New nuclide ^{263}Ha

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(Received 17 April 1991)

A new nuclide ^{263}Ha was produced in the bombardment of a ^{249}Bk target with 93-MeV ^{18}O ions. It was detected via spontaneous fission counting and was shown to have a half-life of about 0.5 min. This activity was also separated from the reaction products by automated rapid chemical separations using cation-exchange chromatography in 0.05M α-hydroxyisobutyric acid. After chemical separation, ^{263}Ha was found to decay by spontaneous fission (57^{+13}_{-15}%) and by α emission ($E_\alpha = 8.35$ MeV, 43%) with a half-life of 27^{+10}_{-7} s. The spontaneous fission fragment energy spectrum is compatible with an average total kinetic energy of about 200 MeV.

PACS number(s): 23.90.+w, 25.70.Jj, 25.85.Ca, 27.90.+b

I. INTRODUCTION

Decay properties of nuclides with neutron numbers greater than 157 are pivotal in determining future trends in the stability of nuclei beyond the upper bounds of the chart of nuclides. In 1971 Ghiorso *et al.* [1] reported the discovery of the heaviest known isotope of element 105, 40±10-s α-emitting isotope ^{262}Ha, produced by the ^{249}Bk(^{18}O,5n) reaction. In these experiments they also detected a 25±10-s spontaneous-fission (SF) activity which they stated could arise from a SF branch in ^{262}Ha or from an electron-capture (EC) branch to ^{262}Rf which fissions with a very short half-life. They also stated they could not rule out the possible assignment of the 25-s SF activity to ^{263}Ha or EC decay of ^{263}Ha to ^{263}Rf which might decay via SF. Since that time, there have been several attempts to determine the decay modes and half-life of the undiscovered isotope ^{263}Ha. In 1980 Druin *et al.* [2] carried out ^{18}O bombardments of ^{249}Bk in an attempt to observe SF of the unknown isotope which should be formed in the ^{249}Bk(^{18}O,4n) reaction. They were unable to observe a spontaneous-fission activity with the yield expected for the (^{18}O,4n) reaction. Dougan *et al.* [3] tried to use the ^{254}Es(^{16}O,α3n) reaction to produce ^{263}Ha. They planned to detect the mother-daughter sequence

$$^{263}\mathrm{Ha} \xrightarrow{\alpha} {}^{259}\mathrm{Lr} \underset{t_{1/2}=5.4\ \mathrm{s}}{\xrightarrow{E_\alpha = 8.46\ \mathrm{MeV}}} {}^{255}\mathrm{Md}$$

in a multiple-α-detector (MAD) system. Based on an extrapolation of (α3n) systematics, they predicted a formation cross section of 15–20 nb for this reaction. Only four possible mother-daughter sequences were observed [3]. Of these, three had mother-daughter detection intervals too long to agree with the known 5.4-s ^{259}Lr half-life and were attributed to degraded fission fragments. The remaining sequence consisted of an 8.33-MeV mother event occurring 41.6 s after deposition in the detector system, followed 8.8 s later by a daughter event at 8.47 MeV. This was not considered as sufficient evidence to claim the discovery of ^{263}Ha since the 3σ upper limit to the formation cross section was only 5 nb, much lower than the predicted 15–20 nb. The ^{254}Es(^{13}C,4n) reaction [4] to produce ^{263}Ha was subsequently investigated, but no time and α-energy correlated events could be detected. This yielded an upper limit for the formation cross section of 2.7 nb, whereas a cross section of 6–10 nb was expected. α emission appeared to be ruled out as a major decay mode for ^{263}Ha by these experiments. Consequently, Hulet *et al.* [4] performed additional experiments using the same reaction to look for EC as the major decay mode for ^{263}Ha. Their predictions indicated that its daughter ^{263}104 would also decay by EC to ^{263}Lr, which should then decay via SF with an estimated half-life of about 5 h. Therefore, they performed a radiochemical search for SF from the granddaughter ^{263}Lr. No SF events were found [4], and it was concluded that these experiments [3,4] had virtually eliminated α and EC as possible decay modes of ^{263}Ha. The most likely possibility was believed to be SF decay with a half-life well under 1 min [4]. Concurrent searches for the undiscovered isotope by Gregorich *et al.* [5], using the ^{249}Bk(^{18}O,4n) reaction, also remained unsuccessful and were interpreted

to mean that ^{263}Ha probably decays by SF with a half-life as short as a few seconds.

