LETTERS TO THE EDITORS

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Fission Products of U²³⁵

THE first quantitative measurements on the formation of fission products in thermal neutron bombardment of uranium were made by Anderson, Fermi and von Grosse[1]. The fission yield of a fission isotope is the probability of the isotope being formed per thermal neutron fission. In bombardment, the rate of production of any active isotope from fission, that is, the fission rate multiplied by the fission yield, is equal to the disintegration rate of the active isotope referred to saturation of bombardment for the isotope. Thus data required for measurement of the fission yield of an active fission isotope are : (a) number of fissions occurring in a given sample per unit time ; and (b) disintegrations per unit time of the isotope referred to saturation of bombardment.

Grummitt, Gueron, Wilkinson and Yaffe[2] determined the fission yields for Ba¹³⁹ (86 min.), Ba¹⁴⁰ (12·7 days) and La¹⁴⁰ (40 hr.) produced in thermal neutron fission of U²³⁵. The direct determination of fission rate in the bombarded sample was avoided by comparison of U²³⁹ (23 min.) and fission product β⁻ activities : accepting the ratio of the capture (σc) and fission (σf) cross-sections for thermal neutrons on natural uranium, the fission yield Yf is given by the expression

$$\frac{\text{U}^{239} \text{ activity (referred to saturation of bombardment)}}{\text{Fission product activity (referred to saturation of bombardment)}} =$$

$$\frac{\text{Thermal neutron capture rate}}{Yf \times \text{thermal fission rate}} = \frac{\sigma c}{Yf\sigma f}.$$

For Ba¹³⁹ and La¹⁴⁰, values of 6·1 × 10⁻² and 5·8 × 10⁻² respectively were obtained. The fission yield for Ba¹⁴⁰ was 4·3 × 10⁻². In the growth of La¹⁴⁰, from the parent Ba¹⁴⁰, 1·43 times as many La¹⁴⁰ as Ba¹⁴⁰ disintegrations were observed. Corrections were made for self-weakening of β⁻ rays in the samples, and for external absorption of β⁻ rays in the counting arrangement used. The latter was such that β⁻ rays of maximum energy below 0·1 Mev. were not observable. In subsequent work with the β⁻ ray spectrometer, L. G. Elliott[3] has shown that a large fraction of the electrons from Ba¹⁴⁰ are below 32 Kev. These electrons would be completely stopped in the mica window of the counters used. If these are a low-energy β⁻ ray spectrum, and not conversion electrons, they would, of course, give rise to La¹⁴⁰, and could account for the observed growth of La¹⁴⁰. Further indication has been obtained using a windowless counter.

Fission yields of other fission isotopes can be obtained by activity comparisons to Ba¹⁴⁰ in the same counting arrangements using the apparent or reference yield of 4·3 × 10⁻² for Ba¹⁴⁰. The longer-lived fission products of uranium have been studied by this method. If Y_i, A_i and Y_{Ba}, A_{Ba} are respectively the fission yields and activities (referred to saturation of bombardment) of any fission product i and Ba¹⁴⁰, then

$$Y_i = \frac{A_i}{A_{Ba}} \times Y_{Ba}, \text{ where } Y_{Ba} = 4\cdot3 \times 10^{-2}.$$

Fission yields have been obtained in this way for twenty-two fission isotopes.

In the graph, the logarithm of the fission yield is plotted against the mass number of the fission isotope. In addition to the present results, fission yields for krypton and xenon isotopes of masses 83, 84, 86, 131, 132, 134 and 136 are included. These were obtained for mass-
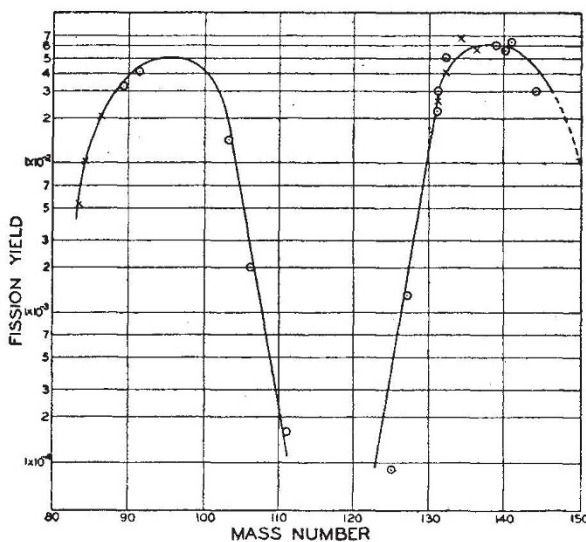
FISSION YIELD *vs.* MASS NUMBER FOR THERMAL NEUTRON FISSION OF U²³⁵

