Nuclear isomers in super heavy elements as stepping stones toward the island of stability

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A long standing prediction of nuclear models is the emergence of a region of long lived or even stable super heavy elements (SHE) beyond the actinides. These nuclei owe their enhanced stability to closed shells in the structure of both protons and neutrons[1-3]. However, theoretical approaches to date disagree with each other and it is left to experiment to explore the shores of the "Island of Stability".

The bulk of experimental effort so far has been focused on direct creation of SHE in heavy ion fusion reactions, leading to the production of elements up to Z=118[4,5]. Recently, detailed spectroscopic studies of nuclei beyond fermium (Z=100) aimed at

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untangling the underlying single-particle structure of SHE have become possible[6,7]. Here we report on an in-depth study of the heaviest nucleus studied in this manner to date, the nobelium isotope ^{254}No with 102 protons and 152 neutrons. We find three excited structures, two of which are isomeric. One of these structures is firmly assigned to a two-proton excitation. These states are highly significant as their location is sensitive to single particle levels above the predicted Z=114 shell gap and thus provide a microscopic benchmark for nuclear models aimed at the understanding of SHE.

One key gap in our understanding of the chemical building blocks of the universe is the question of the heaviest element that can exist. Intimately linked to this question is the problem in nuclear physics of the shell stabilisation of super heavy nuclei. In a simplistic model of the nucleus the repulsive force between more than 100 positively charged protons is sufficient to overcome the attractive strong nuclear force and induce the nucleus to fission. However, protons and neutrons fall into shell structures which, analogous to the enhanced stability of noble gases, give extra binding to nuclei in a closed shell configuration. The largest binding comes from a coincidence of closed shells for both protons and neutrons, as in ^{16}O (Z=N=8), ^{40}Ca (Z=N=20), ^{132}Sn (Z=50, N=82) and ^{208}Pb (Z=82, N=126). The successful explanation of these nuclear magic numbers (Z= N = 2, 8, 20, 28, 50, 82 and N=126) via a large spin-orbit splitting led to the 1963 Nobel prize in physics for Maria Goeppert-Mayer and Hans Jensen. In super heavy nuclei the spin-orbit interaction can open substantial shell gaps, e.g. between the proton $2f_{7/2} – 2f_{5/2}$ spin orbit partners. A nucleus with 114 protons will fill all orbitals up to the

$2f_{7/2}$ shell, and it is the strength of the spin-orbit interaction that determines the size of the Z=114 gap, as schematically shown in Figure 1. Modern theoretical approaches disagree on the size and position of this shell gap. The microscopic-macroscopic models with various parameterisations of the nuclear potential predict Z=114 and N=184. Calculations using self-consistent mean-field approaches have been performed by several authors and broadly fall into two categories, namely relativistic versus non-relativistic approaches. In both cases the splitting between the $2f_{7/2}$-$2f_{5/2}$ spin-orbit partners is not sufficient to open a gap. Most non-relativistic mean-field calculations favour Z=124,126 and N=184, while the relativistic mean-field models show that the effects of magic numbers of single nucleonic configurations valid in lower mass magic nuclei (Sn, Pb, etc) are dissolved in favour of more extended regions of additional shell stabilization[1], centred mainly around Z=120, N=172,184 or Z=126, N=184. Theoretical and experimental progress in this area has recently been reviewed by a number of authors[1-8].

Experimental data for SHE are scarce because the production cross sections for these elements drop rapidly with increasing proton number, down to the 1 picobarn level for element 112[9]. With this cross section, a few atoms per month can be produced. This means that the majority of data available yield integral quantities such as half-lives, decay modes and alpha decay Q-values, but are insensitive to the details of the nucleonic shell structure. However, recently it has become possible to perform spectroscopic studies in nuclei approaching the island of stability, such as ^{254}No (Z=102, N=152). Nuclei in this region are deformed, and the degenerate spherical single particle orbitals split in a well defined and adequately understood manner into components according to the projection of the angular momentum onto the symmetry axis of the nucleus, the K-quantum number. Orbitals originating above the relevant spherical proton shells (such as $2f_{5/2}$) come close to the Fermi level in a deformed nucleus and thus play a key role in the formation of excited states.

