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SOME LIMITATIONS OF DETAILED BALANCE FOR INVERSE REACTION CALCULATIONS IN THE ASTROPHYSICAL p-PROCESS*

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ABSTRACT

p-Process modeling of some rare but stable proton-rich nuclei requires knowledge of a variety of neutron, charged particle, and photonuclear reaction rates at temperatures of 2 to 3×10^9 °K. Detailed balance is usually invoked to obtain the stellar photonuclear rates, in spite of a number of well-known constraints. In this work we attempt to calculate directly the stellar rates for (γ, n) and (γ, α) reactions on ^{151}Eu . These are compared with stellar rates obtained from detailed balance, using the same input parameters for the stellar (n, γ) and (α, γ) reactions on ^{150}Eu and ^{147}Pm , respectively. The two methods yielded somewhat different results, which will be discussed along with some sensitivity studies.

INTRODUCTION

The origins of the rare, proton-rich stable "p-nuclei" have been the subject of numerous investigations¹⁻⁵ in which proton capture or photonuclear reactions or both have been proposed to be the important reactions involved in the p-process, the stellar mechanism for producing such nuclei. The stellar model utilizing the latter reaction type is sometimes called the γ -process.⁴ Such modeling requires stellar photonuclear rates which are usually obtained by detailed balance.⁶ The approach is sound if the forward and reverse reactions involved proceed statistically such that, for the case $A+a=[C]=B+b$, the compound system $[C]$ has the same distributions in energy, angular momentum, and parity. However, there are a number of well-recognized constraints and in this paper we will try to determine the magnitude of the error introduced by some of these unfulfilled assumptions. We are able to attempt to analyze the validity of detailed balance for a variety of situations because our STAPLUS(Hauser-Feshbach plus spin-dependent preequilibrium) code⁷ allows photons in the entrance channel and also permits the spin-parity distributions in the first compound nucleus to be supplied as input.

CALCULATIONAL METHOD

The calculations presented here are for the target nucleus, ^{151}Eu ; the separation energies from ^{151}Eu are: $S_n = 7.937$ MeV, $S_p = 4.891$ MeV and $S_\alpha = -1.964$ MeV. We have chosen europium for this effort because more experimental information exists here than for the light Gd-Sm nuclei, which are of more astrophysical interest. We wrote a code called SUNBURN to calculate photonuclear transitions from target ^{151}Eu states available by the Boltzmann distribution at a given temperature, from some lower excitation energy (such as the ground state) up to an arbitrary maximum target excitation, the neutron separation energy for ^{151}Eu . In most of this work, this range has been divided up into 0.1 MeV energy bins. The photonuclear transitions

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proceed from these target bins to states above the specific reaction threshold up to another arbitrary maximum energy, which we have chosen to be 15 MeV. This is slightly above the $(\gamma, 2n)$ threshold. Actually, we compute the photon absorption cross section, and multiply this by the branching fraction for the reaction of interest. This absorption cross section is computed from the user's choice of multipole strength functions, $f_{xI}(E_\gamma)$, and so is used with each target-state spin, $J(E_t)$, to produce the spin and parity distribution in the ^{151}Eu compound nucleus:

$$\sigma_{\gamma ab}^{J\pi}(E_\gamma) = 3.85 \times 10^3 \left(\frac{2J+1}{2J(E_t)+1} \right) \sum_{xI} E_\gamma^{2I-1} f_{xI}(E_\gamma) \text{ barn.} \quad (1)$$

Multiplying this by the appropriate energy-, spin-, and parity-dependent branching fraction greatly simplifies the programming, and allows easy problem modifications.

