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EVIDENCE FOR LONG-LIVED PROTON DECAY NOT FAR FROM THE β-STABILITY VALLEY PRODUCED BY THE $^{16}O + ^{197}Au$ REACTION AT 80 MeV

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The reaction $^{16}O + ^{197}Au$ has been studied at a bombarding energy of 80 MeV. Long-lived proton decays with half-lives of about $5.8^{+2.2}_{-1.2}$ h and $67.3^{+46.4}_{-19.5}$ h, proton energies of several MeV and production cross-section in the nb region have been observed. The existence of long-lived high energy isomeric state(s) which decay by protons, directly or by delayed protons, is evident from the data. The possible connection with the second minimum in the potential energy curve is discussed.

Proton radioactivity from the ground state or from an isomeric state and β-delayed proton radioactivity are known for some neutron-deficient nuclei where the lifetimes of the proton emitters or their precursors are in the region of 1 μsec to about 100 sec.[1,2] However, recently[3] evidence for the existence of a long-lived isomeric state ($t_{1/2} \sim 90$ min) which decays by strongly enhanced alpha particles, probably to superdeformed band, has been found in the study of $^{16}O + ^{197}Au$ reaction at 80 MeV. Since, for the evaporation residue nuclei produced in this reaction, the expected excitation energies[4] of the heads of the superdeformed bands (the second minima in the potential energy curves) are above the proton separation energies,[5] the emission of protons from such isomeric states or their descendants is in principle possible. This letter describes the observation of long-lived proton decay produced by $^{16}O + ^{197}Au$ reaction at 80 MeV. Preliminary results of this work have been published before.[6]

The experimental conditions were somewhat similar to those described in Ref. 3. A 900 $\mu g/cm^2$ gold target followed by two 150 $\mu g/cm^2$ C catcher foils were irradiated with 80 MeV ^{16}O beam. All the evaporation residue nuclei and only a small part of the fission and the transfer reaction products were caught on the foils. (A list of the identified nuclei and the measured production cross-sections was given in Refs. 3b and 3e). The catcher foils were then transferred to Jerusalem and placed together in between a $\Delta E \sim E$ telescope (for particle identification) from one side, and a 500 mm^2, 10 mm thick, thin window Ge(Li) detector from the other side. The $\Delta E \sim E$ telescope consisted of a 100 mm^2, 24.6 μ thick, ΔE silicon surface barrier detector, and a 450 mm^2, 300 μ thick, E detector. The solid angle of the telescope was about 12% and of the γ-ray detector 16%. Singles spectra as well as coincidences between the ΔE and E signals and between the ΔE and the γ-ray detectors were recorded. Three runs were performed. In the first and third runs the irradiation time was about 21 h and the total integrated current in each run was about 6000 pμC. In the second run the irradiation length was about 44 hours and the total integrated current was about 13000 pμC. However, in this run the current during the last 15 hours was about 26% higher than the average one, and it was found later on that the target was overheated. The measurements were always started about 100 min after the end of the corresponding irradiation. The coincidence resolving time between the ΔE and E detectors was 0.2 μsec and between the ΔE and the Ge(Li) detector was 0.2 μsec for the first two runs and 1.0 μsec in the third run.

