Key Resonances in the ^{30}P(p, γ)^{31}S Gateway Reaction for the Production of Heavy Elements in ONe Novae

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Material emitted as ejecta from ONe novae outbursts is observed to be rich in elements as heavy as Ca. The bottleneck for the synthesis of elements beyond sulphur is the ^{30}P(p, γ)^{31}S reaction. Its reaction rate is, however, not well determined due to uncertainties in the properties of key resonances in the burning regime. In the present study, gamma-ray transitions are reported for the first time from all key states in ^{31}S relevant for the ^{30}P(p, γ)^{31}S reaction. The spins and parity of these resonances have been deduced, and energies have been measured with the highest precision to date. The uncertainty in the estimated ^{30}P(p, γ)^{31}S reaction rate has been drastically reduced. The rate using this new information is typically higher than previous estimates based on earlier experimental data, implying a higher flux of material processed to high-Z elements in novae, but it is in good agreement with predictions using the Hauser-Feshbach approach at higher burning temperatures.

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The accretion of hydrogen-rich material onto the surface of a compact white dwarf drives the thermonuclear runaway that powers classical nova events. They are the second most frequent explosive astrophysical events observed in our Galaxy after x-ray bursts [1]. Their energies are surpassed only by supernovae and x-ray bursts. The nature of nova outbursts depends strongly on the composition of the underlying white dwarf. More massive oxygen-neon (ONe) novae achieve significantly larger peak temperatures than reached in more frequent carbon-oxygen novae. ONe nova models [2,3] suggest that larger peak temperatures lead to the synthesis of nuclei up to $A \approx 40$, which is supported by infrared and ultraviolet observations of novae ejecta [4,5]. Theoretical models of nucleosynthesis in nova environments have been used to estimate the impact of individual reaction rate uncertainties on final nova yields [2,3]. For ONe novae, the ^{30}P(p, γ)^{31}S reaction has been shown to be the key gateway for the production of elements more massive than sulphur, with the uncertainty in the ^{30}P(p, γ)^{31}S reaction rate leading to large uncertainties in the predicted composition of nova ejecta [1].

Presolar grains from primitive meteorites are classified as being of nova origin if they display a large (relative to solar) ^{30}Si/^{28}Si ratio [6,7]. The ^{30}Si abundance, and the reliable astrophysical classification of these grains, depends critically on the ^{30}P(p, γ)^{31}S reaction rate. The present Letter reports first observations of gamma-ray transitions from all the ^{31}S resonances playing a key role in the ^{30}P(p, γ) reaction. The spin and parity (J^{π}) of these key resonances are determined, and their energies are measured with improved precision over previous studies, reducing uncertainties in the ^{30}P(p, γ)^{31}S reaction rate. The rate from this present work is typically higher than what was estimated previously for nova burning conditions, and implies a higher flux of material processed toward high-Z elements.

At temperatures relevant for ONe novae, the ^{30}P(p, γ)^{31}S reaction rate is expected to be dominated by low-spin resonances in ^{31}S close to the ^{30}P + p threshold {S_p = 6130.9(4) keV [8]}. A direct measurement of the reaction rate for these low-energy resonances is currently not possible due to the low intensities of ^{30}P beams. Therefore, an indirect approach is required to reduce uncertainties in the rate. Previously, the states in ^{31}S have been populated via transfer [9], charge exchange [10–12], and heavy-ion fusion-evaporation reactions [13,14]. In the latter gamma-ray spectroscopy study [13,14], transitions were observed from the high-spin states identified above the proton threshold, but none were observed for the key low-spin levels. In the present study, a light-ion fusion reaction was used to selectively populate low-spin states of interest above the proton threshold.