In nuclei with even numbers of protons and neutrons, the ground state always has K=0. Configurations with large values of K thus require a decay path to the ground state that changes this projection gradually. If such intermediate configurations do not exist, the high-K state becomes isomeric[10]. This gives a very clear and unique experimental signature because the isomeric states can readily be identified from their decay times and paths. It has been suggested that in super heavy nuclei the occurrence of isomeric states may be a key factor in the enhanced stability of these nuclei, with isomeric lifetimes exceeding those of the ground states[1,11].

In this work we have chosen the Z=102 nucleus ^{254}No as it provides an ideal laboratory for these studies. It is produced with a reasonable cross section of 2 microbarns, which allows a production rate of 200 atoms per hour, sufficient for detailed spectroscopic studies. Previous measurements[12] in the 1970's tentatively proposed an isomeric configuration in ^{254}No whose existence was recently corroborated through the observation of conversion electron cascades[13]. ^{254}No also has attracted considerable theoretical interest[14-17] since it became the first transfermium nucleus whose rotational structure was established in 1999[18].

In view of the profound implications of this isomer for nuclear models it is important not only to verify its existence and determine the half-life, but also to elucidate the decay path and determine its configuration. We report here on the findings of experiments performed at the Accelerator Laboratory of the University of Jyväskylä, Finland. Similar, but not identical, results on the isomers in ^{254}No have been obtained from experiments at Argonne National Laboratory[19].

The ^{254}No ions were produced via the ^{208}Pb (^{48}Ca, 2n)^{254}No reaction at a beam energy of 219 MeV. They were separated from the beam and unwanted reaction products in the gas-filled recoil separator RITU[20] before being implanted in a double sided position sensitive Si detector (DSSD) at the heart of the GREAT spectrometer[21]. Long lived isomeric nuclei then decayed to the ground state, emitting gamma rays, X rays, and conversion and Auger electrons. Conversion and Auger electrons were detected in the same pixel of the DSSD where they summed to a total deposited energy of up to 600 keV. This provided a clean signal for the decay of the isomer, a method originally suggested by Jones[22]. The gamma and X rays were detected in prompt coincidence with this electron signal in a large segmented planar Ge detector in close proximity to the DSSD and a large Clover Ge detector outside the detector chamber. Finally the ground state of ^{254}No decays via its characteristic 8.1 MeV α decay with a half-life of $T_{1/2} = (51.2 \pm 0.4)$ s and is recorded in the same DSSD pixel. It is this characteristic sequence of implanted recoil, one or more isomeric decays followed by the characteristic alpha decay all in the same detector pixel that allows us to firmly assign the observed isomeric decays to ^{254}No.

Figure 2 shows the time distribution of the electron signal following the recoil implantation. The long-lived isomer with a half-life of $T_{1/2} = (266 \pm 2)$ ms is clearly seen (Inset in Fig. 2a). In addition a new, short-lived isomer with a half-life of $T_{1/2} = (184 \pm 3)$ μs is observed (Inset in Fig. 2b) to feed the 266 ms isomer. From the electron energy distributions (Fig. 2a,b) and the gamma ray spectra (Fig. 2c,d) associated with these isomers one can deduce the decay path in some detail and we propose the level scheme shown in Figure 3. The 266 ms isomer decays via a 53 keV E1 transition into an excited two-quasi particle band feeding the 7^+ rotational band member. The E1 character of this transition is deduced from the observed very small internal conversion coefficient. From the branching ratios between stretched E2 and mixed E2/M1 transitions $R_1 = I_\gamma(81$ keV$)/I_\gamma(150$ keV$)$ and $R_2 = I_\gamma(69$ keV$)/I_\gamma(126$ keV$)$ a value for the g-factor of the band can be extracted assuming the validity of the rotational model. This g-factor is sensitive to the nucleonic configuration. We find $g_K = (0.87 \pm 0.14)$.

This band in turn decays via low energy M1 and E2 transitions to the band head before decaying to the ground state band via two prominent transitions at 841 and 943 keV. The band head spin was earlier determined to be J = 3 [23]. Here the multipolarity of the 841 and 943 keV gamma transitions has been determined to be M1, which firmly establishes the band head spin and parity as $J^\pi = 3^+$. As the full decay path is known, the isomer can be placed at an excitation energy of 1293 keV.

