

EUR 307.e

EUROPEAN ATOMIC ENERGY COMMUNITY – EURATOM

**ABSOLUTE DETERMINATION
OF THE U-235/U-238 RATIO
WITH AN OPTICAL INTERFEROMETER (HYPEAC)**

by

C. BERTHELOT and K.F. LAUER

1963



**Joint Nuclear Research Center
Geel Establishment - Belgium**

Central Nuclear Measurements Bureau

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ABSOLUTE DETERMINATION OF THE U-235/U-238 RATIO WITH AN OPTICAL INTERFEROMETER (HYPEAC)

SUMMARY

An optical method for the absolute determination of the isotope ratio of U-235/U-238 is described. The recording of the homologous lines of uranium is effected by using a photoelectric Fabry-Perot interferometer (Hypeac). A series of samples of uranium oxides containing from 3 to 93% of U-235 (NBS standards) has been analyzed and the results agree to within $\pm 0.70\%$ with those given by the National Bureau of Standards. The relative precision of the method is of the same order, i.e. about $\pm 1\%$ at the 99% confidence level.

1 — INTRODUCTION

Emission-spectroscopic methods of determining the isotope ratios of fissile materials have been used independently of radiochemical or mass-spectrometric methods [1]. The isotope shift is about 0.31 \AA for a wave length of 5144.97 \AA [2] and about 3.77 \AA [3] for the 1.903μ line. In most cases this isotope shift for uranium-235 and 238 does not exceed 0.1 \AA , which means that one is forced to use high-resolution equipment for such determinations by emission spectroscopy.

2 — CHOICE OF THE OPTICAL EQUIPMENT AND THE LIGHT SOURCE

To obtain a separation of about 0.05 \AA in the spectrum range $4,000$ to $7,000 \text{ \AA}$, two types of instrument can be used (theoretical resolution $80,000$ to $140,000$):

- a) a grating spectrograph operating at high order [4];
- b) an interferometric spectrometer such as the Fabry-Perot type [5].

The latter is much smaller than a grating spectrograph or even a prism-echelle spectrograph [6]. If the Fabry-Perot is perfect, its intensity transmission is:

$$I = I_0 \tau_A \left[1 + \frac{4R}{(1-R)^2} \sin^2 \left(\frac{2\pi\mu e}{\lambda} \cos i \right) \right]^{-1}$$

where the transmission factor of multi-dielectric coatings τ_A (0.5-0.9) is given by the relation: $\tau_A = (T/1-R)^2$.

I_0 = initial intensity

A = absorption coefficient

R = reflector power and $T = 1 - R - A$

μ = refractive index of the refracting matter between the two Fabry-Perot plates

e = distance between the plates

λ = wavelength of the monochromatic light

i = angle of incidence of the light beam.

According to P. Jacquinot [7] the product of the resolution and the transmitted light flux is given by:

$$(R\phi)_{F.P.} = 1.2 \pi \tau B S$$

τ = effective transmission factor (0.3 - 0.6)

B = brightness of the source

S = area of the Fabry-Perot;

in the case of a grating we obtain:

$$(R \phi)_{\text{grating}} = \tau B S \beta (\sin i_1 + \sin i_2)$$

τ = effective transmission factor (0.25 - 0.75)

S = grating area

β = angular slit height ($\approx 1/50$)

i_1 = angle of incidence of the beam

i_2 = angle of emergence of the beam.

In general it can be said that, at equal resolving power, the Fabry-Perot interferometer combined with a premonochromator transmits about 30 times as much light as the grating spectrometer, which itself is much better than a prism spectrograph [8].

$$\frac{\phi_{\text{prism}}}{\phi_{\text{grating}}} = \frac{\lambda \cdot \frac{d\mu}{d\lambda}}{2 \sin \varphi} \leq 0.13$$

with

$$\sin i_1 + \sin i_2 = 2 \sin \varphi \quad (\text{Littrow mounting})$$

The most frequently used sources for this kind of analysis are the DC or AC arc [9], which are very luminous but quite unstable, the high-frequency discharge [10] and the hollow cathode [11]. The last-mentioned is very often preferred, although its discharge is 10 to 20 times less luminous than an electrodeless discharge. One very important reason for this is that the hollow cathode, when cooled with liquid nitrogen, gives very sharp and fine lines because the Doppler effect

$$\Delta\lambda_D = \frac{\lambda}{c} (8 RT \log 2/M)^{1/2}$$

then becomes negligible. The stability is such that slow scanning of the spectrum is easily possible and it is quite superior to that of a high-frequency discharge (fig. 1).

3 — THE "HYPEAC" INTERFEROMETER

The instrument we used, known as "Hypeac", was designed and developed by Prof. P. Jacquinot [12] at the Bellevue laboratories of the "Centre National de la Recherche Scientifique" *.

The instrument comprises the following three parts:

3.1 — The vacuum system (*Vacuac*)

A complete pumping system composed of a primary and a secondary pump and the pressure-measuring system ($P \leq 10^{-4}$ Torr), which serves for the degassing of the hollow cathode; the gas-filling system and the Fabry-Perot chamber. The gas-filling system

* Constructed by Soc. Jobin-Yvon under licence from the CEA.

