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Connections between high-K and low-K states in the s-process nucleus ^{176}Lu

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Gamma-ray branches that connect high-K states to low-K states in the s-process nucleus ^{176}Lu were observed, thus providing a link between the 58 Gyr, 7^- ground state and the 5.3 h, 1^- isomeric state. High sensitivity and unambiguous placement were achieved through the study of the decay of the 58 μs $K^\pi = 14^+$ isomer using γ-γ-coincidence measurements. The large number of decay paths from the isomer provides a means of populating a broad selection of states from above, resulting, paradoxically, in higher sensitivity than in cases where low-spin input reactions are used. The out-of band decay widths important for excitation processes in stars are quantified.

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Nature's heaviest naturally occurring odd-odd isotopes, ^{176}Lu and ^{180}Ta, have been the subject of considerable attention, occupying a special place in nuclear-structure and nuclear-astrophysics investigations. They are well deformed and manifest related structures at or near the ground state, formed by coupling the unpaired neutron and proton angular momenta. In this region of the periodic table, orbitals with a relatively high projection on the symmetry axis of the individual particle angular momentum (Ω) occur near both proton and neutron Fermi surfaces. Thus, high and low-K states can be formed at comparable energies by parallel or antiparallel coupling of the individual nucleons to give the total projection $K = |\Omega_p \pm \Omega_n|$.

^{180m}Ta is unique being both the least abundant isotope and the only naturally occurring nuclear isomer (excitation energy of 77 keV). It is formed by aligning the (Nilsson) orbitals to produce a $K^\pi = 9^-$ state from the $\pi 9/2^-[514] \otimes \nu 9/2^+[624]$ configuration. Its lifetime of $\tau_m > 1.2 \times 10^{15}$ years is caused by the fact that its β and γ decays are severely restricted. A transition to the ground state, for example, will involve a low energy and very high multipolarity transition, resulting in a classic "spin" isomer [1]. In contrast, the $K^\pi = 1^+$ ground state formed from the $\pi 7/2^+[404] \otimes \nu 9/2^+[624]$ configuration with $K = |\Omega_p - \Omega_n|$, β decays with a lifetime of a few hours.

The observed abundance and creation of ^{180m}Ta through stellar nucleosynthesis remains the subject of conjecture (see Ref. [2]). Furthermore, laboratory experiments [3,4] reveal substantial cross sections for photon excitation from the 9^- isomeric state to higher lying states (presumably of intermediate K), which have fast decays to the short-lived ground state, making its survival sensitive to the conditions that produce intense fluxes of photons in stellar environments [4,5].

In ^{176}Lu, the situation is similar, but inverted in that the $K^\pi = 7^-, \pi 7/2^+[404] \otimes \nu 7/2^-[514]$ configuration forms the long-lived ground state ($\tau \sim 10^{11}$ years) with a slow β decay to ^{176}Hf, while the antiparallel coupling of the same configuration gives a 123 keV 1^- state (with $K^\pi = 0^-$) that β decays with a mean life of about five hours. As a result of its long ground-state lifetime and being shielded from the r process by the stable nuclide ^{176}Yb, ^{176}Lu was proposed as an s-process chronometer, but its reliability in that context was questioned [6–10]. However, it remains a possible s-process thermometer [7,11].

Its survival and abundance is complicated by the probability for its destruction following the direct population of the short-lived β-decaying state in neutron capture. Also the possibility of excitation from the 1^- state to intermediate states that then decay to the long-lived ground state could increase its abundance, while the opposite path, excitation from the ground state to the short-lived 1^- state, could reduce that abundance. The properties of purported intermediate states will control the sensitivity of the equilibrium between ^{176g}Lu and ^{176m}Lu to the stellar temperature [9]. A 5^- excited state at 839 keV (a member of the $K^\pi = 4^-$ band at 723 keV) with an $E2$ decay to the ground state and a path through the 4^- band to the 1^- state was examined as a candidate for such a state [9,10] and continues to be a focus [12], but no other clear connecting paths between the ground state and low-K structures have been established. Factors that were considered recently include possible mixing through chance degeneracies [13], but the implied γ-ray transitions have not been observed.

