Properties of the 5\(^{-}\) state at 839 keV in \(^{176}\)Lu and the s-process branching at \(A = 176\)

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(Dated: March 23, 2009)

PACS numbers: 26.20.Kn,23.35.+g,25.20.Dc,27.70.+q

I. INTRODUCTION

Several studies have been devoted to the s-process branching at mass number \(A = 176\) in the last years \([1, 2, 3, 4]\). \(^{176}\)Lu and \(^{176}\)Hf are s-only nuclei and shielded from the r-process by stable \(^{176}\)Yb. At first view, the abundance ratio between the unstable \(^{176}\)Lu and the stable \(^{176}\)Hf seems to be a perfect chronometer for the s-process because of the long \(\beta\)-decay half-life of \(^{176}\)Lu of about 40 giga-years. However, this ground state half-life of \(^{176}\)Lu may be dramatically reduced under stellar conditions because of the coupling of the long-living \(J^\pi = 7^-; K = 7\) ground state to a low-lying, short-living \(1^-; 0\) isomer at \(E_x = 123\) keV via so-called intermediate states (IS) at higher energies. The \(1^-\) isomer in \(^{176}\)Lu also \(\beta\)-decays to \(^{176}\)Hf with a short half-life of \(t_{1/2} = 3.66\) h. The coupling depends sensitively on temperature which turns the chronometer into a thermometer for the helium shell flashes of AGB stars which are the commonly accepted stellar site of s-process nucleosynthesis \([5]\). The s-process branching at \(^{176}\)Lu is schematically shown in Fig. 1.

Under s-process conditions most of \(^{176}\)Lu (\(\approx 86\%\)) is produced in the isomeric state in the \(^{175}\)Lu(n,\(\gamma\))\(^{176}\)Lu reaction \([4]\). At low temperatures the coupling between the low-\(K\) isomer and the high-\(K\) ground state is weak, and the isomer, i.e. almost all produced \(^{176}\)Lu, \(\beta\)-decays to \(^{176}\)Hf. At higher temperatures the increasing coupling depopulates the isomer and produces \(^{176}\)Lu in its long-living ground state, thus increasing the \(^{176}\)Lu abundance and the \(^{176}\)Lu(n,\(\gamma\))\(^{177}\)Lu branch. At very high temperatures the production of \(^{176}\)Lu is reduced again because of the repopulation of the isomer when approaching thermal equilibrium.

Properties of candidates for IS have been carefully studied in a series of experiments. Photon scattering \([6]\) and photoactivation of \(^{176}\)Lu \([7]\) have been measured using bremsstrahlung at the Stuttgart dynamitron, and earlier experiments with bremsstrahlung have been performed using medical and technical electron accelerators \([8, 9, 10, 11]\) which are known to provide high photon

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intensities\textsuperscript{12}. Activation after Coulomb excitation was studied at the Tandem accelerator at IPN, Orsay\textsuperscript{13}, and photoactivation experiments with various radioactive sources were reported in\textsuperscript{10,11,16,17}. High-resolution gamma-spectroscopic studies were performed using the $^{175}\text{Lu}(n,\gamma)^{176}\text{Lu}$ reaction at the GAMS spectrometer at ILLE, Grenoble\textsuperscript{18,19,20}, using HPGe detectors\textsuperscript{19,21}, and using the $^{176}\text{Yb}(p,n)^{176}\text{Lu}$ reaction at the Berkeley cyclotron\textsuperscript{22}. The $^{175}\text{Hf}(t,\alpha)^{176}\text{Lu}$ reaction was studied at Los Alamos using a Q3D magnetic spectrograph\textsuperscript{23}, and the $^{176}\text{Lu}(d,p)^{176}\text{Lu}$ reaction was analyzed using the Q3D spectrograph at the Munich tandem accelerator\textsuperscript{19}. Obviously, the s-process branching at $A = 176$ depends also on the neutron capture cross sections. These cross sections have been measured with high accuracy, see\textsuperscript{1,4} and references therein, and will not be analyzed here again.

This paper focuses on experimentally confirmed properties of $^{176}\text{Lu}$ and the well-established IS at an excitation energy of $E_x = 839\text{keV}$ with $J^\pi = 5^-; 4$. It will be shown that the coupling between the high-$K$ 7$^-$; 7 ground state and the low-$K$ 1$^-$; 0 isomer in $^{176}\text{Lu}$ via the IS at 839 keV is significantly stronger than adopted in the latest analysis\textsuperscript{11}. A further enhancement of the coupling has been suggested from K-mixing of two almost degenerate 7$^-$ states at $E_a = 725\text{keV}\textsuperscript{2}$. This paper is organized as follows: In Sect.\textsuperscript{II the formalism for the coupling of low-$K$ and high-$K$ states in a stellar photon bath will be reviewed. This paper focuses on experimental results for the IS at 839 keV in Sect.\textsuperscript{III}, here conclusions can be drawn which are firmly based on experimental data. Special attention will be given to a recent photoactivation experiment\textsuperscript{11}. Contrary to these experimentally based results, the recent analysis of K-mixing for the two 7$^-$ states at 725 keV\textsuperscript{2} has to rely on theoretical considerations. The interpretation of these experimental results for the 839 keV state and the transition from results under laboratory conditions to stellar conditions will be given in Sect.\textsuperscript{IV}. Sect.\textsuperscript{V} lists further candidates for low-lying IS. Astrophysical consequences will be discussed in Sect.\textsuperscript{VI}. Finally, conclusions are given in Sect.\textsuperscript{VII}.