In the course of a series of experiments to investigate further the chemical properties [6] of element 105 using the well-known isotope 34-s ^{262}Ha produced in the ^{249}Bk(^{18}O,5n) reaction, we have developed and used a very efficient aqueous-phase chemical separation procedure for hahnium. In the present work, we have applied this procedure to the products of a ^{249}Bk bombardment at a lower projectile energy, thought to be optimum for the production of ^{263}Ha in the 4n reaction.

II. EXPERIMENT

A 0.54-mg/cm^2 ^{249}Bk target was bombarded with 112-MeV $^{18}O^{5+}$ ions at the LBL 88-Inch Cyclotron. The $^{18}O^{5+}$ beam, after passing through a 2.37-mg/cm^2 Be window, 0.38 mg/cm^2 of N_2 cooling gas, and the 2.49-mg/cm^2 Be target backing, had an energy of 93 MeV in the target material. This energy corresponds to the maximum of the excitation function for the ^{249}Bk(^{18}O,4n) reaction, according to statistical evaporation calculations using the code HIVAP [7]. The beam current was about 0.5 particle μA. The reaction products recoiling out of the target were stopped in He gas at 1.2 bar, which had been loaded with KCl aerosols. The reaction products were attached to the aerosols and swept out of the recoil chamber with the gas in a polyvinyl chloride capillary (1.34 mm inner diameter). They were transported 5 m to the collection site, which was either a tape system equipped with silicon detectors for on-line α and SF counting or the automated rapid chemistry apparatus ARCA II [8].

In the former case, the products were collected by a magnetic tape system. In a collecting chamber held at 16 mbar, the KCl particles exiting the transport capillary were deposited on the surface of the magnetic tape. Every 10 s the tape was stepped to transport the deposited activity through an array of six counting chambers covering a total decay time of 60 s. Each of these chambers was equipped with a 450-mm^2 passivated ion-implanted planar silicon (PIPS) detector. This allowed counting of the samples in nearly 2π geometry. The typical alpha-energy resolution was 25 keV [full width at half maximum (FWHM)]. All alpha and SF events were recorded and analyzed in an event-by-event mode using the PSI tandem data-acquisition system [9]. For on-line display, the CERN PAW software [10] package was used.

In the chemical separation experiments, the transport gas exiting the capillary was pumped through a polyethylene frit by a mechanical pump. The activity-bearing aerosols were collected on that frit. After 1-min collection time, the frit was shuttled into position on top of either of two microchromatography columns (1.6 mm$\times$8 mm) filled with a strongly acidic cation-exchange resin (Aminex A6, particle size 17.5$\pm$2 μm). The activity was washed from the frit onto the column with unbuffered 0.05 mol α-hydroxyiosbutyric acid (α-HiB), where pentavalent metal ions, such as Ha^{5+}, were complexed to such an extent that they were immediately eluted from the columns in about 50 μl, while tetra- or trivalent metal ions remained on the column. The α-HiB effluent was quickly evaporated to dryness on a Ta disk, which produced a thicker source than that on the magnetic tape. α and SF fragment pulse-height analyses were performed on each sample using a system of ten 300-mm^2 PIPS detectors. Typical α-energy resolution was 60 keV (FWHM). Each event was stored along with the time after the start of counting and detector identification. The start of counting was $\approx$40 s after the end of collection, and the total counting time was 450 s.

III. RESULTS AND DISCUSSION

The search for the undiscovered isotope ^{263}Ha began with collection of the reaction products on magnetic tape. The tape was moved at 10-s intervals so as to move the collected products consecutively into position in front of the six detectors for α and SF counting. In this on-line search, identification of isotopes by α spectroscopy was hampered by intense α activities from several polonium isotopes. These nuclides, mostly ^{211}Po^m and ^{212}Po^m, were produced in transfer reactions with lead impurities in the target. During the experiments, noise which caused fissionlike events was noted in the last two detectors of the tape system. The total number of SF's recorded in each of the first four detectors for a beam dose of 3.68×10^{16} particles is shown in Fig. 1. A maximum-likelihood fit was made to the data from the first four detectors only. This yielded a half-life of 27^{+14}_{-8} s. From a similar experiment at a higher bombarding energy of 99 MeV, it was concluded that at stepping times as short as 10 s, long-lived SF activity due to ^{256}Fm is negligible. From the data of Chasteler *et al.* [12], the production cross section for the transfer product ^{256}Md, which decays to ^{256}Fm, is expected to decrease by a factor of 5 between 99 and 93 MeV. A calculation for our experiment at 93 MeV results in a contribution to the data shown in Fig. 1 of less than one SF from ^{256}Fm.