Figure 1a shows the response of the $\Delta E \sim E$ telescope to knock-out protons produced by neutrons emitted from a ^{227}Ac-Be source and a 2 mm thick polyethylene foil situated in front of the ΔE detector. Since only coincidences between the ΔE and E detectors were recorded all the events are to the right of the diagonal line. In a background measurement taken for 139.6 hours, one event, at the very edge of the proton region near the lower broken line, was observed. Figure 1b shows a typical ΔE spectrum which corresponds to the third run and was recorded for about 172 hours. The peak around 5.0 MeV is due to α-particles from ^{205}Po ($t_{1/2} = 1.66$ h; $\alpha/(\beta+\text{EC}) = 0.04\%$) and ^{206}Po ($t_{1/2} = 8.8$ d; $\alpha/(\beta+\text{EC}) = 5.45\%$)[3] which were totally stopped in this detector. It consists of about 1/3 of the whole intensity which is mainly due to the 6.0 MeV group of ^{210}Rn ($t_{1/2} = 2.4$ h). This group shows two components; a peak around 6 MeV due to totally absorbed α-particles, and a broad region which is extended to below 4 MeV due to partially absorbed particles. About 7.5×10^6 counts were recorded in this spectrum. Figure 1c shows the spectrum obtained with the E detector which corresponds to the ΔE spectrum shown in Fig. 1b. The high intensity low energy peak is mainly due to electrons and the broad region up to about 2 MeV is due to α-particles which passed the ΔE detector. Figure 1d shows a two-dimensional $\Delta E \sim E + \Delta E$ spectrum obtained by summing up the results of the three $^{16}\text{O} + {}^{197}\text{Au}$ experiments. In each experiment the measurement took place for about 170 hours and was started about 100 min after the end of the corresponding irradiation. The groups at total energies of about 5, 6 and 7.4 MeV are due to channeling of α-particles with these energies[3] through the ΔE detector. (For instance, 6 MeV α-particles should leave at least 4 MeV in the ΔE detector and the low energy signals at the ΔE detector are due to the channeling effect which was estimated to be about 1% of all the α-particles.) 27 events are seen in the proton region far from the diagonal line, while 20 of them are in the region where most of the protons from the test experiment (Fig. 1a) were seen before. Seven events are seen near the lower broken line.