\section{II. Stellar Transition Rates Between Low-$K$ and High-$K$ States}

Direct transitions between low-$K$ and high-$K$ states are highly suppressed by $K$-selection rules. Typically, one finds suppression factors $F_K = \tau(\text{exp})/\tau(\text{W.u.})$ of the order of 100 per degree $\nu$ of $K$-suppression ($\nu = |\Delta K - L|$) where $\tau(\text{W.u.})$ is the Weisskopf estimate for the lifetime $\tau$ of an ELC or MLC electromagnetic transition\textsuperscript{24}. It has been shown that thermal equilibrium is achieved within the low-$K$ states and within the high-$K$ states on a short timescale of by far less than one second\textsuperscript{2,25} which is much shorter than typical timescales in s-process nucleosynthesis\textsuperscript{5,26}, whereas transition rates between low-$K$ states and high-$K$ states are much slower and depend critically on temperature. Consequently, $^{176}\text{Lu}$ has to be considered as two different species, one with low-$K$ and one with high-$K$, in nucleosynthesis calculations for the s-process. The stellar transition rate $\lambda$ for transitions between the high-$K$ and the low-$K$ species of $^{176}\text{Lu}$ is given by

$$\lambda^*(T) = \int c n_\gamma(E,T) \sigma(E) dE,$$

and the energy-integrated cross section $\sigma$ for an IS at excitation energy $E_{IS}$

$$\sigma = \int \sigma(E) dE = \frac{2J_{IS} + 1}{2J_0 + 1} \left( \frac{\pi \hbar c}{E_{IS}} \right)^2 \times \frac{\Gamma_{IS\rightarrow low-K}}{\Gamma_{IS\rightarrow high-K}} \times \frac{\Gamma_{IS\rightarrow low-K}}{\Gamma_{IS\rightarrow high-K}}$$

may also be written as $b_{IS\rightarrow low-K} \times b_{IS\rightarrow high-K} / \Gamma$ or $b_{IS\rightarrow low-K} \times b_{IS\rightarrow high-K} / \hbar / \tau$ where $b_{IS\rightarrow low-K}$ and $b_{IS\rightarrow high-K}$ are the total decay branchings of the IS and $\tau = \hbar / \Gamma$ is its lifetime.

The total stellar transition rate in Eq. (2.1) is given by the sum over all IS; however, from the exponential dependence of the photon density in Eq. (2.2) it is obvious that only very few low-lying IS – and often only the lowest IS – dominate the stellar transition rate. It will be shown that the experimentally confirmed IS at 839 keV plays a key role which may be superseded if there is strong K-mixing between the two 7$^-$ states at 725 keV\textsuperscript{2}.

Finally, it has to be noted that the principle of detailed balance applies under stellar conditions to the transition rates from high-$K$ to low-$K$ states and its inverse from low-$K$ to high-$K$ states:

$$\frac{\lambda^*(\text{low}-K \rightarrow IS \rightarrow \text{high}-K)}{\lambda^*(\text{high}-K \rightarrow IS \rightarrow \text{low}-K)} \approx \frac{2J_{g.s.} + 1}{2J_{iso} + 1} \exp(\frac{E_{iso}}{kT})$$

with $J_{g.s.} = 7^-$ (high-$K$), $J_{iso} = 1^-$ (low-$K$), and $E_{iso} = 123\text{keV}$ for $^{176}\text{Lu}$. This ratio of reaction rates in
Eq. (2.4) is independent of the properties of the IS. Under typical stellar conditions of the s-process the transition rate from the low-K isomer at 123 keV to the high-K ground state is much larger than its inverse rate which populates the isomer because \((2J_{\text{C, is}} + 1)/(2J_{\text{iso}} + 1) = 5\) for \(^{176}\text{Lu}\) and \(\exp(E_{\text{iso}}/kT) \gg 1\) at typical s-process temperatures. E.g., at \(kT = 23\) keV – typical for the \(^{22}\text{Ne}(\alpha, n)^{25}\text{Mg}\) neutron source during helium shell flashes – a ratio of about 1000 is found. For the lower temperature of about 8 keV – typical for the \(^{13}\text{C}(\alpha, n)^{16}\text{O}\) neutron source – this ratio is even higher. However, at this low temperature the reaction rate is negligibly small, and the low-K states and the high-K states in \(^{176}\text{Lu}\) are decoupled.

III. AVAILABLE EXPERIMENTAL DATA AND INTERPRETATION

As already mentioned in the introduction, various types of experimental data are available for the odd-odd nucleus \(^{176}\text{Lu}\). In particular, because of the stable neighboring \(^{175}\text{Lu}\) it is possible to use the \((n, \gamma)\) reaction for a detailed spectroscopic study which has been performed at the ILL using the ultra-high resolution spectrometer GAMS \([18, 19, 20]\). In addition, neutron transfer in the \(^{175}\text{Lu}(d, p)^{176}\text{Lu}\) reaction has been used in combination with a high-resolution Q3D magnetic spectrograph \([19]\), and the \(^{176}\text{Yb}(p, n)^{176}\text{Lu}\) reaction has been combined with high-resolution spectroscopy of HPGe detectors \([22]\). The obtained information is compiled in the ENSDF online data base \([27]\) which is based on \([28]\). Further information on IS can be deduced from a Coulomb excitation and activation experiment \([13]\) which has detected the activity of the isomer in \(^{176}\text{Lu}\) recoil nuclei after Coulomb excitation by a \(^{32}\text{S}\) projectile, and from various photoreactivation experiments using bremsstrahlung and radioactive sources \([8, 9, 10, 11, 13, 15, 16, 17]\). Finally, a photoreactivation experiment has been performed \([7]\) using the high photon flux of the bremsstrahlung setup of the tandem accelerator at Stuttgart \([29]\). Although a final analysis of these data is unfortunately not available, the data analysis in \([7]\) is sufficient to derive the integrated cross section of the IS at 839 keV unambiguously in combination with the other available data \([13, 18, 19, 20]\).

The integrated cross section for the transition between low-K and high-K states depends on the lifetime \(\tau\) and the branchings \(b\) of the IS, see Eq. (2.3) in Sect. II and the following text. Thus, two different approaches have been followed to determine the integrated cross section \(I_\sigma\) of the IS at 839 keV. For illustration, a partial level scheme of \(^{176}\text{Lu}\) is shown in Fig. 2.

In a first approach, see Sect. III C, high- and ultra-high \(\gamma\)-spectroscopic studies have been performed to measure \(\gamma\)-ray branching ratios. This first approach clearly identifies IS from the measured branching ratios \(b\) to low-K and high-K states; however, an additional measurement of the lifetime \(\tau\) of the IS has to be performed which is difficult for the relevant lifetimes of the order of several picoseconds. Lifetime measurements are further complicated by feeding in the complex decay scheme of the heavy odd-odd nucleus \(^{176}\text{Lu}\) \([21]\). It turns out that this first approach is ideal for the identification of IS, but of limited applicability for the determination of integrated cross sections.