It is this isomer whose existence was inferred from indirect evidence by Ghiorso et al.[12]. In their work the ^{254}No ions were produced in a fusion-evaporation reaction and captured in a He gas jet which deposited them on the surface of a wheel. The wheel was stepped past a number of detection stations which recorded the characteristic 8.1 MeV α-decays from ^{254}No. They also noticed the detection of 8.1 MeV α-decays in detectors which were no longer adjacent to an irradiated spot on the wheel. From this they deduced the existence of an isomeric state with a half life of $T_{1/2} = (280 \pm 40)$ ms, which decays via the emission of a high energy (≈ 1 MeV) gamma ray. They argued that this gamma ray imparts sufficient recoil momentum to the ^{254}No ion to allow it to leave the surface of the wheel for subsequent deposition onto the surface of a detector. The subsequent α decay of the ground state could then be recorded independent of the position of the wheel. The present half-life of $T_{1/2} = (266 \pm 2)$ ms agrees very well with the previous value[12]. In addition, the high energy gamma rays (841 and 943 keV) observed in this work can provide the recoil momentum to the ^{254}No ions necessary to explain the tentative findings of Ghiorso et al[12] and thus provide a solution for a thirty year old experimental puzzle.

Tantalising first glimpses of a short-lived isomer were seen in our earlier experiments[24], but due to low statistics we were unable to deduce a half-life or a decay scheme. In this work the short-lived 184 μs isomer has been unambiguously identified. It feeds the 266 ms isomer via a cascade of transitions including a prominently visible 606 keV gamma ray (see Fig 2d). Other experimental signatures essential to untangling the complex decay path of this isomer are given by the rather large total energy emitted in the form of conversion and Auger electrons (Fig 2b) and the presence of strong L and K X rays which clearly show that the majority of these transitions are highly converted and therefore do not proceed via gamma-ray emission. Taking into account the measured total energy this isomer must lie at least 1.2 MeV higher in excitation energy than the 266 ms isomer. We have also modelled the decay path of this isomer to determine a lower limit on the spin of this state and find the best agreement of the calorimetric electron signal if one assumes a decay path that changes the total angular momentum by 6-8 units, suggesting a spin of $J \geq 14$ for this isomer.

From the single particle orbitals predicted around the Fermi surface both a two neutron structure $\{7/2^+[624]_\upsilon \times 1/2^+[620]_\upsilon\}^{(3+)}$ and a two proton structure $\{1/2^-[521]_\pi \times 7/2^-[514]_\pi\}^{(3+)}$ can be formed. To distinguish between these configurations experimentally the g-factors of both these configurations can be calculated from the g-factors of the individual orbitals to give $g_K^{\upsilon\upsilon}=0.530$ and $g_K^{\pi\pi}= 0.824$. The experimental value of $g_K^{exp}=(0.87 \pm 0.14)$ band clearly identifies the K=3 bandhead as a two-proton excitation involving the $1/2^-[521]_\pi$ orbital stemming from the spherical $2f_{5/2}$ orbital above the Z=114 shell. Thus it is now possible for theoretical models to use this firm assignment as a stepping stone to constrain their parameterisations used in the prediction of the spherical super heavy nuclei.

A unique feature of this mass region is the occurrence of both two-proton and two neutron high-K isomers with spin and parity $J^\pi=8^-$ at low excitation energies. The structure of the 266 ms isomer has been calculated by a number of authors[11,12,25,26]. A two-quasi proton state with a structure $\{7/2^-[514]_\pi \times 9/2^+[624]_\pi\}^{(8-)}$ is found in all calculations. The low-lying isomeric two quasi neutron state is generally calculated with structures $\{7/2^+[624]_\upsilon \times 9/2^-[734]_\upsilon\}^{(8-)}$ and $\{7/2^+[613]_\upsilon \times 9/2^-[734]_\upsilon\}^{(8-)}$. In all calculations the structures are predicted to lie at excitation energies between 1 and 2 MeV. We compare the predictions and recent calculations using the Projected Shell Model (PSM)[27,28] with the experimental data in Figure 4. The majority of calculations consistently predict the two-neutron configuration to lie lowest in energy, although the two proton configuration should be favoured for the 8^- state over the two neutron configurations as the decay from the isomeric $\{7/2^-[514]_\pi \times 9/2^+[624]_\pi\}^{(8-)}$ state to the $\{1/2^-[521]_\pi \times 7/2^-[514]_\pi\}^{(3+)}$ state only involves the transition of a single proton rather than that of two protons and two neutrons. However, the long half-life of the isomer might be partially due to just such a change. This highlights the problems faced by experiments if they have to rely on guidance from theory to make structural assignments in this region: theoretical calculations still cover a wide range and it is precisely the kind of detailed data presented here that will have a major impact on the development of these models.

