Modern Physics Letters A, Vol. 11, No. 11 (1996) 861–869

DISCOVERY OF STRONGLY ENHANCED LOW ENERGY ALPHA DECAY OF A LONG-LIVED ISOMERIC STATE OBTAINED IN ^{16}O + ^{197}Au REACTION AT 80 MeV, PROBABLY TO SUPERDEFORMED BAND

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Received 15 February 1996

The reaction ^{16}O + ^{197}Au has been studied at a bombarding energy of 80 MeV. A group of 5.20 MeV α particles with a half-life of about 90 m has been found in coincidence with characteristic X-rays of At and with γ-rays. The γ-ray energies fit predicted energies for superdeformed band. The data are interpreted as due to formation of a long-lived isomeric state which decays by low energy α-particles to SD band. The calculated probability for decay via a barrier of a superdeformed nucleus was found to be consistent with the experimental results.

The relative relationship between the energies of various α-particle groups and their corresponding half-lives usually fits, to within an order of magnitude, a simplified barrier penetration probability picture.[1] In this letter the observation of a low energy α-particle group whose decay is enhanced by about five orders of magnitude is described, and a possible explanation of this phenomenon is given. Reports on these results have been given.[2]

In various experiments 900 μg/cm^2 gold targets followed by two 150 μg/cm^2 C catcher foils were irradiated with 80 MeV ^{16}O beam. All the evaporation residue nuclei but only a small part of the fission and the transfer reaction products were caught on the foils, which were then transferred to Jerusalem and placed together in between a 300 mm^2, 50 μ thick, Si surface barrier detector and a 500 mm^2, 10 mm thick, thin window Ge(Li) detector. The solid angle of the Si detector was about 27% and of the Ge(Li) detector 16%. The internal efficiency of the γ-ray detector for photo-peak detection was 100% up to about 120 keV, and reduced gradually to 26% at 220 keV. Its resolution (FWHM) was around 900 eV.

In a first group of experiments we measured α-particle and X- and γ-ray spectra and determined the lifetimes of the various produced activities. The α-particle spectra were measured separately from thick C catcher foils (two foils of 150 μg/cm^2

each measured in two different detectors) to obtain absolute cross-sections, and from a thin C catcher foil (40 μg/cm^2) to obtain good resolution of about 60 keV. ^{210}Rn, ^{205}Po, ^{206}Po, ^{211}Po, ^{207}At and ^{209}At were identified in the α spectra, and ^{201}Bi, ^{205}Po, ^{205}At, ^{206}At, ^{209}At, ^{199}Tl and ^{109}Pd in the X- and γ-ray spectra.

Cross-sections (in mb) of $7.2 \pm 20\%$, $7.0 \pm 20\%$, $0.4 \pm 50\%$, $0.2 \pm 50\%$ and $> 0.014 \pm 50\%$, for the $3n$, $4n$, $p2n$, $\alpha 2n$ and $2n$ channels respectively, were determined. (In the last case ambiguity with the pn channel exists and if the activity seen is due to this channel then its cross-section is $0.003 \pm 50\%$ mb.) The measured cross-sections of ^{199}Tl (transfer reaction) and ^{109}Pd (fusion-fission reaction) are 0.5 and 0.1 mb, respectively, being, of course, only a small part of the total production cross-sections for them.

In a second group of experiments singles spectra as well as coincidences between the α-particles and the X- and γ-rays were measured. Two runs were performed. In the first one the irradiation time was 168 m, the average beam intensity 36.2 pnA and the resolving time in the coincidence measurements 1.0 μs. In the second run we irradiated for 188 m with beam intensity 52.1 pnA, and coincidence resolving time 0.2 μs. The resolving times were determined using a random pulse generator in the α-detection system and a ^{137}Cs source in front of the γ detector. The randoms seen in coincidence with the large 6.0 MeV α-particle group (see Figs. 1c–e) confirm these resolving times. A 24 h background measurement gave zero counts in the α-γ coincidence spectrum. The various experimental conditions are summarized in Table 1. Figure 1a shows a typical α-particle spectrum with a resolution of about 250 keV (FWHM) due to the thickness of the source. Figure 1b represents a typical γ-ray spectrum, with a resolution (FWHM) of about 900 eV.