⊙, From activity comparisons

×, Calculated from mass-spectrographic abundance data for krypton and xenon

spectrographic determinations of isotopic abundances in fission product gases by H. G. Thode et al.[4], using as a reference the fission yield of I¹³¹ obtained by activity comparison to Ba¹⁴⁰, with the assumption that I¹³¹ decays directly to Xe¹³¹.

(A) Two symmetrical groups of fission products are formed with maxima at mass numbers 96 and 138 and a fission yield of ~ 6 × 10⁻². The sum of the maxima is approximately 234, inferring that on the average, between one and three secondary neutrons are emitted per fission.

(B) The masses at a fission yield of 1 × 10⁻² are 89 and 104 in the 'light group' and 130 and 151 in the 'heavy group'.

(C) The total of fission yields is 0·9 for each group. The divergence from unity is almost certainly due to a low value for the reference yield of Ba¹⁴⁰. A value of 4·8 × 10⁻² would remove the discrepancy and place the maxima at about 6·5 × 10⁻².

(D) The greater part of the heavy group lies in the rare earth region, and fission isotopes up to europium might be expected.

(E) After about 100 days, the measured gross activity of U²³⁵ fission products is accounted for by activities of the separated isotopes.

(F) Several previously unreported isotopes have been observed during the course of the present work, namely, Sn > ¹²⁹ (136 days, 1·2 Mev.); Sn > ¹³⁰ (17·5 days, 1·7 Mev.); Sn > ¹²⁶ (7·0 days, 1·8 Mev.); Sb⁽¹²⁶⁾ (several years, 0·56 Mev.); Sb > ¹²⁶ (28 days, 1·86 Mev.); Cs⁽¹³⁶⁾ (~ 20 days). Fission yields of these and the following β⁻ active isotopes were measured : Sr⁸⁹ (54·5 days, 1·5 Mev.); Sr⁹⁰ (~ 70 years, 0·6 Mev.); Y⁹⁰ (72 hr., 2·2 Mev.); Y⁹¹ (61 days, 1·4 Mev.); Zr⁹⁵ (65 days, 0·5 Mev.), Nb⁹⁵ (33 days, 0·15 Mev.); Ru¹⁰³ (37 days, 0·25 Mev.); Ru⁽¹⁰⁶⁾ (290 days), Rh⁽¹⁰⁶⁾ (30 sec., 3·3 Mev.); Ag¹¹¹ (7·5 days, 0·9 Mev.); Sb¹²⁷ (95 hr., 0·8 Mev.), Te⁽¹²⁷⁾ (90 days I.T.), Te¹²⁷ (9·3 hours, 0·77 Mev.); Te*¹²⁹ (32 days I.T.), Te¹²⁹ (72 min., 1·5 Mev.); I¹³¹ (8·1 days, 0·6 Mev.); I¹³² (2·30 hr., 1·35 Mev.); Cs⁽¹³⁶⁾ (~ 100 years, 0·8 Mev.); Ba¹³⁹ (86 min., 2·2 Mev.); Ba¹⁴⁰ (12·7 days, 0·92 Mev.), La¹⁴⁰ (40 hr., 1·4 Mev.); Ce⁽¹⁴¹⁾ (18 days); Ce¹⁴⁴ (290 days, 0·4 Mev.), Pr¹⁴⁴ (17 min., 2·5 Mev.).

A detailed account of the experimental work is in preparation.