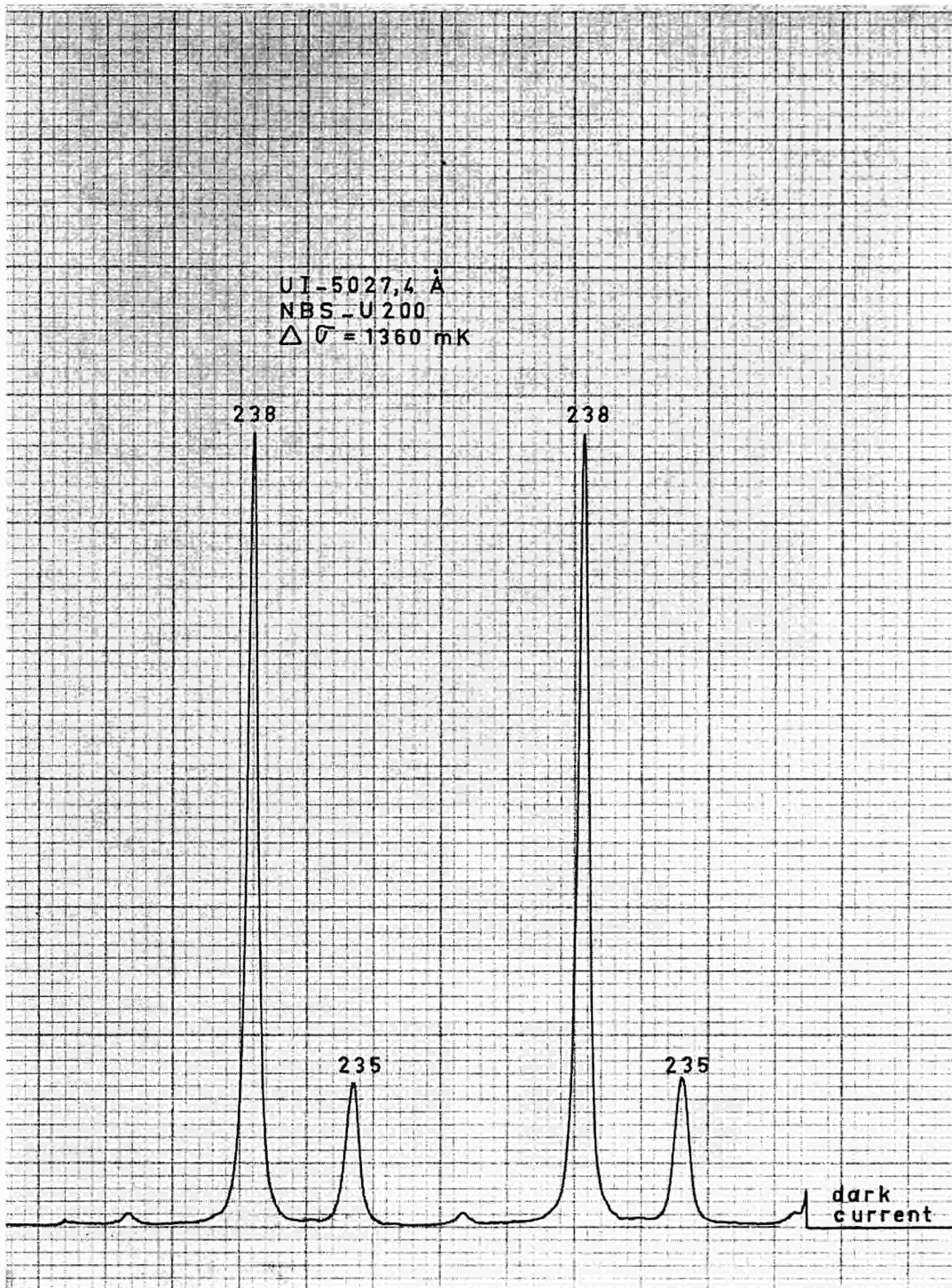


Fig. 1. — Uranium lines U-238 - 5,027.398 Å and U-235 - 5,027.295 Å excited in a liquid-nitrogen-cooled hollow cathode lamp.

consists of the gas container (1 l at 50 atm) and a pressure-reduction system by means of which this pressure can be reduced to about 0.5-1 Torr. For the spectrographically pure neon which we are currently using, this is the optimum pressure range. An indium 'O'-ring secures the vacuum seal of the Schüller-type cathode lamp at the liquid nitrogen temperature.

3.2 — *The optical system*

(a) The grating Ebert-Fastie type premonochromator has symmetrically curved entrance and exit slits. The grating has about 1,200 lines/mm and is 65 mm long. The blaze angle is $36^{\circ}52'$, which gives maximum intensities in the second order at about $5,000 \text{ \AA}^*$. This monochromator isolates a spectrum range of about 1 \AA which is then analyzed by the Fabry-Perot interferometer;

(b) The lenses and mirrors forming the converging system of the optical arrangement (fig. 2);

(c) The Fabry-Perot interferometer consists of two quartz plates separated by three quartz wedges of equal thickness accurate to $\lambda/20$. It is enclosed in a vacuum-tight container. Three fairly strong springs press on these wedges and so keep the plates parallel. The plates which are 70 mm in diameter are cut perpendicularly to the axis from crystalline quartz. The flatness is better than $\lambda/60$. Each of these plates is covered with 7 alternating layers of zinc sulphide (high refractive index, $\mu=2.35$) and cryolite (low refractive index, $\mu=1.35$), the optical thickness of each layer being $\lambda/4$. For a wavelength of $5,000 \text{ \AA}$ the reflection factor R is over 0.9 and the transmission T is of the order of 3 to 4%, which means that the background of the instrument is negligible [13].

From the flatness of the plates it follows that in the region of maximum transmission the limiting "finesse" is about 30 [14]. For the uranium-238 line $5,027.398 \text{ \AA}$, which has no hyperfine structure, we have in fact found a mean finesse of the same order.

At a very low scanning speed of the uranium spectrum, excited by a liquid nitrogen-cooled hollow cathode working at about 20 mA, the broadness of the lines can be measured. The recording was made with a very fine entrance slit of the monochromator and with a low time constant ($RC \ll 1s$), so as to avoid broadening of the lines owing to the instrument function. The width at half intensity of the U-238 line $5,027.389 \text{ \AA}$ is 39 mK. The width at half intensity of the U-235 line $5,027.295 \text{ \AA}$ is 54 mK. The free interval between two orders is 1,360 mK (fig. 3). The spectrum is explored by varying the pressure in the enclosure of the Fabry-Perot. First this enclosure is evacuated and then a leak is produced. The interferometer equation is given by:

$$n\lambda = 2 \mu e \cos i$$

in which n =order of interference.

When the air enters, the refractive index changes ($\Delta\mu=0.000293$ from 0 to 1 atm.) and gives:

$$\frac{\Delta\lambda}{\lambda} = \frac{\Delta\mu}{\mu}$$

In this way several orders can be scanned.

* Bausch and Lomb Optical Co.