In Figs. 1c–e the coincidence events with $E_\alpha = 5.8$ MeV are consistent with random coincidences due to the 6.04 MeV group of ^{210}Rn. A few true coincidences are seen around $E_\alpha = 4.98$ MeV. In Fig. 1c, for α-particles of 4.7 to 5.1 MeV and X- or γ-rays of 70–90 keV, five events are seen while 0.9 random coincidences are expected. The energies of the γ-rays in this region are 70.8, 74.8, 78.9, 81.6 and 86.3 keV with only the 74.8 keV event matching in energy with a rather large peak in the singles spectrum of the γ-rays ($K_{\alpha 2}$(Bi) + $K_{\alpha 1}$(Pb)), while the singles intensity around, for instance 81.6 keV, is about 20 times smaller than the largest peak at 77.1 keV ($K_{\alpha 1}$(Bi) + $K_{\alpha 2}$(Po)). On the other hand, one notices that 81.6 and 78.9 keV (the events surrounded with squares) fit very well with $K_{\alpha 1}$ and $K_{\alpha 2}$ X-rays of At (see below). In the region of γ's between 95 to 210 keV and α's of 4.7 to 5.1 MeV the expected number of random coincidences is 0.25 while two events are seen at 105.3 and 203.0 keV.

In Fig. 1d the results of a long measurement (including also the data of Fig. 1c, see Table 1) are given. Coincidence events between 4.9 MeV α-particles and γ-rays at 26.8, 35.1, 70.8, 114.4, 141.0, 191.9 keV and 71.8, 74.0 keV which fall into minima between the large X-ray lines, were found. The rest (four events at around 75.0 keV and three events around 77.1 keV, and the events around 10.8 and 13.0 keV) may be within statistics, due to random coincidences.