STELLAR PHOTO-REACTION RATE

Assume Boltzmann-type thermal equilibrium

$$\lambda_{\gamma x} = \text{CON} / (2J_0 + 1) * \int_{E_l}^{E_s} dE_t * \frac{\rho(E_t) * \text{BF}(E_t) * (2J(E_t) + 1)}{Q / (2J_0 + 1)} \int_{E_x - E_t}^{E_h - E_t} dE_\gamma * \text{PLNK}(E_\gamma) * \sigma_{\gamma ab}(E_\gamma) * \quad (E_x - E_t \geq 0)$$

$$Q = \sum \rho(E_t) * \text{BF}(E_t) * (2J(E_t) + 1) \quad \left(\frac{\sigma_{\gamma x}(E_\gamma + E_t)}{\sigma_{\gamma ab}(E_\gamma)} \right) \text{ sec}^{-1}$$

$\rho(E_t)$ = target level density MeV^{-1}

$\text{CON} = 2.9979\text{e}+10 * 1.\text{e}-24 \text{ cm}^3 \text{ sec}^{-1} \text{ b}^{-1}$

$J(E_t)$ = most probable J at target E_t

$\text{BF}(E_t)$ = Boltzmann factor at E_t

$\text{PLNK}(E_\gamma)$ = Planck spectrum photons $\text{cm}^{-3} \text{ MeV}^{-1}$

$\sigma_{\gamma ab}$ = photon absorption cross section b

$\sigma_{\gamma x}(E)$ = (γ, x) cross section at $E = E_\gamma + E_t$

E_t is target energy where photon transitions start

E_l is lowest target energy (MeV)

E_h is highest target energy (MeV)

E_x is (γ, x) threshold (MeV) E_s = neutron separation energy (MeV)

Fig. 1. Equation for computing stellar photonuclear reaction rates that is incorporated in the computer code SUNBURN.

The stellar photonuclear rate expression is given in Fig. 1. All calculations were performed on an energy grid with 0.1 MeV steps. The excitation functions $\sigma_{\gamma ab}(E_\gamma)$ and $\sigma_{\gamma x}(E)$ were calculated with the Hauser-Feshbach plus precompound evaporation code STAPLUS,⁷ and interpolated onto the energy grid using a piece-wise cubic Hermite polynomial routine;⁸ the integrations were done using the trapezoidal rule. The integration accuracy was checked at $T_9=2$ by integrating the product of the level density and the Boltzmann factor, from 0 to 8 MeV, and comparing the answer with that obtained in closed form. The agreement was better than 1%, which may not be sufficiently accurate. We hope to try a Gaussian quadrature method in the future.

The neutron transmission coefficients were obtained using the coupled channel code ECIS⁹ and a deformed optical model potential.¹⁰ The proton¹¹ and alpha particle¹² potentials were both spherical. The alpha particles presented the greatest problem when computing transmission coefficients. None of our spherical or deformed potential codes were accurate for transmission coefficients below about 10⁻¹⁵ in magnitude. We intend to modify our codes to incorporate double-precision routines that compute Coulomb wave functions with greatly increased accuracy. In the present work, we graphically extended both $\sigma(\alpha, \gamma)$ and $\sigma(\gamma, \alpha)$ by the barrier penetration method from $E_\alpha=6$ down to about 4.5 MeV. This will not affect our results at $T_9=3$, but might at $T_9=2$. The multipole strength functions we favor for dipole and isoscalar E2 transitions have been described elsewhere.¹³ Our unadjusted strength functions produce photoneutron reactions on ¹⁵¹Eu and ¹⁵³Eu that compare well with the experimental data.¹⁴ Our unadjusted calculations agree very well with the ENDF/B evaluations for (n,2n) and (n,3n) reactions and with the neutron capture data¹⁰ for the (n, γ) reactions on targets of both ¹⁵¹Eu and ¹⁵³Eu. Testing the choice of alpha particle optical potentials is quite important; using the Igo volume potential¹⁵ yields a (γ, α) excitation function that is between 3 and 6 times the magnitude generated using the McFadden surface derivative potential¹². We compared our calculations with data¹⁶ for the (α, n) and (α, γ) reactions on ¹³⁹La and found the results to be quite satisfactory.