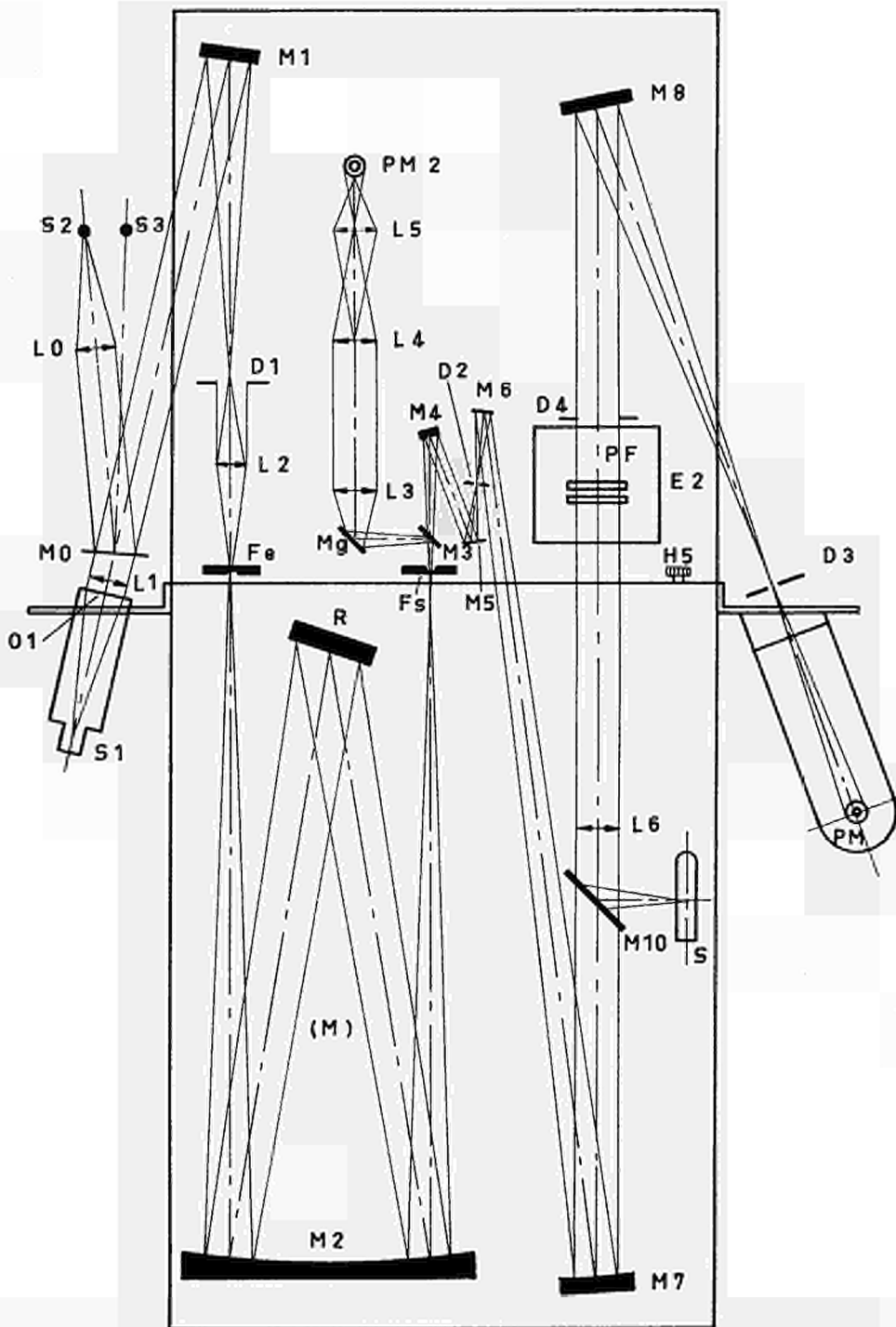


Fig. 2. — Optical arrangement of the Hyposac; S₁ hollow cathode source; S₂ mercury lamp; F, entrance slit and F, exit slit of the monochromator are conjugated with O₁ and PF; R grating with 1,200 grooves/mm; PF Perot-Fabry etalon enclosed in a pressure-tight box; S cadmium lamp; PM photomultiplier tube cooled in liquid nitrogen; PM2 auxiliary phototube.