The structure of the 184 μs isomer has not been discussed in the literature so far. We propose that this state is built on a two-proton two-neutron four quasiparticle configuration. One attractive choice is the $[\{7/2^-[514]_\pi \times 9/2^+[624]_\pi\}^{(8-)} \times \{7/2^+[624]_\upsilon \times 9/2^-[734]_\upsilon\}^{(8-)}]^{(16+)}$ calculated to lie at 2.75 MeV, i.e. the product of the two-proton and two-neutron choices for the 8^- isomer, analogous to the well-known 16^+ isomer[29] in ^{178}Hf. This assignment is favoured by PSM calculations, but it must be stressed that this assignment on experimental grounds alone is highly tentative and may change as the experimental situation improves.

The unique key result of our work is the firm experimental assignment of a state in the Z=102 nucleus ^{254}No involving the crucial $2f_{5/2}$ proton orbital at the heart of the question of shell stabilised super heavy elements. We have shown experimental evidence for two isomeric states in ^{254}No, determined their half-lives and decay paths and assigned two- and four-quasi particle configurations to them. Similar two-quasiparticle high-K isomeric states are expected in neighbouring nuclei, e.g. ^{252}No[30] and ^{248,250}Fm[12]. The production cross sections for these nuclei are lower than for ^{254}No but the prospect of establishing a systematic picture of the dominant quasiparticle energies in this region is essential to arrive at an understanding of super heavy elements. The states studied in this work provide the most stringent test cases for the predictive power of model calculations in the deformed Z~100 region so far.

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Acknowledgements

We thank T.L. Khoo, S.K. Tandel, P.D. Stevenson and P.M. Walker for many fuitful discussions. This work has been supported by the European Union Fifth Framework Programme “Improving Human Potential – Access to Research Infrastructure and the EU 6th Framework programme "Integrating Infrastructure Initiative - Transnational Access" (EURONS). I.G. Darby, A. Steer and M. Venhart gratefully acknowledge a European Community Marie Curie Fellowship. Support by the Academy of Finland under the Finnish Centre of Excellence Programme 2000-2005 is gratefully acknowledged. Funding is also gratefully acknowledged from the UK EPSRC. This work was supported in part by the NSF and the U.S. Department of Energy, Office of Nuclear Physics.

Figure Captions

Figure 1: Schematic illustration of the spherical single proton orbitals in the region of super heavy elements. The strength of the spin-orbit interaction determines the size of the gaps at 114 and 120. Left: large spin-orbit coupling. Right: reduced spin-orbit coupling.

Figure 2: The calorimetric electron signals from the two isomer decays are shown in panels a) and b) respectively while the insets show the time distribution between the recoil implantation and the decay of the isomer. The different decay paths result in different energy signatures with endpoint energies of ~ 400 keV for the 266 ms isomer and ~ 600 keV for the 184 μs isomer. In panels c) and d) the gamma rays observed in prompt coincidence with these electron signals are shown. The difference in the ratio of the K and L X rays is helpful for unravelling the decay paths.

Figure 3: Proposed level scheme of ^{254}No. The 266 ms 8^- isomer is connected to the ground state via an excited 3^+ two quasi particle $\{1/2^-[521]_\pi \times 7/2^-[514]_\pi\}^{(3+)}$ band. The 184 μs (16^+) isomer populates the 8^- isomer band. Details of the exact decay path are tentative, but include a prominent 606 keV gamma ray and a cascade of conversion electrons which establish its position in the level scheme.

Figure 4 Configurations proposed by several authors of the 266 ms isomer. The lowest configurations are the two-neutron $\{7/2^+[624]_\upsilon \times 9/2^-[734]_\upsilon\}^{(8-)}$ (a) and $\{7/2^+[613]_\upsilon \times 9/2^-[734]_\upsilon\}^{(8-)}$ (b-e) two-quasi particle configurations. The two-quasi proton $\{7/2^-[514]_\pi \times 9/2^+[624]_\pi\}^{(8-)}$ configuration lies higher in energy in nearly all cases. Column a) shows calculations from Y Sun (this work), b) from F.R. Xu[11], c) from V.G. Soloviev[25] and d) from Yu.A. Lazarev[26].

