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Thermal Equilibration of ^{176}Lu via K -Mixing

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In astrophysical environments, the long-lived ($T_{1/2} = 37.6$ Gy) ground state of ^{176}Lu can communicate with a short-lived ($T_{1/2} = 3.664$ h) isomeric level through thermal excitations. Thus, the lifetime of ^{176}Lu in an astrophysical environment can be quite different than in the laboratory. We examine the possibility that the rate of equilibration can be enhanced via K -mixing of two levels near $E_x = 725$ keV and estimate the relevant γ -decay rates. We use this result to illustrate the effect of K -mixing on the effective stellar half-life. We also present a network calculation that includes the equilibrating transitions allowed by K -mixing. Even a small amount of K -mixing will decrease the timescale for equilibration during an s -process triggered by the ^{22}Ne neutron source.

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I. INTRODUCTION

The nucleus ^{176}Lu is produced almost exclusively by slow neutron capture in the s -process. It was originally thought that by virtue of its long half-life ($T_{1/2} = 37.6$ Gy), ^{176}Lu could serve as a chronometer of galactic nucleosynthesis. However, it has been shown [1, 2] that the ground state of ^{176}Lu can reach at least a quasi-equilibrium with a short-lived isomeric state at $E_x = 122.9$ keV ($T_{1/2} = 3.664$ h; all excitation energies and other nuclear structure parameters are taken from the ENSDF compilation [3]) through a mediating level at $E_x = 838.6$ keV. As a result, the astrophysical half-life of ^{176}Lu is strongly dependent on temperature. This precludes the use of ^{176}Lu as a *chronometer*, but does imply that it may serve instead as an s -process *thermometer*.

The s -process in the in $A = 176$ mass region is shown in Figure 1. Production of the isomer, either by neutron capture or by thermal excitation of the ground state leads directly (via β -decay) to ^{176}Hf . In contrast, the ground state, which is also produced directly by neutron capture or by thermal excitation of the isomer, will undergo neutron capture to ^{177}Lu , which then β -decays to ^{177}Hf . This temperature-dependent branching in the reaction flow should be reflected in the abundances in this mass region. In fact, consideration of isotopic abundances strongly suggests that the isomer and ground state must communicate during the s -process: As discussed by Klay *et al.* [1] (and references therein), measurements of the partial neutron capture cross section producing the isomer show that it is too large to be consistent with the observed abundance of ^{176}Lu in the ground state. Recent measurements reinforce this conclusion [4]. Studies of the branching of the s -process at $A = 176$ also indicate that there is more s -process flow through the

ground state than predicted by the production rate of the ground state [5] via neutron capture in the classical picture. Taken together, these observations suggest the presence of an additional creation mechanism for the ground state. Neutron capture produces an isomer to ground state ratio of approximately 6 [6, 7]. At typical *s*-process temperatures (about $1\text{--}3 \times 10^8$ K, or $T_9 = 0.1\text{--}0.3$), the thermal equilibrium ratio is $1.3 \times 10^{-7}\text{--}1.7 \times 10^{-3}$. In other words, if thermal equilibrium pertains, then most of the *s*-process flow would ultimately pass through the ground state.

FIG. 1: The s -process path in the vicinity of ^{176}Lu . Stable (or extremely long-lived) nuclei are shown in grey. The ground state of ^{176}Lu and the isomeric state are labeled as g and m, respectively.

Measurements of γ -transitions by Klay *et al.* [8], Lesko *et al.* [2] and Vanhorenbeeck *et al.* [9] have uncovered several paths linking the ground state and the isomer, which would be active during an s -process triggered by the ^{22}Ne neutron source. In this paper we will examine the possibility that K -mixing provides an additional route for the ground state and isomer to reach equilibrium. Interactions on the atomic scale might be sufficient to influence

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equilibration between the ground state and isomer during the s -process. This mechanism is not limited to the case of ^{176}Lu and should operate in other s -process nuclei.