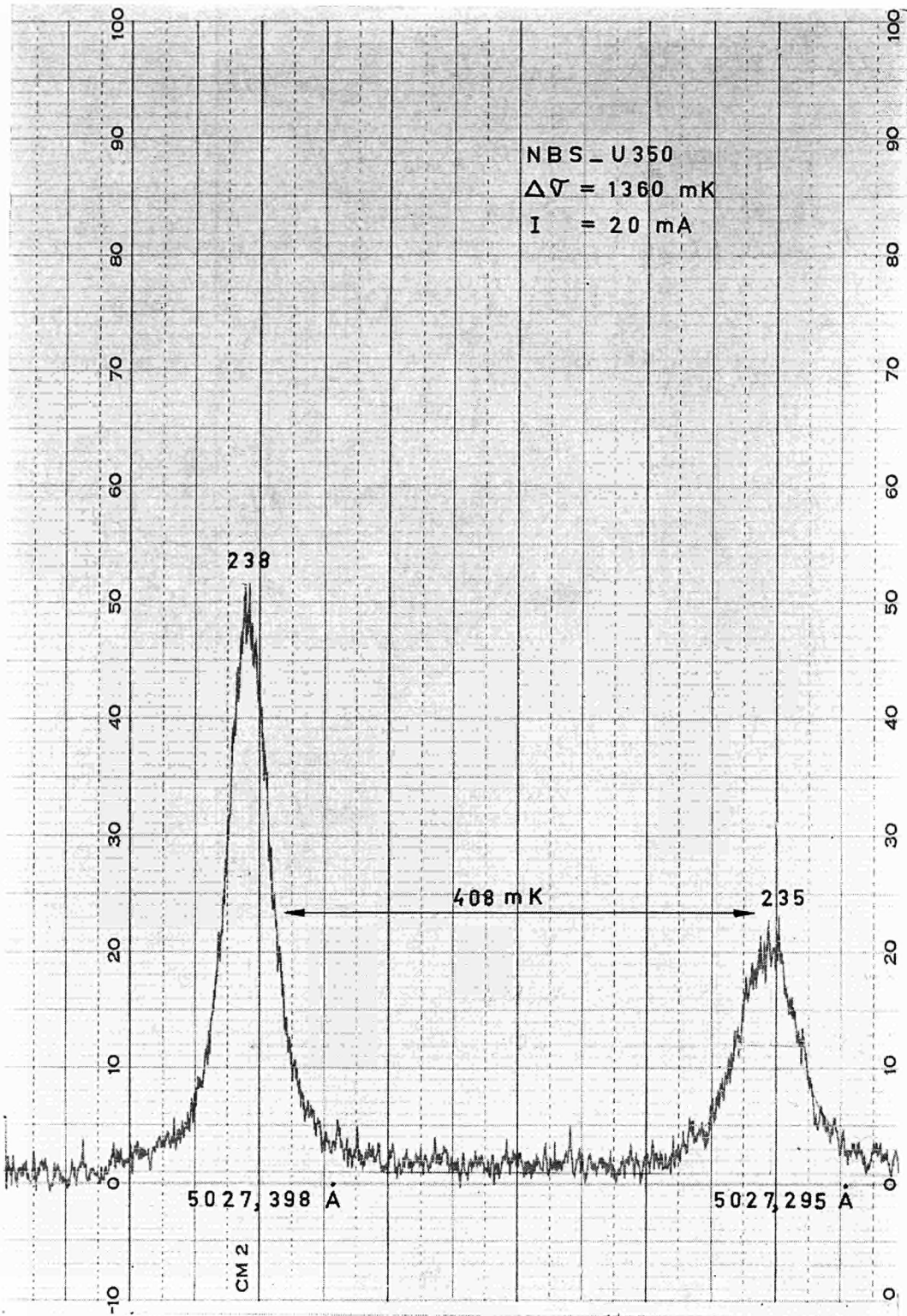


Fig. 3. — Typical pattern of spectra recorded with a very small time constant ($RC \ll 1s$) and low-speed pressure scanning. The width at half intensity is different for the two isotopes.

If the width $\Delta\nu$ of the lines were known, we could calculate the maximum diameter D of the diaphragm which allows only the central interference fringe to pass through. If the angle of incidence is small, we get:

$$\mu e i^2 = 2 \mu e - n\lambda.$$

For an angular diameter of the ring $\Omega = 2i$ and for a focal distance F of the deflecting mirror, we can write:

$$D = F\Omega = F \left(8 \cdot \frac{\Delta\nu}{\nu} \right)^{1/2}$$

If F is taken as 1,000 mm, the value of D for uranium line 5,027.398 Å is:

$$D_0 = 0.1(8 \times 0.039 \times 5,027.4)^{1/2} = 3.96 \text{ mm.}$$

3.3 — The electronic system (*Electronac*)

(a) The current generator for the hollow cathode:

open-circuit voltage: 800 V current: 0 to 250 mA.

(b) The amplifying and recording system. The current amplifier contains a series of resistances and capacitors with which the time constant of the recording system can be adjusted. A potentiometer recorder traces the isotope lines (speedomax type).

(c) The stabilized high-voltage supply for the photomultiplier (0 to 3,000 V).

The light transmitted by the Fabry-Perot falls on a photomultiplier RCA 1 P 21. In this way a much stronger signal is obtained than by photographic recording [15]. Only the light of the central ring of the interference fringes passes through a diaphragm into the photocathode, which is cooled in liquid nitrogen. This arrangement renders the thermoionic component of the dark current almost negligible. Measurements of the dark current at the output end of the photomultiplier have shown it to be about 6×10^{-13} A. The nine stages of the multiplier 1 P 21 have a maximum amplification of about 2×10^6 , which means that the dark current at the photocathode is less than 3×10^{-19} A. When the instrument is focused on the maximum of the uranium-238 line 5,027.398 Å and the hollow cathode is working at 30 mA and charged with 10 mg of U_3O_8 (enrichment 35% U-235), a 0.30 mm entrance slit of the monochromator gives a current of the order of 3×10^{-10} A at the output end of the photomultiplier tube (supply: 840 V).

4 — EXPERIMENTAL CONDITIONS

4.1 — Choice of homologous lines

The uranium spectrum, as excited in the hollow cathode, has been recorded with the aid of an auxiliary photomultiplier placed at the output end of the premonochromator. This auxiliary system can also be used for checking the stability of the hollow cathode. Slow rotation of the grating permits recording of the spectrum range between 4,500 and 5,500 Å. This corresponds to the transmission region of the Fabry-Perot, whose maximum reflecting power is at about 5,000 Å.

The most intense lines we found were: 5,027.40 Å; 5,280.40 Å; 5,481.22 Å. The line 5,481.22 Å has no hyperfine structure, but unfortunately it is just at the limit of the effective wavelength range of the Fabry-Perot. The isotopic shift for U-235 and U-238

of the line 5,280.39 Å is 273 mK but the line of the 235 isotope has a hyperfine structure which would entail precision losses when measuring the area of the line (fig. 4). We finally chose the line 5,027 Å, which has very little hyperfine structure [16], and the highest product of $B \cdot \Delta i$ (B =source brightness, Δi =isotope shift).

4.2 — Chemical state of the samples

The chemical state of the uranium has a marked influence on the intensity of the lines emitted from the hollow cathode and therefore also on the overall precision of the determination.

Uranium metal has the strongest emission, i.e. about 5-6 times that of oxides. It is also possible to obtain good results by evaporation of nitrate or acetate solutions in the hollow cathode. In this case the even and fine distribution of the residue seems to enhance the emitted intensity. For certain practical reasons we worked with uranium oxide and were nevertheless able to obtain quite satisfactory results (fig. 5).

Table I gives the experimental conditions used in the absolute determinations of the U-235/U-238 ratios.

TABLE I

Current intensity of the Al hollow cathode cooled by liquid nitrogen:	20 - 45 mA (*)
Spectre neon pressure:	0.5 - 1 mm Hg (**)
Weight of the sample (for 50 measurements):	5 mg
Homologous lines:	U-238 - 5,027.398 Å U-235 - 5,027.295 Å
Isotopic shift:	408 mK
Entrance slit of the monochromator:	300 μ
Grating of the monochromator (Ebert-Fastie):	1,200 lines/mm
Blaze angle:	37°
Fabry-Perot interferometer, effective range:	5,000 Å ± 400 Å
Free spectrum range between two orders:	1,360 mK
Photomultiplier cooled in liquid nitrogen:	1 P 21 RCA
Relative sensitivity of the photomultiplier:	70% at 5,000 Å
Stabilised high voltage of the photomultiplier:	620 - 840 V
Potentiometric recorder:	-0.2 to +2mV
Scanning time for two orders:	8 to 20 min.
Precision of the planimeter:	0.25%

Remarks: (*) When water-cooling only is used, the pressure should be about 5-10 mm Hg; the lines will then be enlarged because of a strong Doppler effect.