The production cross section for the fission activity shown in Fig. 1 is 5.5$\pm$2.5 nb. This value results from an assumed 50$\pm$10% gas-jet transport yield, a detection efficiency of 70$\pm$10%, and a transport time of 5 s for the

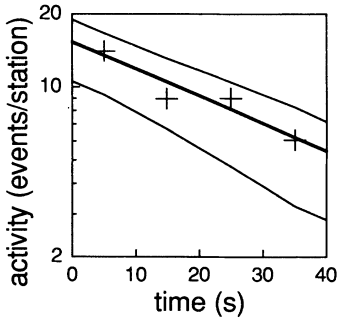

FIG. 1. Decay curve of the SF activity measured with the magnetic tape system using four detector stations and a stepping time of 10 s. The numbers of counts in each detector station are indicated by the crosses. The center line is the most probable fit to the data, and gives a half-life of 27^{+14}_{-8} s. The upper and lower lines are the limits which encompass 68% of the probability in a Poisson distribution centered on the number of counts in the most probable fit.

products from the target chamber to the tape system. This cross section is at variance with the 1.2±0.3 nb given by Druin *et al.* [2] for a $34.0^{+28.6}_{-13.6}$-s SF activity at 93 MeV bombarding energy.

Even though the half-life of the observed SF activity is nearly the same as that of the 5*n* product 34-s ^{262}Ha, which is reported to have a SF branch of about 50% [6], an assignment of the activity to this isotope was excluded because the cross section for production of ^{262}Ha at 93 MeV was expected to be less than 1 nb (see later discussion). Therefore, it was assumed that the new SF activity is associated with the decay of the previously undiscovered 4*n* product ^{263}Ha and that, because of the similar half-lives, the SF decay of ^{263}Ha was nearly indistinguishable from SF of ^{263}Ha in earlier experiments.

The chemical procedure involving cation-exchange separations with α-HiB solutions was developed with 34-s ^{262}Ha from the ^{249}Bk(^{18}O,5*n*) reaction at an ^{18}O energy of 99 MeV. This chemical separation was found to be very specific for element 105. The sum of all α-particle spectra from these experiments at 99 MeV is shown in Fig. 2(a). Apart from contamination by ^{249}Cf sputtered from the target (this material is not dissolved in the weakly acidic α-HiB solution and is washed mechanically through the column) and by small amounts of Bi and Po activities produced by transfer reactions on a Pb impurity