II. EQUILIBRATION OF ^{176}Lu

Direct γ -ray decay of the isomer ($J^\pi = 1^-, K^\pi = 0^-$) to the ground state ($J^\pi = 7^-, K^\pi = 7^-$) is strongly suppressed by their large difference in angular momentum. Also, because of the approximate conservation of the K quantum number, γ -decay is further hindered by a factor of approximately 100 per degree of K -forbiddenness [10] (i.e., by a factor $10^{2\nu}$, where $\nu \approx |\Delta K - \lambda|$ and λ is the γ -ray multipolarity). In general, the approach to equilibrium under stellar conditions can be inferred by comparing the time scales for γ -ray absorption and emission to the time scales for nuclear reactions and decays. States that are strongly linked by downward (and therefore upward) transitions, such as members of a given rotational band, will quickly reach full thermal equilibrium and will evolve together. States that are more weakly connected may not reach thermal equilibrium if other nuclear processes are faster. In the case of ^{176}Lu , the members of the ground-state band ($\pi 7/2 [404] + \nu 7/2 [514]$, $K^\pi = 7^-$) will quickly reach thermal equilibrium. For example, at $T_9 = 0.3$, the 184.1-keV, $J^\pi = 8^-$ state will equilibrate with the ground state with a time constant of 21 ns. Side bands that feed the ground-state band will also equilibrate with the ground-state band. Similarly, the states in the isomer band ($\pi 7/2 [404] - \nu 7/2 [514]$, $K^\pi = 0^-$) and associated side bands will rapidly form an equilibrium cluster. At this point ^{176}Lu can be thought of as a quasi-equilibrium ensemble consisting of two separate, equilibrated clusters (the ground-state and isomer clusters). New groups of states in local equilibrium will form, presumably via K -allowed transitions (either upward or downward) from states in the original equilibrium clusters to those in other bands. If the time scale for linking transitions is short (compared to the time scale for neutron capture or β -decay), then these new states will reach equilibrium with the original cluster. Otherwise, they will form separate groups in local equilibrium. Eventually, the clusters originating from the ground state will reach those based on the isomer and produce quasi-equilibrium between the ground state and isomer. This process will be discussed in more detail in Sec. V.

The previous γ -decay studies of ^{176}Lu , [2, 8] clearly show that the $J^\pi = 5^-$ state at $E_x = 838.6$ keV ($K = 4$) decays directly to the ground state via a strong E2 transition and via cascades that ultimately end in the isomer (more recent measurements [9] show evidence for mediating states at somewhat higher excitation energy). In a stellar plasma, this state would be populated from the isomer in the manner described above, via 3–4 interband transitions, and primarily via a single photon from the ground state. Interestingly, several of the strongest transitions from this level (including the ground-state branch)

are K -forbidden [11], which implies that the 838.6-keV state may be mixed with other nearby 5^- states.

Klay *et al.* [1] discuss the possibility that weak, undetected K -forbidden transitions may also provide an equilibration path (but conclude that they are most likely negligible). In particular, they consider a $\Delta K = 7$ E2 transition from the 5^- state at 437.3 keV (in the isomer band) to the ground state, which could compete with equilibration via the 838.6-keV state if the E2 strength is enhanced to the level observed in interband transitions (in this case, $B(E2) = 200$ Weisskopf units). However, *intra*band transitions linking different intrinsic states (as is the case here) will not be similarly enhanced. Furthermore, simple Coriolis ($\Delta K = 1$) or vibrational ($\Delta K = 2, 3$) mixing can not account for a low-lying $\Delta K = 7$ transition. Indeed, the lower-energy members of both the ground and isomer bands have ratios of dynamic to kinematic moments of inertia that are nearly unity (indicating little rotational perturbation), and thus K should be approximately conserved in the absence of accidental mixing with a nearby 5^- state. Since there are no other 5^- states in the vicinity of the 437.3-keV state, this possibility must be discounted. A hindrance factor of 100 per degree of K -forbiddenness is an empirical estimate, but is reasonably well supported by the data [10]. Systematics of K -forbidden transitions in the $A = 180$ region seem to show a trend versus $N_p N_n$ [12] which suggests a factor of about 72 per degree of K -forbiddenness for ^{176}Lu . At this level, the 437.3-keV state will not provide an equilibration route during the s -process. Additional, low-lying transitions were also considered by Lesko *et al.* [2], but apparently without regard to the fact that they would be K -forbidden.

However, the strength of any such transition could be enhanced by K -mixing, which would largely circumvent the K -selection rule. As mentioned above, this mechanism appears to be at work for equilibration through the 838.6-keV state and there is clear evidence for K -mixing at higher excitation energies [13]. Chance degeneracies can also lead to K -mixing and thus it seems at least plausible that the 7^- states at $E_x = 724.7$ keV ($\pi 7/2 [404] - \nu 7/2 [514]$, $K^\pi = 0^-$) and 725.2 keV ($\pi 5/2 [402] - \nu 7/2 [514]$, $K^\pi = 6^-$) will mix. The former state is a member of the isomer band whereas the latter state is a member of a band that can be populated by thermal excitation of the ground state. If these states do indeed mix, then the ground state and isomer are linked by a series of K -allowed transitions. As pointed out by Kondev *et al.* [14] and McGoram *et al.* [13], even small amounts of mixing can significantly increase the rate of transitions that would otherwise be K -hindered. These transitions could be much weaker than the unmixed transitions and thus would have escaped notice in previous decay studies. However, since the mediating states would be at lower excitation energy, ≈ 725 keV rather than at 839 keV, they would be produced more efficiently from the stellar photon bath. This scenario for equilibration is illustrated in Figure 2.

FIG. 2: Proposed equilibration scheme. States are listed according to their J^π and excitation energies (in keV). The numbers in parentheses denote the states of interest. Downward γ -transitions have been observed for the links represented by black arrows. The matrix element that describes the mixing of the 7^- states [labeled (5) and (7)] in the $K^\pi = 6^-$ and $K^\pi = 0^-$ bands is denoted by V . The 2 bands are potentially linked by γ -transitions between states (5) and (6) and between (3) and (7) (shown in grey).