(**) The optimum pressure changes with the geometry of the hollow cathode, especially with the diameter of the hole in the Al base.

5 — RESULTS

For the absolute determination of the isotope ratio in uranium oxides two methods of evaluating the data were used:

- a) Measurement of the areas under the homologous lines after tracing the background line and correcting the feet of the spectrum lines;
- b) Measurement of the height of the homologous peaks.

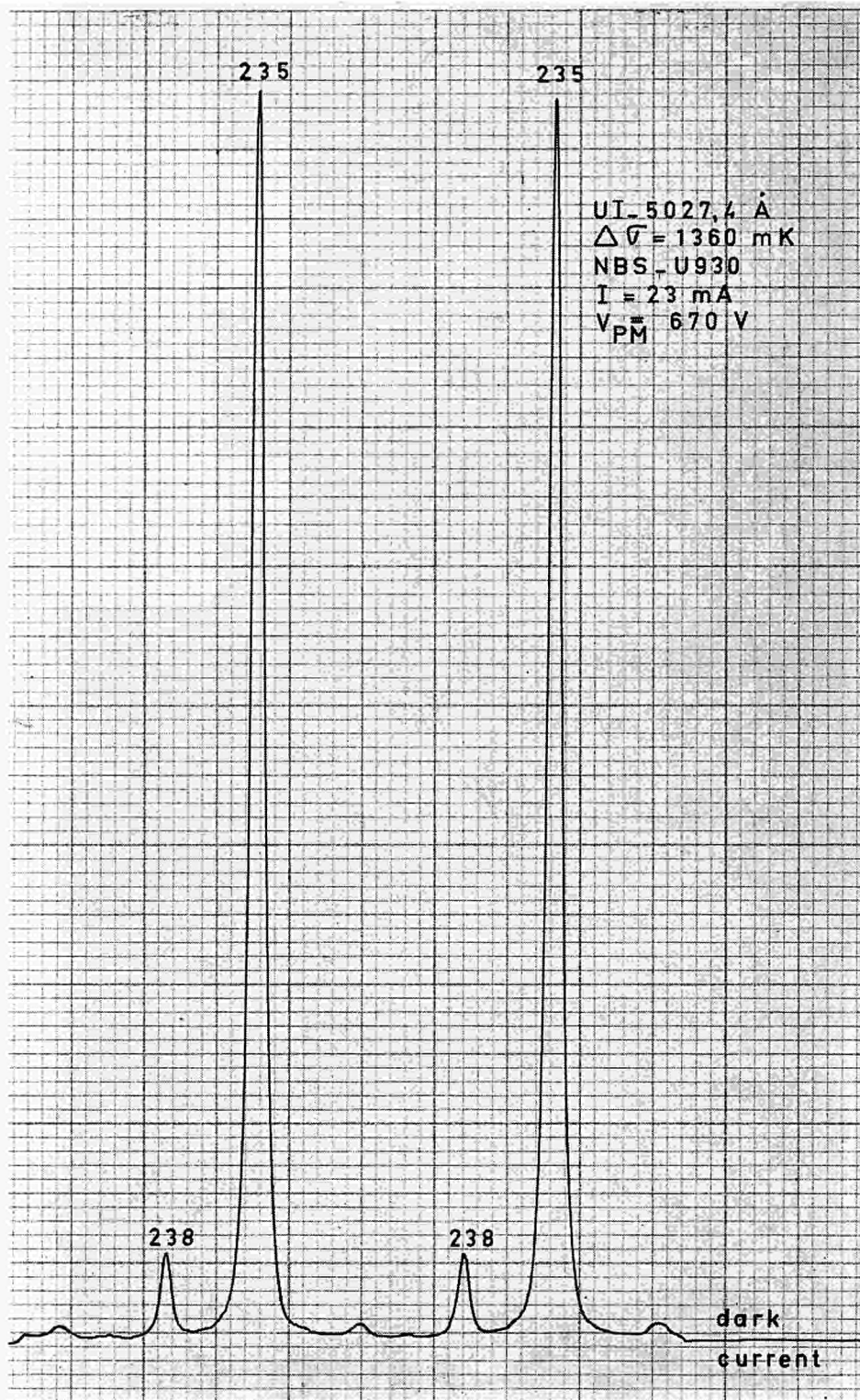


Fig. 5. — Recorder trace of UI - 5,027.4 Å in the hollow cathode, loaded with U_3O_8 containing 93.27 % of U-235 and 5.44 % of U-238.

In order to obtain an idea of the reliability of our method, we used the isotopic uranium standards of the National Bureau of Standards (Washington) as criteria for our measurements. The absolute and relative discrepancies with respect to these criteria are listed in the attached tables. If there is no self-absorption, the intensity ratio of the homologous lines $r = \frac{I - 5,027.398 \text{ \AA}}{I - 5,027.295 \text{ \AA}}$ is a direct measure of the ratio of the number of U-235/U-238 atoms that have been excited in the hollow cathode. This ratio is an accurate indication of the real isotope ratio since the excitation energies and the transition probabilities are the same for both the lines used. The same logic applies to the partition functions if the hyperfine levels are ignored.

The weight concentration of the 235 isotope is given by the following equation:

$$C = \frac{(235 + 238)\%}{\frac{238}{235}r + 1}$$

in which the percentage (U-235 + U-238)% is less than 100% because our samples contain both U-234 and U-236 at different concentrations. Each spectrum is measured 5 to 10 times with the planimeter, so that each single value is the mean of 5 to 10 area measurements.

In order to compare the experimental values with the certified values, we corrected our results by using the U-234 and U-236 concentrations given by the NBS. Table II gives the isotopic concentrations of the NBS standards as measured with the Hypeac by determining the areas under the homologous lines.