In a second approach, see Sect. III D, photoactivation experiments have been performed. The photoactivation yield is directly proportional to the integrated cross section \(I_\sigma^{\text{ab}}\) of the contributing IS, i.e. this measurement provides integrated cross sections \(I_\sigma\) but does not provide the ingredients for \(I_\sigma\) which are the branching \(b\) and the lifetime \(\tau\). Because of the typically broad spectral shape of the incoming photons it is difficult to assign the photoactivation yield directly to a particular IS. This argument holds for experiments with radioactive sources, bremsstrahlung, and virtual photons in Coulomb excitation.

It will be shown in Sect. III D that the combination of the high-resolution \(\gamma\)-spectroscopic studies (first approach) and photoactivation results (second approach) allows the first unique determination of the integrated cross section \(I_\sigma\) of the IS at 839 keV. The combined analysis leads to a reduction of the uncertainty of \(I_\sigma\) from
about two orders of magnitude to about a factor of two.

A. Photoactivation at the Stuttgart dynamitron

The recent photoactivation experiment performed at the Stuttgart dynamitron accelerator plays an essential role in this analysis. We briefly review the results given only in conference proceedings up to now [7]. Details of the experiment will be published elsewhere [30].

The high-current dynamitron accelerator at the IIS Stuttgart provides high bremsstrahlung intensities up to energies of 4 MeV and allows for very sensitive photoactivation studies [31, 32]. Details of the accelerator and the experimental set-up are given in [29]. Lutetium samples with natural isotopic composition and masses of about 9 mg of $^{176}$Lu (total mass about 350 mg) were irradiated with bremsstrahlung at endpoint energies from about 800 keV to 3 MeV. Small steps in the endpoint energy were used to assign the photoactivation yield to particular IS which is otherwise a serious limitation of photoactivation experiments (see above, “second approach”).

The isomer population via IS was detected by decay $\gamma$-spectroscopy of the 88 keV line from the $^{176}$Lu$^{\text{m}}$ $\rightarrow ^{176}$Hf decay at higher endpoint energies. However, a practically constant background in this $\gamma$-ray line is found from the decay of the long-living $^{176}$Lu ground state. To improve the sensitivity, $\beta$-spectroscopy has been used to detect electrons with energies above the endpoint of the $^{176}$Lu ground state decay. From both $\beta$- and $\gamma$-spectroscopy data the half-life of the isomer could be determined with $t_{1/2} = 3.63 \pm 0.02$ h ($\gamma$-spectroscopy) and $3.640 \pm 0.004$ h ($\beta$-spectroscopy); these results are in good agreement with the adopted half-life of $3.664 \pm 0.019$ h [27]. The measured events in the $\gamma$- and $\beta$-detectors have been properly corrected taking into account the decay of $^{176}$Lu$^{\text{m}}$ during the irradiation, the short waiting time between irradiation and counting, and during the counting time. The derived experimental yield is shown in Fig. 3 in arbitrary units.

The experimental photoactivation yield from a particular IS is proportional to the integrated cross section $I_\sigma$ and increases almost linearly with the endpoint energy of the bremsstrahlung. As long as the endpoint energy remains below the lowest IS, the experimental yield obviously vanishes. When the first IS is reached, the experimental yield starts to increase roughly linearly. Whenever the endpoint energy exceeds a further IS, the slope of the yield curve increases because of the additional contribution of this IS. A schematic view of these properties of the yield curve is shown in Fig. 1 of [29].

The experimental yield is shown in Fig. 3 for endpoint energies between 800 keV and 2 MeV. A clear evidence for IS is found from the kinks of the yield curve which are located at 880 keV, 1060 keV, 1330 keV, and 1660 keV. The uncertainty of the energies of the IS is about 30 keV which is composed mainly of the uncertainty of the experimental endpoint energies and of the finite energy steps in the yield curve in Fig. 3. This limited energy resolution complicates the assignment of the kinks in the yield curve to individual IS in $^{176}$Lu. The energy difference between the lowest IS and the second IS is $\Delta E = 180 \pm 15$ keV. The smaller uncertainty of about 15 keV for the energy difference can be obtained because the error in the endpoint assignment is approximately the same for all measurements between 800 keV and 1200 keV.

From the changes of the slope in the yield curve between IS (see Fig. 3) a rough estimate for the relative strength of the integrated cross sections $I_\sigma$ for the lowest IS can be derived. The cross sections behave like $1 : 0.4 : 0.7 : 14.3$ for the first four IS in $^{176}$Lu at 880 keV, 1060 keV, 1330 keV and 1660 keV.

B. High-resolution $\gamma$-spectroscopic data (“first approach”)

The dominating decay of the IS is the direct E2 transition from the $5^-$ IS at 839 keV to the $7^-$ ground state. Firmly assigned is the intraband transition from the $5^-$ IS to its $4^-$ band head of the $K = 4$ band at 723 keV. This $4^-$ state decays via a $\gamma$-ray cascade down to the $1^-$; 0 isomer at 123 keV (see Fig. 2). Thus, the properties of the $5^-$ state at 839 keV as IS are experimentally clearly confirmed. Further, there are several tentative assignments for transitions from the $5^-$ IS at 839 keV to low-$K$ states which finally end at the $1^-$ isomer at 123 keV. Depending on these tentative assignments, the $\gamma$-ray branching to the low-$K$ isomer is

$$0.021 \leq b_{IS \rightarrow \text{low}-K} \leq 0.105$$ (3.1)
Using these $\gamma$-ray branchings and theoretically estimated conversion coefficients \[33\], this leads to
\[0.065 \leq b^{+CE}_{IS \rightarrow low-K} \leq 0.149 \quad (3.2)\]
for the branching $b^{+CE}$ including conversion electrons in neutral $^{176}$Lu. The branching under stellar conditions $b^*$ will be in between the lower limit of the $\gamma$-ray branching $b^*$ and the upper limit of the branching including conversion electrons $b^{+CE}$:
\[b^*_{\text{min}} \leq b^* \leq b^{+CE} \quad (3.3)\]
\[0.021 \leq b^* \leq 0.149 \quad (3.4)\]
The precise value of $b^*$ depends on the degree of ionization at a given stellar temperature $T$. It can be estimated using the formalism in \[34\] which leads to about $n_{K}^{*} \approx 0.4$ electrons in the K-shell at $kT = 23$ keV instead of $n_K = 2$ for neutral atoms.