A 22 MeV, 10-pnA beam of ^{4}He ions from the Argonne ATLAS accelerator bombarded a 120 μg/cm^2-thick ^{28}Si target to produce ^{31}S nuclei via the one-neutron evaporation channel. The main fusion reaction channels corresponded to one-proton (^{31}P) and one-alpha (^{28}Si) particle evaporation. Gamma decays were detected with a Gammasphere [15,16] operated with a trigger requirement of two coincident gamma rays. Data were sorted off-line into γ-γ matrices and γ-γ-γ cubes. A spectrum produced by placing a gate on the 1249-keV transition in the γ-γ matrix is shown in Fig. 1. Energy and efficiency calibrations were performed using

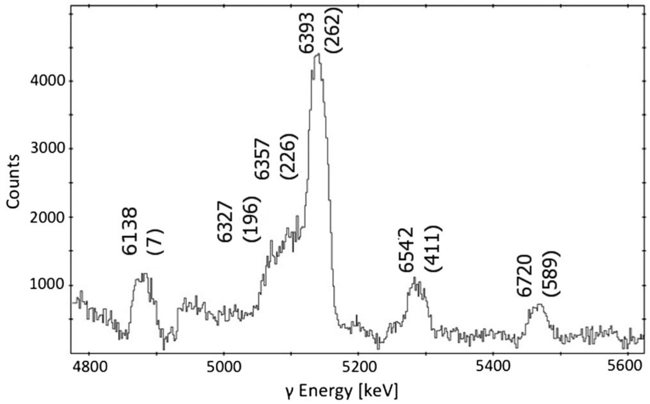

FIG. 1. Gamma-ray energy spectrum gated on the 1249-keV ($3/2_1^+$ to ground state) transition in ^{31}S. Numbers above peaks represent excitation energies of states in ^{31}S, and those in parentheses represent the resonance energies in the ^{30}P + p system.

^{152}Eu and ^{56}Co sources and a single high-energy line in ^{16}O. For strong transitions, intensities were fitted as a function of detection angle with respect to the beam axis with the function $W(\theta) = A_0\{1 + a_2P_2(\cos\theta) + a_4P_4(\cos\theta)\}$. Positive values for a_2 with $a_4 = 0$ signify $\Delta J = 0$ transitions, a negative value for a_2 and $a_4 = 0$, $\Delta J = 1$, and a positive value for a_2 and negative for a_4, $\Delta J = 2$, with values depending on the initial and final spins of the states. Due to the choice of a light-ion fusion evaporation channel with a relatively low degree of spin alignment, an additional angular correlation analysis was performed. $R_{\rm DCO}$ is defined as the ratio of the gamma-ray intensity at forward and backward angles to the intensity at 90° [$R_{\rm DCO} = \frac{I(\approx 0°)+I(\approx 180°)}{I(\approx 90°)}$]. Values for $\Delta J = 0$[1.05(3)], $\Delta J = 1$ [0.55(2) for pure $E1$, rising to ~0.80 for mixed $M1/E2$], and $\Delta J = 2$ [1.30(2)] transitions were determined by fitting known transitions observed in the study, as were the a_2, a_4 coefficients.

Table I presents a summary of the properties of the excited states above the proton threshold in ^{31}S from the present work. The excitation energies are in general significantly more precise and in good agreement with previous values. The gamma-ray transitions from six states are reported for the first time, including all of the key resonances for the ^{30}P(p, γ)^{31}S reaction rate in novae. The $1/2^+$ state at 6259 keV reported in a number of light-ion reaction studies [9–12] is not observed because it decays with a single direct transition to the ground state, which the experiment is not sensitive to. The $T = 3/2$ level at 6283 keV is also not observed because its population is isospin forbidden by the one-neutron evaporation reaction mechanism. Since it is also isospin forbidden for the ^{30}P(p, γ)^{31}S reaction, this state is unimportant for the reaction rate. The following discussion concentrates on the assignment of the key astrophysical resonances deduced from the newly reported gamma-ray transitions.

The 6327-keV state was first identified by Wrede *et al.* [10] using the ^{31}P(^{3}He, t)^{31}S charge-exchange reaction and tentatively assigned to either a $3/2^-$ or $7/2^+$ state. This level is observed here to decay via the 5078-keV gamma ray to the $3/2_1^+$ state (see Fig. 1). Angular distribution (AD) measurements of the 5078-keV gamma ray agree with a $\Delta J = 0$ transition from a $J = 3/2$ state, and are consistent with other similar transitions observed from known, low-lying $J = 3/2$ states in this study. There is only one candidate for a $3/2^+$ state (at 6233 keV) in the

TABLE I. Properties of excited states in ^{31}S. Level energies are corrected for the recoil of the compound nucleus. The first column lists the most precise energy measurement of the state from previous work.