TABLE II

N°	NBS Standard (235+238)% by weight	Mass. conc.	Determ. with Hypeac	Difference absolute	relative %	Number of determinations
U 030	99.96	3.01 (235)	3.02 ₃	+0.013	0.43	41
		96.95 (238)	96.93 ₇	-0.013	0.013	
U 050	99.93	4.95 (235)	4.94	-0.01	0.20	21
		94.98 (238)	94.99	+0.01	0.01	
U 100	99.89	10.07 (235)	10.00 ₂	-0.068	0.67	30
		89.82 (238)	89.88 ₈	+0.068	0.075	
U 150	99.84	15.13 (235)	25.10 ₇	-0.023	0.15	27
		84.71 (238)	84.73 ₃	+0.023	0.027	
U 200	99.66	19.80 (235)	19.79 ₄	-0.006	0.03	28
		79.86 (238)	79.86 ₆	+0.006	0.007	
U 350	99.58	34.89 (235)	35.07 ₅	+0.185	0.53	30
		64.69 (238)	64.50 ₅	-0.185	0.28	
U 750	99.15	75.12 (235)	75.13 ₈	+0.018	0.024	35
		24.03 (238)	24.01 ₂	-0.018	0.075	
U 800	99.09	80.07 (235)	80.02	-0.05	0.062	24
		19.02 (238)	19.07	+0.05	0.26	
U 850	98.99	84.99 (235)	84.89 ₂	-0.098	0.11	45
		14.00 (238)	14.09 ₃	+0.098	0.70	
U 900	98.90	90.10 (235)	90.10 ₁	+0.004	0.004	37
		8.80 (238)	8.79 ₆	-0.004	0.045	
U 930	98.71	93.27 (235)	93.24 ₅	-0.021	0.022	23
		5.44 (238)	5.46 ₁	+0.021	0.38	

Remarks: The relative deviation from the certified values is always less than 0.70%.

The precision of the measurements for each standard is given with a confidence level of 99%,

using the relation $\frac{2.6\sigma}{\sqrt{n}}$ in which σ is the standard deviation.

Certain anomalous values are eliminated by using Chauvenet's criterion:

$$\frac{2h}{\sqrt{\pi}} \int_0^{\infty} e^{-h^2c^2} dc = \frac{1}{2n}$$

with $h = 1/\sigma \sqrt{2}$

The above calculations were performed on an electronic computer (Ferranti, type Mercury).

Table III shows the type of print-out that we obtain for a series of measurements.

TABLE III

URANIUM ISOTOPE ANALYSIS BY OPTICAL METHOD (HYPEAC)				
NBS U-930				
U-235 ± U-238 = 98.71 weight %				
3	238	235	0	
REEKS 1				
<i>Mol-procenten</i>				
1	5.3340	93.3760	1.2900	
2	5.4480	93.2620	1.2900	
3	5.4520	93.2580	1.2900	
4	5.4400	93.2700	1.2900	
5	5.4020	93.3080	1.2900	
6	5.4610	93.2490	1.2900	
7	5.4300	93.2800	1.2900	
8	5.4140	93.2960	1.2900	
9	5.5930	93.1170	1.2900	
<i>Verworpen metingen</i>				
<i>Nr.</i>	<i>Meting</i>	<i>MU</i>	<i>Afwijking</i>	<i>Toegelaten afw.</i>
238	9	5.4416	2.21222, 0	1.92000, 0
238	1	5.4226	-2.16870, 0	1.87000, 0
<i>Er zijn 7 aangenomen metingen</i>				
<i>Nr.</i>	<i>MU</i>	<i>S</i>	<i>S'</i>	<i>PR</i>
238	5.4353	2.12659,-2	8.03775,-3	2.0697,-2
235	93.2747	2.12659,-2	8.03776,-3	2.0697,-2
0	1.2900	4.02377,-9	1.52084,-9	3.9162,-9
<i>Maximale afwijkingen</i>				
238	5	-3.32858,-2	-6.12401,-3	
235	5	3.32849,-2	3.56848,-4	
0	2	3.72529,-9	2.88782,-9	

REEKS 2*Mol-procenten*

1	5.6290	93.0810	1.2900
2	5.4060	93.3040	1.2900
3	5.3840	93.3260	1.2900
4	5.4090	93.3010	1.2900
5	5.4630	93.2470	1.2900
6	5.4880	93.2220	1.2900
7	5.4500	93.2600	1.2900
8	5.4090	93.3010	1.2900
9	5.4270	93.2830	1.2900

Verworpen metingen

Nr.	Meting	MU	Afwijking	Toegelaten afw.
238	1	5.4517	2.39629, 0	1.92000, 0

Er zijn 8 aangenomen metingen

Nr.	MU	S	S'	PR
238	5.4295	3.47111,-2	1.22722,-2	3.1601,-2
235	93.2805	3.47111,-2	1.22722,-2	3.1601,-2
0	1.2900	3.98250,-9	1.40803,-9	3.6257,-9

Maximale afwijkingen

Nr.	Meting	Max. afwijking	Rel. afw.
238	6	5.85000,-2	1.07745,-2
235	6	-5.85001,-2	-6.27141,-4
0	2	-3.72529,-9	-2.88782,-9

REEKS 3*Mol-procenten*

1	5.4520	93.2580	1.2900
2	5.4170	93.2930	1.2900
3	5.4660	93.2440	1.2900
4	5.6820	93.0280	1.2900
5	5.4660	93.2440	1.2900
6	5.3870	93.3230	1.2900
7	5.5840	93.1260	1.2900
8	5.6620	93.0480	1.2900

Er zijn 8 aangenomen metingen

Nr.	MU	S	S'	PR
238	5.5145	1.12768,-1	3.98694,-2	1.0266,-1
235	93.1955	1.12768,-1	3.98694,-2	1.0266,-1
0	1.2900	1.43591,-8	5.07671,-9	1.3073,-8

Maximale afwijkingen

Nr.	Meting	Max. afwijking	Rel. afw.
238	4	1.67500,-1	3.03745,-2
235	4	-1.67501,-1	-1.79730,-3
0	4	-1.86265,-8	-1.44391,-8

Vergelijking van reeksen

Nr.	MU	S	S'	PR
238	5.4608	7.85130,-2	1.63711,-2	4.21556,-2
235	93.2492	7.78404,-2	1.62308,-2	4.17944,-2
0	1.2900	7.36112,-5	1.53490,-5	3.95236,-5

Symbols used for the results given by the electronic computer.