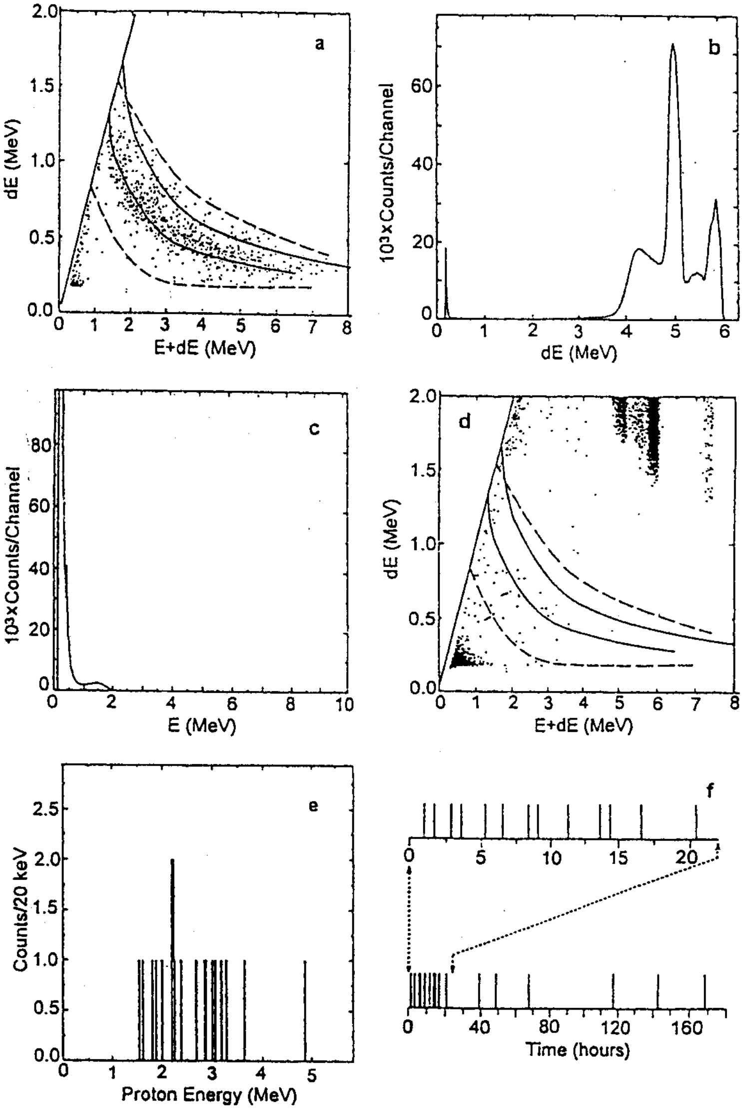

Fig. 1. (a) A two-dimensional ΔE vs $E + \Delta E$ spectrum obtained with knock-out protons due to neutrons emitted from a ^{227}Ac-Be source and a 2 mm thick polyethylene foil situated in front of the ΔE detector. The region in between the two continuous lines is the expected region for protons according to their known stopping power and range values.[7] The two broken lines give the limits where all the protons were seen. The diagonal line represents the $\Delta E = E + \Delta E$ ($E = 0$) dependence. (b) A typical singles ΔE spectrum (see text). (c) A typical singles E spectrum (see text). (d) Similar to (a) but for the combined spectra obtained from the three ^{16}O + ^{197}Au experiments. The two small arrows show the two correlated events with a time difference of 278 ms between them (see text). (e) A proton spectrum obtained from the 20 identified proton events seen in (d) far from the diagonal line and having both the characteristic stopping powers and energies of protons (see text). (f) A time sequence plot where the times at which the individual protons events were observed.

Although they appeared in the region where protons were seen before in Fig. 1a, their relative number is larger. A possible explanation for them is that they are also proton events which however gave too small signals in the ΔE detector, because of channeling or, more likely, due to the large charged space produced by the high α-particle intensity, which exists in the experiment (Fig. 1d). and did not exist in the test experiment (Fig. 1a). Their ΔE and E values could not fit at all with α-particles (ΔE and E pulses much too low) or with electrons (ΔE and E pulses much too large). A statistical analysis[3] shows that the probability that out of 32 events seen in the whole region between the α-particles and the electrons, taking out the region near the diagonal line, 20 will fall accidentally in the specified proton region is very low: $\binom{32}{20} \times 0.22^{20} \times 0.78^{12} = 8 \times 10^{-7}$. 0.22 is the relative area of the strip where protons are expected as compared to the total area. (This probability increases to 1×10^{-4} if one assumes, without a particular reason, an area ratio of 0.3.) Lacking clear identification of the seven events near the lower broken line they were not considered below, although including them in the analysis does not change the conclusions. In addition to the events seen in the region far from the diagonal line, approximately 15 events are seen in the proton region near to this line. The number of random coincidences was estimated to be about 1 in the region far from the diagonal line and about 6 in the region close to this line. The times of occurrence of single coincidence events were recorded in the experiment and it was found that the 1.96 MeV event was followed by the 1.49 event with a time difference of 278 ms. (These two events are marked with small arrows in Fig. 1d.) No triple coincidences between the events seen in Fig. 1d in the proton region far from the diagonal line and γ-rays were found in the experiment. However, four coincidence events in the proton region not far from the diagonal line were found also in coincidence with γ-rays of 72.7, 74.5, 90.1 and 161.7 keV in energy. We were not able to find candidates for the 90.1 and 161.7 keV. The 74.5 and 72.7 keV fit with X-rays of Bi and Pb and they can be random because of the large intensity of these groups in the γ spectra.

Figure 1e shows the obtained proton spectrum using the 20 events seen in the center of the proton region having both the stopping powers and energies of protons. Protons with energies up to 3.3 MeV and one event each at 3.60 and 4.84 MeV are seen in this spectrum. A sharp peak of five events is seen at $E_p = 2.19$ MeV. All the energies of these five events (2.161, 2.177, 2.182, 2.200 and 2.210 MeV) fall within 50 keV, the characteristic width for protons (taking into account the thickness of the source). The probability that by chance 5 out of 20 events, distributed smoothly over 3.5 MeV, will cluster in a peak of 50 keV is: $\binom{20}{4} \times (0.05/3.5)^4 \times (1\text{–}0.05/3.5)^{16} = 1.6 \times 10^{-4}$. (Since one does not know *a priori* where the peak will occur, the first event is not significant. Similar estimate taking into account 19 events, distributed over 2.5 MeV, gives 4.6×10^{-4}.)

In Fig. 1f a time sequence plot, where the times at which the 20 proton events were recorded, is given. Two distinct groups are seen. By using the formulas of

Ref. 8, where the lifetime τ is determined as the arithmetic mean of the individual lifetimes at which the events were observed, half-lives of about $5.8^{+2.2}_{-1.2}$ h and $67.3^{+46.4}_{-19.5}$ h were deduced for these two groups. The deduced half-life of the 2.19 MeV group is $36.9^{+30.0}_{-11.4}$ h which is consistent with the half-life of the long group. The total production cross-section is in the region of 1 nb.