In order to compare our calculated stellar photonuclear reaction rates or rate factors with those computed via detailed balance, we follow the usual treatment⁶ and calculate the capture cross sections using the discrete target levels (14 to 32) weighted in their true sequence by the Maxwell-Boltzmann and (2J+1) factors and the partition function. In the continuum we use a representative capture cross section derived from the above weighting sequence. In standard notation, the detailed balance expression for the stellar photonuclear reaction rate factor $N_A \langle \bar{\sigma}_{\gamma X} v \rangle^*$ is related to the stellar rate factor for the (x, γ) capture reaction, $N_A \langle \bar{\sigma}_{X\gamma} v \rangle^*$, by

$$N_A \langle \bar{\sigma}_{\gamma X} v \rangle^* = \left(\frac{g_I g_X}{g_L} \right) \left(\frac{Q_I}{Q_L} \right) \left(\frac{A_I A_X}{A_L} \right)^{3/2} \left(\frac{kT}{2\pi \hbar^2 N_A} \right)^{3/2} N_A \langle \bar{\sigma}_{X\gamma} v \rangle^* \exp[-(S_X + E_X)/kT]. \quad (2)$$

We have changed the symbol for the partition function from G to Q, and that for the Q-value to $S_X + E_X$ to conform with Fig. 1. Here, S_X is the separation energy of particle X from the compound nucleus (say, ¹⁵¹Eu) and E_X is that particle's incident energy in the center of mass system. Note that the stellar photonuclear rate factor is equal to the actual stellar rate for nuclei at thermal equilibrium exposed to a Planck photon distribution, i.e., the rate $\lambda_{\gamma X}$ sec⁻¹ in Fig. 1 is numerically equal to the rate factor $N_A \langle \bar{\sigma}_{\gamma X} v \rangle^*$.

We may see that the two rate factors in Eq. 2 are linearly proportional; if Igo's transmission coefficients had been used instead of McFadden's, the stellar photonuclear reaction rate would have been a factor of 3 to 6 times faster than the rate we calculate. Of particular interest is the effect produced by low-energy charged particles trying to penetrate the Coulomb barrier in the (x, γ) reaction. For example, alpha particles on ¹⁴⁷Pm, producing the compound system ¹⁵¹Eu, are extremely insensitive to the photon widths, $\Gamma_\gamma(E_\gamma)$, and therefore to either the photon transmission coefficients or the level density in ¹⁵¹Eu. Either of the two may be divided by one or more orders of magnitude without changing the $\sigma(\alpha, \gamma)$ cross section, because of the very small size of $\Gamma_\alpha(E_\alpha)$. Below the neutron separation energy in ¹⁵¹Eu, the $\sigma(\alpha, \gamma)$ cross section is only the alpha particle reaction cross

section. There is essentially no information concerning photons contained within $N_A \langle \bar{\sigma}_{\alpha\gamma} v \rangle^*$, and yet Eq. 2 predicts a stellar (γ, α) reaction rate! The explanation, of course, is that Eq. 2 predicts one number, for a given temperature, that is the convolution of the Planck spectrum with some photonuclear excitation function, properly weighted and summed over all available target states. There are an infinite number of multipole strength-function combinations that will yield the same numerical rate, even if concurrent and complimentary variations in the ^{151}Eu level density are ignored.

One final comment should be made concerning Eq. 2. Assume for a moment that only dipole transitions are important for photonuclear reactions. These will produce a very narrow compound nucleus spin and parity distribution, as compared with that produced by incident particles, particularly the large number of partial waves brought in by alpha particles. To make the comparison of the rates calculated by the equation in Fig. 1 with that derived from detailed balance, we have utilized the feature of the STAPLUS code to read as input the normalized spin and parity distributions that a particle would produce in the compound nucleus and to distribute the photon absorption cross section accordingly, before allowing the photonuclear reactions to proceed.

RESULTS

In Fig. 2a, we show the calculated excitation function for $^{147}\text{Pm}(\alpha, \gamma)^{151}\text{Eu}$, with photon transmission coefficients from our systematics, and with these decreased by a factor of 10. Below the neutron separation energy in ^{151}Eu there is no visible difference between the two calculations. The same effect is found when the ^{151}Eu level density is decreased by the same amount. As mentioned in the previous section, the alpha particle is primarily exploring the Coulomb barrier, while the exponentially decreasing tail of the nuclear wave function interacts only slightly with the nucleus.