A — For each series:

MU : mean value $\mu = \sum_i x_i/n$

S : Standard deviation $\left\{ \sum_i (x_i - \mu)^2 / n - 1 \right\}^{1/2}$

S' : $\frac{S}{\sqrt{n}}$

PR : 2.575 S' (99% confidence level)

B — For a group of series:

MU : mean of the mean values $\mu = \sum_i \mu_i p_i n_i / \sum_i p_i n_i$

μ_i = mean value of the i^{th} series

p_i = weight of this series $\left(\frac{1}{S^2} \right)$

n = number of measurements of the series

S : standard deviation

S' : dispersion of the mean value

PR : uncertainty on the mean value at 99% limit.

In table IV the precision of our measurements calculated as above is compared with that of the measurements given by the NBS.

TABLE IV

No	NBS Standards	Hypac Measurements	$2.6 \sigma / \sqrt{n}$ (prob. 99%)	Relative precision %	Number of determinations	Nb. of determinations rejected *
U 030	3.01 ± 0.015	3.02 ₃	$1.08 \cdot 10^{-2}$	0.38	41	0
	96.95 ± 0.0485	96.93 ₇		0.01		
U 050	4.95 ± 0.025	4.94	$3.95 \cdot 10^{-2}$	0.80	21	0
	94.98 ± 0.0475	94.99		0.04		
U 100	10.07 ± 0.05	10.00 ₂	$8.35 \cdot 10^{-2}$	0.83	32	2
	89.82 ± 0.054	89.88 ₈		0.09		
U 150	15.13 ± 0.075	15.10 ₇	$9.26 \cdot 10^{-2}$	0.61	32	5
	84.71 ± 0.085	84.73 ₃		0.11		
U 200	19.80 ± 0.099	19.79 ₄	$1.13 \cdot 10^{-1}$	0.57	29	1
	79.86 ± 0.16	79.86 ₆		0.14		
U 350	34.89 ± 0.174	35.07 ₅	$1.67 \cdot 10^{-1}$	0.45	30	0
	64.69 ± 0.129	64.50 ₅		0.26		
U 750	75.12 ± 0.15	75.13 ₈	$2.47 \cdot 10^{-1}$	0.33	40	5
	24.03 ± 0.12	24.01 ₂		1.03		
U 800	80.07 ± 0.080	80.02	$1.93 \cdot 10^{-1}$	0.24	24	0
	19.02 ± 0.095	19.07		1.01		
U 850	84.99 ± 0.085	84.89 ₂	$1.39 \cdot 10^{-1}$	0.16	47	2
	14.00 ± 0.070	14.09 ₈		0.98		
U 900	90.10 ± 0.09	90.10 ₄	$8.36 \cdot 10^{-2}$	0.093	37	0
	8.80 ± 0.044	8.79 ₆		0.95		
U 930	93.27 ± 0.0466	93.24 ₆	$4.19 \cdot 10^{-2}$	0.045	26	3
	5.44 ± 0.027	5.46 ₁		0.77		

* Rejected values on the basis of Chauvenet's criterion.

It can be seen that the relative precision varies from 0.01% to 1.03%, depending on the isotopic concentration. Under the actual experimental conditions we can therefore obtain a precision of 1% for the absolute determination in the range from 3 to 93% U-235. By carrying out at least 100 measurements and using several charges of hollow cathodes it will be possible to increase this precision. The determination of the area of the peaks with the planimeter is extremely tedious, and consequently we were unable to make these required additional determinations. We are planning to use an electronic integrator instead of the recorder and thus to speed up the measurements.

As it is much easier to measure the height of a peak, we attempted to use this method; the results are given in table V.

TABLE V
EVALUATION BY HEIGHT RATIO

NBS Standard	Isotopes	Determination with the Hypeac	Difference absolute	relative %	Number of determinations
U 030	235	3.03 ± 0.05	+0.02	0.67	25
U 050	235	5.00 ± 0.11	+0.06	1.2	21-5 = 16
U 100	235	10.06 ± 0.10	-0.01	0.1	39
U 150	235	15.02 ± 0.14	-0.11	0.73	45
U 200	235	29.87 ± 0.20	+0.07	0.35	33
U 350	235	34.81 ± 0.18	-0.08	0.23	62
U 750	238	27.20 ± 0.20	+3.17	13.2	54
U 800	238	22.46 ± 0.22	+3.44	18.1	20
U 850	238	17.36 ± 0.19	+3.36	24	17
U 900	238	11.61 ± 0.11	+2.81	31.9	11
U 930	238	8.17 ± 0.14	+2.73	50	23

It should be noted that the higher the U-235 content the greater is the difference between the experimental and the certified values; this is clearly apparent from fig. 6.

For U-235 concentrations of less than 35%, the absolute analysis is possible with about the same accuracy as is obtained with the planimeter method. The precision remains roughly the same over the whole range.

6 — POSSIBILITIES OF DETERMINING THE U-234 AND U-236 ISOTOPES BY AN OPTICAL METHOD

Contrary to the situation with mass-spectrometric measurements, in the case of emission-spectrometric measurements the determination of these two isotopes presents certain problems. One of the difficulties is their normally low concentration in samples of enriched U-235. R.J. Murphy [17] and G.V. Wheeler [18] were able to photometer the 4,244 Å lines for U-236 concentrations above 2%. D.D. Smith [19] gives the wavelength of the uranium isotopes as follows:

