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LAWRENCE RADIATION LABORATORY
UNIVERSITY of CALIFORNIA BERKELEY

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THE NEW ELEMENT HAHNIUM, ATOMIC NUMBER 105*

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ABSTRACT

An isotope of element 105 with mass number 260 has been formed by bombarding ^{249}Cf with ^{15}N ions. The Z and A of the nuclide have been unambiguously identified by recoil-milking the 30-second ^{256}Lr daughter. $^{260}105$ has a 1.6±0.3 second half-life and decays by alpha-particle emission with groups at 9.06 (55%), 9.10 (25%), and 9.14 MeV (20%). Branching decay by spontaneous fission is less than 20%. Some comments are made on the earlier results of the Dubna group concerning a few alpha particle events at 9.4 and 9.7 MeV which they claim to be due to element 105.

Using the same target of ^{249}Cf that was instrumental in the discovery of the alpha-emitting isotopes of element 104,[1] rutherfordium,[2] we have produced with moderate yield a 1.6-second 9.1-MeV alpha-particle activity by bombardment with ^{15}N ions at the HILAC. By the alpha-recoil milking of a known isotope of element 103 we have obtained evidence that assigns this new radioactivity unambiguously to an isotope of element 105.

The procedures used in these new experiments were similar to those described in our previous communications.[1,2,3] The 300 microgram/cm^2 target of ^{249}Cf, which had been prepared by the molecular-plating method in

October 1968, is still in excellent shape and appears to be unchanged by several thousand microamperehours of heavy-ion bombardment.

The element-105 reaction recoils were ejected from the target a short distance into helium gas at 620 torr. They were pumped through a small orifice into a rough vacuum to impinge upon the periphery of a vertically mounted wheel which acted as a carrier. The wheel was periodically rotated to place the collected transmutation products next to a series of solid-state Si-Au surface-barrier detectors in order to measure their alpha-particle spectra. Half-life information was derived both from the relative numbers of counts detected at each station and from the decay of the activities while the wheel was stationary at each position.

To measure the alpha-recoil daughters of these activities each detecting crystal facing the wheel was periodically shuttled to a position opposite another similar detector where at high geometry the two detectors together could analyze the daughter alpha-particle activity which had recoiled off the wheel into the crystal. At each detecting station there were four detectors, two "mother" crystals which alternately faced the wheel and two "daughter" crystals to alternately face the "mother" crystals when they were shuttled off the wheel. Five stations were arranged at 39° intervals so that the same position on the 45-cm diameter wheel would not be re-examined by the detectors until all steps of the digital motor had been used.

The information from each of the many detectors was amplified by modular units developed in our laboratory and processed and stored by a PDP-9 computer and ancillary devices. Alpha-particle spectra were analyzed in 512-channel segments covering the range from 6 to 12 MeV with spontaneous-fission discriminators set to detect pulses greater than 30 MeV. The spurious count level was essentially eliminated by the use of judicious shielding and electronic-gating techniques.

The bombardments were made at a beam level of 4 microamperes measured as $^{15}N^{+7}$ through the 4.7-mm diameter target. The 156-MeV beam energy of the HILAC was reduced to the proper energy, usually about 85 MeV, by the use of beryllium degraders mounted very close to the target. This energy was measured by detecting the particles scattered at 30^o from the target through a thin window in the target chamber.

The alpha-particle spectra displayed in Fig. 1 resulted from a series of bombardments of the ^{249}Cf target with ^{15}N ions. The individual spectra show the total of counts recorded at each of the five stations by the two movable detectors when facing the wheel. The sum of the five spectra is plotted topmost. The wheel-cycle rate was one second and the shuttle period 50 seconds.

The complex group of peaks above 9 MeV is assigned to $^{260}105$; by use of the SAMPO computer program[4] it can be resolved into alpha-particle groups at 9.06 (55%), 9.10 (25%), and 9.14 MeV (20%). For alpha-energy calibration the 6.773-MeV peak of ^{213}Fr and the 7.443-MeV peak of ^{211}Po were used. The absolute accuracy of the energy values is estimated to be 0.02 MeV. Calculations based on spin-independent (ℓ=o) equations of Preston[5] give hindrance factors 7, 20, and 33, respectively, for these transitions. The half-life of this activity is 1.6±0.3 seconds. The branching by spontaneous fission is less than 20%, or alternatively, assuming that ^{260}Rf is a very short-lived fission emitter, the electron-capture branching is less than 20%.