Because of the low intensity of protons and the large intensity of the α-particles as seen in Fig. 1b and Fig. 1a of Ref. 3, the possibility of proton production due to the (α, p) processes on the carbon foils and the surroundings of the source in the vacuum chamber should be considered. The main components in the α-spectra are the 6.04 MeV due to ^{210}Rn ($t_{1/2} = 2.4$ h) and 5.22 MeV due to ^{206}Po ($t_{1/2} = 8.8$ d). In addition ^{211}Po (a daughter of the ^{211}Rn ($t_{1/2} = 14.6$ h)–^{211}At ($t_{1/2} = 7.2$ h) chain) with α-particles of 7.45 MeV is produced in the reaction with a total intensity of α-particles of about 400 times smaller than the 6.04 MeV group of ^{210}Rn. First one notices that the measured half-lives of the protons of $5.8^{+2.2}_{-1.2}$ h and $67.3^{+46.4}_{-19.5}$ h are different from the lifetimes of any of the above-mentioned α-particle groups and in particular from 2.4 h, the half-life of the main group of the α-particles. Secondly it is estimated below that at the most only 1 proton event could have been due to the various (α, p) processes. In principle protons could be produced due to (α, p) processes on the C foils, the Si of the ΔE detector, the Be back window of the chamber, the Al frames of the C foils, the H in the epoxy on the edge of the detector and H, N and O contaminations in the C foils and the Be window. However, because of negative Q values the minimum energy needed for (α, p) reactions on ^{9}Be, ^{12}C, ^{13}C, ^{16}O, ^{17}O and ^{18}O are 9.95, 6.62, 9.70, 10.14, 6.987 and 6.842 MeV, respectively. All of them are above the energies of the main groups of α-particles mentioned above. In addition the expected energies of protons from the ^{27}Al$(\alpha, p)^{30}$Si reaction is between 6 and 7 MeV which is out of the region where protons were found in the experiment. In Table 1 the expected number of protons which could have been seen in the experiment due to the various possible (α, p)

Table 1. Estimates for the possible production of protons due to various (α, p) processes.

E_α (MeV)	Target	$\sigma(\alpha, p)$ (mb)	Impurity (%)	Np
6.04	^{28}Si	10[a]		0.68
6.04; 5.2	^{14}N (in C)	200[a]	0.1	0.02
6.04; 5.2	^{14}N (in Be)	200[a]	0.02[b]	0.02
6.04; 5.2	^{1}H (in C)	1000	0.1	0.06
6.04; 5.2	^{1}H (in edge of detector)	1000		0.32
7.45	^{12}C			0.0[c]
7.45	^{28}Si	150[a]		0.03
				1.13

[a]Refs. 9–11.

[b]Manufacturer specifications.

[c]Energies of proton outside the measured range ($E_p(\max) = 1.1$ MeV).