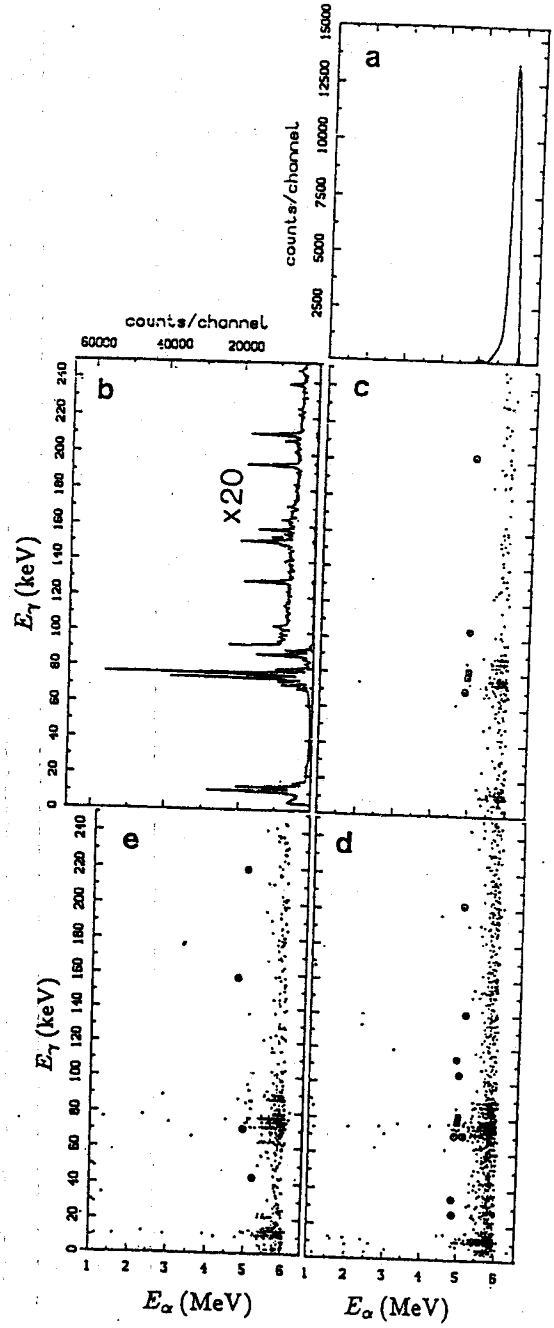

Fig. 1. Various spectra obtained from the catcher foils: (a) and (b) are singles α- and γ-ray spectra, respectively; (c)–(e) are α–γ coincidence spectra. The coincidence events surrounded with squares fit the $K_{\alpha 1}$ and $K_{\alpha 2}$ X-rays of At. The coincidence events surrounded with circles fit predicted transitions of SDB. (See text and Tables 1 and 2.)

Table 1. The experimental conditions of the various results given in Fig. 1.

Figure No.	Type of meas.	Run No.	Starting time[a] of meas.	Stopping time[a] of meas.
1a	singles α	1	198	252
1b	singles γ	1	198	252
1c	coin. α-γ[b]	1	198	252
1d	coin. α-γ[b]	1	100	993[d]
1e	coin. α-γ[c]	2	100	421

[a] Time in minutes after end of irradiation.

[b] Resolving time 1.0 μs.

[c] Resolving time 0.2 μs.

[d] Except from 252 to 315 m where no data was taken.

In Fig. 1e the results of the second run are given. The beam intensity was about 1.4 times larger and the resolving time 5 times shorter, (0.2 μs). In the α particle range of 4.7–5.2 MeV and γ rays of between 60–110 keV 16 events are seen while 4.7 random coincidences are expected, and for γ-rays of 130–230 keV three events are found, while 0.8 accidental coincidences are expected. The three events at around 77.0 keV, and the one event each at 74.7 keV and 87.6 keV could be random coincidences due to the large $K_{\alpha 1}$(Bi), $K_{\alpha 2}$(Bi)$+K_{\alpha 1}$(Pb) and $K_{\beta 1}$(Bi) peaks. The rest, at energies of 70.8, 71.7, 74.2, 75.9, 82.5, 85.4, 86.5 (two events); 92.5, 95.7, 102.3, 136.8, 157.8 and 219.5 and also at 43.6 keV should be true coincidences. 70.8 (two events), 71.8, 74.0 and 86.3 have also been seen (Fig. 1d). Altogether 29 true coincidence events (14 in the first run and 15 in the second run) between about 5 MeV α-particles and various γ-rays have been observed.

The energy of the α-particle group corrected for the thickness of the catcher foils is $5.20^{+0.05}_{-0.25}$ MeV. Its estimated half-life is about 90 m with an uncertainty of about 50%. This estimate was obtained by using the formulas of Ref. 3 and also by a best fit analysis of the data to the relationship $\Delta N = A_0(e^{-\lambda t_1} - e^{-\lambda t_2})$. The production cross-section of this group is about 30 nb, if one assumes high multiplicity of the γ's as suggested by the α-γ coincidence data (see below), which leads to about unit detection probability in the γ-ray detector, despite its rather low total efficiency. (For γ multiplicity of 1, one finds about 200 nb).

This group of 5.2 MeV α-particles with about 90 m half-life could not be identified with any known activity. The only transition seen with similar energy and lifetime is the g.s. to g.s. transition of ^{205}Po, which however would not give α-γ coincidences. Furthermore known weak α branches cannot explain the data. The only strong candidate which has an α branching energy and lifetime not too far away from our measured values is ^{210}Rn ($t_{1/2} = 2.4$ h). Its 5.35 MeV α's (branching ratio 5.6×10^{-5}) are in coincidence with Comptons from the 701 keV. This leads to 0.04 coincidence events for the γ region of 70–90 keV in Fig. 1c where in fact five are seen. In conclusion the α activity must come from unknown isomeric state(s).

