Measurement of ^{25}Mg$(p,\gamma)^{26}$Al^g resonance strengths via accelerator mass spectrometry

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The strengths of resonances located at center-of-mass energies of $E_{\mathrm{c.m.}} = 189$, 304, 374, and 418 keV for the ^{25}Mg(p,γ) reaction have been measured for the first time with an off-line method: Mg targets were firstly activated with protons at the resonance energies and the produced ^{26}Al^g nuclei were counted by means of highly sensitive accelerator mass spectrometry (AMS). Thus, the production of ^{26}Al in its ground state is determined independently from the γ-decay branching ratio. While the 304, 374, and 418 keV resonances show fair agreement with previous measurements, the 189 keV resonance yield a significantly less strength. In addition, an experimental upper limit for the $E_{\mathrm{c.m.}} = 92$ keV resonance was determined.

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I. INTRODUCTION

In the last 20 years radioactive ^{26}Al became a key isotope to understand and validate models of stellar nucleosynthesis. It was as long as 50 years ago introduced into astrophysics by Urey [1] as a radioactive heat source capable of melting asteroids (see also Ref. [2]). This assumption led to the prediction of an isotopic excess of ^{26}Mg in those bodies caused by the β-decay of ^{26}Al. This ^{26}Mg excess was found in Al-rich inclusions of the Allende meteorite [3], which motivated the search for live extraterrestrial ^{26}Al as the main target of γ-ray astronomy. The ^{26}Al ground state ($t_{1/2} = 0.716$ Myr) decays via β^+ and EC to the 2^+ first excited state in ^{26}Mg, whose deexcitation exhibits a γ-ray of 1.809 MeV. The isomeric state of ^{26}Al at $E_x = 228$ keV ($t_{1/2} = 6.3$ s) decays exclusively to the ground state of ^{26}Mg and it is not associated with the emission of γ-rays.

In 1982 the cosmic 1.809 MeV γ-ray line was experimentally detected and identified by the HEAO-C experiment [4], triggering considerable interest in determining the sources of interstellar ^{26}Al. The 1.809 MeV γ-ray was in fact the first extra-solar radioactivity ever detected and, due to the cosmologically short half-life of ^{26}Al, it is considered as a proof of ongoing nucleosynthesis within the Milky Way. The observed flux, $\phi_\gamma = 4 \times 10^{-4}\,\mathrm{cm}^{-2}\,\mathrm{s}^{-1}\,\mathrm{rad}^{-1}$, is consistent with an amount of 3.4 $M_\odot$ of ^{26}Al in the interstellar medium. Nowadays, γ-ray telescopes map the 1.809 MeV emissions in our Galaxy, enabling the identification of active regions, and thus representing a valuable tool for the validation of nucleosynthesis models [5].

Stellar ^{26}Al is produced within the Mg-Al chain through the ^{25}Mg(p,γ) reaction at high temperature H- or C-Ne-burning. Hydrostatic H-burning occurs at temperatures in the range of 30–90 MK in massive stars (40 $M_\odot < M < 120\ M_\odot$) during their Wolf-Rayet (WR) phase or in less massive stars ($M < 9\ M_\odot$) during their Asymptotic Giant Branch (AGB) phase, while explosive conditions (0.1 GK < T < 0.4 GK) are met in white dwarfs that accrete H-rich material and lead to nova outbursts. ^{26}Al is also produced in hydrostatic C-Ne burning prior to a supernova (SN) (T $\sim$ 1 GK), and to a lesser extent, during the explosive phase itself (T $\sim$ 2–3 GK). Although some evidence favors WR stars [6], the sources of ^{26}Al are still not fully determined.

For a better understanding of the possible sources of ^{26}Al more accurate rates for the ^{25}Mg(p,γ) reaction are required. At the temperatures mentioned above, the ^{26}Al production rate depends on very weak resonances whose determination poses a very difficult task to the on-line detection of the prompt γ-ray emission [7,8].

In this work we present a new and independent approach to measure the strengths of resonances that dominate the ^{25}Mg$(p,\gamma)^{26}$Al^g reaction at temperatures in the 0.1–1.5 GK range (see also Refs. [9,10]): ^{26}Al was firstly produced by proton bombardment of ^{25}Mg targets at the corresponding resonance energies, and the resulting ^{26}Al^g nuclei were counted off-line by means of the AMS technique.