Figs. 2a,b. Change in the $^{147}\text{Pm}(\alpha, \gamma)^{151}\text{Eu}$ and $^{151}\text{Eu}(\gamma, \alpha)^{147}\text{Pm}$ cross sections due to 1/10 decrease in photon transmission coefficients.

Suppressing spin and parity,

$$\sigma_{\alpha\gamma}(E_\alpha) \approx \pi\lambda^2 \left(\frac{2\pi\Gamma_{\alpha 0}}{D} \right) \left(\frac{\Gamma_\gamma}{\Gamma_\gamma + \Gamma_\alpha + \Gamma_n} \right). \quad (3)$$

Recall that at excitation energies in ^{151}Eu below the neutron separation energy, $\sigma_{\alpha\gamma}(E_\alpha)$ is merely the alpha particle reaction cross section, $\sigma_{\alpha R\alpha}(E_\alpha)$. The symbol $\Gamma_{\alpha 0}$ represents the transition from the ground state of ^{147}Pm to the continuum in ^{151}Eu , and we have observed that $\Gamma_{\alpha 0}$ is usually $\geq 90\%$ of the total width, Γ_α . Because Γ_γ is many orders of magnitude larger than Γ_α , very large changes in Γ_γ produce no noticeable effect. In Fig. 2b, the reverse reaction is shown. Following the above argument concerning the relative sizes of the widths Γ_α and Γ_γ , it is easy to show why changing Γ_γ when Γ_n is zero produces no effect, but at higher excitation energies in ^{151}Eu , accessed by photons, a decrease in their transmission coefficients by a factor of 1/10 results in essentially the same decrease in $\sigma(\gamma, \alpha)$. For temperatures in the range of $T_9=2$ or 3, the Planck photon distribution convolutes primarily with the low-energy portion of the (γ, α) excitation function, and detailed balance calculations should be impacted only slightly. At higher temperatures a correct representation of the multipole strength becomes steadily more important, until such high temperatures are reached that multiple-particle emission occurs, such as $(\gamma, 2n)$ or $(\gamma, \alpha n)$, at which point the detailed balance approach is no longer useful. The approach of calculating directly the photonuclear reactions and their stellar rates, does not suffer from these restrictions, and clearly is useful in signaling the energy threshold where some of the restrictions of detailed balance begin to appear and hence the temperature range where detailed balance becomes less accurate.

Perhaps a more interesting illustration of the fact that detailed balance provides only a minimal constraint on the multipole strength functions, $f_{\lambda l}(E_\gamma)$, involves a comparison of the results using a giant dipole resonance shape for E1 strengths with the energy-independent f_{E1} from the Weisskopf single-particle approach. We applied a hindrance factor of 1×10^{-3} to the Weisskopf f_{E1} and normalized the $f_{E1} + f_{M1}$ such that $2\pi\Gamma_\gamma/D = 3.95$ at the neutron separation energy in ^{151}Eu - the same value our dipole systematics produced. Figs. 3a and 3b show the anticipated effects on the (γ, α) reaction of the two vastly different functional forms for f_{E1} , even when both Γ_γ 's were set equal at S_n . The effect on $\sigma(\alpha, \gamma)$ is barely discernable. This complete dominance by the Coulomb barrier may also be observed in the calculation of $N_A \langle \sigma_{\alpha\gamma} \rangle^*$, the forward rate factor in the detailed balance equation shown in Eq. 2. For ^{147}Pm , we used 14 discrete levels up to 0.807 MeV. Figure 4a presents a selection of $^{147}\text{Pm}(\alpha, \gamma)^{151}\text{Eu}$ excitation functions. For target spins of 0.5 to 5.5, and for level energies from zero to 0.807 MeV, the excitation functions overlap so well that a single curve was used to represent the collection in the convolution with the Maxwell-Boltzmann function. The order of the spin sequence in the $(2J+1)$ weighting was immaterial. This was not true in the (n, γ) case, as may be seen in Fig. 4b. Because of the Boltzmann factor weighting of the $(2J+1)$ term, the order of the spin sequence with excitation energy should not be neglected; the Maxwell-Boltzmann averaged capture cross section from each of the 27 levels in ^{150}Eu varied significantly.