The 8.87-MeV peak as well as its 8.81-MeV satellite belong to 0.7-sec ^{257}Lr and the complex peak at 8.6-MeV belongs to 4.0-sec ^{258}Lr.[6,7,8] Several of the peaks with a lower alpha energy are present because of lead and mercury impurities in the target. Bombardment of lead and mercury targets with ^{15}N ions insured that the new activity was not produced by these impurities. Measurements were also made of the extent to which atoms carried by the gas jetting out of the orifice could be deposited directly on the crystal faces of the detecting stations. The abundantly produced ^{214}Ra was used as a tracer and it was found that stopping the wheel reduced the observed amount of activity by more than a factor of 10^5.

The measured relative cross sections for the 9.1, 8.87, and 8.6-MeV alpha activities at four different bombarding energies are plotted in Fig. 2. The peak production rate of the 9.1-MeV activity is about 1.5 alpha counts per microamperehour which corresponds to a cross section of $3 \cdot 10^{-33}$ cm^2 assuming a recoil-collection yield of 50%. Both the absolute cross-section value and the shape of the excitation curve agree with the predicted ones for the $^{249}Cf(^{15}N,4n)^{260}105$ reaction.[9]

We have recently discovered certain isomeric transitions in the heavy-element region that are able to transfer their ground-state daughters from the wheel to the mother detectors. This is accomplished by the feeble recoil energy imparted to them by the photons or electrons emitted in such transitions. We have also detected this transfer in some cases of electron-capture. We felt that it was important to prove that the recoil transfer involved in this experiment was due to the much greater energy (some 10^5 times) imparted by the emission of an alpha particle. We found that we could reduce the I.T.-recoil by a factor of more than 10 by biassing the wheel negative a few volts relative to the adjacent detector faces and by adding a modest gas pressure (ca 10 torr) of argon in this region. The mother-daughter experiments were conducted in this fashion. In addition, one experiment was conducted in which the potential was reversed and the gas removed to enhance the possibility of detecting a 1.6-second I.T. in ^{256}Lr and none was found.

The alpha-particle spectra shown in Fig. 3 were recorded simultaneously with those displayed in Fig. 1, but by the detectors in the off-wheel position, i.e., they arose from the decay of alpha-recoil-daughter atoms embedded in the movable detectors. We believe that the alpha-particle events with an energy of 8.2 to 8.6 MeV belong to the daughter of the 1.6-sec 9.1-MeV activity for the following reasons: (1) the number of recorded events at successive detector stations diminishes with a half-life of 2 ± 1 seconds; and (2) the ratio of counts in the 9.1 MeV peak in the mother spectrum to those between 8.2 to 8.6 MeV in the daughter spectrum is 234:84 = 2.8 ± 0.4 and agrees well with the calculated value 2.7. The 8.4-MeV daughter activity decays with a half-life of 30 ± 10 seconds, which value is based on the distribution of counts in the four 12.5-sec time subgroups of the 50-second shuttle period. In the inset above the sum spectrum in Fig. 3 there is shown an alpha spectrum of 30-sec ^{256}Lr produced by the ^{249}Cf(^{11}B,4n)^{256}Lr reaction[6]. Because of the similarity of the sum spectrum with the spectrum in the inset, and the good agreement of the half-lives, the daughter activity is assigned to ^{256}Lr and therefore the 9.1-MeV mother activity has to be ^{260}105. Our knowledge of the decay properties of ^{255}Lr is not detailed enough to rule it out definitively as a daughter possibility, but its alpha-spectrum in known to differ from that of ^{256}Lr to the extent that it is unlikely to produce the spectrum in Fig. 3. Another argument against the mother-daughter pair being $^{259}105 \longrightarrow {}^{255}Lr$ is that we did not observe the 9.1-MeV activity in a 36-μAhr bombardment of ^{249}Cf with ^{14}N ions.