II. DETERMINATION OF THE ^{25}Mg$(p,\gamma)^{26}$Al^g REACTION RATE

The stellar reaction rate $N_A\langle\sigma v\rangle$ at a given temperature T is calculated as [11]

$$N_A\langle\sigma v\rangle = N_A\sqrt{\frac{8}{\pi\mu(kT)^3}}\int_0^\infty \sigma E\, \mathrm{e}^{-E/kT}\, dE, \quad (1)$$

where N_A is the Avogadro's number, μ the reduced mass of the system, k the Boltzmann constant, σ the cross section, v the relative velocity, and E the energy in the center-of-mass system. As shown by [12] contributions from direct capture process and from the high-energy tail of the $E_R = -25.7$ keV subthreshold resonance are negligible for $T > 7$ MK. The ^{25}Mg(p,γ) reaction rate is then dominated by

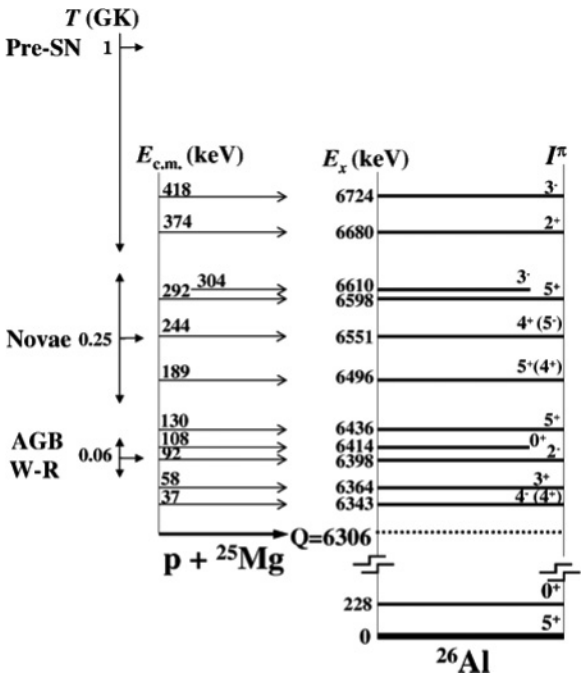

FIG. 1. Level scheme of ^{26}Al. Plotted are the ground state ($t_{1/2}$ = 0.716 Myr), the isomeric state at 228 keV ($t_{1/2}$ = 6.3 s) and excited levels (E_x) above the ^{25}Mg + p threshold (Q = 6306 keV) with the corresponding center-of-mass resonance energies ($E_{\text{c.m.}}$). Typical temperatures and the corresponding Gamow peaks for different astrophysical scenarios are also shown. For a given temperature, the Gamow peak indicates *a priori* which resonances might have the larger contribution to the reaction rate.

narrow resonances (total width $\Gamma = \Gamma_\gamma + \Gamma_p$ much smaller than its resonance energy E_R, where Γ_γ and Γ_p denote the partial widths for the γ and proton decay channels, respectively). The relevant resonances lie just above the proton capture threshold, corresponding to reaction energies of few hundreds keV, far below the Coulomb barrier of this reaction (E^{CB}= 2.86 MeV) (see Fig. 1).

Hence, the reaction rate can be calculated as [11]

$$\langle \sigma\, v \rangle = \hbar^2 \left(\frac{2\pi}{\mu k T}\right)^{3/2} \sum_i (\omega\gamma)_i \, \mathrm{e}^{-E_R^i/kT}, \qquad (2)$$

where $(\omega\gamma)_i = \frac{2J_R^i+1}{(2J_p+1)(2J_T+1)} \frac{\Gamma_p^i \Gamma_\gamma^i}{\Gamma_p^i+\Gamma_\gamma^i}$ are the resonance strengths. Here, J_R, J_p, and J_T are the angular momentum of the excited level of the compound nucleus, the projectile, and the target nucleus, respectively. The experimental determination of the resonance strengths is usually done by proton bombarding of thick ^{25}Mg targets (meaning that the energy loss of the particles in the target is much larger than the resonance widths, i.e. $\Delta E_{\text{target}} \gg \Gamma$). In such cases the resonance strengths are obtained as

$$\omega\gamma = \frac{2}{\lambda^2} \frac{m_T}{m_T + m_p} \varepsilon Y, \qquad (3)$$

where λ is the center-of-mass de Broglie wavelength of the projectile, m_T and m_p are the masses of the target and the projectile, respectively, $\varepsilon = \frac{dE}{n\,dx}$ is the stopping power, and Y the yield of products [11].