The insensitivity of the stellar (γ, α) rate, below the (γ, n) threshold, to variations in the multipole strength-function modeling is not observed in the stellar (γ, n) rate because Γ_γ cannot be neglected as compared with Γ_n until the ^{151}Eu nucleus is several MeV above the (γ, n) threshold. Figure 5 displays the effect produced in the calculated $^{151}\text{Eu}(\gamma, n)^{150}\text{Eu}$ reaction by a 1/10 decrease in the photon transmission

Figs. 3a,b. Change in the $^{147}\text{Pm}(\alpha,\gamma)^{151}\text{Eu}$ and $^{151}\text{Eu}(\gamma,\alpha)^{147}\text{Pm}$ cross sections due to different dipole strength functions.

Figs. 4a,b. $^{147}\text{Pm}(\alpha,\gamma)^{151}\text{Eu}$ and $^{150}\text{Eu}(n,\gamma)^{151}\text{Eu}$ excitation functions for a selection of discrete level target states.

Fig. 5. Change in $^{151}\text{Eu}(\gamma,n)^{150}\text{Eu}$ cross section due to 1/10 decrease (--) in multipole strength functions.

coefficients; the decrease in $\sigma(\gamma,n)$ is also about 1/10, while the (n,γ) capture cross section decreases only about 40%. The stellar photoneutron rates show a factor-of-two decrease for $T_9=2$ to 3. Because the Planck distribution decreased by about 10^5 at $T_9=2$, for ^{151}Eu excitation energies of 8 to 10 MeV, we are not sure that our integration of the equation in Fig. 1 in steps of 0.1 MeV is sufficiently accurate. From 8.5 to 9.5 MeV of excitation, for example, the branching fraction $\sigma(\gamma,n)/\sigma_{\gamma ab}$ only changes from 0.59 to 0.77, but is 0.014 at 8.0 MeV. Not allowing γ -ray competition in the first compound nucleus increases the stellar photoneutron rate by almost a factor of 5. We plan to study other integration methods in the near future.

In Figs. 6a and 6b, we show our calculated stellar rates for the (γ,n) and (γ,α) reactions on ^{151}Eu , for temperatures of $T_9 = 2$ to 3. The curves show the rates vs target excitation energy, after which the total integrated rate is given. The Gamov peak values for the (α,γ) reaction at temperatures of $T_9=2.0, 2.5$, and 3.0 are, respectively, 5.5, 6.9, and 8 MeV; for the corresponding E_α energies add 2 MeV. These peak values are reasonable for the convolution of the Maxwell-Boltzmann function with the (α,γ) excitation function, whereas for the stellar (γ,α) rate functions shown in Fig. 6a the skewing of the curves' peak values to much lower excitation energies emphasizes the dramatic effect due to the shape of the Planck photon distribution. Hence, very low energy alpha particles contribute greatly to the stellar photonuclear rates. In Fig. 6b, our arbitrary cut off of ^{151}Eu target states at 8 MeV might appear to be too low an energy. However, an examination of the curve values near 8 MeV indicates little of the total integral was lost. Also, the behavior of the curves at low energies shows that discrete level information for the target nucleus is essentially not needed due to the moderation of the Boltzmann factor by the rapidly increasing level density function. Should one wish to compute stellar photonuclear rates at even higher temperatures, rate curves such as these, along with the calculated values of the neutron widths, should help decide the highest energy in ^{151}Eu where states are sufficiently long lived to be considered available target states.