E_x (keV) (previous)	E_x (keV) (present)	E_r (keV)	E_γ (keV)	a_2/a_4	$R_{\rm DCO}$	ΔJ	Assignment
6134(2)[a]	6138.3(21)	7.4(21)	2785.7(20)[c]			0, 2	$(3/2, 7/2)^+ \to 7/2^+$
			4889.5(6)[c]	0.15(12)/0.03(14)	1.12(24)	0, 2	$(3/2, 7/2)^+ \to 3/2^+$
6160.2(7)[b]	6158.5(5)	27.6(6)	1707.6(3)	0.22(2)/ − 0.07(3)	1.08(7)	0	$7/2^+ \to 7/2^-$
			2873.9(6)			1	$7/2^+ \to 5/2^+$
6259(1)[a]		129.1(10)					$1/2^+$
6283(2)[a]		152.1(21)					$3/2^+$; $T = 3/2$
6327(2)[a]	6327.0(5)	196.1(6)	5077.7(5)[c]	0.14(7)/0.10(9)	0.90(24)	0	$3/2^- \to 3/2^+$
6357(2)[a]	6357.3(2)	226.4(5)	5108.0(2)[c]	−0.25(5)/ − 0.01(7)	0.49(11)	1	$5/2^- \to 3/2^+$
6376.9(5)[b]	6376.9(4)	246.0(6)	1925.7(2)	−0.71(1)/0.03(1)	0.44(2)	1	$9/2^- \to 7/2^-$
			3025.4(3)	−0.46(1)/ − 0.01(2)	0.58(2)	1	$9/2^- \to 7/2^+$
	6392.5(2)	261.6(5)	5143.1(2)[c]	−0.25(3)/ − 0.09(3)	0.75(3)	1	$5/2^+ \to 3/2^+$
6393.7(5)[b]	6394.2(2)	263.3(4)	1091.2(4)	−0.37(5)/ − 0.01(1)	0.47(2)	1	$11/2^+ \to 9/2^+$
			3042.9(1)	0.26(1)/ − 0.26(1)	1.33(3)	2	$11/2^+ \to 7/2^+$
6543(2)[a]	6541.9(4)	411.0(6)	5292.5(4)[c]	0.13(4)/ − 0.03(6)	0.94(15)	0	$3/2^- \to 3/2^+$
6585(2)[a]	6583.1(20)	452.2(21)	3298.0(20)[c]			0, 1	$(5/2, 7/2)^- \to 5/2^+$
6636.3(15)[b]	6636.1(7)	505.2(8)	2184.9(4)			1	$9/2^- \to 7/2^-$
			3284.7(2)	−0.48(1)/ − 0.02(2)	0.49(3)	1	$9/2^- \to 7/2^+$

[a]Ref. [11].
[b]Ref. [14].
[c]Gamma rays observed for the first time in this work.

well studied mirror nucleus ^{31}P in this excitation energy region, but this would imply a large negative Coulomb energy difference for the analogue level (see Fig. 2 for the matching of ^{31}S levels with their mirror partners). Such negative energy shifts are not observed for positive-parity states in this region of the *sd* shell [17,18], and we therefore conclude that this is a $3/2^-$ resonance. In a more recent study of the ^{31}P(^{3}He, t)^{31}S charge-exchange reaction by Parikh *et al.* [12], an assignment of $1/2^+$ was deduced for the 6327-keV state using a coupled channels analysis of the particle angular distribution. However, this paper [12] also confirmed previous assignments of another $1/2^+$ level at 6259 keV discussed above. The observation of two $1/2^+$ states, however, is not consistent with the known level structure of the mirror partner ^{31}P, nor with shell model calculations [19] that indicate the existence of only one $1/2^+$ level in this region (at 6337 keV in ^{31}P) with a single, dominant gamma-decay transition to the ground state [20]. We therefore link the known $1/2^+$ state at 6259 keV in ^{31}S with the 6337-keV level in ^{31}P.