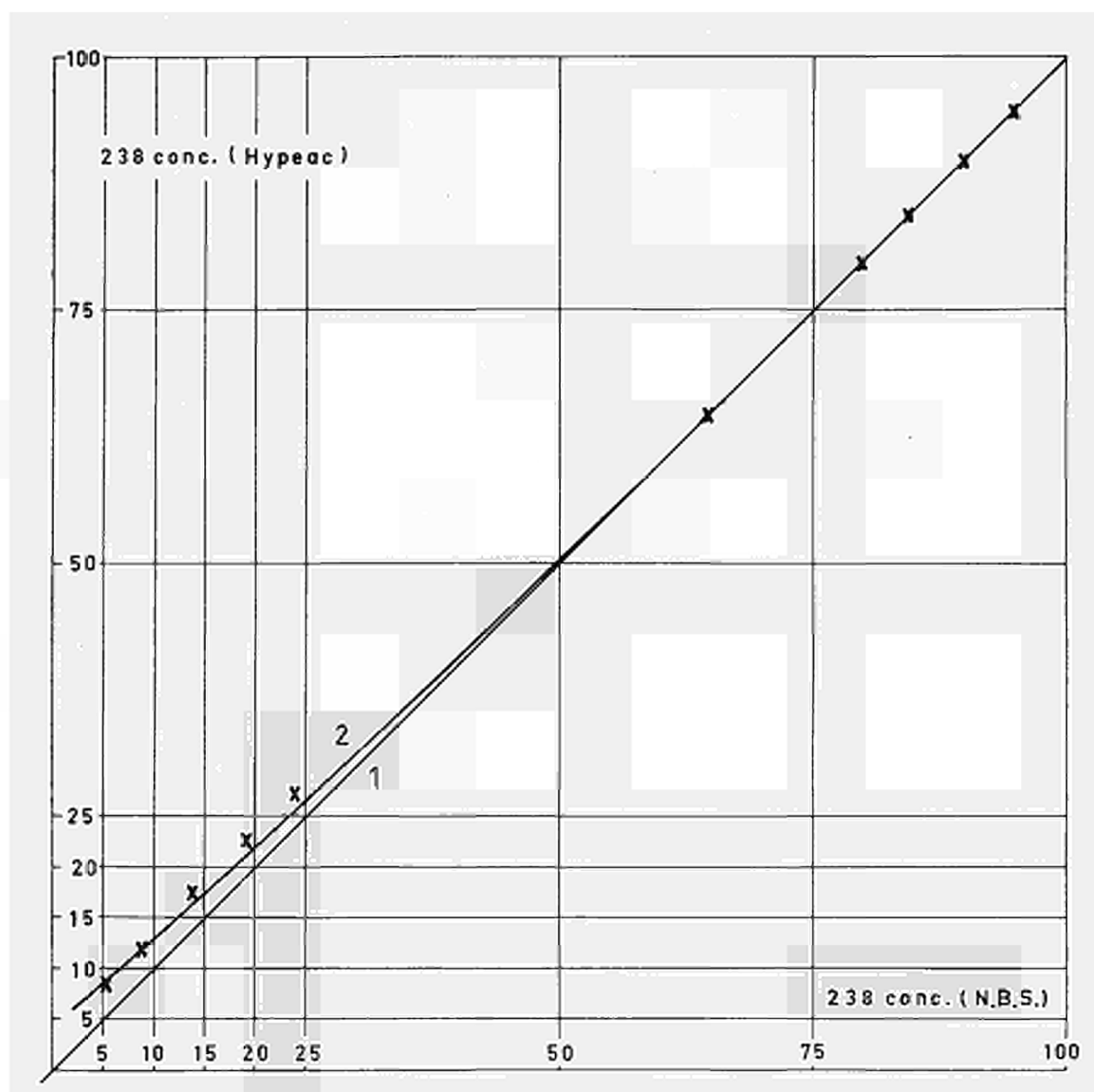


Fig. 6. — Systematic deviation between: 1) planimetric evaluation, and 2) evaluation by height ratio.

U-238	4,244.373 Å
U-236	4,244.226 Å
U-235	4,244.122 Å
U-234	4,244.075 Å
U-233	4,243.977 Å

E.W. Richards [20] has also given a list of lines that could be used for the analysis of U-236. J.L. Saunderson, E. Dubois, A.O. Malley and P. Flynn were able to determine contents of the order of 1 % with an accuracy of within a few percent [21].

Using a Fabry-Perot spectrometer with a resolving power of 10^6 it should be possible to resolve the lines for the 234, 235, 236 and 238 isotopes.

7 — CONCLUSION

The actual precision of the determination of the U-235/U-238 ratio by absolute measurement of the areas under the resolved lines is 1% or better. The time needed for such an analysis is 5-6 hours. By mechanizing the evaluation of the spectra it should be possible to increase the number of determinations made on a single sample and consequently to approach the precision obtainable in mass spectrometry. Within the precision cited our absolute values agree with those given by the NBS.

In all probability this method can also be used for lower U-235 contents after accurate calibration of the attenuation factors of the instrument. By using other lines it might be possible to extend the analysis to the U-236 isotopes and perhaps to U-234 and U-233.

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ADDENDUM

Experiments with an electronic integrator connected directly after the first amplification stage of the HYPEAC have been started. The Meci recorder and the automatic electronic integrator Perkin Elmer are connected in parallel in such a way that the trace of the spectrum lines on the recorder and the digital values of their surface are obtained simultaneously.

The electronic integrator Perkin Elmer D2 has a time constant of about 10 ms and therefore the resolving power of this unit is very high. It also contains an automatic range changer which can decrease the sensibility by the factors 1 : 10 : 100. The integrated values are printed out by a digital printer giving 7 digits (Kienzle). The correction for changing ranges is automatic.

In the following table the first results with enriched uranium are given, the time for recording these values was about one hour.

ISOTOPE ANALYSIS OF URANIUM WITH THE HYPEAC AND THE AUTOMATIC
ELECTRONIC INTEGRATOR

NBS U 200

U-235 + U-238 = 99.66 at. %

No.	Integrated Values		Concentration	
	5,027.295 Å U-235	5,027.398 Å U-238	atom %	
			U-235	U-238
1	235,208	935,503	20.02 ₄	79.63 ₆
2	157,633	635,115	19.81 ₇	79.84 ₃
3	150,546	618,182	19.51 ₈	80.14 ₂
4	158,383	653,521	19.44 ₂	80.21 ₈
5	327,805	1,302,637	20.04 ₀	79.62 ₀
6	339,607	1,336,394	20.19 ₄	79.46 ₆
7	342,974	1,334,244	20.38 ₀	79.28 ₀
8	334,918	1,339,870	19.93 ₀	79.73 ₀
9	330,817	1,327,723	19.88 ₀	79.78 ₀
10	339,728	1,380,158	19.68 ₈	79.97 ₂
11	301,578	1,183,221	20.26 ₄	79.39 ₆
12	299,301	1,233,890	19.45 ₆	80.20 ₄
Mean Values			19.88 ₆	79.77 ₄
NBS Values			20.00	79.66
Absolute Difference			0.11 ₄	
Relative Difference			0.57 %	
Standard Deviation			= 0.31 ₅	
Standard Deviation of the mean value (99 % confidence level)			0.27 ₈	

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