We have also made time-correlation measurements to show that the 30-second 8.4-MeV alpha particles follow the emission of the 1.6-second 9.1-MeV mother activity on the wheel. Preliminary results do indeed confirm this assumption but, unfortunately, a relatively high background from 3.2-second 8.4-MeV ^{256}No interferes. Further measurements are being made in which this interference is substantially reduced by a more elaborate mode of operation.

In 1968 there was published a paper by G. N. Flerov et al.,[10,11] which purported to show the discovery of two alpha-emitting isotopes of element 105 produced by the bombardment of ^{243}Am with ^{22}Ne ions. The transmutation products were carried by a gas stream through an annular solid-state detector to a collecting surface. In the gross spectrum they observed peaks with energies of 8.3, 8.7, 9.0, and 11.6 MeV which were ascribed to known reaction products from lead and americium. Delayed coincidences were observed between alpha-particle pulses of 8.8 to 10.3 MeV with those from 8.35 to 8.6 MeV, a region which is occupied by ^{256}Lr and, supposedly, ^{257}Lr. In particular they seemed to find a statistically meaningful correlation for "peaks" at 9.4 and 9.7 MeV. They came to a preliminary conclusion that they might be detecting $^{261}105$ with $E_\alpha = 9.4 \pm 0.1$ MeV; $0.1 < T\text{-}1/2 < 3$ seconds and $^{260}105$ with $E_\alpha = 9.7 \pm 0.1$ MeV; $T\text{-}1/2 > 0.01$ seconds.

The rate of production of these events was extremely low; only 10 delayed coincidences were observed in 400 microamperehours. We have shown in Fig. 4 a compilation of their data on the coincident events arranged according to their energy range. Their gross alpha spectrum is also shown and for comparison we have plotted the high energy part of some of our data on the same energy scale. There appears to be a similar continuum above 9.2 MeV in both cases but they are not necessarily due to the same effect. In our experiments this high energy tail for the most part is due to one or more very light nuclides produced by the interaction of the ^{15}N ions with the Be substrate of our target. We have searched for delayed coincidences between these high energy alpha particles and the various lawrencium peaks. We found none that were statistically significant.

In addition to these negative findings it is unlikely that the 9.4 and 9.7 MeV "groups" can be due to $^{260}105$ or $^{261}105$ for the following reasons: (1) our present work shows that $^{260}105$ has an energy of ca 9.1 MeV and (2) the daughter of $^{261}105$, ^{257}Lr, was excluded from the Dubna delayed-coincidence measurements because its energy and half-life are not the same as ^{256}Lr, as assumed. In view of these considerations it is difficult for us to ascribe any significance to the meager data of the Dubna group regarding alpha-emitting isotopes of element 105.

In honor of the late Otto Hahn we respectfully suggest that this new element be given the name hahnium with the symbol Ha.

In a complicated research effort such as this we obviously are indebted to many people but in particular we would like to express our gratitude for the continued essential and patient assistance provided by R. G. Leres, A. A. Wydler, C. A. Corum, A. E. Larsh, and D. F. Lebeck. The experiments were made possible by the excellent performance of the accelerator and for this we must thank F. S. Grobelch and the HILAC operating and maintenance staffs. As always we appreciate the interest and suggestions of G. T. Seaborg.