It is obvious that the low-energy bremsstrahlung in the photoactivation experiment \[6\] is not able to produce highly ionized $^{176}$Lu. The same argument holds for all photoactivation experiments with radioactive sources \[10, 13, 16, 17\] and for the activation experiment after Coulomb excitation where the recoiling $^{176}$Lu nucleus has relatively low energies. Thus, activation experiments in the laboratory determine $b^{+CE}$ instead of $b^*$ or $b^*$ (see also Sects. II C and IV). Note that the conversion coefficient for low-energy transitions is mainly defined by the strongest bound K-shell electrons.

It has been attempted to measure the lifetime $\tau$ of the IS at 839 keV in \[18, 19, 20\] which is mainly defined by the strongest transition from the IS to the ground state. An upper limit could be obtained from the measured time distributions using the generalized centroid-shift method \[19\], and a lower limit was obtained from the $\gamma$-ray induced Doppler broadening (GRID) technique \[18\]. The combined result is
\[10 \text{ ps} \leq \tau \leq 433 \text{ ps} \quad (3.5)\]
Note that the half-life $t_{1/2} \leq 300$ ps has been determined in the original work \[18\] which is later cited – probably in error – as lifetime $\tau \leq 300$ ps \[18, 20\]. A recent photon scattering experiment \[6\] did not see the IS at 839 keV; from the experimental limits $\tau \geq 1.5$ ps could be derived which does not further restrict the allowed range for the lifetime $\tau$ in Eq. 3.5.

The combination of the branching ratios $b^*$, $b^{+CE}$, and $b^*$ in Eqs. (3.1), (3.2), and (3.3), and the lifetime $\tau$ in Eq. (3.5) leads to the following integrated cross sections
\[12.5 \text{ meV fm}^2 \leq I_{\gamma}^{1/2} \leq 2479 \text{ meV fm}^2 \quad (3.6)\]
\[37.1 \text{ meV fm}^2 \leq I_{\gamma}^{1/2} \leq 3344 \text{ meV fm}^2 \quad (3.7)\]
with the value $I_{\gamma}^{1/2}$ calculated for neutral $^{176}$Lu using the branching $b^{+CE}$ including conversion electrons, and the value $I_{\gamma}^{1/2}$ for fully ionized $^{176}$Lu using the $\gamma$-ray branching $b^*$. Again, the stellar value $I_{\gamma}$ will be in between the laboratory result and the result for fully ionized $^{176}$Lu.

Besides the IS at 839 keV, further IS at higher energies have been detected in the $\gamma$-spectroscopic experiments \[19, 22\]. Decay branches to the low-K and to the high-K part of the $^{176}$Lu level scheme have been reported for the $5^{-}: 4$ state at 922 keV, the $6^{-}: 4$ state at 963 keV, the $5^{-}: 3$ state at 1032 keV, and the $5^{-}: 3$ state at 1069 keV. Only for the 922 keV IS an upper limit of $\tau < 290$ ps is available \[19\] which translates together with the measured branching ratios to a lower limit of the integrated cross section $I_{\gamma} \gtrsim 100$ meV fm$^2$. No lifetime information is available for the IS at 963 keV, 1032 keV, and 1069 keV; it is impossible to derive the integrated cross sections $I_{\gamma}$ for these IS from the $\gamma$-spectroscopic data. Furthermore, the IS at 963 keV and 1069 keV cannot be seen in photoactivation because of the missing ground state branch \[27\].

Summarizing the high-resolution $\gamma$-spectroscopic experiments, a clear assignment of IS is possible but the integrated cross section $I_{\gamma}$ of the IS at 839 keV remains uncertain by about two orders of magnitude, and $I_{\gamma}$ for other IS cannot be determined.

### C. Photoactivation experiments (“second approach”)

Because of the dominating ground state decay of the IS at 839 keV it is possible to measure the integrated cross section $I_{\gamma}$ by photoactivation experiments \[7, 13\] (see also Sect. IV). Other branches from the IS at 839 keV to high-K states in $^{176}$Lu contribute only by about 10%; this marginal correction to the integrated cross section $I_{\gamma}$ is neglected in the following.

Photoactivation experiments have been performed with bremsstrahlung, radioactive sources, and virtual photons in Coulomb excitation. There is general agreement between the various types of activation studies that the experimental yield rises steeply with energy. This indicates that there are IS with large integrated cross sections at higher energies. However, these IS at higher energies are not relevant under s-process conditions. Consequently, it is not possible to derive properties of low-lying IS from bremsstrahlung photoactivation experiments with endpoint energies of several MeV \[8, 9, 11\]. There is also general agreement that the lowest IS is located at energies above 650 keV because photoactivation of $^{176}$Lu could not be observed after irradiation with $^{137}$Cs sources ($E_{\gamma} = 662$ keV) \[10, 16, 17\] and in an experiment with bremsstrahlung with an endpoint energy of 600 keV \[10\]. A clear photoactivation signal has been observed using higher energy $\gamma$-ray sources $^{60}$Co ($E_{\gamma} = 1332$ keV and 1173 keV) \[10, 15, 16, 17\] and $^{24}$Na ($E_{\gamma} = 2754$ keV and 1369 keV) \[10\]. All these findings are nicely confirmed by the measured yield curve in Fig. 3.

In general, the yield in photoactivation experiments with broad incoming photon spectra determines a weighted sum of integrated cross sections $I_{\gamma}$ for all IS within the photon spectrum. The contribution of a par-
D. Combination of the two approaches

Although the photoactivation experiment using bremsstrahlung [7] does not provide integrated cross sections yet, this experiment does provide rough information on the excitation energies of the lowest IS from the kinks in the measured yield curve (see Fig. 3). The lowest IS is located at 880 ± 30 keV, and the next IS is found at 1060 ± 30 keV. The energy difference between the first and second IS is much better defined by ΔE = 180 keV with an estimated uncertainty of about 15 keV. The comparison with the known IS from the γ-spectroscopic studies [19, 22] leads to the clear conclusion that only the IS at 839 keV and 1032 keV with ΔE = 193 keV have been seen in the photoactivation experiment [6] whereas the state at 922 keV does not show up in the photoactivation yield curve. From the yield curve in Fig. 3, an upper limit for the integrated cross section of the IS at 922 keV can be estimated which is about a factor of five lower than the integrated cross section for the 839 keV state. As will be shown below, this finding is in agreement with the lower limit for I_γ ≥ 100 meV fm² for this IS from the γ-spectroscopic data.

