangle/\langle x_p\rangle$ is 1110, 111, and 11.0 at the energies 10, 100, and 1000 eV, respectively.

nucleus with $I=0$ it follows that the average value of $g\Gamma_n$ is independent of J for p-wave resonances; thus, the quantity $g\Gamma_n/\langle g\Gamma_n\rangle = \Gamma_n^l/\langle\Gamma_n^l\rangle$ is expected to obey the Porter-Thomas[10] distribution for both the s-wave and p-wave resonances, even though we cannot determine the g values of the individual p-wave resonances. Now Eq. (3) reduces to the explicit form[11]

$$P(p,g\Gamma_n)=\{1+\tfrac{1}{3}(\langle g\Gamma_n\rangle_p/\langle g\Gamma_n\rangle_s)^{1/2}\exp[\tfrac{1}{2}(x_p-x_s)]\}^{-1}$$
$$=\{1+cE_0^{1/2}\exp[\tfrac{1}{2}(x_p-x_s)]\}^{-1}, \qquad (4)$$

where $x\equiv g\Gamma_n/\langle g\Gamma_n\rangle$ and c is a constant that is independent of E_0 and $g\Gamma_n$. For the conditions and parameters assumed above, $c=3.16\times10^{-3}$ when E_0 is in eV.

The second form of Eq. (4) provides a clear description of the dependence of $P(p,g\Gamma_n)$ on various factors. First, notice that when x_p and x_s are small, $P(g,g\Gamma_n)$ depends mainly on E_0 and its value is almost unity for $E_0<100$ eV. Second, as x_p increases beyond a value of 5 or 10, the exponential term becomes important and $P(p,g\Gamma_n)$ decreases rapidly. Thus, for the low-energy resonances with which we are concerned, there is only a rather small range of $g\Gamma_n$ within which the value of $P(p,g\Gamma_n)$ does not provide a meaningful determination of the orbital angular momentum. This behavior of $P(p,g\Gamma_n)$ is shown graphically in Fig. 3. Also, the figure emphasizes the importance of low energy for an effective discrimination between s-wave and p-wave resonances on the basis of the neutron width.

The calculated values for the probability $P(p,g\Gamma_n)$ that the observed U^{238} resonances are p-wave resonances are given in column 5 of Table I. These values indicate that there is a high probability that all of the narrow resonances listed in the upper part of the table are p-wave resonances—even the rather wide one at 89.5 eV. Also, all the resonances in the lower part of the table are almost certainly s-wave resonances. These conclusions are not inconsistent with previously published information except with regard to the narrow resonance at 10.25 eV. Chrien *et al.*[12] have concluded that this is an s-wave resonance because its capture γ-ray spectrum is roughly similar to that of resonances that are surely s wave. Although we agree that their γ-ray data do provide a strong indication that the 10.25-eV resonance is excited by s waves, it also seems to us that the unavoidably marginal quality of the γ-ray spectrum of this weak resonance leaves room for doubt.

In an effort to remove some of the remaining uncertainty about the l values of the narrow resonances, we have examined their shapes to see if they are inconsistent with the asymmetrical shape that would be expected of s-wave resonances. The results obtained are given in column 6 of Table I. No useful information could be obtained for the narrow resonances because the expected difference between s-wave and p-wave resonances is too small. However, a definite asymmetry was observed for each of the resonances for which $g\Gamma_n$ is large enough to result in a small value of $P(p,g\Gamma_n)$; thus, these are surely s-wave resonances.

The l-value assignments that follow from the data of Table I and from the above discussion are given in the last column. It must be understood, of course, that there is some small probability that any one of the p-wave assignments is in error.

If the above conclusions about the nature of the small transmission dips are accepted, one may use the data of Table I to deduce some interesting average properties of p-wave resonances of U^{238}. First, consider the level spacing. The most reliable data for this purpose are provided by the resonances below the large s-wave resonance at 21 eV. Here four spacings cover an interval of 15.2 eV in an energy range within which almost all of the p-wave resonances should be observable. Thus, the average spacing is roughly 3.8 eV—a value that, in view of the small size of the sample of spacings, is not inconsistent with the expected value of one-third the s-wave spacing of 18 eV.

A rough value of the strength function S_1 of p-wave resonances may also be obtained. By an obvious extension of the definition given in Eq. (1), the best value

[10] C. E. Porter and R. G. Thomas, Phys. Rev. **104**, 483 (1956).