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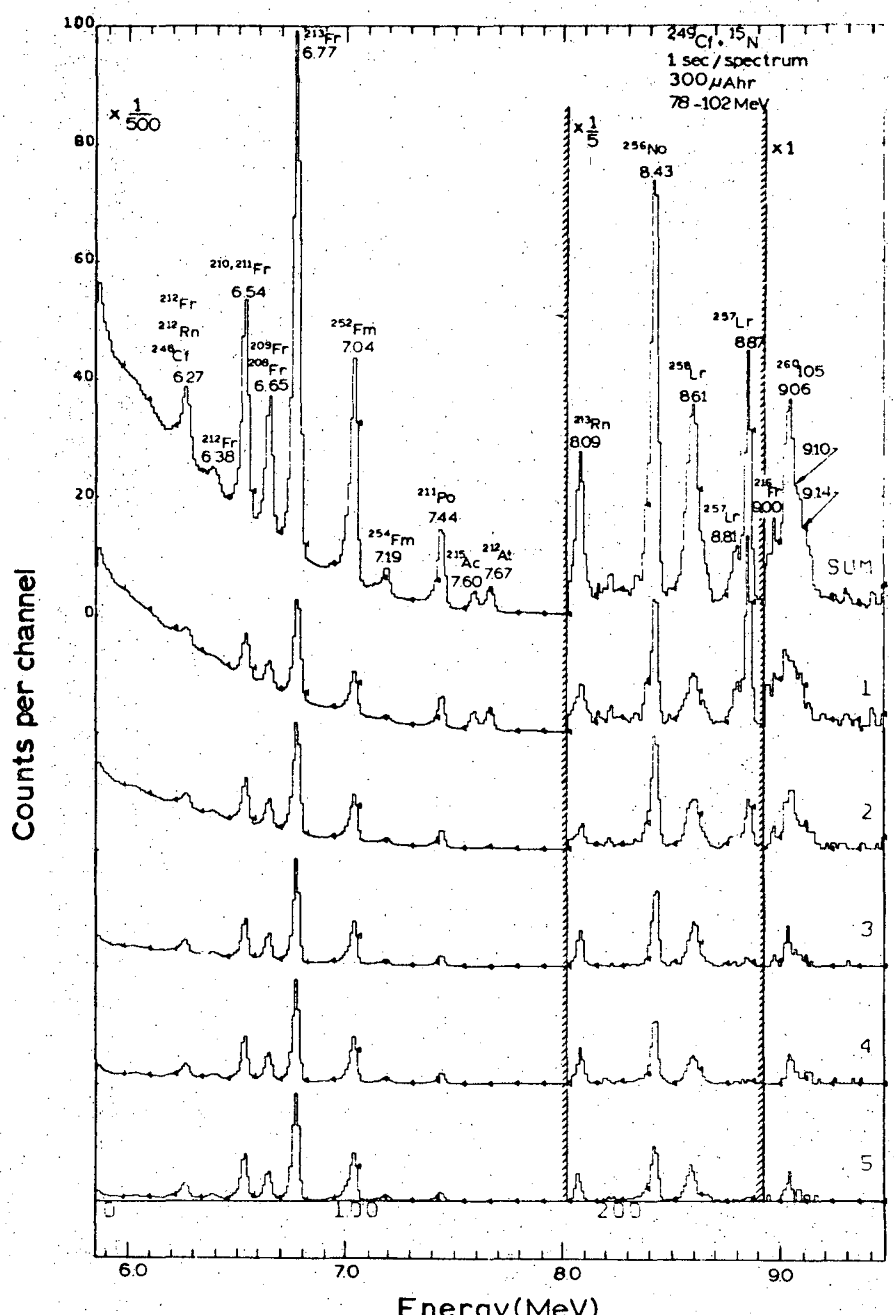
FIGURE CAPTIONS

Fig. 1. A series of alpha-particle spectra produced by bombardments of ^{249}Cf with ^{15}N ions. The alpha peak at 9.00 MeV is that of ^{216}Fr, the daughter of 23-msec ^{220}Ac produced from the lead impurity in the target. Most of the ^{211}Po derives from electron-capture of 7.2-hr ^{211}At.

Fig. 2. Excitation curves for Lr and element-105 activities produced in bombardments of ^{249}Cf with ^{15}N ions.