The experimental difficulty in identifying and characterizing the states of interest, variously called "relay levels," "gateway states," "intermediate states," and so on is that ^{180}Ta and ^{176}Lu are not accessible by conventional (high spin) heavy-ion fusion-evaporation reactions. While low-spin reactions (such as neutron capture) are valuable, these are less amenable to the γ-γ coincidence techniques that are useful in establishing the placement of γ-ray transitions, particularly in odd-odd nuclei where the level densities are high and chance coincidences in γ-ray energies are common. Although the states of importance are likely to be of intermediate spins, in terms of spectroscopy, the ideal experimental situation is to be able to populate them from higher-spin states above and thus enable

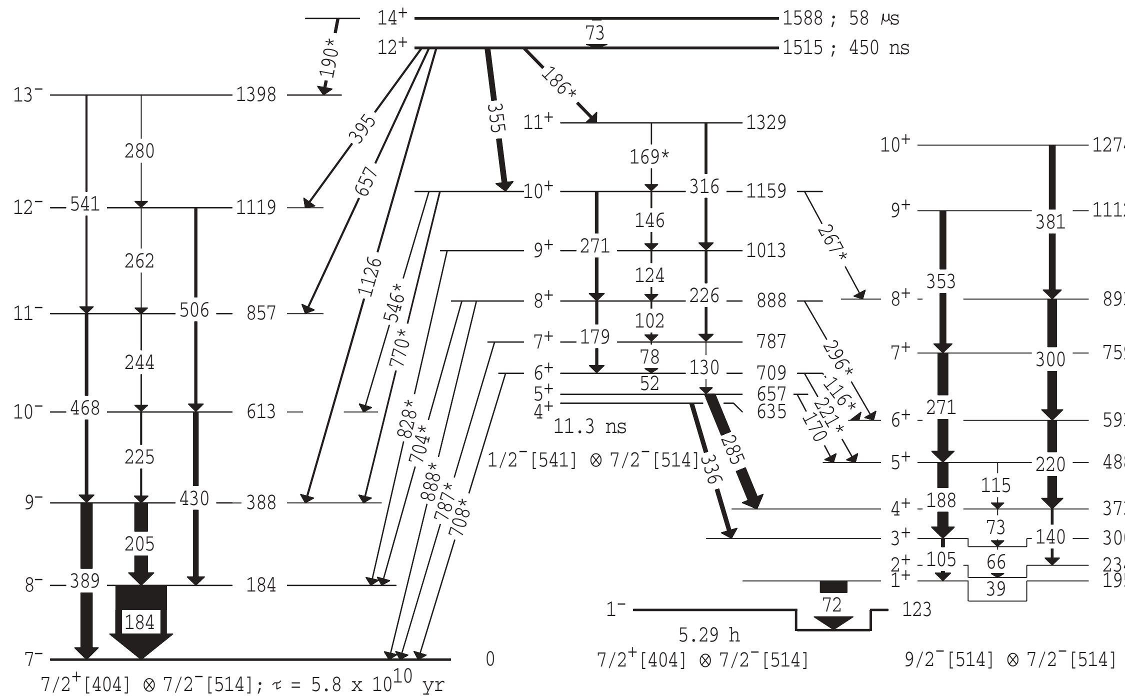

FIG. 1. Partial scheme showing selected states populated in the decay of the $K^{\pi} = 12^{+}$ and 14^{+} isomers in ^{176}Lu and higher members of the 1^{+} band. Asterisks indicate the newly identified transitions. See Ref. [14] for other branches from the isomers.

γ-γ measurements. By gating on transitions that feed a particular state, it is possible to identify branches out of the state, unambiguously. Hence, significant population of higher-spin states is an advantage provided that the population is not too selective.

The presence of a long-lived four-quasiparticle isomer ($K^{\pi} = 14^{+}$) that predominantly decays to a four-quasiparticle 12^{+} isomer, which itself decays via numerous branches to both intrinsic states and rotational levels of intermediate spin, provides just that situation in ^{176}Lu. These isomers and an extensive level scheme for ^{176}Lu were identified using the incomplete fusion reaction ^{176}Yb(^{7}Li,α3n)^{176}Lu and various time-correlated spectroscopic techniques, as partly reported in Ref. [14].