These mirror systematics indicate the existence of a second $3/2^-$ state in this energy range. We observe a transition at 5108 keV decaying toward the $3/2_1^+$ level (see Fig. 1) that shares the same AD characteristics as the 5078-keV transition from the $3/2^-$ level at 6327 keV. This second transition represents an excitation energy of 6542 keV and agrees with the energy of a level first reported by Wrede *et al.* [10,11]. However, no definite J^π assignment had previously been reported for this level.

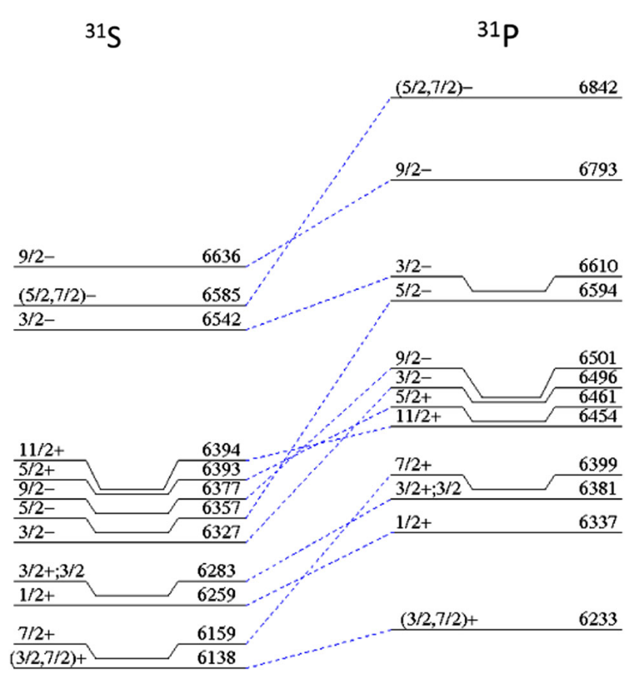

FIG. 2 (color online). Mirror diagram for states within 500 keV of the ^{30}P + p threshold. Excitation energies and J^π values in ^{31}S are from the present work with ^{31}P from Refs. [20,21,25].

In Fig. 2, the $3/2^-$ states at 6327 and 6542 keV in ^{31}S are paired with the 6610 and 6496 keV $3/2^-$ analogues in ^{31}P, which also have dominant decay branches to the $3/2_1^+$ level.

A new level is reported here at 6393 keV. The AD of the 5143-keV γ decay to the $3/2_1^+$ state is characteristic of a $\Delta J = 1$ transition. Since the AD is anisotropic, ruling out $J = 1/2$ states, a $J = 5/2$ level is proposed. The $R_{\rm DCO}$ value is inconsistent with a pure $E1$ transition and consistent with a mixed $M1/E2$ transition; therefore, we conclude that the 6393-keV level is a $5/2^+$ state and pair it with the one known $5/2^+$ state in this energy range in ^{31}P (at 6461 keV). The 6393-keV level has probably not been observed before because it would not have been resolved from the neighboring 6394-keV state in light-ion charge-particle reaction studies. Indeed, Wrede *et al.* [10,11] required the addition of a new, unresolved and unassigned level around 6400 keV in their (^{3}He, t) study to account for the fit to their data in the region of the known 6394-keV level. Parikh *et al.* [12] supported this observation in their later (^{3}He, t) study. With the introduction and assignment of the new state at 6393 keV, all known isobaric analogue states in the stable nucleus ^{31}P are now paired with levels in the region above the proton threshold in ^{31}S.