processes, are given. For the Si case an effective thickness of 1 mg/cm^2, which attenuate the energy of the α-particles by about 0.5 MeV, was assumed. Since the energy of the α-particles are below the Coulomb barrier, further reduction of the energy reduces the cross-section considerably. In the case of knock-out protons, one has to take into account only the protons which are produced at such angles where their accompanied α-particles do not reach the detector. (The α-particles emitted in this process are above about 2 MeV. They will leave a very large pulse in the ΔE detector, as compared to 0.2–1.0 MeV. the characteristic energy loss of protons (see Figs. 1a and 1d). It is seen in Table 1 that only about 1 event, out of 20(27) seen, could be accounted for by the various (α, p) processes. Out of them only a total of 0.1 events may be due to the assumed contaminations of H and N. To explore this question even further an attempt was made to study the response of the experimental system to α-particles from an emanation ^{212}Pb(ThB) source. Two α-particle groups are emitted from this source at 8.741 MeV (64%) and at 6.05 MeV (36%). To simulate the experimental situation described above the source was collected on a 12.5 mg/cm^2 Au foil and in addition a 12.5 mg/cm^2 Au degrader was used during the measurements. The Au backing from one side and the degrader from the other side reduced the energy of the 8.74 MeV group to 6.1 MeV very close to the energy of the 6.04 MeV group of ^{210}Rn mentioned above. However, the 6.05–6.09 MeV group from the source was degraded to 2.8 MeV with a large tail down to zero energies. As a result many random coincidences were added to the $\Delta E \sim E$ coincidence measurements as compared to the measurement with the C catcher foils, where the intensity in this region of energies was very low (see Fig. 1b). Therefore the measurements were limited to rather weak sources. Two 150 μg/cm^2 C catcher foils which were used in the original experiments were situated on the source to study their possible contribution to the proton events. Two experiments were performed and in each of them 1 coincidence event was recorded in the proton region of the $\Delta E \sim E$ coincidence spectra, while 1.3 ± 0.4 and 1.8 ± 0.6 events due to random coincidences were estimated. (The estimation of 1.3 and 1.8 events mentioned above was done by a comparison with measurements where stronger sources and hence a larger number of random coincidences were measured, and by following their decay with time.) Assuming that 1 of the 2 observed events (which could very well be also a random event) be due to (α, p) processes and scaling according to the different intensities, one gets an upper limit of 3 events (out of 20 or 27 seen) in the ^{16}O + ^{197}Au experiments which may have been due to these processes.

Thus mostly all the identified protons seen in the experiment could not be due to (α, p) processes in the source or its surroundings and one is led to conclude that they are due to decay from highly excited long-lived isomeric states which were produced by the ^{16}O + ^{197}Au reaction, or from their descendants. The excitation energies of the proton emitting states should be above the proton separation energies. The lowest proton separation energy for the evaporation residue nuclei or their descendants is in ^{209}Fr at 1.43 MeV.[5] As seen in Fig. 1e, protons with energies of

1.5 to 3.3 MeV were seen in the experiment (and perhaps also at 3.6 and 4.8 MeV). Therefore the excitation energies of the proton emitting states should be at 2.9 to 4.73 MeV or higher. (The two correlated events at 1.96 and 1.49 MeV (if it is a proton) with a time difference of 278 ms indicate that the excitation energies might be considerably higher, above about 8.5 MeV.) The expected[1] half-lives for protons of 1.5 to 3.3 MeV for nuclei with, for instance, $Z = 87$ are very short, from 2.5×10^{-4} to 6×10^{-14} s. (The calculated value for 2.19 MeV protons is 2×10^{-9} s.) The exact origin of the protons and the character of the isomeric state(s) with lifetime in the hour region is not clear and further work is needed. Recently[3] a long-lived isomeric state, which decays by low energy α-particles probably to superdeformed band states in the second well of the potential energy curve, has been found using the same reaction. It was argued that this isomeric state may be a high spin isomer in the second well of the potential energy curve. The observed protons may perhaps be due to similar phenomenon, namely a decay of a high spin isomer(s) in the second well of the potential energy curve, directly, or by delayed protons, to the normal states. The expected excitation energies of the second minima of the nuclei produced in the reaction are quite high.[4] Proton decay from such states or from their descendants, may lead to different states in the daughter nuclei, resulting with various proton energies as seen in Fig. 1e. The situation is similar to the known β-delayed proton radioactivity,[2] rather than to proton radioactivity from the ground state.[1]

It should be mentioned that unidentified low energy particle groups were found before[12,13] in studies of actinides produced by secondary reactions.[14] Some of them may perhaps be strongly enhanced α-particle groups,[3] but the very low energy ones like for instance the 3.0 and 4.0 MeV particle groups seen with the Am source[12,15] may be due to protons.

In summary, long-lived proton decays with lifetimes of about 6 and 70 hours have been seen in the $^{16}O + ^{197}Au$ reaction at 80 MeV. The existence of long-lived highly excited isomeric state(s) which decay by protons, directly or by delayed protons, is evident from the data. The character of the isomeric state(s) is not clear, and the possibility of proton transitions from states in the second well of the potential energy curve to the normal states is raised.

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