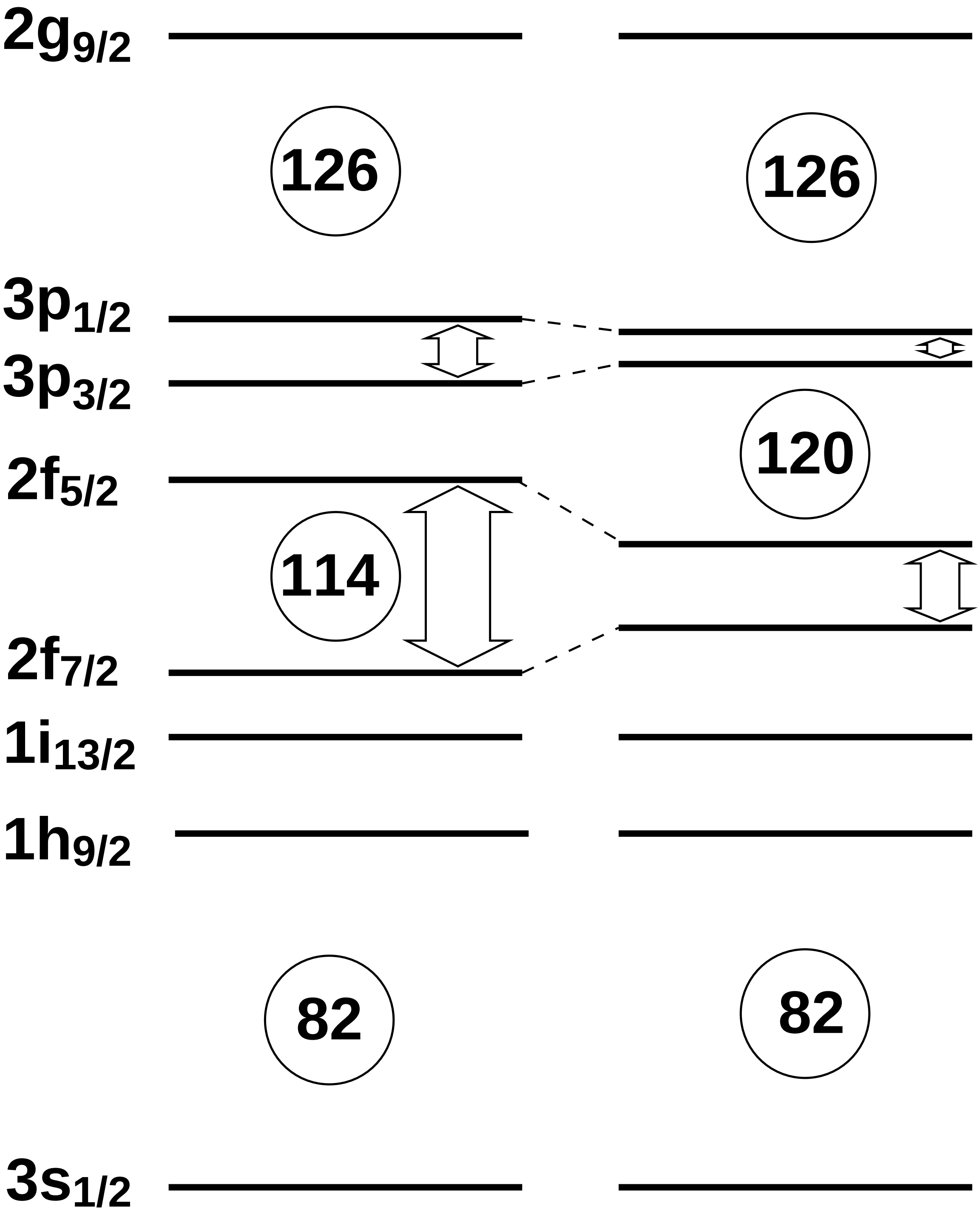

Figure 1

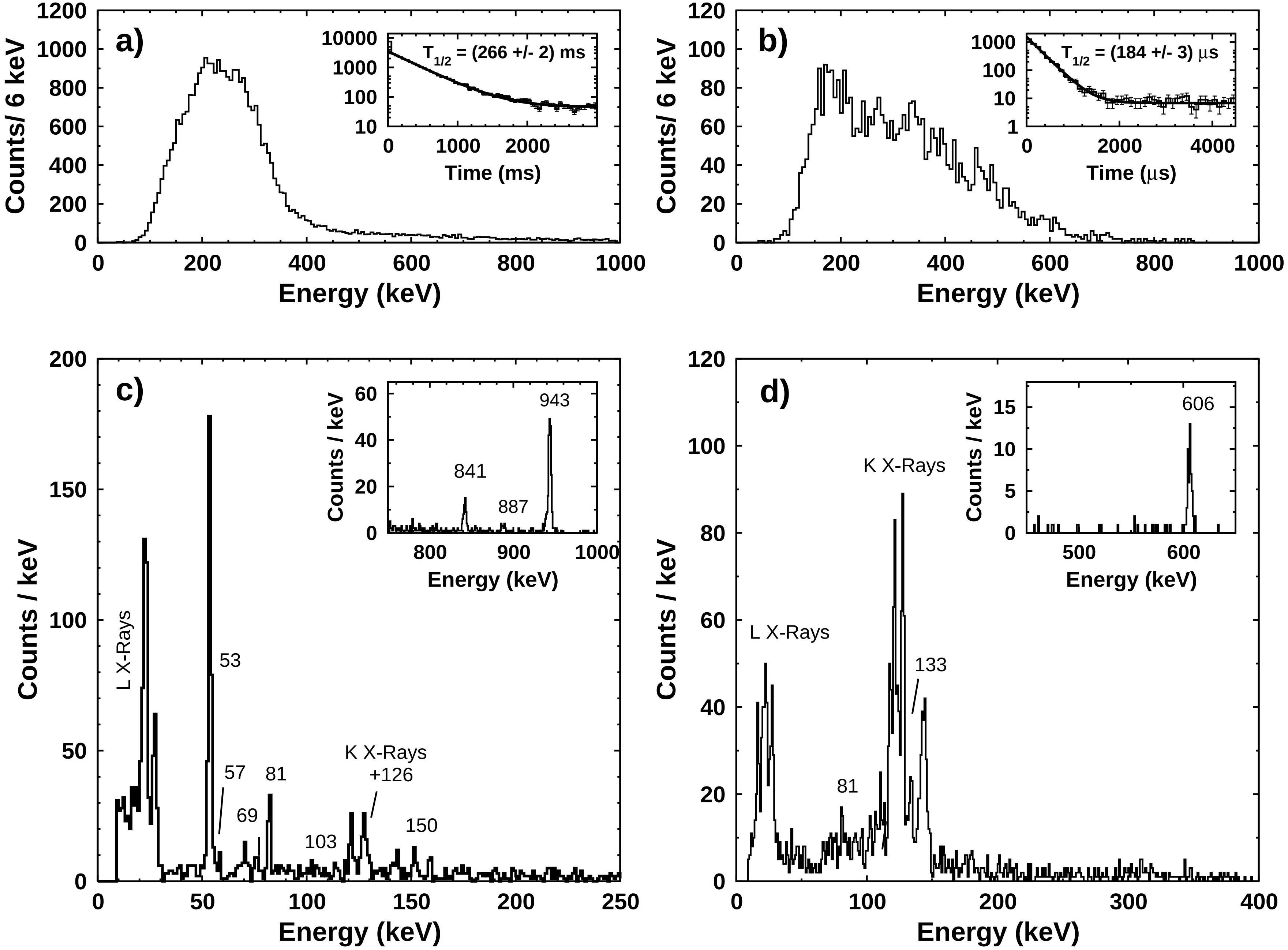
a)
Counts/ 6 keV
1200
1000
800
600
400
200
0
0
200
400
600
800
1000
Energy (keV)
$T_{1/2}$ = (266 +/- 2) ms
10000
1000
100
10
0
1000
2000
Time (ms)
b)
Counts/ 6 keV
120
100
80
60
40
20
0
0
200
400
600
800
1000
Energy (keV)
$T_{1/2}$ = (184 +/- 3) µs
1000
100
10
1
0
2000
4000
Time (µs)
c)
Counts / keV
200
150
100
50
0
0
50
100
150
200
250
Energy (keV)
L X-Rays
53
57
69
81
103
K X-Rays
+126
150
Counts / keV
60
40
20
0
800
900
1000
Energy (keV)
841
887
943
d)
Counts / keV
120
100
80
60
40
20
0
0
100
200
300
400
Energy (keV)
L X-Rays
81
K X-Rays
133
Counts / keV
15
10
5
0
500
600
Energy (keV)
606