It should be pointed out that the mediating transitions occur between excited states and thus would not be observed in photo-absorption measurements. A similar situation has been described in the case of ^{180}Ta [15].

III. K -MIXING OF 2 LEVELS AND γ -DECAY

If we assume that 2 states of a given J^π , but different K (K_1 and K_2) are mixed, then the resultant wavefunction for each state can be written schematically as

$$|J^\pi, \bar{K}_1\rangle = \alpha |J^\pi, K_1\rangle - \beta |J^\pi, K_2\rangle \quad (1)$$

and

$$|J^\pi, \bar{K}_2\rangle = \alpha |J^\pi, K_1\rangle + \beta |J^\pi, K_2\rangle, \quad (2)$$

where

$$\alpha^2 + \beta^2 = 1. \quad (3)$$

In our case, $J^\pi = 7^-$; $\bar{K}_1$ and $\bar{K}_2$ refer to the mixed- K nature of the states (where K is no longer a good quantum number); the pure K states have $K_1 = 6$, and $K_2 = 0$.

The matrix element for the E2 transition linking e.g., states (5) and (6) in Figure 2 [henceforth, transition a $\rightarrow$ b will be denoted as (ab)] is

$$\begin{aligned} \langle 5^-, 0 \| M(E2) \| 7^-, \bar{K}_1 \rangle &= \alpha \langle 5^-, 0 \| M(E2) \| 7^-, 6 \rangle \\ &- \beta \langle 5^-, 0 \| M(E2) \| 7^-, 0 \rangle \end{aligned} \quad (4)$$

where $M(E2)$ is the electric quadrupole operator. The first term represents a K -forbidden decay, which can be ignored. Assuming that $T_{1/2}^\gamma/T_{1/2}^W = 100^\nu$ (where $T_{1/2}^W$ is the Weisskopf estimate for the partial half-life and ν is the K -hindrance factor introduced above), $T_{1/2}^\gamma(56) = 488$ ms, which is much longer than for the allowed decays calculated below. Since

$$B(E2; J, K \rightarrow J-2, K) = \frac{1}{2J+1} |\langle J-2, K \| M(E2) \| J, K \rangle|^2, \quad (5)$$

we have

$$B(E2; 7^-, \bar{K}_1 \rightarrow 5^-, 0) = \beta^2 B(E2; 7^-, 0 \rightarrow 5^-, 0). \quad (6)$$

From the relationship between $B(E2)$ and the partial half-life for the transition, we finally obtain

$$T_{1/2}^\gamma(56) = \left(\frac{E_\gamma(76)}{E_\gamma(56)} \right)^5 \frac{T_{1/2}^\gamma(76)}{(\beta/\alpha)^2} \approx \frac{T_{1/2}^\gamma(76)}{(\beta/\alpha)^2}. \quad (7)$$

In other words, the K -mixed (56) transition can be calculated from the partial half-life for the observed (76) transition if the mixing amplitude β can be determined.

A similar approach can be used for the transition between levels (7) and (3). However one difference is that the (73) and (53) transitions can be of mixed M1 + E2

character. In this case

$$\begin{aligned} \frac{1}{T_{1/2}^\gamma(73)} &= \frac{1}{T_{1/2}^\gamma(M1; 73)} + \frac{1}{T_{1/2}^\gamma(E2; 73)} \\ &= \frac{1 + \delta^2(73)}{T_{1/2}^\gamma(M1; 73)}, \end{aligned} \quad (8)$$

where $\delta(73)$ is the E2/M1 mixing ratio. Assuming that $\delta(73) = \delta(53)$, then

$$T_{1/2}^\gamma(73) = \left(\frac{E_\gamma(53)}{E_\gamma(73)} \right)^5 \frac{T_{1/2}^\gamma(53)}{(\beta/\alpha)^2} \approx \frac{T_{1/2}^\gamma(53)}{(\beta/\alpha)^2}. \quad (9)$$

IV. ESTIMATED γ -DECAY RATES

The partial half-lives for the observed (53) and (76) transitions have not been measured and so we have calculated them using a semi-empirical approach. For the (76) E2 transition, we use the relationship between $B(E2)$ and the intrinsic quadrupole moment Q_0 :

$$B(E2; J, K \rightarrow J-2, K) = \frac{5}{16\pi} Q_0^2 |\langle JK20 | J-2K \rangle|^2 \quad (10)$$

where $Q_0 = 7.35(4)$ eb is calculated using the measured $Q = 4.92(3)$ eb [3] for the ground-state band and the relationship

$$Q = \frac{3K^2 - J(J+1)}{(J+1)(2J+3)} Q_0. \quad (11)$$

The remaining term in Eqn. 10 is a Clebsch-Gordan coefficient. This yields $B(E2) = 1.734$ (eb)², which implies $T_{1/2}^\gamma(76) = 16.7/\alpha^2$ ps. For the (53) M1 transition, we make use of

$$\begin{aligned} B(M1; J, K \rightarrow J-1, K) &= \\ \frac{3}{4\pi} (g_K - g_R)^2 K^2 |\langle JK0 | J-1K \rangle|^2. \end{aligned} \quad (12)$$