With that knowledge as a base, we used deep inelastic reactions and Gammasphere [15] to obtain new information. The combination of a long lifetime, good population, and high-efficiency instrumentation, thus allowing time and multiple-γ constraints to be applied, provided the ability to isolate and identify weak decays. The present results are from measurements made using 6.0 MeV/nucleon ^{136}Xe beams delivered by the ATLAS facility at Argonne National Laboratory. Nanosecond pulses, separated by 825 ns, were incident on a metallic Lu target enriched to 47% in ^{176}Lu, ~6 mg/cm^2 thick, with a 25 mg/cm^2 Au foil directly behind. The target thickness was such as to integrate down over the main yield of inelastic processes from ~20% above the Coulomb barrier. During this experiment, Gammasphere had 100 detectors in operation. Triple coincidences were required and the main data analysis was carried out with γ-γ-γ cubes with various time-difference conditions and also with time constraints relative to the pulsed beam to select different out-of-beam regimes. More details on the experimental conditions and analyses are reported in Ref. [16].

Measurements were also carried out using a macroscopically chopped beam with (beam on)/(beam off) conditions of 100μs/300μs. Out-of-beam dual coincidence events were recorded in reference to a precision clock and γ-γ matrices were constructed for six contiguous out-of-beam time regions. This allowed states following the 58 μs isomer to be examined with dual rather than triple coincidences, providing an independent measure of branching ratios. It was also possible to reduce the contamination from long isomers in other nuclei in some of the gates by subtracting equivalent gates obtained from the later time regions.

The new level scheme is given in Fig. 1. This figure differs from that given in Ref. [14] in that the two excited 8^{+} bands that are fed by the isomers are not shown and the new decays to the 4^{+} band based on the 635-keV state are now incorporated. (The 8^{+} structures were confirmed, but they are not relevant to the present discussion.) New transitions are indicated by asterisks. These include a 190-keV transition directly from the 14^{+} isomer to the 13^{-} state of the 7^{-} band confirming the placement of the isomer, but more importantly, multiple branches from members of the 4^{+} band to both the 7^{-} and 1^{+} bands, the latter of which decays directly to the 1^{-} β-decaying isomer at 123 keV. (The 4^{+} band is known up to spin 14^{+} and was assigned by McGoram *et al*. [14,17] to the $\pi 1/2^{-}[541] \otimes \nu 7/2^{-}[514]$ configuration, as indicated in the level scheme.)

A sample of the γ-γ coincidence information used to establish the scheme is given in Fig. 2. Figure 2(a) is a

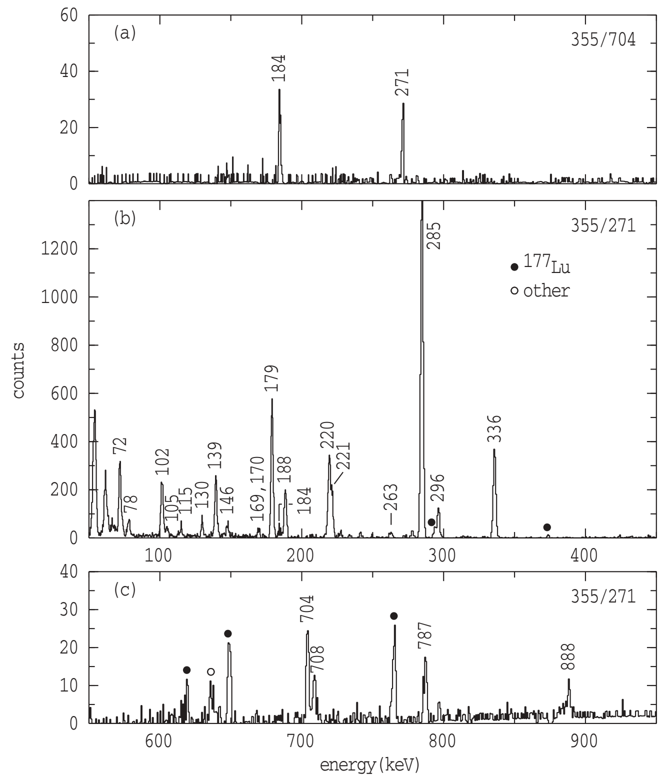

FIG. 2. Double-gated γ-ray spectra in the out-of-beam time region, with energy gates as indicated. Identified contaminants are marked by filled and open circles.

spectrum obtained with a double gate on the 355 and 704-keV transitions. The presence of only the 184 and 271-keV γ rays leads to an unambiguous placement of the 704-keV transition as the $E1$ decay connecting the 4^+ band member (888 keV) to the 8^- state at 184 keV. The lower two panels [Figs. 2(b) and 2(c)] are the low and high-energy regions of the spectrum obtained with a double gate on the 355 and 271-keV transitions, the most intense cascade feeding the 888-keV state. Although weak (note the difference in the vertical scale between the two spectra), the 704, 708, 787, and 888-keV links to the 7^- band are clear, as are, for example, the 296 and 221-keV connections to the 1^+ band. Most contaminant lines in this spectrum arise from the decay of the high-spin isomer in ^{177}Lu, which has coincident transitions at 273 and 355 keV [18].