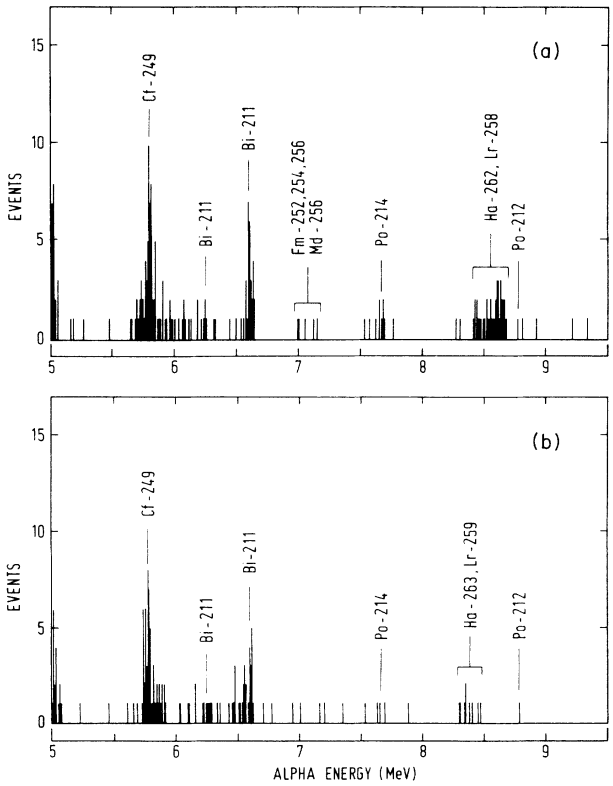

FIG. 2. (a) Sum spectrum of all α particles observed in the bombardment of ^{249}Bk with 22.3 μA h of 99-MeV $^{18}O^{5+}$ ions (Ha fractions from cation-exchange separations in 0.05 mol α-HiB). (b) Sum spectrum of all α particles observed in the bombardment of ^{249}Bk with 15.6 μA h of 93-MeV $^{18}O^{5+}$ ions. The chemical procedure was the same as for (a).

in the target, the spectrum is extremely clean and compatible with the complex spectra of ^{262}Ha and its daughter ^{258}Lr. Besides 41 α events from ^{262}Ha and ^{258}Lr, among them 9 correlated pairs of parent-daughter decays with the correct half-life of $4.2^{+1.5}_{-1.1}$ s for the correlated daughters, 23 fissions attributable to the decay of ^{262}Ha were also detected. These event rates are consistent with the detector efficiency and the known cross section for the production of ^{262}Ha at 99 MeV [6].

Application of the same chemical separation and counting techniques to the products from a 93-MeV $^{18}O^{5+}$ bombardment resulted in the α spectrum shown in Fig. 2(b). The absence of α particles with energies above 8.5 MeV indicates that the ^{262}Ha/^{258}Lr pair is no longer present. Based on the measured cross section for ^{262}Ha at 99 MeV of 8 nb [6] and on calculations with the code HIVAP, the cross section for production of ^{262}Ha at 93 MeV is expected to be <1 nb. At the same time, HIVAP predicts a cross section for the 4*n* product ^{263}Ha at 93 MeV on the same order of magnitude as for the 5*n* product ^{262}Ha at 99 MeV. We have detected 9 α particles and 18 SF's that we attribute to the new isotope ^{263}Ha and its daughter ^{259}Lr. A maximum-likelihood technique [11] was used to fit the α-decay lifetimes with a single half-life. The fit to the 9 α's with lifetimes less than 200 s resulted in a half-life of 20^{+11}_{-6} s. A similar fit to the 18 SF events resulted in a half-life of 34^{+15}_{-9} s. Taking these half-lives to be consistent, a two-component fit (allowing for a long-lived background) was used for all the α and SF lifetimes shorter than the measurement time of 450 s. This resulted in the ^{263}Ha half-life of 27^{+10}_{-7} s. The error limits are the values at which the likelihood function drops to one-half of its maximum value and include the covariance effects of the other free parameters. They represent approximately 70% confidence limits. In all experiments the time between chemical separation of Ha from Lr to the start of counting was sufficient for ^{259}Lr to approach transient equilibrium. Table I shows our interpretation of the α events.

The α-particle energy for ^{263}Ha resulting from the data shown in Table I is 8.355±0.027 MeV; that for its daughter ^{259}Lr is 8.445±0.029 MeV. While the mother energy is compatible with expectations based on α-energy

TABLE I. α events between 8.3 and 8.5 MeV in the 93-MeV ^{18}O bombardment of ^{249}Bk.

Detector	Energy (MeV)	Lifetime (s)	Assignment
5	8323	0.90	^{263}Ha
1	8369	7.95	^{263}Ha
9	8414	4.62	^{259}Lr
8	8463	34.67	^{259}Lr
9	8318	17.50	^{263}Ha
2	8484	63.20	^{259}Lr
9	8358	66.88	^{263}Ha
4	8395	39.15	^{263}Ha[a]
9	8366	16.85	^{263}Ha[b]

[a]Correlated with a 72-MeV fission event 6.00 s later.
[b]Correlated with a 106-MeV fission event 8.58 s later.