In order to avoid the drawbacks arising from interferences caused by γ-ray background when applying an on-line detection method, we have measured the accumulated number of ^{26}Al^g produced in the proton irradiation by means of AMS. This technique is capable of measuring isotope ratios down to 10^{-16} with an efficiency in the range of 10^{-2}–10^{-5} depending on the radioisotope under investigation. In the case of ^{26}Al detection, this sensitivity allows a quantitative AMS-measurement of resonance strengths down to values of 10^{-7} eV after one day of activation assuming a 1 mA beam.

We note that when using this method only the production of the astrophysically-observable ^{26}Al^g (ground state) is measured. The ^{26}Al^m ($t_{1/2}$ = 6.3 s) isomer has decayed long before the AMS counting takes place.

III. SAMPLE IRRADIATION AND AMS MEASUREMENTS

The targets exposed to the proton irradiation were made of MgO, deposited on glass-carbon plates. These targets were irradiated using two different implanters at the Forschungszentrum Rossendorf, Germany. In both implanters the beam current was monitored by a precision current transducer and integrated to determine the total irradiated dose with negligible uncertainty.

For the resonances at $E_{\text{c.m.}}$ = 304, 374 and 418 keV a 500 kV, 10 μA ion implanter was used. Natural MgO targets (140–200 μg/cm^2 thick) were irradiated for a few hours with doses of about 5×10^{18} protons. For these three resonances approximately 10^6–10^7 ^{26}Al nuclei were produced.

For the $E_{\text{c.m.}}$ = 189 keV resonance, a different ion implanter (210 kV, 1 mA) was utilized. In this case, four ^{25}MgO targets (93% enrichment, 45–64 μg/cm^2 thick) were irradiated in different runs with a total irradiation time of 10 days and a dose of 1.1×10^{21} protons, producing about 10^5 ^{26}Al nuclei. To avoid the loss of MgO due to sputtering during the proton bombardment, the targets have been covered with thin tungsten protection layers (16–17 μg/cm^2). The energy loss caused by these layers were estimated to be about 1.8 keV, with a (1 s.d.) straggling of 0.7 keV. Therefore, the beam energy was set at 203 keV to assure that all protons enter the MgO layer with an energy above the resonance value,[1] E_{lab} = (197.1 ± 0.1) keV. Moreover, the thickness of the MgO layer, equivalent to an energy loss of about (20 ± 2) keV, ensures that, on the one side, all protons meet the resonance energy within this layer and, on the other side, that ^{26}Al recoils (with a maximum range of 6 μg/cm^2) are retained within the MgO layer. These conditions are fulfilled provided the uncertainties in the target thickness (about 10%) and in the location of the resonance energy, and taking into account the maximum deviation in the absolute value of the beam energy (2 keV). The corresponding calculations have been performed applying SRIM2003 [13,14].

In addition, an enriched ^{25}MgO target was irradiated with 4×10^{20} protons at the resonance energy $E_{\text{c.m.}}$ = 92 keV using

[1] Resonance energies were calculated from the excited levels given in Ref. [15] with Q = (6306.45 ± 0.05) keV [16].

TABLE I. Relevant experimental parameters for the determination of the resonance strengths. The $^{26}Al/^{27}Al$ ratio of samples irradiated at resonance energies of $E_{c.m.} = 304.0$, 374.0 and 417.8 keV were measured at both AMS facilities, in Munich (M) and at the VERA laboratory in Vienna (V). For those cases the weighted mean value (mean) is also indicated. For the resonance at $E_{c.m.} = 189.5$ keV four different targets were independently irradiated (samples *a* to *d*). Those samples and the one irradiated at $E_{c.m.} = 92.2$ keV were measured at VERA only. Quoted $^{26}Al/^{27}Al$ ratios are corrected for background.