W. E. GRUMMITT
G. WILKINSON

Atomic Energy Division,
National Research Council,
Chalk River Laboratory,
Chalk River, Ontario.
June 1.

[1] Anderson, H., Fermi, E., von Grosse, A., *Phys. Rev.*, **59**, 52 (1941).
[2] Grummitt, W. E., Gueron, J., Wilkinson, G., Yaffe, L., unpublished work (Montreal, 1944).
[3] Elliott, L. G., unpublished work (Montreal, 1945).
[4] Thode, H. G., Graham, R. L., Harkness, A. L., unpublished work (Hamilton, Ont., 1945).

Polyatomic Electronic Spectra : Further Analysis of the Vibrations of the ¹B₂ᵤ State of Benzene

IN a previous communication[1] we indicated how, in the example of the ¹B₂ᵤ state of C₆H₆ and C₆D₆, the product theorem and computations of force constants could be used to calculate or verify vibrational frequencies in electronically excited molecules. The discussion was almost entirely confined to the out-of-plane vibrations of both molecules, as insufficient experimental data were then available to permit calculations of the product ratios or force constants of the in-plane vibrations. It has now, however, been found possible to apply these methods to the in-plane vibrations, thus completing the first determination of a force system of an excited polyatomic molecule.

It was pointed out that Sponer's assignment of the frequencies 2565 cm.⁻¹ in C₆H₆[2] and 1821 cm.⁻¹ in C₆D₆[3] to the A_{1g}(H) vibration of the excited state, when combined with the known A_{1g}(C) frequencies, 923 cm.⁻¹ and 879 cm.⁻¹, of excited C₆H₆ and C₆D₆ respectively, was not in agreement with the product theorem. We now propose that the C₆H₆ progression previously regarded as

$$0-0 + 520 (\overset{+}{E_g})' + 2565 (A_{1g})' + \nu_1' \times 923 (A_{1g})'$$

and the corresponding C₆D₆ progression

$$0-0 + 499 (\overset{+}{E_g})' + 1821 (A_{1g})' + \nu_1' \times 879 (A_{1g})'$$

should be formulated

$$0-0 + 3085 (\overset{+}{E_g})' + \nu_1' \times 923 (A_{1g})'$$

and

$$0-0 + 2320 (\overset{+}{E_g})' + \nu_1' \times 879 (A_{1g})'. \text{ respectively.}$$

The frequencies 3085 cm.⁻¹ in C₆H₆ and 2320 cm.⁻¹ in C₆D₆ are assigned to the $\overset{+}{E_g}$ (H-stretching) vibration of the excited state. The frequencies of the same vibration in the electronic ground-state are 3047 cm.⁻¹ in C₆H₆ and 2264 cm.⁻¹ in C₆D₆. The assignment thus implies that the hydrogen-stretching force constant is higher in the excited state than in the ground-state, unlike other force constants, which we find to be reduced as a result of excitation. If the electron which is excited is one of the π-electrons then it seems plausible, not only that forces involving deformations of C—C bonds should be reduced, but also that the C—H bonds may slightly increase their order at the expense of the weakened ring system, with the result that the hydrogen-stretching constant becomes increased.

An immediate consequence is that the A_{1g}(H) frequencies should be raised. Consistent values are obtained from the hitherto unassigned progressions

$$0-0 + 520 (\overset{+}{E_g})' + 3130 (A_{1g})' + \nu_1' \times 923 (A_{1g})'$$

in the absorption spectrum of C₆H₆ and

$$0-0 + 499 (\overset{+}{E_g})' + 2361 (A_{1g})' + \nu_1' \times 879 (A_{1g})',$$

in that of C₆D₆. The frequencies 3130 and 2361 cm.⁻¹ are assigned to the A_{1g}(H) vibration of the excited state. The product ratio,

$$\Pi A_{1g}(C_6H_6)'/\Pi A_{1g}(C_6D_6)' = 923 \times 3130/879 \times 2361 = 1\cdot392,$$

is in satisfactory agreement with the ground-state value 1·404.