We now wish to compare our stellar photonuclear reaction rates calculated directly with those obtained via detailed balance. First we must discuss the spin-parity distribution in the first compound nucleus, which must be the same for the forward and reverse reactions if detailed balance is to obtain. Photons produce a narrow distribution regardless of photon energy; an extreme example would be dipole transitions from a spin-zero target--only $J=1$ states could be produced. Neutrons incident on the $(Z,N-1)$ nucleus can produce a much broader distribution, even for relatively low incident energies. Alpha particles can bring in many partial waves and

Figs. 6a,b. Stellar photonuclear rates vs target excitation energy for the $(\gamma, \alpha)^{147}\text{Pm}$ and $(\gamma, n)^{150}\text{Eu}$ reactions, respectively, plus total rates.

produce very broad distributions. As noted above, our STAPLUS code allows us to supply a spin and parity distribution for the first compound nucleus, normalized to the actual (photon) reaction cross section. We then repeated our (γ, n) and (γ, α) excitation function calculations, changing the initial spin distribution to that which would have been produced by a neutron or an alpha particle of the appropriate center-of-mass energy, for ^{151}Eu excitation energies of 15 MeV down to the reaction threshold. The results appear in Figs. 7a and 7b, where the ratio of the directly calculated stellar rates to those obtained via detailed balance are given for the two types of photonuclear calculations--the normal calculation using the photon-produced spin distribution and the modified calculation using the particle-produced distribution as input. For the first case, both the (γ, α) and (γ, n) rates are about twice those inferred from detailed balance. When the particle-produced distributions were used, the (γ, α) ratios differed not greatly from unity, whereas the (γ, n) ratios decreased about 20% or so.

From our results so far, it cannot be said unequivocally that the direct calculation of stellar photonuclear rates is a more accurate procedure than the traditional detailed balance approach. However, because both the (γ, α) and (γ, n) stellar rates differ from the detailed balance results by about the same factor, and the reactions are so vastly different, there is some suggestion that the direct calculational approach is more reliable. Recall the function $J(E_J)$ in the equation in Fig. 1. For ^{151}Eu this function ranges from 1.5 at low target excitation energies to 5.5 for target energies near 8 MeV. As such, it tends to resemble the neutron distribution (although the higher spins are not represented), but it does not emulate the alpha particle distribution. Therefore, when the latter distributions are inserted into the (γ, α) calculations, Fig. 7a shows that the results reproduce those from detailed balance. Because the neutron distributions are more accurately reproduced by the $J(E_J)$ function, in the direct stellar rate calculation, their insertion as input produces only the small change shown in Fig. 7b. This suggests to us that the detailed balance requirement of identical

spin-parity distributions for the forward and reverse reactions is not being fulfilled--certainly not in our calculations and probably not in nature.

Figs. 7a,b. Stellar rates for $^{151}\text{Eu}(\gamma, \alpha)^{147}\text{Pm}$ and $^{151}\text{Eu}(\gamma, n)^{150}\text{Eu}$ reactions obtained from direct calculations ratioed to those inferred from detailed balance.

CONCLUSIONS

Using ^{151}Eu as the target isotope and with realistic nuclear parameters, we have compared the direct calculations of the stellar photo-reaction rates with those inferred from detailed balance. Because interest in the astrophysical p-process initiated this study, the temperature range was confined to $T_9 = 2$ to 3. A number of conclusions may be drawn:

1. The directly computed stellar photonuclear rates for the (γ, α) and (γ, n) reactions were about 2 to 3 times the rates inferred from detailed balance. It is argued, but not proved, that the direct calculational approach is the more accurate.
2. The direct computational approach is not limited to a single emitted particle; multiple particle or no particle emission may be treated as easily, i.e. $(\gamma, 2n)$, $(\gamma, \alpha n)$, a ground state populating an isomer, an isomeric state depopulated back to the ground state, etc.
3. The direct calculation is much less dependent on knowledge of the low-energy nuclear level structure than the usual approach.
4. The photonuclear excitation functions themselves provide useful information concerning the temperature at which the inverse reaction may begin to deviate from the restrictions imposed by detailed balance. Further, when they are used to produce stellar photonuclear rates as a function of the target excitation energy, one obtains additional information on the best choice of the energy range of the integrals shown in Fig. 1.
5. The p-process nuclei are stable while their predecessors often are not. Photonuclear reactions on stable nuclei should be easier to compute than capture reactions on unstable nuclei, simply because more experimental data are available to supply input to the nuclear reaction calculations.

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