The gyromagnetic ratios $g_K(\pi 5/2[402]) = 1.57$ and $g_K(\nu 7/2[514]) = 0.30$ were calculated using deformed Woods-Saxon wave functions with deformation parameters $\beta_2 = 0.278$, $\beta_4 = -0.071$, and $\beta_6 = -0.009$ [16]. The collective gyromagnetic ratio g_R was calculated from

$$\mu = g_R J + (g_K - g_R) \frac{K^2}{J+1} \quad (13)$$

with $\mu = +3.1692(45) \mu_N$ [3] and $|(g_K - g_R)| = 0.175(7)$ [17]. The resulting value, $g_R = 0.30$ was assumed to be the same for all bands. From this, we obtain $B(M1) = 0.298 \mu_N^2$ and $T_{1/2}^\gamma(M1) = 31.5$ ps. However, since this transition can be of mixed M1 + E2 multipolarity,

$$T_{1/2}^\gamma = \frac{T_{1/2}^\gamma(M1)}{1 + \delta(53)^2}, \quad (14)$$

where

$$\delta(53) = 0.933 \frac{Q_0}{(g_K - g_R)} \frac{E_\gamma(53)(MeV)}{\sqrt{J^2 - 1}} = 0.3. \quad (15)$$

Therefore, $T_{1/2}^\gamma(53) = 28.9/\alpha^2$ ps.

The partial half-lives for the K -mixed (56) and (73) transitions can be calculated using Eqns. 7 and 9. The mixing matrix element V and the amplitudes α and β are related by

$$V = \alpha \times \beta \times \Delta E_x, \quad (16)$$

where ΔE_x is the energy difference between the two 7^- levels (517 eV). Given that $\alpha^2 + \beta^2 = 1$, Eqn. 16 can be solved for β^2 :

$$\beta^2 = \frac{2 \left(\frac{V}{\Delta E_x} \right)^2}{1 + \left[1 - 4 \left(\frac{V}{\Delta E_x} \right)^2 \right]^{1/2}}. \quad (17)$$

Unfortunately, V is unknown and can take values up to $\Delta E_x/2 = 258.5$ eV. Consequently, we have calculated partial half-lives for different values of V , which are listed in Table I. Large values of V or β^2 can be ruled out

TABLE I: Mixing matrix elements and partial half-lives for K -mixed transitions

V (eV)	β^2 (%)	$T_{1/2}^\gamma(76)$ (ps)	$T_{1/2}^\gamma(56)$ (ps)	$T_{1/2}^\gamma(53)$ (ps)	$T_{1/2}^\gamma(73)$ (ps)
258.5	50.0	3.34e+01	3.34e+01	5.78e+01	5.78e+01
150	9.28	1.84e+01	1.80e+02	3.19e+01	3.11e+02
100	3.89	1.74e+01	4.29e+02	3.01e+01	7.43e+02
50	0.944	1.69e+01	1.77e+03	2.92e+01	3.06e+03
10	3.74e-02	1.67e+01	4.47e+04	2.89e+01	7.73e+04
5	9.40e-03	1.67e+01	1.79e+05	2.89e+01	3.09e+05
1	3.74e-04	1.67e+01	4.47e+06	2.89e+01	7.73e+06

by the existing γ -ray data. For example, if we assume that the uncertainties in the intensities for the K -allowed transitions measured by Klay *et al.* [8] are a measure of their sensitivity to weak, neighboring transitions, then we can eliminate branching ratios for the K -forbidden transitions of greater than about 10% of the K -allowed strength. This implies $V \lesssim 153$ eV or $\beta^2 \lesssim 9.7\%$. We will assume these upper limits in the discussion that follows.

V. EFFECTIVE DECAY RATE IN A STELLAR PLASMA

The time evolution of the states shown in Figure 2 can be calculated by solving the set of coupled differential equations that describe the behavior of each state. Since various transition rates can differ by many orders of magnitude at s -process temperatures, significant care must be exercised to control the size of numerical errors.