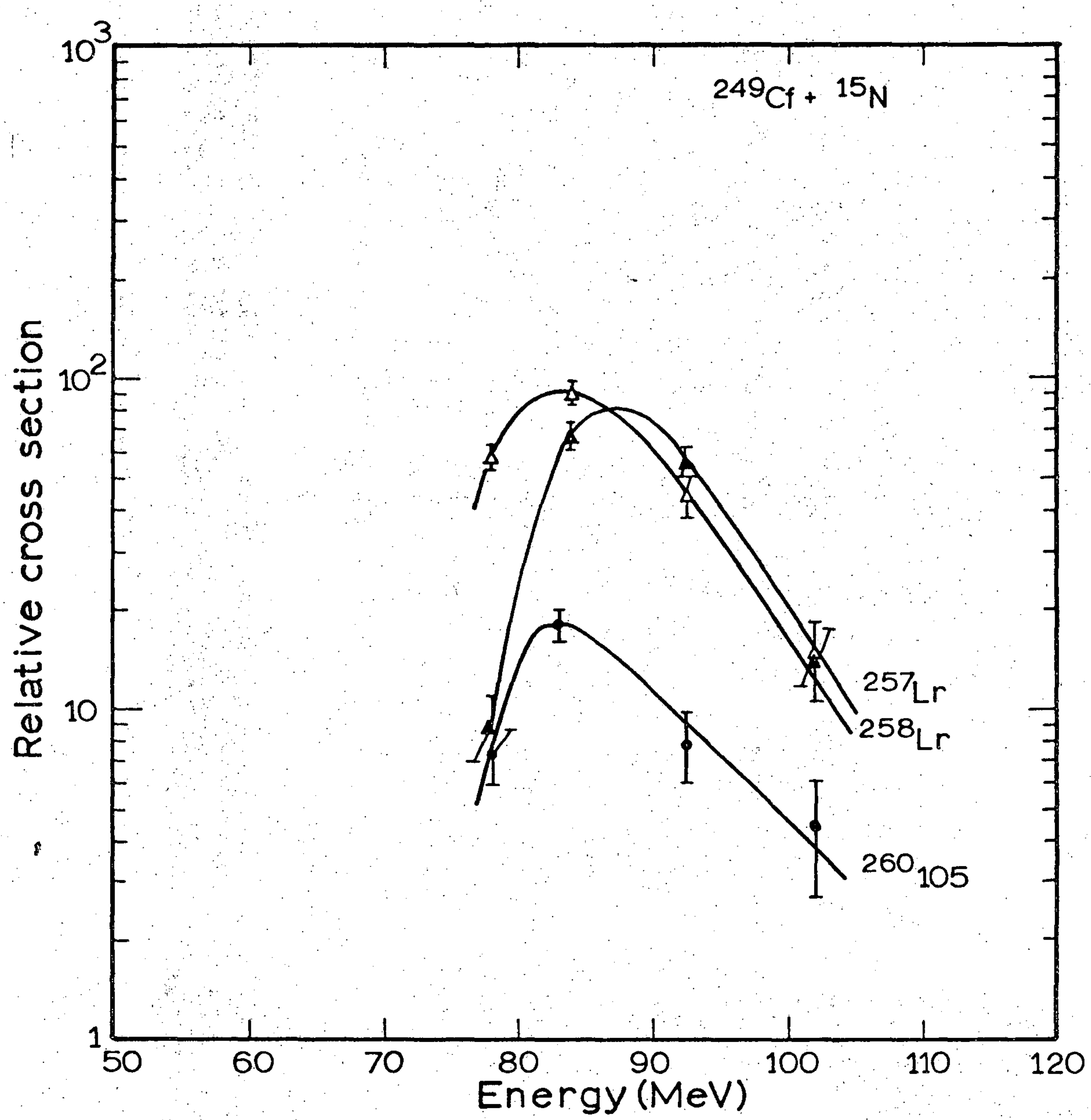
Fig. 3. A series of alpha-particle spectra from the same bombardment as those in Fig. 1, but recorded by the detectors in the off-wheel position. The spectrum in the inset is that of ^{256}Lr produced by the $^{249}Cf(^{11}B,4n)^{256}Lr$ reaction. The energy scale in the inset is the same as that in the main figure, but the full scale for counts per channel is 100.

Fig. 4. A comparison of the alpha-particle spectra produced in the Dubna experiments in 1968 with those reported in this paper.