AD measurements for the 5108-keV gamma ray from the 6357-keV state, are consistent with a $\Delta J = \pm 1$ transition to the $3/2_1^+$ level. Since $1/2^+$ states decay isotropically and the AD is anisotropic, these are ruled out, and as there are no candidates left for $J^\pi = 5/2^+$ states, we propose that it is a $5/2^-$ state and pair it with the known analogue level at 6461 keV in ^{31}P (see Fig. 2). The $R_{\rm DCO}$ value is in good agreement with a pure $E1$ transition. The only previous definite assignment of this state was $3/2^+$ in the (^{3}He, t) study of Parikh *et al.* [12]; however, our gamma-ray AD data would require a positive value for a_2 of $\sim$0.2 which can be ruled out at the 9σ level. Furthermore, there is no credible available $3/2^+$ candidate in the relevant excitation range in the mirror nucleus ^{31}P [20]. There is a state at 6233 keV in ^{31}P assigned $(3/2\text{–}9/2)^+$ [20], but this would imply a very large negative Coulomb energy shift. Here we pair this with the 6138-keV level from which we identify gamma transitions to the $3/2_1^+$ and $7/2_1^+$ levels. The AD coefficients for the main transition to the $3/2^+$ level favors a $3/2^+$ assignment but the statistics are relatively poor, meaning a $\Delta J = 2$ transition from a $7/2^+$ level cannot be ruled out. Parikh *et al.* [12] report a $J = 9/2$ assignment for the 6138-keV level. However, in this event no gamma-ray transition would be observed to the $3/2_1^+$ level since an $E3/M3$ transition would not compete with $E1$, $M1$, or $E2$ transitions to other levels, in contradiction with the current observation of such a transition.

Prior to the work of Wrede *et al.* [10,11], the presence of a 6583-keV level was tentative; it is observed here in coincidence with the deexcitation of the $5/2_2^+$ state. In the ^{31}P mirror system, the only state in the energy range 6.4–7 MeV to exhibit such a decay branch is the 6842-keV level [21]. The weakly observed gamma-ray branch does not allow for an AD measurement; therefore, we adopt the J^π of the mirror as $(5/2, 7/2)^-$ from the compilation of Ref. [22].

For the estimation of the $^{30}P(p,\gamma)^{31}S$ reaction rate in Fig. 3(a) using the new resonance assignments, we follow the procedure described by Wrede *et al.* [10,11]. The spectroscopic factors of all but a few states are unknown. In keeping with Refs. [9–11,14], we choose $C^2S = 0.10$ for even-parity resonances, and $C^2S = 0.02$ for odd-parity ones when no other information is available. For temperatures in the region $0.10 \leq T \leq 0.25$ GK, the rate is dominated by resonances at 196 and 226 keV, with J^π assignments of $3/2^-$ and $5/2^-$, respectively, allowing for $l_p = 1$ capture to both states. For peak temperatures reached in ONe novae events, $0.25 \leq T \leq 0.42$ GK, the contribution of two resonances again dominates the rate. The resonance at 411 keV is assigned $J^\pi = 3/2^-$ and for the largest nova temperatures, $l_p = 1$ capture to this state is found to strongly influence the total rate. The J^π of the neighboring resonance at 452 keV is uncertain and is presently assigned as $(5/2, 7/2)^-$. Assuming $J^\pi = 5/2^-$ for the calculation implies an enhanced rate as the resonance may be populated by $l_p = 1$ capture, taking a $7/2^-$ assignment restricts the population to $l_p = 3$ protons and reduces the rate in the $0.25 \leq T \leq 0.42$ GK temperature region by a factor ~2.