Resonance energy $E_{c.m.}$ (keV)	Proton dose N_p ($\times 10^{18}$)	Al-carrier C_{Al} (μg)	^{26}Al events N_{26}^{det}	Background events	$^{26}Al/^{27}Al$ ratio r ($\times 10^{-15}$)	
92.2	390	100	0	0.1 ± 0.05	<5	(V)
189.5	(a) 230	500	2	0.6 ± 0.2	2.1 ± 1.5	(V)
	(b) 337	250	4	0.4 ± 0.1	12 ± 6	(V)
	(c) 223	400	9	1.4 ± 0.3	3.0 ± 1.1	(V)
	(d) 352	400	9	1.5 ± 0.3	2.8 ± 1.1	(V)
304.0	5.4	500	109	0.8 ± 0.4	844 ± 120	(M)
			110	0.1 ± 0.02	591 ± 58	(V)
				mean:	640 ± 65	
374.0	1.0	300	53	2.5 ± 1	364 ± 64	(M)
			50	0.1 ± 0.01	314 ± 45	(V)
				mean:	333 ± 37	
417.8	4.5	500	120	0.4 ± 0.2	1527 ± 207	(M)
			1075	0.4 ± 0.06	1510 ± 55	(V)
				mean:	1511 ± 53	

the 210 kV implanter, achieving an experimental upper limit for this resonance (see Table I for details).

The irradiated targets were chemically treated as follows: (a) The target material was dissolved in acid; (b) 250–500 μg Al carrier material, already dissolved in the acid, was added and homogenized together with the produced ^{26}Al; (c) both Al isotopes (^{27}Al and ^{26}Al) were separated from the MgO target matrix, eliminating the Mg down to a concentration of about 5 ppm; (d) the remaining material was converted into Al_2O_3 in order to be used as sample material for the negative-ion sputter source (see Ref. [17] for further details). It is assumed that no fractionation of the Al isotopes occurs during the chemical treatment.

The $^{26}Al/^{27}Al$ ratios of the samples (in the range of 10^{15}–10^{-12}) were determined by means of AMS: the ^{27}Al current was measured with Faraday cups while the ^{26}Al ions were identified and counted with a heavy-ion detection system. The absolute isotope ratios were achieved by comparison with well-known $^{26}Al/^{27}Al$ standards. The background level of the Al measurements was determined by means of blank samples made out of the same Al material and of nonactivated MgO, as well.

Samples for resonances at $E_{c.m.} = 304$, 374, and 418 keV were measured using the Munich 14-MV tandem accelerator. In this case, $^{26}AlO^-$ and $^{27}AlO^-$ ions were extracted from the ion source and accelerated by applying terminal voltages of 12.5 and 12.0 MV, respectively. These molecules were dissociated in a carbon stripper foil, spurious ions were eliminated by a Wien filter and the $^{26}Al^{7+}$ were selected by the analyzing magnet. We extracted AlO^- ions from the ion source as they are formed by a higher efficiency than Al^- (about a factor of 20). Usually $^{26}Al^-$ is used for the AMS measurements since the corresponding stable isobar ^{26}Mg does not form negative ions, while ^{26}MgO does. Hence, in this case it was necessary to suppress the isobar $^{26}Mg^{7+}$ by about nine orders of magnitude, which was achieved by the combined use of a gas-filled magnet and a multi-ΔE ionization chamber [18]. This technique requires high beam energies (>3 MeV/nucleon) which are available with the Munich 14 MV facility.

As a crosscheck, the samples measured in Munich were also independently measured at the Vienna Environmental Research Accelerator (VERA) laboratory. This dedicated AMS facility is based on a 3-MV tandem accelerator and offers a high overall efficiency through the whole system. In addition, a ^{26}Al detection limit as low as 6×10^{-16} for the $^{26}Al/^{27}Al$ isotope ratio was demonstrated [19]. A fast switching system between the radioisotope ^{26}Al and the stable isotope ^{27}Al results in high precision data [20]. Al^- ions were extracted from the negative ion source (SNICS II equipped with a 40-sample target wheel), therefore, there were no isobaric interferences from ^{26}Mg in this case. Selecting the 3^+ charge state after the tandem accelerator resulted in a particle transmission of 50% from the low energy side to the particle detector. The overall efficiency at VERA was determined to be 5×10^{-4}.