A half-life of about 90 m is very short for 5.2 MeV α-particles for nuclei in the Fr region. Using for instance the formulas of Ref. 4 for ^{210}Fr one calculates 2.7×10^7 m, about 3×10^5 times longer than experimentally observed. In principle this discrepancy could be resolved assuming that the seen α-particles are a small branch of much stronger main decay mode. However in this case the production cross-section of the isomeric state should be 30 nb (or 200 nb) $\times\ 3 \times 10^5 = 9$ mb (or 60 mb). Such large cross-sections are excluded by the measured α-particle and X- and γ-ray spectra where cross-sections down to 3 μb were measured.

More information on the origin and character of the 5.2 MeV α-particle group comes from the measured energies of the γ-rays. The 81.6 and the 78.9 keV γ-rays (surrounded by squares in Figs. 1c and 1d) fit very well with the $K_{\alpha 1}$ and $K_{\alpha 2}$ X-rays of At of 81.52 and 78.95 keV, respectively. The parent nucleus may be an isotope of Fr. The events with X-ray energies of Bi and perhaps also of Pb are most likely random α-γ coincidences. The three events at 70.8 keV, in principle may be due to $K_{\alpha 1}$ of Hg. However, in this case 1.8 events at 68.9 ($K_{\alpha 2}$ of Hg) and one event at 80.2 ($K_{\beta 1}$ of Hg) are expected, while zero events are seen. According to Poisson statistics the probability to see zero events when 2.8 are expected is only 6%. In addition, X-rays of Hg would imply α decay from Pb. But no decay from any Pb isotope was seen. It is impossible to identify consistently the other observed γ-rays with any known transitions. On the other hand, one notices that almost all of the measured γ-rays in the clean regions outside the region of X-rays, those surrounded with circles in Figs. 1c, 1d and 1e, follow nicely a $J(J+1)$ law and fit well with predicted γ-rays of superdeformed bands.[5] A best fit analysis to an $AJ(J+1)$ law gives for A the value of 4.40 keV with a χ^2 value of 0.62 while such a fit to an $AJ(J+1)+BJ^2(J+1)^2$ gives $A = 4.41$ keV and $B = -3.6\times10^{-5}$ keV with $\chi^2 = 0.53$. The value of 4.40 keV fits very well with 4.38 keV, the average of the two values, 4.31 and 4.45 keV, obtained from the superdeformed bands of[6] ^{240}Pu and[7] ^{194}Tl (band 1a), and scaled to $A = 206$ according to the $A^{5/3}$ law. The observed energies of the γ-rays as compared to the transitions assuming $E_x = 4.40 \times J(J+1)$ are given in Table 2. It is seen that the average deviation is 0.35 keV and the maximum is 0.6 keV. As mentioned above the total number of true coincidence events in the first run (Fig. 1d, see above) were 14, and in the second run 15 (Fig. 1e, see above). This fits within the measurement of the relative beam intensity which was about 1.4 times larger in the second experiment. However, the total number of events which fit with transitions in the SDB were eight and four in the first and second runs respectively. This could be due to the poor statistics, but more likely should be attributed to the shorter (by 5 times) resolving time in the second run. The other γ-rays which do not match with transitions in the SDB, and in particular those that repeat themselves in the two runs like 71.8, 74.1 and 86.4 are probably due to other transitions in the same nucleus.

Despite the fact that only a small number of events and no γ lines were seen, the assignment to a $J(J+1)$ law is very significant. First, as mentioned above

Table 2. The energies of the γ-rays in coincidence with the 5.2 MeV α-particles (the circled events in Figs. 1c–e as compared to transitions assuming $E_x = 4.40 \times J(J+1)$.