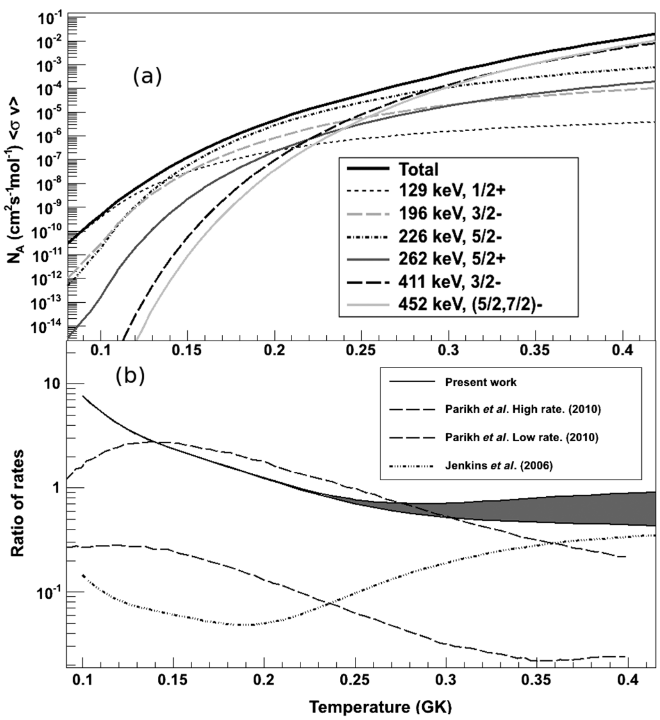

FIG. 3. (a) The $^{30}P(p,\gamma)^{31}S$ reaction rate estimated for temperatures relevant to ONe novae assuming the choice of spectroscopic factors discussed in the text. Individual contributions of resonances dominating the total rate are also shown. (b) Present estimated reaction rate as a ratio to the statistical model rate used in nova models [3,23]. The shaded region indicates the variation in reaction rate depending on whether a $5/2^-$ or $7/2^-$ assignment for the 452-keV resonance is taken. Also shown is the ratio estimated from the previous gamma-ray study [14], and the ratios corresponding to the upper and lower limits estimated in the study of Parikh *et al.* [12].

Figure 3(b) provides the estimated higher and lower $^{30}P(p,\gamma)^{31}S$ reaction rates from the present work taking the limiting l_p cases for the 452-keV resonance into account. They are presented as a ratio to the statistical model estimate used in the novae model calculations of José *et al.* [3]. It can be seen that there is now good agreement in the key astrophysical burning temperature region for ONe novae (from ~0.2–0.42 GK). A comparison is also given with the most recent estimates [12] that have a large spread between high and low $^{30}P(p,\gamma)^{31}S$ rates. Quoting Ref. [12], these "can be attributed directly to the uncertainties in the exact J^π values." The proton partial widths used in Ref. [12] were based on a Monte Carlo treatment, and the upper and lower reaction rate limits shown in Fig. 3(b) also incorporate estimated uncertainties in these widths. There are significant differences in the shapes of the rate ratios in Fig. 3(b). The rate ratios calculated by Parikh *et al.* [12] reach a maximum for $T \sim 0.15$ GK and then decrease gradually. However, in the present work, $1/2^+$ and $3/2^+$ assignments of Parikh *et al.* for the resonances at 196 and 226 keV are ruled out and are replaced with $l_p = 1$ captures in the lower temperature regime. Most of the difference can be attributed to this since Parikh *et al.* note that the rate above 0.08 GK is dominated by these states being populated by *s*-wave capture. A comparison is also provided in Fig. 3(b) with the most recent gamma-ray spectroscopy study of ^{31}S [14]. This heavy-ion fusion reaction did not identify the low-spin resonances populated in the light-ion fusion reaction utilized here, and the large increase in the predicted rate in the present study can be wholly attributed to the identification of these low-spin resonances.

The increased $^{30}P(p,\gamma)^{31}S$ reaction rate produces significantly higher yields of heavy elements in ONe novae ejecta in stellar model calculations. Iliadis *et al.* [2] performed reaction rate sensitivity calculations of elemental abundances in ejecta for different stellar models at the peak ONe novae burning temperature of ~0.25 GK. At this temperature, our estimated rate is ~10 times greater than both the lower limit set by Parikh *et al.* [12] and the value estimated by Jenkins *et al.* [14]. For example, this increased rate can lead to an increase of over a factor ~2 in the production of sulphur isotopes in ONe novae ejecta depending on the stellar model, and significant increases also for Cl and Ar isotopes [2]. However, the most notable consequence of the newly determined rate concerns the production of ^{30}Si, which has implications for mass ratio measurements in the classification of presolar grains of nova origin [6]. Here the factor $\sim$10 increase in the $^{30}P(p, \gamma)^{31}S$ reaction rate typically produces a factor $\sim$10 reduction in the expected abundance of ^{30}Si [2].