This becomes cumbersome once ^{176}Lu is placed into a larger network of nuclei. However, in a stellar plasma, ^{176}Lu will rapidly evolve into a configuration of 2 quasi-equilibrium clusters, one of which contains the states in the ground-state and $K = 6$ bands, while the other is composed of the states in the isomer band. Within the clusters, the states rapidly achieve thermal equilibrium, as described in Sec. II. To illustrate this point, we have calculated the time evolution of the states shown in Figure 2 for $T_9 = 0.25$ and mixing amplitude $\beta^2 = 9.40 \times 10^{-3}\%$. Only the 184.1-keV and 388.9-keV states have tabulated lifetimes [3] and so those for the remaining states were calculated using the procedures described above. The results are shown in Figure 3, assuming that only the ground state is populated at $t = 0$ and that there is no destruction of ^{176}Lu via β -decay. The top panel shows the states that equilibrate quickly with the ground state. This cluster reaches thermal equilibrium within about 0.1 ns. The bottom panel shows the cluster built on the isomer, which reaches equilibrium somewhat later than the ground-state cluster (here the steady-state relative abundances are reached after about 1 ns). The rise in the abundances of these states is a consequence of leakage out of the ground-state cluster. At about 10^5 s, the isomer cluster reaches equilibrium with the ground-state cluster and all of the relative abundances assume their steady-state values. This time scale is about 14-15 orders of magnitude longer than the time scales for equilibration within the clusters. Thus, on the time scale of equilibration between the ground state and the isomer, equilibration within a cluster can be considered to be instantaneous. Therefore, the only γ -ray transitions that have to be considered explicitly are those between the clusters, i.e. $5 \leftrightarrow 6$ and $3 \leftrightarrow 7$. The total rates for these transitions must also include internal conversion, which we have calculated using the procedures described in Ref [18]. This addition is negligible for the $5 \rightarrow 6$ transition, but the $7 \rightarrow 3$ transition has conversion coefficients $\alpha_{M1} = 0.95$ and $\alpha_{E2} = 0.59$. However, at s -process temperatures and densities, the average occupancy of the K-shell of ^{176}Lu is $n_K \approx 0.4$ [19] as opposed to $n_K = 2$ in the laboratory. Since $\alpha_{eff} \approx \alpha_{nK}/2$ [19], under stellar conditions the conversion coefficients are reduced to effective values $\alpha_{eff} = 0.17$ (M1) and $\alpha_{eff} = 0.065$ (E2). This changes the transition rate obtained from Table I by about 11%, which is negligible for our purposes.

In the 2-cluster approximation, the time evolution of the clusters can be described by 2 coupled equations:

$$\frac{d}{dt} \left(\sum_i n_i \right)_g = -n_1 \lambda_\beta^1 - n_3 \lambda_{37} - n_5 \lambda_{56} + n_6 \lambda_{65} + n_7 \lambda_{73}, \quad (18)$$

which describes the ground-state cluster and

$$\frac{d}{dt} \left(\sum_i n_i \right)_m = -n_2 \lambda_\beta^2 - n_6 \lambda_{65} - n_7 \lambda_{73} + n_3 \lambda_{37} + n_5 \lambda_{56} \quad (19)$$

FIG. 3: The time evolution of the states shown in Figure 2 for $T_9 = 0.25$ and $\beta^2 = 9.40 \times 10^{-3}\%$, assuming no destruction of ^{176}Lu via β -decay. The top panel shows states in the ground-state cluster, labeled by their excitation energies (in keV). The isomer cluster is shown in the bottom panel. The two clusters equilibrate after about 10^5 s, which is 14-15 orders of magnitude longer than the equilibrium times within the clusters. The dots show the abundance of the isomer calculated using the 2-level approximation described in the text.

for the isomer cluster. Here n_i refers to the abundance of state i in Figure 2; The subscripts g and m denote the equilibrium clusters built on the ground state and isomer, respectively; λ_{ij} are the $i \rightarrow j$ transition rates, and λ_β^1 and λ_β^2 are the laboratory β -decay rates for the ground state and isomer. Although the β -decay rates depend on the degree of ionization, the temperatures and densities of the s -process are low enough that this effect may be neglected. The transition rate linking an initial state i with a higher-lying final state f via absorption of a photon is

$$\lambda_{if} = \frac{g_f}{g_i} \frac{\lambda_{fi}^s}{e^{E_{if}/kT} - 1}, \quad (20)$$

where $g = 2J + 1$, λ_{fi}^s is the rate for spontaneous decay from f to i , E_{if} is the energy difference between i and f , and k is Boltzmann's constant. The corresponding total decay rate between i and f (including stimulated emission) is

$$\lambda_{if} = \frac{\lambda_{fi}^s}{1 - e^{-E_{if}/kT}}. \quad (21)$$

Since the states within a cluster are in thermal equilibrium, their relative abundances are given by

$$C_{ij} \equiv \frac{n_i}{n_j} = \frac{g_i}{g_j} e^{-(E_{xi} - E_{xj})/kT}. \quad (22)$$