The kinks in the yield curve of Fig. 3 at higher energies (1330 keV and 1660 keV) cannot be assigned to IS from γ-spectroscopy because the highest energies in the γ-spectroscopic studies were 1130 keV [19] and 902 keV [22]. A very tentative assignment can be suggested for the kink at 1330 keV. It may correspond (i) to the 1332 keV level seen in photon scattering and thus coupled to the ground state [6] or (ii) to a level at 1301 keV seen in the 176Yb(p,n)176Lu reaction [22] which is coupled to the isomer. A few levels around the IS at 1660 keV are reported in [27] at 1617 keV, 1655 keV, 1679 keV, 1689 keV, and 1693 keV; however, for two of these levels not even a spin/parity assignment has been adopted in [27].

The photoactivation experiment after Coulomb excitation [13] is not able to resolve individual IS. The given result in [13] of bγ+CE × (1 − bγ+CE)/τ = 12 ± 10 × 10⁹ s⁻¹ translates to an integrated cross section I_σ^lab = 3164 ± 2637 meV fm² for the strength of IS below 1 MeV excitation energy in 176Lu with an uncertainty of a factor of two. Because the γ-spectroscopic experiments [19] find only two IS below 1 MeV excitation energy, namely at 839 keV and 922 keV, the measured activation cross section must be the sum of the cross sections of both states. The next IS is reached at a significantly higher energy of 1032 keV which is more than 100 keV above the 922 keV state and 193 keV above the 839 keV state.

Because the 922 keV state does not contribute to the photoactivation yield in the bremsstrahlung experiment, this state also cannot contribute to the Coulomb activation yield. Thus, the integrated cross section I_σ^lab = 3164 meV fm² from the Coulomb excitation experiment [13] is dominated by the contribution of the IS at 839 keV; more than 80% of the strength in [13] can be assigned to the 839 keV IS. This result is at the upper end of the allowed range of I_σ^lab in Eq. 3.7 which has been determined from γ-spectroscopy experiments [18, 19]. Combining the limits of the γ-spectroscopy experiments and the integrated cross section from the Coulomb excitation and photoactivation experiments leads to the conclusion that the branching bγ+CE must be at the upper end of the allowed range in Eq. 3.2, and the lifetime τ of the IS at 839 keV must be close to the lower limit of 18 ± 19. A consistent set of parameters for the IS at 839 keV is bγ+CE ≈ 0.1 and τ ≈ 12 ps leading to an integrated cross section I_σ^lab ≈ 2000 meV fm² with an upper limit of about 3500 meV fm² from the branching and the lifetime from γ-spectroscopic studies [18, 19, 20, 22] and a lower limit of about 1200 meV fm² from the activation experiments [6, 12]. In total, an uncertainty of a factor of two for the integrated cross section I_σ^lab seems to be a careful realistic estimate.

This result is also in reasonable agreement with the photoactivation using 60Co sources with a maximum energy of E = 1332 keV. The integrated cross sections of the 839 keV, (922 keV), 1032 keV, and 1330 keV IS scale like 1 : (≤ 0.2) : 0.4 : 0.7 (see Fig. 3 and Sect. 3.1A), i.e., about one half of the measured cross section of 6–7 eV fm² must be assigned to the lowest IS at 839 keV. However, this assignment to the lowest IS has a rather large uncertainty because the IS at 1330 ± 30 keV is located very close to the 60Co energy of 1332 keV. Additionally, a tentative state in 176Lu has been seen in photon scattering [6] at 1332 keV. If there is accidental overlap between the primary 60Co energy and the excitation energy of an IS in 176Lu, then the yield in the 60Co photoactivation experiments [10, 13, 16, 17] may be strongly affected by this IS at 1330 keV.
At first view, from the allowed ranges of $I_{\text{lab}}^{\text{lab}}$ from γ-spectroscopy in Eq. 3.7 and the result $I_{\sigma}^{\text{lab}} = 31647^{+65}_{-582}$ meV fm$^2$ from the Coulomb activation experiment 13, a slightly higher result of $I_{\sigma}^{\text{lab}} \approx 3000$ meV fm$^2$ would be derived. However, the limit from γ-spectroscopy is a combined limit from two independent measurements of the branching ratio and the lifetime. A result of $I_{\sigma}^{\text{lab}} \approx 3000$ meV fm$^2$ would require that both quantities are very close to their experimental limits which is statistically not very likely. Taking into account the relatively large uncertainties of the Coulomb activation experiment, $I_{\sigma}^{\text{lab}} \approx 2000$ meV fm$^2$ seems to be a more realistic estimate for the integrated cross section of the IS at 839 keV in $^{176}$Lu. A similar result is obtained from the average of the combined lower and upper limits from γ-spectroscopy and Coulomb activation.

IV. LABORATORY AND STELLAR REACTION RATES

In general, dramatic changes may be found for photon-induced reaction rates under laboratory and under stellar conditions. Whereas under laboratory conditions the target nucleus is in its ground state (except the case of $^{180}$Ta which is found in nature in its long-living isomeric state), under stellar conditions excited states are populated in thermal equilibrium according to the Boltzmann statistics. Consequently, the transition rate under laboratory conditions depends on the direct decay width $\Gamma_{IS-\text{g.s.}}$ to the ground state, whereas under stellar conditions the rate depends on the total γ-decay width to all states which finally cascade down to the ground state; in the case of $^{176}$Lu this width is given by $\Gamma_{IS-\text{high}-K}$ to all high-$K$ states. A detailed discussion of the importance of thermally excited states under stellar conditions is given in 25, 30.

For the case of $^{176}$Lu the influence of thermally excited states is unexpectedly low. The dominating IS at 839 keV decays preferentially to the ground state, and the transition rate under stellar conditions is only moderately enhanced because $\Gamma_{IS-\text{g.s.}} \approx \Gamma_{IS-\text{high}-K}$. Compared to other uncertainties, this moderate enhancement of the order of 10 − 20 % can be neglected in this work.

However, there is another effect that has to be analyzed. The only firmly assigned transition from the 5− IS at 839 keV to low-$K$ states in $^{176}$Lu is the intraband transition to the band head of the $K = 4$ band at 723 keV with a relatively small transition energy of 116 keV. This M1 or E2 transition is enhanced by internal conversion for neutral $^{176}$Lu atoms with conversion coefficients $\alpha_{M1} = 2.42$ and $\alpha_{E2} = 1.93$ 33. This transition has been tentatively assigned to (M1) in 19. Because of the relatively small difference of the M1 and E2 conversion coefficients, only the M1 assignment is used in the following discussion.