Transition	E_γ (expt.)[a] (keV)	E_γ (theor.) (keV)	ΔE (keV)
3 ⇒ 2	26.8	26.4	+0.4
(2 ⇒ 0)[b]			
4 ⇒ 3	35.1	35.2	−0.1
5 ⇒ 4	43.6	44.0	−0.4
(3 ⇒ 1)[b]			
8 ⇒ 7	70.8[c]	70.4	+0.4
12 ⇒ 11	105.3	105.6	−0.3
13 ⇒ 12	114.4	114.4	0.0
(7 ⇒ 5)			
16 ⇒ 15	141.0	140.8	+0.2
18 ⇒ 17	157.8	158.4	−0.6
12 ⇒ 10	203.0	202.4	+0.6
13 ⇒ 11	219.5	220.0	−0.5

[a] The peak to total ratio was 100% up to about 120 keV and reduced gradually to 26% at 220 keV.

[b] Highly converted.

[c] Three events.

and noted in Table 2, the photo-peak to total ratios for these low energy γ-rays were 100% or close to 100% for almost all of them. In principle these γ's could be Compton (due to some higher unknown γ lines) rather than photo-peak events. However in the whole clean regions outside the X-ray region (20–60 keV and 100–250 keV) there are in total 13 events in coincidence with 5.0 MeV γ's, where nine of them follow the $J(J+1)$ law. The probability for 9 out of 13 events which are distributed evenly due to Compton effect to fall into nine specified energy positions within the energy resolution of the γ detector (0.9 keV) is

$$\binom{13}{9} p^9(1-p)^{(13-9)} = 4.3 \times 10^{-8}\,.$$

p is equal to $16 \times 0.9/190$ where 16 is the number of possible γ transitions up to 250 keV between possible state with $E_x = 4.40 \times J(J+1)$ keV (excluding the range 60–100 keV), and $190 = (60 - 20) + (250 - 100)$ the total energy range. Therefore one has to conclude that the measured energies of the γ-rays are photo-peak γ's and not Compton γ's from unknown energetic γ's being in coincidence with the 5.2 MeV α-particle group.

According to our spin assignments out of the 10 γ-rays which could be related to transitions in the SDB 8 (or 7) are dipole transitions and 2 (or 3) are quadrupole transitions. The dipole transitions should be $E1$ rather than $M1$ since the internal conversion coefficients (δ) for $M1$ transitions in $Z = 85$ nuclei with energies of, for instance, 26 or 35 keV, are about 130 and 55, respectively,[8] and one does not see the relatively high intensity of L X-rays expected for such high δ-values. The 114.4 keV may be an $E1$ or $E2$ transition. $E1$ transitions mean those between positive and negative parity states. In order that $E1$ transitions could compete with $E2$ transitions their half-lives should be about the same or even shorter. Assuming that the $E2$ transitions in the SDB are enhanced by a factor of 10^3 with respect to the Weisskopf estimates,[9] the $E1$ transitions should then be retarded by a factor of 100. This is a rather low retardation factor. However, strong competitions between $E1$ and $E2$ transitions were seen before, for instance in[10] ^{222}Th and[11] ^{163}Er and interpreted as due to coupling to octupole degrees of freedom. From the observed X-rays of At one may infer that the parent nucleus is an isotope of Fr. The assigned SD γ-rays sequence give transitions between integer spins and therefore the daughter nucleus should be an odd–odd nucleus. The largest production cross-sections are for the $3n$ and the $4n$ channels and therefore the SDB may be in ^{206}At. Around $Z = 86$ and $N = 116$ a doubly magic region of SD shapes is predicted (for small and large spins) when octupole deformations are included.[12,13] When reflection symmetry does not exist the spins and parities in rotational bands in even nuclei are[14] $I = 0^+, 1^-, 2^+, 3^-, \ldots$, for the simplex quantum number[13,14] $S = 1$ and $I = 0^-, 1^+, 2^-, 3^+, \ldots$ for $S = -1$, and $E1$ transitions are relatively enhanced.[13]