The main results are summarized in Table I. The γ-ray decay widths listed in the penultimate column were obtained by scaling to the widths for the in-band $E2$ transitions calculated from the rotational model formulas with $K = 4$ and $Q_0 = 7.35$ eb (see, for example Ref. [13]). For such a well-deformed nucleus, these should be reliable to a level of $\sim$20%, the main uncertainty being the choice of K (lower-K values give higher $E2$ strengths). The 635 keV band will have mixed K and exhibits rotation alignment because of Coriolis mixing within the proton $h_{9/2}$ orbitals, of which the $1/2^-[541]$ orbital is the lowest. This should also be borne in mind in interpreting the transition strengths, given in the last column of Table I. Less inhibition will occur in the $E1$ transitions than might be expected on the basis of the nominal forbiddenness of $\nu = \Delta K - \lambda = (7 - 4) - 1 = 2$. Taking into account all branches and internal conversion, the lifetimes of the 4^+ band members based on this approach would be 23 ps for the 10^+ state, 49 ps for the 9^+ state, 95 ps for the 8^+ state, and 93 ps for the 7^+ state. (The decay of the 6^+ state is not included in the table because the main in-band decays are of low energy and highly converted. Hence, extraction of a decay width is more problematic and more sensitive to the K value because

TABLE I. Branching ratios, partial γ-ray decay widths, and transitions strengths for members of the $K^\pi = 4^+$ band.

E_i I_i^π	I_f^π	$\sigma\lambda$	E_γ (keV)	I_γ [a]	Γ_γ [b] (eV)	$B(\sigma\lambda)$ (W.u.)
1159	9^+	$M1$ [c]	146	34(1)	$5.1(2) \times 10^{-6}$	
10^+	8^+	$E2$	271	100(1)	$1.47(3) \times 10^{-5}$	209(4)
	10^-	$E1$	546	1.2(3)	$1.7(4) \times 10^{-7}$	$5(1) \times 10^{-7}$
	9^-	$E1$	770	2.0(1)	$2.9(3) \times 10^{-7}$	$3.1(3) \times 10^{-7}$
	8^+	$E2$	267	2.1(2)	$3.0(3) \times 10^{-7}$	4.7(5)
1013	8^+	$M1$ [c]	124	46(2)	$2.4(2) \times 10^{-6}$	
9^+	7^+	$E2$	226	100(5)	$5.1(4) \times 10^{-6}$	185(13)
	9^-	E1	828	7(1)	$3.4(4) \times 10^{-7}$	$2.8(3) \times 10^{-7}$
888	7^+	$M1$ [c]	102	76(5)	$1.00(7) \times 10^{-6}$	
8^+	6^+	$E2$	179	100(3)	$1.33(6) \times 10^{-6}$	153(7)
	8^-	$E1$	704	10(1)	$1.36(13) \times 10^{-7}$	$1.84(17) \times 10^{-7}$
	7^-	$E1$	888	7(1)	$0.94(14) \times 10^{-7}$	$0.63(9) \times 10^{-7}$
	6^+	$E2$	296	28(2)	$3.7(3) \times 10^{-7}$	3.5(2)
787	6^+	$M1$ [c]	78	392(15)	$7.6(9) \times 10^{-7}$	
7^+	5^+	$E2$	130	100(11)	$2.0(3) \times 10^{-7}$	111(17)
	7^-	E1	787	54(7)	$1.04(18) \times 10^{-7}$	$1.01(18) \times 10^{-7}$

[a]Normalized to the in-band $E2$ transition at 100 units.
[b]Absolute widths scaled to the calculated $\Delta I = 2$ in-band $E2$ transitions assuming $K = 4$ and $Q_0 = 7.35$ eb.
[c]Mixed $M1/E2$. Separate $M1$ and $E2$ strengths are not listed but are taken into account in deducing total widths.

of the proximity to the bandhead. Noting these qualifications, an estimate of $\Gamma_\gamma \sim 2 \times 10^{-8}$ eV for the 708 keV $E1$ branch is obtained.)