[11] The arguments that lead to Eq. (4) may also be used to derive a useful expression for $P(p,g\Gamma_n)$ when $I\neq0$. In the general case the ratio π_s/π_p is not necessarily $\frac{1}{3}$; instead, it is $\frac{1}{3}$, 4/9, and $\frac{1}{2}$ when I is 0, $\frac{1}{2}$, and ≥1, respectively. Also, when $I\neq0$ the density function ρ_p is not expected to be a simple Porter-Thomas distribution but rather a distribution formed by the superposition of two or more Porter-Thomas distributions with somewhat different mean values. Nevertheless, if the factor $\frac{1}{3}$ is replaced by the appropriate value of π_s/π_p, Eq. (4) is as accurate as is meaningful for most nuclides, in view of our limited knowledge of the J dependence of $\langle g\Gamma_n\rangle$.

[12] R. E. Chrien, M. R. Bhat, O. A. Wasson, and D. L. Price, Bull. Am. Phys. Soc. **12**, 105 (1967).

of S_1 is

$$S_1=\frac{1}{3\Delta E}\sum P(p,g\Gamma_n)g\Gamma_n^1. \qquad (5)$$

In this sum let us restrict the data to the range 0–100 eV. Also, assume that a quarter of the 100-eV range is excluded from observation by the presence of the large s-wave resonances. Then we obtain the value

$$S_1=(2.14_{-1.0}^{+2.0})\times10^{-4},$$

where the errors designate 80% confidence limits calculated under the assumption that the reduced width $g\Gamma_n^1$ is distributed according to the Porter-Thomas distribution. If the contribution from the 10.25-eV resonance is dropped, the value of S_1 becomes 1.6×10^{-4}. These values are in excellent agreement with Utley's value of 2.5×10^{-4}.

Nothing useful can be learned about the distribution of neutron widths, because there is a high probability that a resonance is not detected if $g\Gamma_n/\langle g\Gamma_n\rangle<0.1$. The dearth of p-wave resonances with small values of $g\Gamma_n/\langle g\Gamma_n\rangle$ is apparent from the table.

Perhaps the most interesting resonance parameter that can be deduced from the narrow resonances is the total radiation width for a p-wave resonance, since few such widths have been reported previously for individual p-wave resonances in a heavy nuclide. The most favorable transmission dip for this purpose is the one at 4.41 eV. By curve fitting these data we obtain $\Gamma_\gamma=0.017\pm0.007$ eV, a result that does not differ significantly from the value 0.026 ± 0.002 eV for s-wave resonances.[4] Unfortunately, the error cannot be reduced enough to provide an accurate comparison of the two widths because the shape of the necessarily small transmission dip is dominated by the Doppler width.

The authors are indebted to Dr. J. E. Moyal for a helpful discussion of Bayes's theorem.

PHYSICAL REVIEW VOLUME 171, NUMBER 4 20 JULY 1968

Production of ^{7}Be, ^{22}Na, and ^{24}Na Fragments from Heavy Elements at 3, 10, and 30 GeV*

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Formation cross sections of ^{7}Be, ^{22}Na, and ^{24}Na from tantalum, lead, and uranium targets irradiated with 3-, 10-, and 30-GeV protons have been measured. Similar measurements have been made with silver and gold targets at 30 GeV. At 10 and 30 GeV, the yield of ^{24}Na increases regularly with target mass, whereas at lower energies plots of cross section versus target mass show minima. From the ^{7}Be data, it seems probable that the excitation energy transferred to the struck nucleus by the incident particle reaches a maximum even for the heaviest nuclei at $E_p\leq10$ GeV. Very little, if any, additional nuclear excitation energy is gained by raising the incident proton energy above this figure.

I. INTRODUCTION

The formation cross sections of ^{7}Be, ^{22}Na, and ^{24}Na have been measured from tantalum, lead, and uranium targets at 3, 10, and 30 GeV and from silver and gold at 30 GeV. These results combined with published data from copper[1,2] and silver[3] targets extend the systematic study of the yields of these fragments as a function of target mass to the highest proton energies presently available. The results from lead and uranium targets presented here were originally included in a survey of formation cross sections of a large number of nuclides from these targets at 3 and 30 GeV.[4]

There are two reasons for interest in the yields of these fragments at high incident energies. In the work of Caretto *et al.*[5] and Crespo *et al.*[6] and others it was observed that plots of cross sections of fragments in the 18 to 30 mass region versus target mass show minima somewhere between silver and tantalum targets. The minima seemed to become shallower as the energy of the incident proton was increased, however, they were still present at 6 GeV. At this energy cross sections for ^{24}Na were lower from silver targets than from either

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[4] G. Friedlander, *Physics and Chemistry of Fission* (International Atomic Energy Agency, Vienna, 1965), Vol. II, p. 265.
[5] A. A. Caretto, J. Hudis, and G. Friedlander, Phys. Rev. **110**, 1130 (1958).
[6] V. P. Crespo, J. M. Alexander, and E. K. Hyde, Phys. Rev. **131**, 1765 (1963).