Therefore, the abundances appearing in Eqns. 18 and 19 can be written as fractions of the ground state or isomer abundances. This allows these equations to be recast as a concise 2x2 coupled system of equations for the abundances of the ground state and isomer:

$$\frac{d}{dt}n_1 = a_{11}n_1 + a_{12}n_2 \quad (23)$$

$$\frac{d}{dt}n_2 = a_{21}n_1 + a_{22}n_2 \quad (24)$$

or equivalently as a matrix equation

$$\frac{d}{dt}\mathbf{n} = \mathbf{A} \cdot \mathbf{n}. \quad (25)$$

The transition matrix elements a_{ij} are

$$\begin{aligned} a_{11} &\equiv \frac{(-\lambda_\beta^1 - C_{51}\lambda_{56} - C_{31}\lambda_{37})}{(1 + C_{31} + C_{51} + \dots)} & a_{12} &\equiv \frac{(C_{72}\lambda_{73} + C_{62}\lambda_{65})}{(1 + C_{31} + C_{51} + \dots)} \\ a_{21} &\equiv \frac{(C_{51}\lambda_{56} + C_{31}\lambda_{37})}{1 + (C_{42} + C_{62} + C_{72} + \dots)} & a_{22} &\equiv \frac{(-\lambda_\beta^2 - C_{72}\lambda_{73} - C_{62}\lambda_{65})}{1 + (C_{42} + C_{62} + C_{72} + \dots)}, \end{aligned} \quad (26)$$

where a_{12} and a_{21} represent the effective $m \rightarrow g$ and $g \rightarrow m$ rates, respectively. The denominators in these expressions are formally a sum over all of the C_{ij} for each cluster. In practice, only the lowest-energy states contribute to this sum and for s -process temperatures, $C_{ij} \ll 1$. Eq. 25 has the solution

$$\mathbf{n}(t) = \mathbf{N}_1 e^{-\lambda_1 t} + \mathbf{N}_2 e^{-\lambda_2 t} \quad (27)$$

where $\mathbf{N}_i$ and $-\lambda_i$ are the eigenvectors (normalized to the correct initial abundances) and eigenvalues of $\mathbf{A}$, respectively. The smallest eigenvalue corresponds to the effective decay rate of the (quasi) equilibrated nucleus. The other eigenvalue (which is approximately equal to a_{22}) is the inverse of the time constant for quasi or full equilibration. The resulting effective half-life of ^{176}Lu is shown in Figure 4 where we have also included the known equilibration path through the 838.6-keV state, with $T_{1/2} \geq 6.9$ ps [11]. The experimental upper limit on the half-life for this state is $T_{1/2} = 300$ ps [1]. However, a conservative, theoretical upper limit is ≈ 35 ps [11] and we have adopted this value. Internal conversion enhances the rate of the transition from this state to the 4^- state at 722.9 keV (with $\alpha_{M1} = 2.49$ [3]) as measured in the laboratory. Therefore, to calculate the effective decay rate under stellar conditions we have used an effective internal-conversion coefficient as described above. The occupancy of the K-shell, n_K was calculated as a function of temperature [20], assuming a constant electron density of $4 \times 10^{26} \text{ cm}^{-3}$. A K -admixture on the order of $10^{-2}\%$ is sufficient to substantially shorten the stellar half-life of ^{176}Lu . This level of mixing corresponds to a matrix element of ~ 5 eV, which is on the scale of atomic interactions.

VI. THE APPROACH TO EQUILIBRIUM

The effective half-life shown in Figure 4 gives an indication of the temperature range where equilibration will occur, but the actual behavior of ^{176}Lu in a stellar environment will depend on the competition between the time scale for equilibration, τ_{eq} (the inverse of the largest eigenvalue in Eqn. 27) and those for neutron capture, τ_n and β -decay, τ_β . If τ_{eq} is smaller than both τ_n and τ_β , then ^{176}Lu will maintain thermal equilibrium with an effective half-life as shown in Figure 4. Otherwise, ^{176}Lu will achieve a quasi-equilibrium in which the relative population of excited states can be quite different from the equilibrium values. In this case, the approach to equilibrium will lag behind abundance changes from n -capture and β -decay and it is necessary to include the equilibrating transitions (a_{12} and a_{21} from Eqns. 23 and 23) explicitly when calculating the time evolution of ^{176}Lu .

During thermal pulses in low-mass AGB stars (with peak temperatures of $T_9 = 0.25 - 0.35$ and neutron densities of $8 \times 10^6 - 3 \times 10^{10} \text{ cm}^{-3}$), the question of whether full or quasi-equilibrium pertains depends on the half-life for the 838.6-keV state and strongly on the K -mixing amplitude that we assume for the 725-keV states. This is shown in Figure 5. For peak temperatures $T_9 \gtrsim 0.30$, ^{176}Lu will be in full thermal equilibrium for the entire range of K -mixing considered here because $\tau_{eq} \ll \tau_n$ and τ_β (here we have assumed a neutron density of $3 \times 10^{10} \text{ cm}^{-3}$). However, this is not the case in the limit of weak K -mixing and lower temperatures. For $T_9 \lesssim 0.2$, $\tau_{eq} > \tau_\beta$ even in the case of maximal mixing and thus ^{176}Lu can not be ascribed an effective decay rate.