Two features stand out from Table I. First, all $E1$ branches lie approximately in the range 1–5 $\times 10^{-7}$ W.u. Second, both the $E2$ branches at 267 keV from the 10^+ ($K = 4$) state and that at 296 keV from the 8^+ ($K = 4$) state have strengths of several single-particle units, despite the fact that both are K forbidden. This is a direct consequence of mixing through a near degeneracy between the 8^+ state of the 4^+ band and the 8^+ level of the 1^+ band that are observed to be 4.6 keV apart. Following the approach detailed in Refs. [14,19], a comparison of the strength of the 179-keV in-band $E2$ transition and the 296-keV intraband $E2$ γ ray (ignoring mixing in the 6^+ states, which are more widely separated) leads to an estimated amplitude of $\beta = 0.109$ in the mixed wave function for the 888 keV state, writing it as $\alpha|K = 4 > +\beta|K = 1 >$ with $\beta = \sqrt{1-\alpha^2}$. This corresponds to a mixing matrix element of $|V| = 0.50$ keV. The complementary admixture of a $K = 4$ component in the 893 keV 8^+ state then enables the 267-keV transition from the 10^+, ($K = 4^+$) state to the 8^+ ($K = 1^+$) level.

With the new information, it is now also possible to make a statement about the expected resonant photoexcitation of ^{176}Lu in the laboratory. The only isolated states that are expected to be strongly excited by electric dipole excitation from the 7^- ground state will have spins and parities 6^+, 7^+, and 8^+. As outlined for the comparable case of ^{180}Ta in Ref. [20], that process will select out such triplets with rotational spacings in successively higher rotational bands.

In the case of an isolated resonance, the Breit-Wigner form of the energy and angle-integrated cross section (s_E) for inelastic photon excitation from an initial state with angular momentum I_m to an intermediate state I_a involves the statistical factor $g = \frac{2I_a+1}{2I_m+1}$, the photon wavelength λ, and the decay widths [21], with

$$s_E = \int_{E_R} \sigma dE = \frac{\lambda^2}{4} g \frac{\Gamma_m \Gamma_0}{\Gamma},$$

where the total decay width of the intermediate state is Γ, the decay width for all paths that reach the 1^-, 123 keV state is Γ_0, and Γ_m is the width for decay back to the initial (7^-) state. The total decay widths (including internal conversion) are 0.71×10^{-5} eV for the 7^+ state at 787 keV, and 0.69×10^{-5} eV for the 8^+ level at 888 keV. Taking the $E1$ decay widths given in Table I results in integrated cross sections of 0.62×10^{-3} eV.b and 0.52×10^{-3} eV.b for the 787 and 888 keV states, respectively.

Mohr *et al.* [12] reported results from as yet unpublished measurements on the photoactivation of ^{176}Lu (focusing partly on the possibility of $E2$ excitation of the 5^-, 839 keV state as a possible pathway, as discussed earlier). Unfortunately Ref. [12] does not show results below an end point energy of 800 keV, nor are resonance strengths given, but the first clear resonance observed is at 880 keV (albeit with an energy uncertainty of ~30 keV), matching the energy of the 888 keV state characterized here. The mixing between the 888 and 893 keV 8^+ states discussed earlier will also lead to a (smaller) resonance at 893 keV, although this would be beyond the energy resolution in such experiments.

Note that the cross sections predicted here are much smaller than those observed for ^{180}Ta [3,4] (which are ~0.06 eV.b). The present $E1$ strengths of $\sim 10^{-7}$ W.u. also provide a perspective on the possible (as yet unobserved) branches in ^{180}Ta. These were conjectured to exist if the resonances in the laboratory photoexcitation were indeed correlated with the position of members of a $K^\pi = 5^+$ band [20]. Furthermore, the analysis of Ref. [20] concluded that for the purported branches to be consistent with measured resonance strengths the $E1$ strengths would have to be $\sim 10^{-5}$ W.u., two orders of magnitude larger than the $E1$ transitions observed here.

In summary, we have established a clear connection between lower members of the $K^\pi = 7^-$ ground-state band and the 1^-, 5.3 hr β-decaying isomer, through a sequence of five members of a $K^\pi = 4^+$ band. The results will have implications for photoexcitation of ^{176}Lu in the laboratory and for the excitation, production, and survival of ^{176}Lu in a stellar environment where equilibrium between the ground state and the isomer may occur at lower temperatures than previously anticipated and where there will be a Maxwellian population of both the ground and excited states.

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