In summary, a light-ion fusion reaction populated low-spin states in ^{31}S above the proton threshold. Their γ decays were studied for the first time. This has led to the assignment of the key astrophysical resonances for the $^{30}P(p, \gamma)^{31}S$ reaction rate in novae. Consequently, there is a large reduction in the uncertainty in the estimated reaction rate. The present estimated rate now agrees well with that based on the statistical model [23] approach used by José *et al.* to calculate abundances of heavy elements ejected by ONe novae [3]. This rate is $\sim$10 higher at peak ONe novae burning temperatures compared to some recent estimates from experimental studies of the level structure of ^{31}S. This produces significant increases in the production of isotopes of the elements S, Cl, and Ar in novae ejecta, dependent on the stellar model, and a decrease of $\sim$10 in the predicted abundance of ^{30}Si in presolar grains of nova origin. Further progress in reducing uncertainties in the $^{30}P(p, \gamma)^{31}S$ reaction rate could be achieved by measurements of the largely unknown proton partial widths of the key resonances using transfer reactions [24].

[1] J. José, Astrophys. J. **560**, 897 (2001).
[2] C. Iliadis, A. Champagne, J. José, S. Starrfield, and P. Tupper, Astrophys. J. Suppl. Ser. **142**, 105 (2002).
[3] J. José, M. Hernanz, and C. Illiadis, Nucl. Phys. **A777**, 550 (2006).
[4] R.D. Gehrz, J.W. Truran, R.E. Williams, and S. Starrfield, Publ. Astron. Soc. Pac. **110**, 3 (1998).
[5] J. Andrea, H. Drechsel, and S. Starrfield, Astron. Astrophys. **291**, 869 (1994).
[6] J. José, M. Hernanz, S. Amari, K. Lodders, and E. Zinner, Astrophys. J. **612**, 414 (2004).
[7] S. Amari and K. Lodders, in *Highlights of Astronomy, Prague, 2006* (Cambridge University Press, Cambridge, 2007), p. 9.
[8] A. Kankainen *et al.*, Phys. Rev. C **82**, 052501(R) (2010).
[9] Z. Ma *et al.*, Phys. Rev. C **76**, 015803 (2007).
[10] C. Wrede, J. A. Caggiano, J. A. Clark, C. Deibel, A. Parikh, and P. D. Parker, Phys. Rev. C **76**, 052802(R) (2007).
[11] C. Wrede, J. A. Caggiano, J. A. Clark, C. M. Deibel, A. Parikh, and P. D. Parker, Phys. Rev. C **79**, 045803 (2009).
[12] A. Parikh *et al.*, Phys. Rev. C **83**, 045806 (2011).
[13] D. G. Jenkins *et al.*, Phys. Rev. C **72**, 031303(R) (2005).
[14] D. G. Jenkins *et al.*, Phys. Rev. C **73**, 065802 (2006).
[15] I. Y. Lee, Prog. Part. Nucl. Phys. **38**, 65 (1997).
[16] R. V. F. Janssens and F. S. Stephens, Nucl. Phys. News Int. **6**, No. 4, 9 (1996).
[17] J. Ekman *et al.*, Phys. Rev. Lett. **92**, 132502 (2004).
[18] G. Lotay, P. J. Woods, D. Seweryniak, M. P. Carpenter, H. M. David, R. V. F. Janssens, and S. Zhu, Phys. Rev. C **84**, 035802 (2011).
[19] B. A. Brown, http://www.nscl.msu.edu/brown/resources/SDE.HTM.
[20] P. M. Endt and C. van der Leun, Nucl. Phys. **A310**, 1 (1978).
[21] E. O. De Neijs, G. D. Haasbroek, M. A. Meyer, R. S. Rossouw, D. Reitmann, Nucl. Phys. **A254**, 45 (1975).
[22] A. C. Wolff and H. G. Leighton, Nucl. Phys. **A140**, 319 (1970).
[23] T. Rauscher and F.-K. Thielemann, At. Data Nucl. Data Tables **75**, 1 (2000).
[24] A. M. Mukhamedzhanov, Phys. Rev. C **84**, 044616 (2011).
[25] A. C. Wolff, M. A. Meyer, and P. M. Endt, Nucl. Phys. **A107**, 332 (1968).