systematics, the daughter energy is in agreement with Eskola *et al.* [13] and with recent work [14], in which ^{259}Lr was produced directly via the ^{15}N+^{248}Cm reaction. These experiments [14] resulted in a half-life for ^{259}Lr of $6.35^{+0.46}_{-0.42}$ s and in the discovery of a previously undetected fission branch of 23±2 % in this isotope. The two α-SF correlations are likely to be due to the $^{263}\text{Ha} \xrightarrow{\alpha} {}^{259}\text{Lr} \xrightarrow{\text{SF}}$ decay sequence. To further assess the probabilities for detecting ^{263}Ha and/or ^{259}Lr, we use the following experimental information or reasonable assumptions: (a) The efficiency for detecting an α particle from the source is 0.35, (b) the efficiency for detecting a SF from the source is 0.70, (c) the source is thin enough for alpha recoils to escape from the source and deposit on the detector surface with an efficiency of 0.35, (d) the efficiency for detecting an α (SF) from the detector face is 0.5 (1.0), (e) the ^{263}Ha half-life is 26.9 s, (f) the ^{259}Lr half-life is 6.35 s, and (g) the fission branch in ^{259}Lr is 0.23. The ten possibilities for detecting ^{263}Ha and/or ^{259}Lr are listed in Table II.

The probabilities listed on the right column of Table II are for a 57% fission branch in ^{263}Ha. Note that the probabilities for the first seven event types add up to 1.0, and the probabilities for the last three add up to 0.133. This is the number expected for transient equilibrium (0.309) multiplied by the α branch in ^{263}Ha (0.43). There were two unknowns in this system: the ^{263}Ha fission branch and the total number of ^{263}Ha atoms which were placed in front of the detectors. We detected 4 events of type 1, 0 events of type 2, 2 events of type 3, 16 events of types 6, 7, and 8, and 3 events of types 5 and 9. Using a Poisson distribution, we find that the most probable values (with error limits at which the probabilities fall to 0.5 of the maximum) are

$$\text{number of } ^{263}\text{Ha atoms} = 34.4^{+8.7}_{-7.4},$$

$$\text{fission branch in } ^{263}\text{Ha} = 57^{+13}_{-15}\ \% .$$

The unnormalized natural logarithm of the likelihood for this most probable fit is $\ln(L) = -8.990$. Monte Carlo simulations were performed in order to see if this value for $\ln(L)$ is typical for fits to similar sets of events. In these simulations the event types and detection probabilities of Table II were used. In 10 000 runs the 34 Ha atoms were randomly distributed between each of the seven different types of events. The number of daughter atoms present at the start of counting was determined with a Poisson distribution about the number of atoms expected for transient equilibrium (4.52). These ^{259}Lr decays were randomly distributed between the three detection possibilities (event types 8, 9, and 10) listed in Table II. A likelihood fit was performed for each of these 10 000 distributions. The most probable likelihood was found around $\ln(L) = -8.2$. This means that our $\ln(L)$ of -8.99 is typical. Also, the widths of the distributions for the number of Ha atoms and fission branches are similar to the error limits determined from the shape of the probability distribution for our experimental events. Thus our results for the discovery of the new isotope ^{263}Ha hold up to all statistical tests. The nonobservation of α-α correlations is compatible with statistics.

The single-fragment kinetic-energy distribution of the 18 fission events detected in the chemically separated samples of ^{263}Ha is shown in Fig. 3(a). It is compared to the kinetic-energy distribution for an electroplated ^{252}Cf source measured with the same detectors. From the α intensity in Fig. 2(b) around 7 MeV, where one expects to see the major α intensity from actinide isotopes produced in transfer reactions, and from the known α-to-SF ratio associated with their decay, we can conclude that there was essentially no SF background to disturb the ^{263}Ha fission measurement. The peak energies [15] in the ^{252}Cf single-fragment energy spectrum were used to determine a linear energy calibration for fission fragments. The average fragment energies for ^{263}Ha and ^{252}Cf were multiplied by 2 to give estimates for their respective average total kinetic energies. An average total kinetic energy for ^{263}Ha SF of 207±7 MeV resulted from a small correction to these estimates which brought the average post-neutron-emission total kinetic energy for ^{252}Cf SF to 181.0 MeV [15]. The fission-fragment kinetic-energy distribution from the first two chambers of the tape system is shown in Fig. 3(b) together with the relevant ^{252}Cf calibration spectrum. The average post-neutron-emission total kinetic energy deduced from this measurement is 199±5 MeV. The two values are consistent with each other and with SF systematics [16]. Because only single SF fragments were detected, it is not possible to draw detailed conclusions about the total kinetic-energy and mass distributions. The data seem to indicate that the mass

TABLE II. Event types and detection probabilities for ^{263}Ha and/or ^{259}Lr.