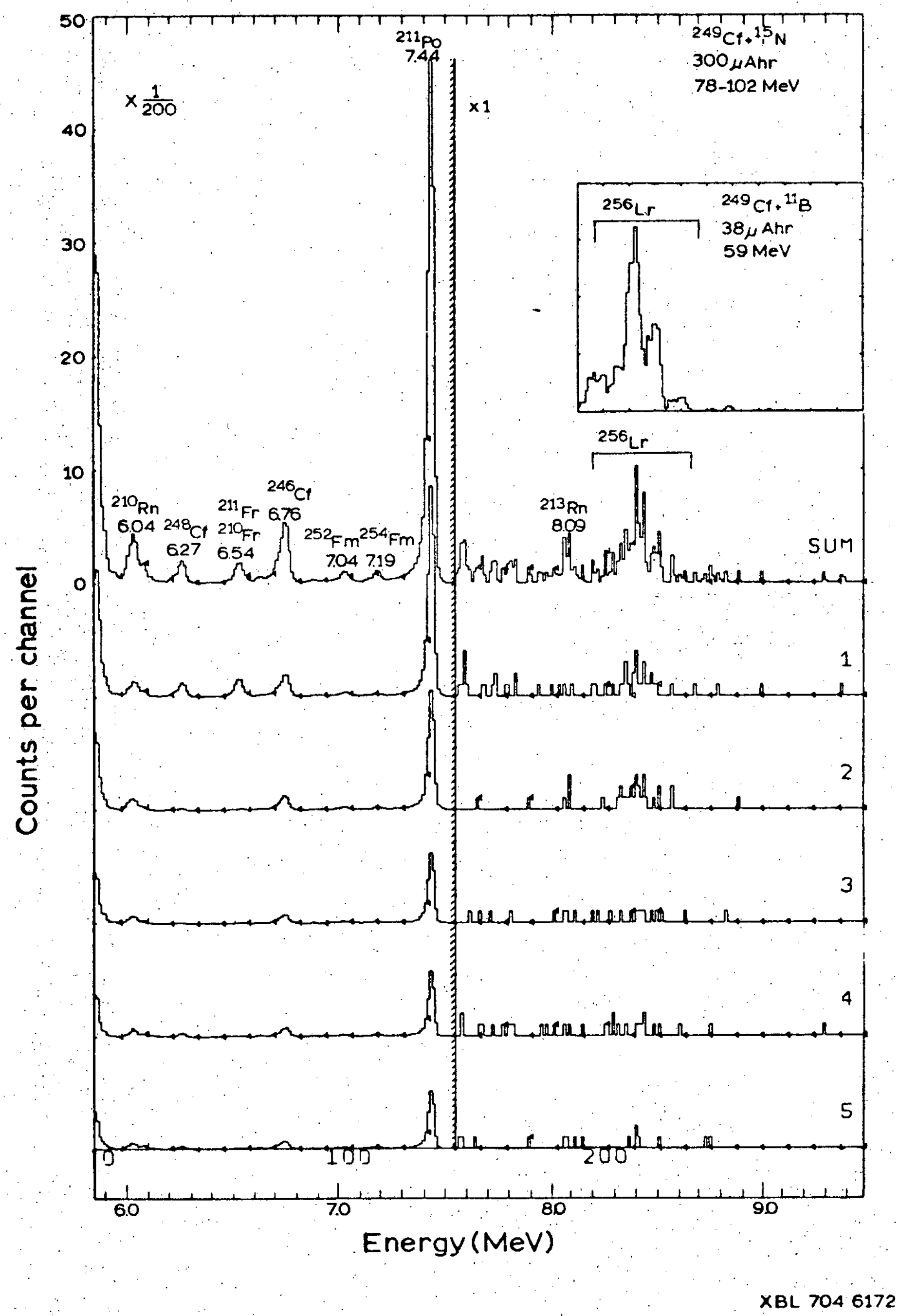
XBL 704 6171

Fig. 1



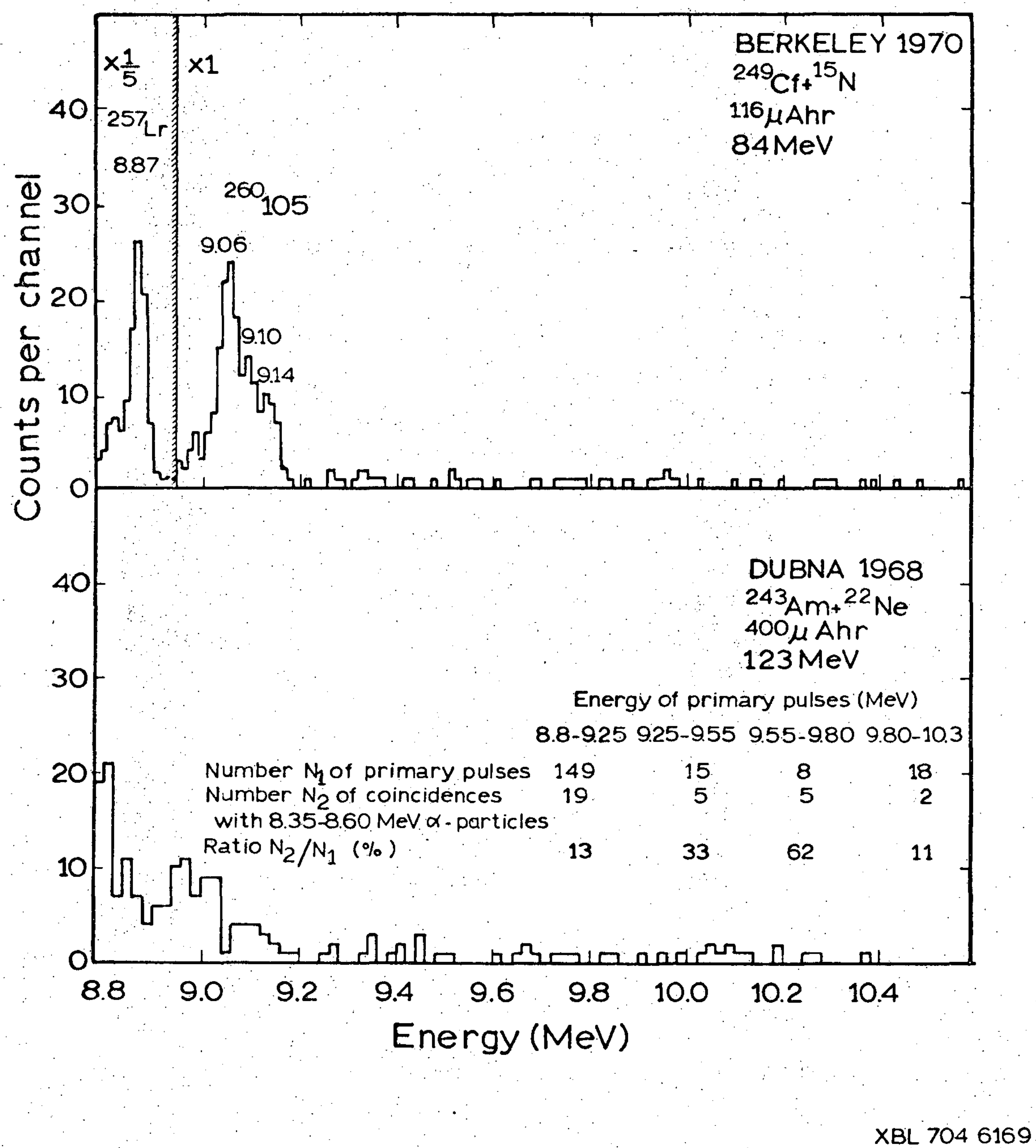
XBL 704 6170

Fig. 2



50
40
30
20
10
0
Counts per channel
x 1/200
x1
211Po 7.44
249Cf+15N
300 μAhr
78-102 MeV
256Lr
249Cf+11B
38 μAhr
59 MeV
210Rn 6.04
248Cf 6.27
211Fr 210Fr 6.54
246Cf 6.76
252Fm 7.04
254Fm 7.19
213Rn 8.09
SUM
1
2
3
4
5
0
100
200
6.0
7.0
8.0
9.0
Energy (MeV)
XBL 704 6172

Fig. 3



BERKELEY 1970
249Cf+15N
116μAhr
84MeV
×1/5
×1
257Lr
8.87
260105
9.06
9.10
9.14
DUBNA 1968
243Am+22Ne
400μ Ahr
123 MeV
Energy of primary pulses (MeV)
8.8-9.25 9.25-9.55 9.55-9.80 9.80-10.3
Number N1 of primary pulses 149 15 8 18
Number N2 of coincidences with 8.35-8.60 MeV α-particles 19 5 5 2
Ratio N2/N1 (%) 13 33 62 11
Counts per channel
Energy (MeV)
0
10
20
30
40
8.8
9.0
9.2
9.4
9.6
9.8
10.0
10.2
10.4
XBL 704 6169

Fig. 4

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