In laboratory experiments the charge state of $^{176}$Lu depends on the experimental conditions. Under stellar conditions $^{176}$Lu is highly ionized depending mainly on the temperature $T$ and weakly on the electron density $n_e$ 34. The M1 conversion coefficient is mainly defined by the K-shell contribution. In all experiments under study in this paper 7, 13, 18, 19, 20, 21, 22, $^{176}$Lu will not be fully ionized. This is obvious for the neutron capture experiments with thermal neutrons 18, 19, 20 and the photoactivation with low-energy bremsstrahlung 7. But also for the $^{175}$Lu(d,p)$^{176}$Lu 19, $^{176}$Yb(p,n)$^{176}$Lu 22, and Coulomb excitation and activation experiments 13, the energies are not sufficient to produce fully ionized $^{176}$Lu. Thus, the derived transition strengths and integrated cross sections $I_{\text{lab}}^{\text{lab}}$, see Eq. 3.7, from the activation experiments can be compared to the γ-spectroscopic results including conversion electrons, and a combined result $I_{\sigma}^{\text{lab}} \approx 2000$ meV fm$^2$ has already been given in Sect. IV B.

At typical stellar s-process conditions (thermal en-
ergy \(kT = 23\text{ keV}\), electron density \(n_e = (3 - 5) \times 10^{26} \text{ cm}^{-3}\) the K-shell of \(^{176}\text{Lu}\) is partially ionized leading to \(n_K^\ast \approx 0.4\) instead of \(n_K = 2\) for neutral atoms. Thus, the decay width for the high-\(K\) \(\rightarrow\) low-\(K\) transition with \(E = 116\text{ keV}\) has to be reduced by the factor \((1 + \alpha_{\text{eff}}) / (1 + \alpha_{\text{lab}}) \approx 0.43\) with \(\alpha_{\text{eff}} \approx \alpha_{\text{lab}} \times n_K^\ast / 2\) leading to an integrated cross section of \(I_\sigma^\ast \approx 850\text{ meV fm}^2\) under stellar s-process conditions.

The reduction factor \((1 + \alpha_{\text{eff}}) / (1 + \alpha_{\text{lab}}) \approx 0.43\) is only valid for the low-energy transition with \(E = 116\text{ keV}\). There are further tentative assignments for transitions from the 839 keV IS to low-\(K\) states with higher transition energies of \(E = 181\text{ keV}, 203\text{ keV},\) and 243 keV. The conversion coefficients for these transitions are significantly smaller with \(\alpha_{\text{lab}} < 1\) compared to the 116 keV transition with \(\alpha_{\text{lab}} = 2.42\). The reduction factor \((1 + \alpha_{\text{eff}}) / (1 + \alpha_{\text{lab}})\) remains close to unity for these tentatively assigned transitions. As the found branching \(b^{\gamma+CE}\) to low-\(K\) states (see at the end of Sect. III B) is about 0.1, it is very likely that not only the firmly assigned transition, but also some tentatively assigned transitions contribute to the total transition strength from the IS at 839 keV to low-\(K\) states. Consequently, the reduction factor from \(I_{\text{lab}}^\ast\) for neutral \(^{176}\text{Lu}\) to the stellar \(I_\sigma^\ast\) for partially ionized \(^{176}\text{Lu}\) is between the minimum value of \(\approx 0.43\) for the 116 keV transition and the maximum value of unity.

Combining all the above information and its uncertainties, the final result for the integrated cross section from high-\(K\) to low-\(K\) states via the IS at 839 keV under stellar s-process conditions is

\[
600\text{ meV fm}^2 \leq I_\sigma^\ast \leq 2500\text{ meV fm}^2 \tag{4.1}
\]

which is entirely based on experimental results and reliably calculated internal conversion coefficients and stellar ionization. In short, this result can also be given as \(I_\sigma^\ast \approx 1200\text{ meV fm}^2\) with an uncertainty of about a factor of two. The resulting reaction rate via the 839 keV state will be compared to other candidates for IS in the next Sect. V.

V. FURTHER CANDIDATES FOR LOW-LYING INTERMEDIATE STATES

The reaction rates from the high-\(K\) ground state to the low-\(K\) isomer in \(^{176}\text{Lu}\) are shown for various IS in Fig. 3. Because of the dominance of the IS at 839 keV state, further candidates for IS are only briefly discussed.

In general, properties of astrophysically relevant IS should be an intermediate \(K\) quantum number and a low excitation energy. The transition rate \(\lambda^\ast\) depends linearly on the integrated cross section \(I_\sigma^\ast\) and exponentially on the excitation energy \(E_{\gamma}\). The discussion in this Sect. V has to rely on theoretical considerations because the relevant properties of most candidates for IS have not been determined experimentally: whereas \(\gamma\)-spectroscopic data \([19, 20]\) clearly identify candidates for IS by their decay branches to low-\(K\) and high-\(K\) states, the integrated cross section \(I_\sigma\) cannot be derived from experiment because of missing lifetime data. Because of the relatively huge uncertainties of the lifetimes from theoretical Weisskopf estimates, a further correction of the data because of weakened internal conversion under stellar conditions – typically of the order of a factor of two or less – is neglected here.

Good candidates for further IS are the band heads of the \(K = 4\) bands at 723 keV and 788 keV. Both \(4^-\) states decay preferentially to a \(3^-\) state at 658 keV which finally cascades down to the \(1^-\) isomer. Both \(4^-\) states may branch to a \(6^-\) high-\(K\) state at 564 keV although this branching has not been observed experimentally. Assuming one Weisskopf unit for this allowed \(\Delta K = 2\) E2 transition, the integrated cross sections are \(I_\sigma^\ast \approx 10\text{ meV fm}^2\) for the 788 keV state and \(I_\sigma^\ast \approx 2\text{ meV fm}^2\) for the 723 keV state. The reaction rate of both \(4^-\) states (dash-dotted lines in Fig. 3) is significantly lower at \(kT = 23\text{ keV}\) than the 839 keV rate which is shown as thick solid line with its uncertainties (thin solid lines). The strength of the \(K\)-allowed direct M3 or E4 transition from the \(4^-\) states to the \(7^-\) ground state is negligible compared to the above mentioned E2 strength.