It should be noted that the measurements performed in Vienna and in Munich are based on two independent AMS facilities using different approaches for the quantification of the radioisotope ^{26}Al.

In addition to the higher energy resonances, samples irradiated at the resonance energies of $E_{c.m.} = 92$ and 189 keV were also measured at VERA. For these low energies, resonance strengths five orders of magnitude lower than for the previous higher-energy resonances were expected, i.e., $^{26}Al/^{27}Al$ isotope ratios of the order of a few times 10^{-15} were obtained. At such low ratios a well quantified background level becomes important. To this end a series of blank samples (up to 11 blank samples per measurement series) were measured continuously in succession to the irradiated samples. Because counting statistics of both blank and irradiated samples dominated the

TABLE II. Resonance strengths of the ^{25}Mg(p,γ) reaction for the formation of ^{26}Al in its ground state, $\omega\gamma^g = \omega\gamma f_0$. Listed values for Refs. [21–23] are calculated from published resonance strengths $\omega\gamma$, and branching ratios to the ground state f_0 taken from Ref. [12] (see also Refs. [7,8]). Errors indicate a 68% (1 s.d.) confidence level.

$E_{\text{c.m.}}$ (keV)	f_0	$\omega\gamma^{\text{g}}$ (eV)		
		Present work	NACRE [21,22]	Powell *et al.* [23]
92.2	(85 ± 1)%	$<2\times10^{-8}$	$(1^{+0.1}_{-0.3})\times10^{-10}$	
189.5	(74 ± 1)%	$(1.1\pm0.2)\times10^{-7}$	$(5.3\pm0.7)\times10^{-7}$	
304.0	(87 ± 1)%	$(2.1\pm0.2)\times10^{-2}$	$(2.7\pm0.2)\times10^{-2}$	
374.0	(67 ± 1)%	$(4.0\pm0.4)\times10^{-2}$	$(4.2\pm0.3)\times10^{-2}$	
417.8	(96 ± 1)%	$(7.1\pm0.2)\times10^{-2}$	$(11.1\pm0.6)\times10^{-2}$	$(9.0\pm0.6)\times10^{-2}$

final uncertainty, the measurements were repeated until the whole sample material was used up in the AMS measurement. Indeed, the high number of blank samples proved a constant and low background level. In addition, they provided enough counts in order to confine the background uncertainty to a level where it did not contribute significantly to the total uncertainty of our final data. Similarly to the Munich measurements, a series of Al standards with well known isotope ratios were used for the determination of the absolute scaling factors of the AMS measurement. A systematic uncertainty of 2% was added to the statistical error for the VERA results (mainly the error of the standard materials). For the results obtained in Munich a systematic uncertainty of 10% was added as a conservative assumption (see Table I).

In principle, two effects might lead to a lower ^{26}Al concentration in the sample: (a) a loss of ^{25}MgO, eventually together with the already produced ^{26}Al atoms contained therein, due to sputtering of the target material induced by the intense proton beam, and (b) an incomplete removal of the ^{25}MgO material from the target backing (glass carbon). The W protection layer withstood the irradiation and it could be totally removed in the chemical treatment of the samples. However, in order to rule out the above mentioned two possibilities of losses, a quantitative measurement of the total amount of Mg extracted in the chemical process was performed. The measurement was performed by inductively coupled plasma-mass spectrometry (ICP-MS). It yielded a Mg concentration in the extracted solution of (13.2 ± 1.1) mg/L, in perfect agreement with the expected value of 12 mg/L. Moreover, to rule out any possible contamination from external Mg, the isotopic composition of the extracted Mg was also determined. The ratio obtained was ^{24}Mg:^{25}Mg:^{26}Mg = 4.5 ± 0.2 : 93.5 ± 0.2 : 2.01 ± 0.03 (2 s.d.), which is again in perfect agreement with the specification of the enriched ^{25}Mg material used in the targets (4.4 : 93.3 : 2.3).