Event type	Designation	Detection probability
1	detect parent α, miss daughter	0.085
2	detect parent α, detect daughter α	0.041
3	detect parent α, detect daughter fission	0.024
4	miss parent, miss daughter	0.330
5	miss parent, detect daughter α	0.075
6	miss parent, detect daughter fission	0.045
7	detect parent fission	0.399
8	detect fission from ^{259}Lr present at start count	0.021
9	detect α from ^{259}Lr present at start count	0.036
10	miss detection of ^{259}Lr present at start count	0.076

distribution is broadly symmetric.

The decay properties of ^{263}Ha, with an overall half-life of 26.9 s, a single α group at 8.35 MeV, a fission branch of 57%, and an α branch of 43%, give an experimental partial half-life for α decay of 62.6 s. According to the most recent alpha-decay systematics [17], the unhindered α decay half-life is 47.3 s, and the resulting hindrance factor is 1.3. It should be noted that the experimental values are lower limits on α-decay half-lives and hindrance factors because we were probably not sensitive to a possible electron-capture branch in ^{263}Ha. For odd-even nuclei the average hindrance factor for favored α decay is 2.11 ± 1.29 [17], and so the experimental half-life agrees well with the expectation, especially in light of the uncertainty in the overall half-life and fission branch. The lower limit for the SF hindrance factor for the odd proton in ^{263}Ha calculated relative to the even-even neighbor ^{262}Rf [18] is 1.0×10^{3}.

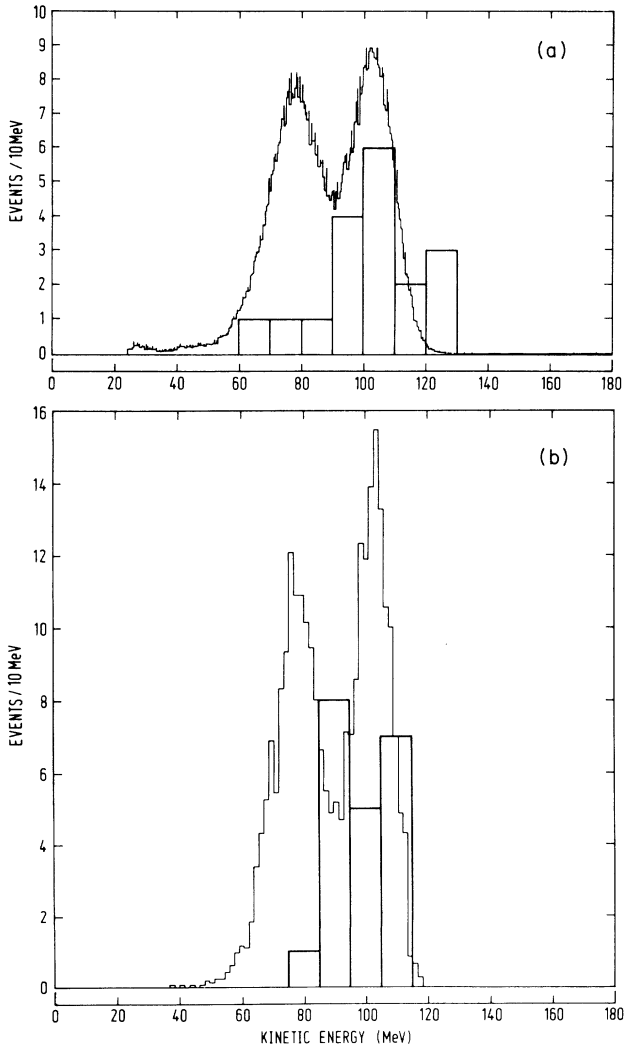

FIG. 3. Fission-fragment kinetic-energy distribution associated with the decay of ^{263}Ha (histogram) compared to the kinetic-energy spectrum of a ^{252}Cf source measured in the same system: (a) after chemical separation and (b) measured on line with the tape system.