![Image of Fig. 4: (Color online) Reaction rates \(\lambda^\ast\) under stellar conditions for the transition from the high-\(K\) ground state to the low-\(K\) isomer in \(^{176}\text{Lu}\) from Eqs. (2.1) and (2.3).](image-url)
The $5^{-}0$ low-$K$ state at 437 keV decays by $\gamma$-cascades to the $1^{-}$ isomer. The strength of the unobserved $K$-forbidden $E2$ transition to the high-$K$ ground state can be estimated from the $K$-forbiddenness $\nu = 5$ leading to $10^{-10}$ W.u. for this transition and $I^*_\gamma = 1.1 \times 10^{-7}$ meV fm$^2$. Although the 437 keV state has a much lower excitation energy than the other IS, this strength is by far not sufficient for a noticeable contribution to the transition rate $\lambda^*$ (dotted line in Fig. 4).

$K$-mixing of two almost degenerate $7^-$ states at 725 keV has been studied in detail in [2]. Based on the results in Tab. I of [2] the transition rate can be calculated; the thin dotted lines and the gray shaded area show the allowed range of $\sigma_{K}$. At the relevant energy of $kT = 23$ keV the contribution of $K$-mixing may be larger or much lower than the experimentally confirmed transition rate of the 839 keV state. It is obvious that the decay branches of these two $7^-$ states have to be studied experimentally before a clear conclusion on the relevance of $K$-mixing can be drawn. From the experimental limits of unobserved $\gamma$-transitions from the two $7^-$ states in [19] a rough estimate for an upper limit of the integrated cross section and transition rate may be estimated which is about one order of magnitude lower than the result for full $K$-mixing shown in Fig. 4 [2, 37].

Additionally, Fig. 4 shows the result of an astrophysical determination – see Sect. VI – of the transition rate of the IS at 839 keV with an integrated cross section $I^*_\gamma = 71$ meV fm$^2$ [4] (dashed line) which is at least one order of magnitude smaller than the experimental result derived in Sects. II and IV.

Higher-lying IS have a negligible contribution to the total reaction rate. From the combination of the $\gamma$-spectroscopic data [19] and the photoactivation yield in Fig. 3 the integrated cross section of the IS at 922 keV is $100$ meV fm$^2 \lesssim I^* \lesssim 500$ meV fm$^2$. Compared to the IS at 839 keV, the contribution of the IS at 922 keV is suppressed by its smaller $I_\gamma$ and the higher excitation energy. A similar consideration for the IS at 1032 keV leads to an integrated cross section of about $I_\gamma \approx 1.4$ eV fm$^2$ with an uncertainty of at least a factor of two. Together with the measured branchings of the IS at 1032 keV a lifetime of $\tau \approx 25$ ps can be derived for this state.

**VI. ASTROPHYSICAL CONSEQUENCES**

$^{176}$Lu and $^{176}$Hf are produced in the so-called main component of the $s$-process which is assigned to thermally pulsing low-mass AGB stars [3, 27, 35]. The $s$-process branching at $A = 176$ is of special interest because this branching depends very sensitively on the temperature during $s$-process nucleosynthesis whereas most other $s$-process branchings depend on the neutron density.

Two neutron sources operate in thermally pulsing AGB stars. The $^{13}$C($\alpha$,n)$^{16}$O reaction operates during the interpulse phase at relatively low temperatures of about $kT \approx 8$ keV for about $10^4$ years; it releases about 90% of the total neutron exposure. The $^{22}$Ne($\alpha$,n)$^{25}$Mg reaction operates during the convective helium shell flashes at temperatures of about $kT \approx 23$ keV which last for about six years; the temperature increases with the number of the thermal pulse and reaches a maximum of about $kT \approx 27$ keV [30].

The lower temperature of about 8 keV in the interpulse phase is not sufficient for a thermal coupling between the high-$K$ ground state and the low-$K$ isomer in $^{176}$Lu. The reaction rate $\lambda^*$ drops below about $10^{-15}/s \approx 3 \times 10^{-9}/y$ already above 12 keV (see Fig. 4) and is thus negligible at 8 keV. The high-$K$ ground state and the low-$K$ isomer have to be treated as two fully separated species in this interpulse phase. However, at the higher temperature during the helium shell flashes the thermal coupling becomes effective.

Because of the sensitivity of the thermal coupling to the temperature, careful $s$-process calculations have been performed in [31] taking into account the neutron and temperature profiles in detail. The convective region was divided into 30 meshes, and the production and decay of $^{176}$Lu was calculated in each mesh. After each time step of less than one hour, the abundances from all zones were averaged to take into account convective mixing. Such time steps are sufficiently short compared to typical reaction rates of the thermal coupling between high-$K$ and low-$K$ states in $^{176}$Lu (see Fig. 4).

It was found in [31] that the production ratio between $^{176}$Lu and $^{176}$Hf changes dramatically during the evolution of a helium shell flash (see Fig. 8 of [31]). At the onset of the flash a large ratio between $^{176}$Hf and $^{176}$Lu is found because of the dominating production of the $^{176}$Lu isomer in the $^{175}$Lu($n,\gamma$)$^{176}$Lu reaction which decays to $^{176}$Hf. As the temperature during the flash increases, $^{176}$Hf is destroyed by neutron capture, but only weakly reproduced by the decay of the $^{176}$Lu isomer because of the thermal coupling of the low-$K$ isomer to the long-living high-$K$ ground state and the bypass of $^{176}$Hf in the subsequent $^{176}$Lu($n,\gamma$)$^{177}$Lu neutron capture reaction. At the end of the flash neutron density and temperature drop down, and the initial ratio of $^{176}$Hf and $^{176}$Lu is almost restored. This means that the final abundances of $^{176}$Hf and $^{176}$Lu depend not only sensitively on temperature, but also on the thermal conditions at the end of the helium shell flashes. This makes predictions of the $^{176}$Hf and $^{176}$Lu abundances extremely difficult and invalidates the simple interpretation of $^{176}$Lu as $s$-process thermometer.