Figure 2

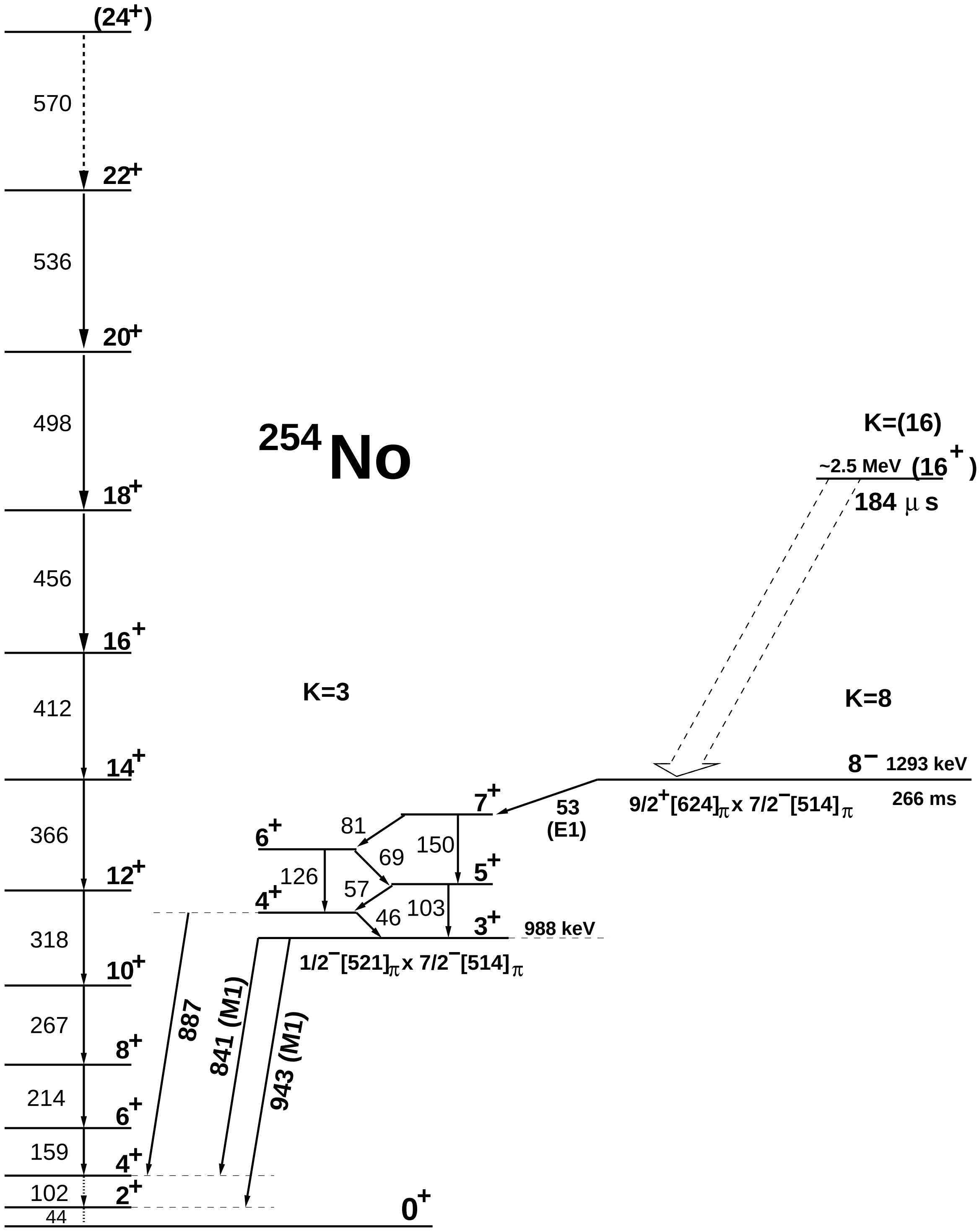
254No
(24+)
570
22+
536
20+
498
18+
456
16+
412
14+
366
12+
318
10+
267
8+
214
6+
159
4+
102
2+
44
0+
K=3
6+
81
7+
150
69
126
5+
57
4+
103
46
3+
988 keV
1/2−[521]π x 7/2−[514]π
887
841 (M1)
943 (M1)
53
(E1)
K=(16)
~2.5 MeV
(16+)
184 μs
K=8
8−
1293 keV
9/2+[624]π x 7/2−[514]π
266 ms