IV. EXPERIMENTAL RESULTS

Resonance strenghts are derived from experimental data according to Eq. (3). The yield of products Y, defined as the ratio between the number of produced nuclei and the number of incident projectiles, is in our case obtained as

$$Y = \frac{N_{26}^{\text{det}}}{N_p\ \varepsilon_{\text{AMS}}} = \frac{r\ C_{\text{Al}}}{N_p}, \tag{4}$$

where N_{26}^{det} is the number of detected ^{26}Al counts, N_p the number of incident protons, $\varepsilon_{\text{AMS}} = N_{27}^{\text{det}}/C_{\text{Al}}$ is the efficiency of the AMS measurement, C_{Al} is the amount of Al carrier used and r is the measured ^{26}Al/^{27}Al isotope ratio.

Table II (see also Fig. 2) lists the results obtained in this work in comparison with values adopted by the NACRE Collaboration (Nuclear Astrophysics Compilation of Reaction Rates) [21,22] and with a value measured by Powell *et al.* [23] (not included in the NACRE compilation). Measured values for the strength of resonances located at $E_{\text{c.m.}} = 304$ and 374 keV are in very good agreement with the values recommended by NACRE, whereas for the resonance at $E_{\text{c.m.}} = 418$ keV a rather lower value was obtained.

In the case of the $E_{\text{c.m.}} = 189$ keV resonance, the strengths obtained from the four AMS measurements were consistent with each other but significantly below the value recommended by NACRE (see Fig. 3). In all cases, intervals indicate a 68% confidence limit.

We note that up to now there is no direct measurement of the strength of the $E_{\text{c.m.}} = 92$ keV resonance. The

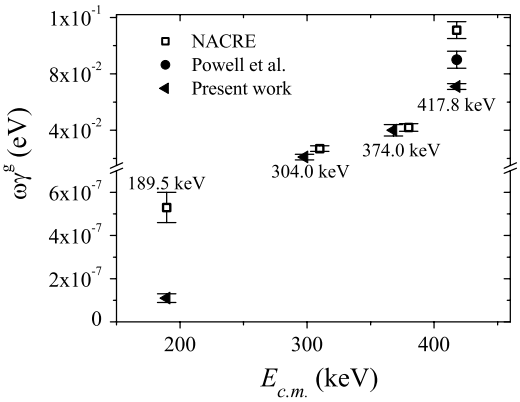

FIG. 2. Strengths of resonances at $E_{\text{c.m.}} = 189$, 304, 374, and 418 keV of the ^{25}Mg(p,γ) reaction for the formation of ^{26}Al in its ground state. Measured values of this work are compared to those recommended by NACRE [21,22] and to a value measured by Powell *et al.* [23].

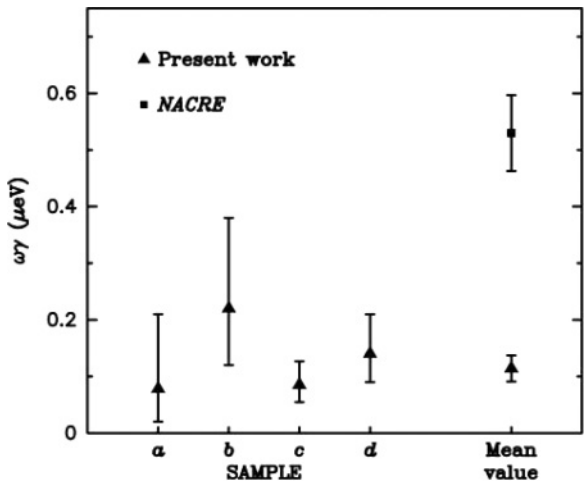

FIG. 3. Results of the four independent resonance-strength measurements (a, b, c, and d) at $E_{\text{c.m.}} = 189$ keV performed in the present work and their mean value, compared to the recommendation of NACRE [21,22].

value recommended by NACRE corresponds to a calculation based on the distorted wave Born approximation using proton partial widths derived from the single-particle transfer reaction ^{25}Mg(^{3}He, d)^{26}Al [24,25].