An ideal stellar $s$-process model should be able to reproduce the so-called overproduction factors $[N_e(176)/N_e(150\text{Sm})]/[N_\alpha(176)/N_\alpha(150\text{Sm})]$ (normalized to the s-only nucleus $^{150}$Sm) of $^{176}$Lu and $^{176}$Hf simultaneously. The slow decay of the long-lived $^{176}$Lu ground state in the interstellar medium prior to the formation of the solar system slightly reduces the abundance of $^{176}$Lu and leaves the more abundant $^{176}$Hf almost unchanged. Thus, overproduction factors of about $1.05 \pm 0.05$% for $^{176}$Lu and $1.00 \pm 0.05$% for $^{176}$Hf
are the acceptable ranges \[1\]. Such a solution, the so-called “best case” with overproduction factors of 1.04 for $^{176}\text{Lu}$ and 0.95 for $^{176}\text{Hf}$, has been found in \[1\] with the parameters $b^* = 0.022$ and $\tau = 80 \text{ ps}$ for the IS at 839 keV which correspond to an integrated cross section \(I^*_{\gamma} = 71 \text{ meV fm}^2\). Already a slightly increased integrated cross section (e.g. $b^* = 0.022$, $\tau = 50 \text{ ps}$, \(I^*_{\gamma} = 114 \text{ meV fm}^2\)) shifts the overproduction factors to their limits (1.08 for $^{176}\text{Lu}$ and 0.90 for $^{176}\text{Hf}$) (see Table 7 of Ref. \[1\]).

The result of the present study shows that the integrated cross section of the IS at 839 keV is roughly one order of magnitude larger than the “best case” of \[1\] (see Sect. IV and Fig. 4). Therefore, the calculations in \[1\] were repeated with the larger integrated cross section derived in this work, i.e. with a significantly stronger coupling between the low-\(K\) isomer and the high-\(K\) ground state in $^{176}\text{Lu}$.

Most of $^{176}\text{Lu}$ is produced in the low-\(K\) isomer under s-process conditions which decays to $^{176}\text{Hf}$. The stronger coupling transforms $^{176}\text{Lu}$ from the low-\(K\) isomer to the high-\(K\) ground state, thus increasing the overproduction factor of $^{176}\text{Lu}$ and reducing the overproduction factor of $^{176}\text{Hf}$. With the stellar model used in Ref. \[1\] it was not possible to find a consistent solution within the given experimental errors of the neutron capture cross sections of the lutetium and hafnium isotopes and the uncertainty of the thermal coupling. E.g., using the measured isomeric production ratio of 0.86 and an integrated cross section \(I^*_{\gamma} = 850 \text{ meV fm}^2\) from Sect. IV the overproduction factors are 1.80 for $^{176}\text{Lu}$ and 0.61 for $^{176}\text{Hf}$ which is far out of the given range of 1.05 ± 10 % for $^{176}\text{Lu}$ and 1.00 ± 5 % for $^{176}\text{Hf}$ (see above).

A further test with a variation of the neutron production rate of the $^{22}\text{Ne}(\alpha,n)^{25}\text{Mg}$ reaction within a factor of two was also not successful. (For a detailed study of the influence of the neutron production rate in the $^{22}\text{Ne}(\alpha,n)^{25}\text{Mg}$ reaction on the s-process nucleosynthesis see \[10\].)

The astrophysical ingredients of the stellar s-process model have to be known with very high precision. Because of the extreme temperature dependence of the stellar transition rate $\lambda^*$ between high-\(K\) and low-\(K\) states in $^{176}\text{Lu}$, a possible solution of the problem may originate from modifications of the temperature profile or convective mixing during the helium shell flashes. It has to be noted that an increase of the integrated cross section \(I^*_{\gamma}\) by one order of magnitude (as determined in this work compared to the “best case” of \[1\]) may be compensated by a minor decrease in temperature by only about 1.5 keV leading to the same transition rate $\lambda^*$ via the IS at 839 keV. Such minor modifications of the temperature profile will have only small influence on other s-process branchings because most branchings are mainly sensitive to the neutron density. Although the temperature, which is “read” from the s-process thermometer $^{176}\text{Lu}$, is only about 1.5 keV lower than in the temperature profile of the latest s-process study of the $A = 176$ branching \[1\], this interpretation of $^{176}\text{Lu}$ as s-process thermometer would by far be too simplistic because of the extremely sensitive interplay of nuclear and stellar thermometer in the final phase of helium shell flashes in AGB stars.

VII. CONCLUSIONS

It has been shown that the IS at 839 keV in $^{176}\text{Lu}$ leads to a much stronger coupling between low-\(K\) and high-\(K\) states in $^{176}\text{Lu}$ than assumed in a recent study \[1\]. This result is firmly based on a variety of experimental data for the IS at 839 keV. A further enhancement of the coupling may come from $K$-mixing of two almost degenerate states at 725 keV; the analysis of the $K$-mixing has to rely on theoretical arguments up to now \[2\]. Further candidates for IS at higher and lower energies do not contribute significantly to the transition rate $\lambda^*$ from high-\(K\) to low-\(K\) states in $^{176}\text{Lu}$ at s-process temperatures.

From the above results it is obvious that the s-process branching at $A = 176$ cannot be well described using the latest s-process model \[1\]. The nuclear physics ingredients of the model seem to be reliable and based on experimental data (except the $K$-mixing of the two 7– states at 725 keV). The neutron capture cross sections in this mass region have been measured carefully in the last years including the isomer branch in the $^{175}\text{Lu}(n,\gamma)^{176}\text{Lu}$ reaction \[1, 4\]. The coupling of high-\(K\) and low-\(K\) states via IS is known from the combined analysis of all available experimental data. However, there is still an unsatisfactory large range of allowed values for the integrated cross section \(I^*_{\gamma}\) under stellar conditions which should be reduced by further experiments (e.g. photoinactivation using quasi-monochromatic $\gamma$-rays or using a quasi-stellar photon spectrum \[11\]; unfortunately, the relevant energy range is not easily accessible at the Hf$\gamma$S facility \[22\]). Such experiments may also address the influence of the suggested \(K\)-mixing \[2\] on the transition rate.

After the nuclear physics input has been considerably improved for the IS at 839 keV, it was found that the astrophysical interpretation of the $^{176}\text{Lu}/^{176}\text{Hf}$ pair as an s-process thermometer is again in question due to a strong overproduction of $^{176}\text{Lu}$. The ultimate solution of this problem requires improved data for the $K$-mixing of the two almost degenerate states at 725 keV, and it will further depend on refinements of the stellar physics in the final phase of helium shell flashes in AGB stars, thus providing deeper insight into the interesting physics of helium shell flashes.

Acknowledgments

Encouraging discussions with A. Champagne and L. Lakosi are gratefully acknowledged.