Figure 3

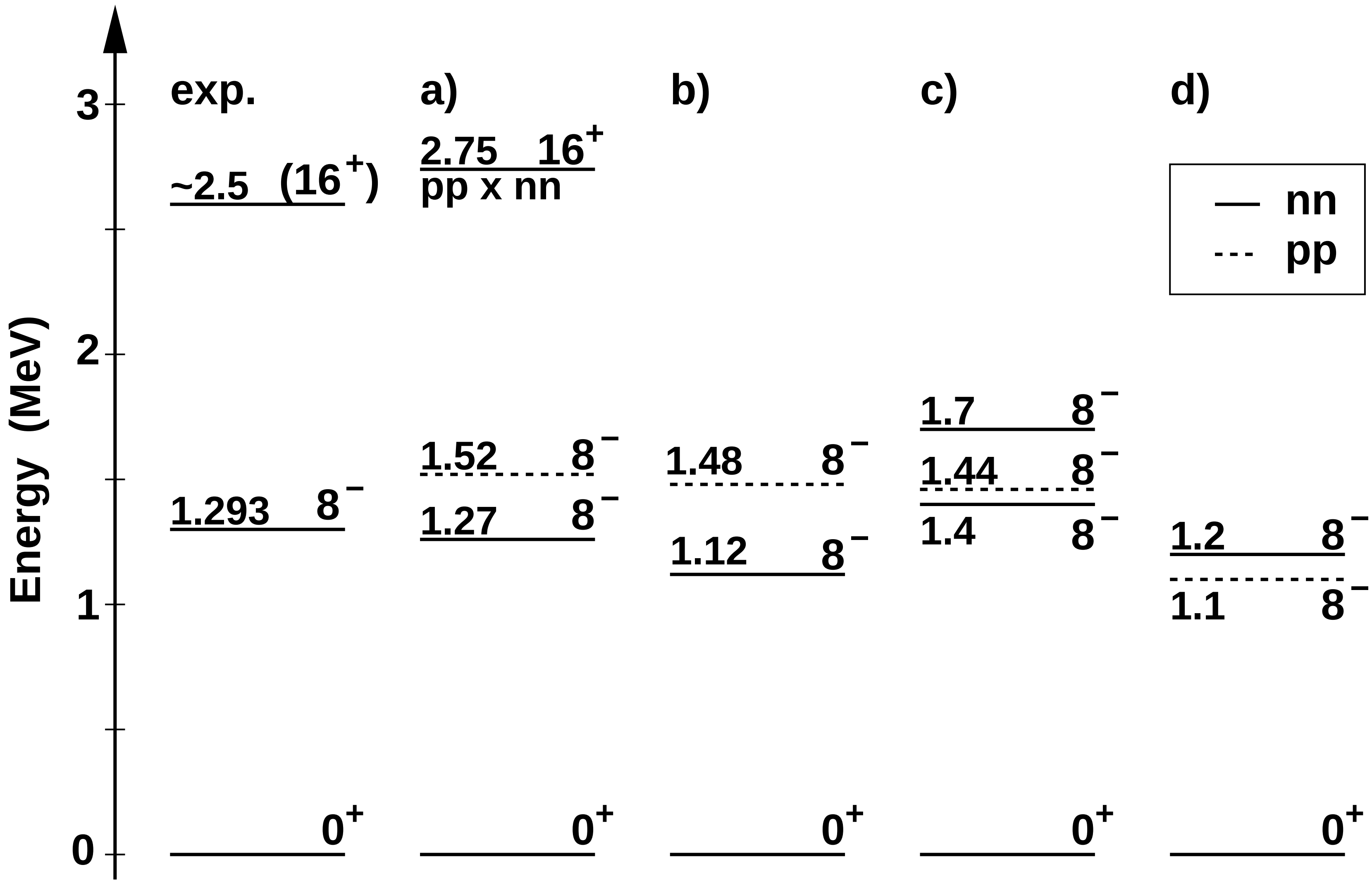
Energy (MeV)
0
1
2
3
exp.
a)
b)
c)
d)
~2.5 (16+)
1.293 8−
0+
2.75 16+
pp x nn
1.52 8−
1.27 8−
1.48 8−
1.12 8−
1.7 8−
1.44 8−
1.4 8−
1.2 8−
1.1 8−
nn
pp

Figure 4