The four measured resonances, $E_{\text{c.m.}} = 189$, 304, 374, and 418 keV, dominate the reaction rate [as expressed in Eq. (2)] in the temperature range from 0.1 to 1.5 GK (see also Fig. 1). Figure 4(a) shows the individual contribution of these resonances (assuming present work values) to the reaction rate and the corresponding total reaction rate (using NACRE recommendations for the other 85 resonances lying between $E_{\text{c.m.}} = 37.5$ and 1920.5 keV). These rates were calculated with the program RATEERRORS [26], developed by Thompson and Iliadis [27]. In order to visualize the differences of the present work rate with that recommended by NACRE Figure 4(b) shows the ratio of these rates with their corresponding 68% lower and upper confidence limits. These limits are derived from the respective uncertainties in the resonance strengths (see Table II).

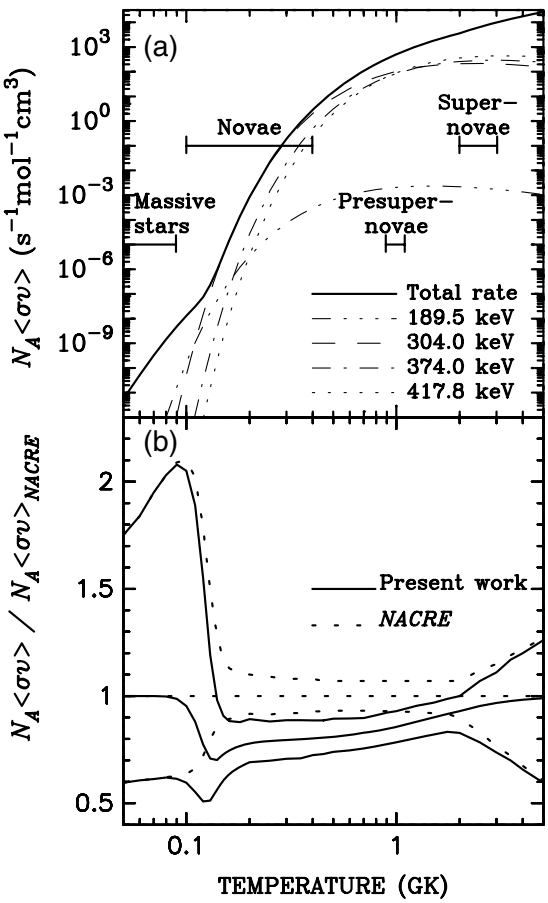

FIG. 4. Dependence of the ^{25}Mg$(p,\gamma)^{26}$Al^g reaction rate with the stellar temperature. (a) Individual contributions of the resonances at $E_{\text{c.m.}} = 189$, 304, 374, and 418 keV to the reaction rate, calculated from the values obtained in this work, and total reaction rate (full line) using values recommended by NACRE [21,22] for other 85 resonances between $E_{\text{c.m.}} = 37.5$ and 1920.5 keV. These calculations were performed with the program RATEERRORS [26]. The characteristic temperature ranges in which ^{26}Al is produced in the various astrophysical scenarios are also shown. (b) Ratio of the total reaction rate obtained in the present work to that recommended by NACRE, with their corresponding 68% lower and upper confidence limits (solid lines). NACRE confidence limits are also plotted (dashed lines).

V. SUMMARY AND CONCLUSIONS

We present new measurements of the strengths of the four resonances which dominate the ^{25}Mg(p,γ) reaction leading to the ground state of ^{26}Al at temperatures in the 0.1–1.5 GK range. They were measured for the first time by an off-line method, independent of the usual prompt γ-ray detection. In this method the formation of the astrophysically-relevant ^{26}Al^g is directly determined without reference to the branching ratio f_0 to the ground state. While there is fair agreement for the resonances at $E_{\text{c.m.}} = 304$, 374, and 418 keV we found a significantly lower strength value at $E_{\text{c.m.}} = 189$ keV when compared to the value recommended by NACRE (basically determined by the on-line measurement performed by Iliadis *et al*. [8]). We extensively investigated several possible effects entering in the determination of the resonance strengths, not finding any reasonable contribution able to increase our measured value at 189 keV by a factor of 5 so as to match the NACRE value.

According to the present results, the production of ^{26}Al via the proton capture reaction ^{25}Mg(p,γ) is significantly lower than the accepted values at stellar temperatures in the range of 0.1–1 GK, which are typical in presupernovae and in novae explosions.

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