Following Ref. [15], the transition rate between the ground state and isomer through an intermediate state (IS) is

$$\lambda(T) = \int cn_\gamma(E, T) \sigma(E) dE, \quad (28)$$

where $n_\gamma(E, T)$ is the thermal photon density

$$n_\gamma(E, T) = \left(\frac{1}{\pi}\right)^2 \left(\frac{1}{\hbar c}\right)^3 \frac{E^2}{\exp(E/kT) - 1}. \quad (29)$$

For s -process conditions, $\lambda(T) \approx cn_\gamma(E_{IS}, T) I_\sigma(E_{IS})$ where E_{IS} is the difference in excitation energies between the intermediate state and the initial state and I_σ is the energy-integrated cross section

$$\begin{aligned} I_\sigma(E_{IS}) &= \int \sigma(E) dE \\ &= \frac{2J_{IS} + 1}{2J_0 + 1} \left(\frac{\pi \hbar c}{E_{IS}}\right)^2 \frac{\Gamma_{IS \rightarrow \text{low-K}} \Gamma_{IS \rightarrow \text{high-K}}}{\Gamma}. \end{aligned} \quad (30)$$

Here J_0 , $\Gamma_{IS \rightarrow \text{low-K}}$, $\Gamma_{IS \rightarrow \text{high-K}}$, and Γ are the spin of the initial state, the partial widths for transitions to low- K and high- K states from the IS, and the total width of

FIG. 4: *Upper panel:* The effective half-life of the ground state of ^{176}Lu as a function of temperature. The solid lines are the effective half-life if ^{176}Lu equilibrates only via the 838.6-keV state, assuming that the half-life of this state is either 6.9 ps or 35 ps. The shaded region includes the additional path that would operate via K -mixing of the 725-keV states. The long half-life limit of the shaded region results from a half life of 35 ps for the 838.6-keV state, combined with the lowest K -mixing fraction considered here ($3.74 \times 10^{-4}\%$); the shortest half-life assumes $T_{1/2} = 6.9$ ps for the 838.6-keV state with our assumed maximal K -mixing fraction of 9.7%. Full thermal equilibrium is denoted by the dashed line. *Lower panel:* An expanded view of the effective half-life showing its dependence on the assumed K -mixing fraction for the 725-keV states (calculated assuming that $T_{1/2} = 6.9$ ps for the 838.6-keV state).

the IS, respectively. When comparing with experimental photo-absorption data, the relevant partial widths are those that correspond to the experimentally accessible transitions. For example, with ^{176}Lu initially in the ground state, $\Gamma_{IS \rightarrow \text{high}-K}$ would be the partial width for the $839 \rightarrow \text{ground}$ transition. Partial widths for decays to high- K excited states would not enter because the corresponding upward transition would not be observable. In contrast, the integrated cross section under stellar con-

ditions, I_{σ}^* would include all possible transitions so that $\Gamma_{IS \rightarrow \text{high}-K}$ would be the *total* decay width to high- K states. In addition, the effect of ionization on transitions with large internal-conversion coefficients must also be considered. These issues have been discussed in detail in Ref. [19] with the result $I_{\sigma}^*(839) = 0.60 - 2.50 \text{ eV fm}^2$ for the 838.6-keV state. If the 725-keV states are mixed, then I_{σ}^* would increase by $1.17 \times 10^{-4} - 2.75 \text{ eV fm}^2$ for mixing at the level of $3.74 \times 10^{-4}\% - 9.7\%$. In other

FIG. 5: The equilibration time constant for ^{176}Lu assuming no production or destruction. The solid lines show the time constants for equilibration through the 838.6-keV state for $T_{1/2} = 6.9$ ps and 35 ps. The dashed lines indicate the time constants if the 725-keV states are mixed at the levels shown (assuming $T_{1/2} = 6.9$ ps for the 838.6-keV state). The dash-dot lines are the time constants for β -decay of the isomer (assuming the laboratory half-life) and n-capture on ^{175}Lu (upper line) and ^{176}Lu (lower line). The time constants for n-capture were calculated using rates from the KADONIS compilation [6] with a neutron density of $3 \times 10^{10} \text{ cm}^{-3}$. For $\tau_{eq} \ll \tau_n$ and τ_β , the isomer and ground state will reach full thermal equilibrium. Otherwise, quasi-equilibrium will ensue.

words, for $\beta^2 \approx 9.7\%$, the resulting $I_\sigma^*(725)$ would be comparable to the maximum for the 838.6-keV state.

VII. ASTROPHYSICAL ASPECTS

Assuming that the K -mixing scenario is in fact valid, our calculation shows that the onset of quasi-equilibrium and the transition to full equilibrium may occur at lower temperatures than previously thought [1]. Full equilibrium through the 838.6-keV state alone occurs for $T_9 \approx 0.28$ whereas an additional path through the 725-keV states will move this transition point downward toward a minimum of $T_9 = 0.22$. To show schematically how the abundance ^{176}Lu will evolve during the s -process we have performed post-processing calculations for a $3 M_\odot$ AGB star of solar composition during the thermal-pulse phase. The temperature and density profiles at the base of the convective shell as well as the abundances of the major isotopes were calculated using the Monash version of the Mount Stromlo stellar-structure code (Ref. [21] and references therein). The initial abundances of the s -process isotopes were calculated for temperatures and densities appropriate to the interpulse phase, during which the $^{13}\text{C}(\alpha, n)^{16}\text{O}$ reaction is the primary source of neutrons. Neutron-capture rates in the Yb-Hf region were taken