Assuming that the head of the SDB in ^{206}At is at 10.89 MeV excitation energy, as predicted[15] for the second minimum of the potential energy curve (shape isomer), and that the α-particles decay to a spin-18 state at 12.4 MeV from our assigned $E_x = 4.40 \times J(J+1)$, then the isomeric state in ^{210}Fr may be at about 11 MeV excitation. (The value of 10.89 MeV was obtained by interpolation from the neighboring even–even nuclei.[15])

The effect of large deformations on α-particle decay with 5.2 MeV from ^{210}Fr was calculated with the potential parameters of Igo[16] but with a deformed radius $R_0(1 + \beta_2 Y_{20}(\theta) + \beta_3 Y_{30}(\theta) + \beta_4 Y_{40}(\theta))$ (Table 3). (A formation probability of[17] $25(4/A)^3$ was used in the calculations.) Only the decay out of the tip ($\theta = 0$) was directly calculated. Solid angle averaging increases the lifetimes by about a factor of 3. For $\beta_2 = 0.7$ (2:1 axes ratio) the lifetime reduces by about four orders

Table 3. Predicted half-lives for the 6.57 MeV ground state to ground state transition and for the 5.2 MeV α-particles emitted from ^{210}Fr as function of various deformations and compared to the experimental values (see text).

E_α	β_2	β_3	β_4	$t_{1/2}$(T)(m)	$t_{1/2}$(T)/$t_{1/2}$(E)
6.57	0.0	0.0	0.0	6.0	1.1
5.2	0.0	0.0	0.0	2.8×10^7	3.1×10^5
5.2	0.7	0.0	0.0	1.3×10^3	1.4×10^1
5.2	0.7	0.07	0.0	3.5×10^2	3.9
5.2	0.7	0.15	0.0	8.2×10^1	0.9
5.2	0.7	0.07	0.06	1.0×10^2	1.1

of magnitude and brings it to within one and a half orders to the experimental value. Further reduction occurs if octupole deformations of the values quoted in Ref. 12 and some hexadecapole deformation are additionally taken into account. A 4% increase in the radius parameter (from $r_0 = 1.17$ to $r_0 = 1.22$), reasonable for the α decay from a highly excited isomeric state, reduces the lifetime by another factor of 4. Thus strongly enhanced α-particle decay can be understood. Let us mention in passing that unidentified low energy particle groups which may be strongly enhanced α's were found before[18,19] in studies of actinides produced by secondary reactions, and that long-lived isomeric states which decay by EC have been found in ^{236}Am and ^{236}Bk.[20]

The origin and character of the isomeric state is not entirely clear. Its decay to high spins indicates that it has high spin. Its strongly enhanced α decay to SDB states indicates that it may be a state in the second potential well, since otherwise it would have been structurally hindered. It is perhaps a high spin isomeric state in the second well of the potential energy curve, similar to the situation with the fission isomers where, in some cases, two such isomeric states were found in the same nucleus.[21]

In summary, a 5.2 MeV α-particle group with a half-life of about 90 m has been found in coincidence with X-rays of At and with γ-rays which fit expected transitions in SDB. This suggests the existence of a long-lived high spin isomeric state, probably in ^{210}Fr, which prefers to decay by α-particles to the SDB. The strongly enhanced decay (the lifetime is about five orders of magnitude shorter than normally expected for such energy and Z values) can be quantitatively understood as due to the large deformations of the nucleus in the SD states. Based on theoretical predictions for the excitation energies of the head of the SDB the excitation energy of the isomeric state may be around 11 MeV. The possibility that the isomeric state is a high spin isomer in the second well of the potential energy curve is raised. The existence of such a long-lived isomeric state, with a half-life much longer than its corresponding ground state, may add a new consideration regarding the stability and the production mechanism of heavy and perhaps also of superheavy elements.[22-24]

Acknowledgments

We are grateful to J. Burde and N. Zeldes for very valuable discussions, G. Hollos, Y. Shahar and the accelerator crew in Rehovot for providing the ^{16}O beam, and S. Gorni, O. Skala and the electronic team of the Hebrew University for technical assistance.

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