The production cross section for ^{263}Ha at 93 MeV, as derived from the ^{249}Bk content in the target, the He/KCl-jet efficiency, the number of ^{263}Ha decays detected, the counting efficiency, and the chemical yield assumed to be the same as for tracer activities of niobium, tantalum, and protactinium, is 10 ± 6 nb. The large uncertainty is due to large fluctuations in the gas-jet transport efficiency during the production run. Nevertheless, this cross section is consistent with the 5.5 ± 2.5 nb for SF measured with the tape system and a SF branch of 57%. From the absence of α events with $E_\alpha > 8.5$ MeV, where the majority of the ^{262}Ha-^{258}Lr decay is located (Fig. 2), we derive an upper limit of ≤ 1.3 nb for the production of the $5n$ product ^{262}Ha at 93 MeV. (This assumes that the observation of no events corresponds to three events at 95% confidence level.)

The cross section for production of ^{262}Ha at 99 MeV reported earlier [6] must be reduced because it is clear now that both isotopes ^{262}Ha and ^{263}Ha are produced at this energy. (This was probably also true for the SF activity reported by Ghiorso *et al.* [1].) This is suggested by evaporation calculations as well as by the data. In the concurrent study of chemical properties of hahnium performed at 99 MeV bombarding energy, we observed a total of 80 α events with $8.3 \leq E_\alpha \leq 8.7$ MeV, with 5 events around $E_\alpha = 8.35$ MeV which must be assigned to ^{263}Ha. From the detection probabilities in Table II, it follows that, in addition, about 5 daughter α decays at $E_\alpha = 8.45$ MeV must have been present too, but are masked by the 8.45-MeV decay branch in ^{262}Ha. Thus 10 α events out of 80 belong to the $4n$ channel and 70 to the $5n$ channel. Based on the relative detection probabilities listed in Table II, we conclude that the 10 α events should have been accompanied by 18 SF events. Subtracting these from the total of 53 SF events attributable to hahnium decays leaves 35 SF events associated with the decay of ^{262}Ha. This reduces the fission branch in ^{262}Ha to about 33%. The new cross sections at 99 MeV are then 6 ± 3 nb for ^{262}Ha and 2 ± 1 nb for ^{263}Ha. The same procedure applied separately to the results of the α-HiB chemistry, which produced the cleanest hahnium spectra, gives cross sections of 5.3 ± 2.8 nb for ^{262}Ha and 1.6 ± 1.0 nb for ^{263}Ha, respectively.

IV. CONCLUSION

We have observed a new α and SF activity which we assign to the decay of a new isotope ^{263}Ha. It decays with a half-life of 27^{+10}_{-7} s and has a 43% α branch and a 57^{+13}_{-15}% SF branch. The α-particle energy is 8.355 MeV. It is not possible to exclude decay via electron capture because our experiments may not have been sensitive to this. Based on measurements of the single-fragment kinetic-energy spectrum, the SF of ^{263}Ha appears to have a broadly symmetric mass distribution with an average post-neutron-emission total kinetic energy of 207 ± 7 MeV. Definitive chemical separations were performed

which show that the new activity should be assigned to element 105. The atomic number identification is supported by the agreement of the observed partial half-life for α decay with that expected for an isotope of element 105 in the region of mass 263, which decays by emission of 8.355-MeV α particles. The assignment to mass 263 is consistent with α-decay energy systematics in this region. This mass assignment is also consistent with the measured production cross section of 10 ± 6 nb at 93 MeV, which is of the same order of magnitude as the $5n$ cross section at 99 MeV, as predicted by evaporation calculations. The mass assignment is also supported by the observation of α particles of the ^{259}Lr daughter in the chemically separated Ha samples and by observation of two events in which the α decay of ^{263}Ha was followed by the SF decay of the ^{259}Lr daughter. The correlation times of these α-SF events are consistent with the 6.35-s half-life of the ^{259}Lr daughter.

ACKNOWLEDGMENTS

The authors are indebted to the Division of Chemical Sciences, Office of Basic Energy Sciences, U.S. Department of Energy, for making the ^{249}Bk target material available through the transplutonium element production facilities at the Oak Ridge National Laboratory. We wish to thank the staff and crew of the LBL 88-Inch Cyclotron for providing the ^{18}O beams and for technical support. The German and Swiss groups have enjoyed the hospitality of the Lawrence Berkeley Laboratory Nuclear Science Division. This work was supported by the German Federal Minister for Research and Technology (BMFT) under Contract No. 03-HE2MAI, by the Office of Energy Research, Office of Basic Energy Sciences, Chemical Sciences Division of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098, and by the Swiss National Science Foundation under Contract No. 2.204-0.86.

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