from the KADONIS compilation [6]. Since both ^{176}Hf and ^{176}Lu are produced almost entirely by the s -process (there is a small p -process contribution to ^{176}Hf , which we will ignore), the ratio $^{176}\text{Hf}/^{176}\text{Lu}$ is a useful s -process diagnostic because its value at the time of the formation of the solar system is reasonably well established. The results of our calculation for the 9th thermal pulse are shown in Figure 6. Here, the peak temperature is $T_9 \approx 0.24$ and thus the ^{22}Ne neutron source is only marginally activated. The neutron burst lasts for about 12 years, which is about 10% of the duration of the entire convective phase. These results are not meant to represent a quantitative depiction of the s -process for this star. We have calculated the s -process for conditions found only at the base of the convective shell and have not included mixing. Our purpose is simply to show how $^{176}\text{Hf}/^{176}\text{Lu}$ behaves for different equilibration rates (a more realistic treatment of nucleosynthesis can be found in Ref. [7]). The general trend shown in Fig. 6 is that $^{176}\text{Hf}/^{176}\text{Lu}$ decreases as the timescale for equilibration rate decreases. In our model, the ground/isomer ratio is 0.062 at the end of the interpulse phase. At the peak of the thermal pulse, this ratio increases to 217 – 722 as equilibrating transitions convert the isomer to the ground state. The lower value assumes that the equilibration path is through the 838.6 keV state with $T_{1/2} = 35$ ps, whereas the upper value assumes $T_{1/2} = 6.9$ ps for this state and a K -mixing fraction of 0.0094% for the 725-keV states. The value for full thermal equilibrium is 2461 for this particular temperature ($T_9 = 0.23$) and thus the ground state and isomer are in a quasi-equilibrium and remain so throughout the neutron burst. However, it can be seen that K -mixing can begin to have an effect for a mixing fraction as low as 0.001%. The case of maximal K -mixing (9.7%) is somewhat different because here the ground state and isomer reach thermal equilibrium within about 3 years and the resulting ground-state fraction is high enough for significant flow via $^{176}\text{Lu}(n, \gamma)^{177}\text{Lu}$, which reduces the overall abundance of ^{176}Lu . In general, high levels of K -mixing appear to be ruled out because they lead to too much ^{176}Lu relative to ^{176}Hf .

The important point here is that K -mixing fractions of 0.001 – 0.0094% may be significant. The corresponding matrix elements are $V = 1.6 - 5$ eV, which are on the order of atomic interactions. At this level, it is fair to wonder if any nuclear state can be considered pure. In fact, the recent observation of $\Delta K = 13$ mixing in ^{174}Lu with $V \approx 19$ eV suggests a minimum V in the range of 10-20 eV [22]. This general conclusion likely applies to other K -isomers in the s -process (e.g. ^{180}Ta [23]). In other words, the degree of K -mixing that exists in rotational nuclei is probably sufficient to allow K -isomers to reach at least quasi-equilibria with their ground states during some phases of the s -process. In the case of ^{176}Lu , quasi- or full equilibrium will be reached for temperatures characteristic of the thermal-pulse phase in low-mass AGB stars [24] or the weak s -process in massive stars [25]. However, as shown in Table I, the branching ratios for

FIG. 6: The evolution of $^{176}\text{Hf}/^{176}\text{Lu}$ as a function of time since the start of the neutron burst for the 9th thermal pulse of a $3 M_{\odot}$ AGB star of solar composition. The time of peak neutron density is indicated by the vertical line. The solid lines indicate the effect of mediating transitions through the 838.6-keV state for $T_{1/2} = 6.9$ and 35 ps. The dashed lines represent the additional equilibration route through the 725-keV states, assuming $T_{1/2} = 6.9$ ps for the 838.6-keV state for the indicated K -mixing fractions. Maximal K -mixing of 9.7% is denoted by the heavy dashed line.

the relevant γ -ray transitions are probably too small to be observed, which complicates the use of ^{176}Lu as an s -process thermometer. As pointed out by Mohr *et al.* [19], $I_{\sigma}^*(839)$ is already too large for the "best-case" s -process in Ref. [7] unless the s -process temperature is reduced by about 1.5 keV from its canonical value of $kT \sim 27$ keV, thereby reducing the thermal photon density. In principle, significant K -mixing would make this matter worse. For example, mixing at the 1% level would require that the s -process temperature be reduced by about 4 keV. A matrix element on the order of several eV implies that K -mixing should at least contribute to the equilibration of ^{176}Lu . However, without experimental verification, it is difficult to predict the amount of ^{176}Lu produced, or

$^{176}\text{Hf}/^{176}\text{Lu}$ for a given temperature regime.

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