Observation of the beta decay of $^{180}Hf^m$

S. E. Kellogg and E. B. Norman*
Nuclear Physics Laboratory, University of Washington, Seattle, Washington 98195
(Received 26 December 1984)

A new γ ray with an energy of 100.7 keV has been observed in the decay of $^{180}Hf^m$. This line is interpreted as the transition between the $I^\pi=8^+$ level at 174 keV and the 9^- isomeric state in ^{180}Ta following the 113-keV endpoint β decay of $^{180}Hf^m$. The laboratory branching ratio is 2.3×10^{-4} compared to the electromagnetic decay mode of $^{180}Hf^m$. A possible second branch to a proposed 7^- level in ^{180}Ta is not seen. Even when the enhancement due to bound state β decay in a stellar environment is included, the observed branch is not sufficient to explain the abundance of $^{180}Ta^m$ in the universe.

I. INTRODUCTION

Doubly-odd ^{180}Ta stands on one of the last nuclear frontiers. Bearing the distinction of being the rarest of the stable isotopes, ^{180}Ta is found in nature in an isomeric level with $t_{1/2}>3\times10^{13}$ years,[1] just 73 ± 2 keV above its ground state.[2,3] The $I^\pi,K=(9^-,9)$ isomer is strongly K inhibited from making both γ and β transitions while the $(1^+,1)$ ground state decays by both electron capture and β^- emission to stable ^{180}Hf and ^{180}W with a total $t_{1/2}=8.1$ h. This unique position renders $^{180}Ta^m$ difficult to produce via "standard" astrophysical production mechanisms. Beer and Ward[4] suggested that $^{180}Ta^m$ may be populated through the fractional β^- decay of the $t_{1/2}=5.5$ h, $I^\pi,K=(8^-,8)$ isomeric state in ^{180}Hf which lies 214 ± 6 keV above the high-spin $^{180}Ta^m$. The direct $8^-\to9^-$ isomer to isomer allowed Gamow-Teller transition was first studied by Gallagher *et al.*[5] and is the subject of a new experimental investigation.[6] A partial level diagram is shown in Fig. 1.

Recently, proton and neutron transfer reactions have been used to identify low-lying rotational bands in ^{180}Ta. Warde *et al.*[7,8] have observed a multiplet at 181 ± 10 keV above the ground state which they decomposed into the $I^\pi,K=(4^+,1)$ member of the ground state rotor, and two band heads with $I^\pi,K=(8^+,8)$ and $(7^-,7)$. Independently, Dewberry and Naumann[9] have observed the same line which they resolved into the $(4^+,1)$ and $(8^+,8)$ states at 183 ± 4 and 168 ± 7 keV, preferring to place the $(7^-,7)$ band head at 462 keV of excitation. Simple theoretical models which attempt to construct the odd-odd nuclei by superimposing mass $(A-1)$ odd-neutron and odd-proton quasiparticle states place the $(7^-,7)$ at about 180 keV in ^{180}Ta, just below the $(8^+,8)$ level.[10] The $(8^+,8)$ and perhaps the $(7^-,7)$ states lie about 100 keV below the $(8^-,8)$ isomer in ^{180}Hf and K-allowed β decays to these levels are energetically possible. Therefore, one or two weak 100 ± 10 keV γ transitions to the $(9^-,9)$ $^{180}Ta^m$ should be observable in the decay of $^{180}Hf^m$. Because the β decay of the $(8^-,8)$ to $(7^-,7)$ state is unhindered, this branch might play a crucial role in the astrophysical production of $^{180}Ta^m$.

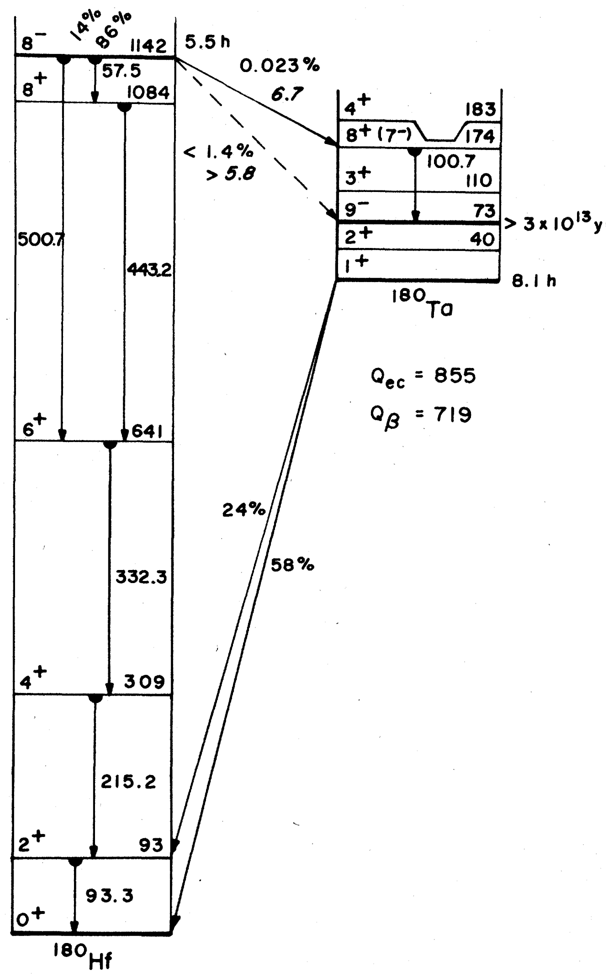

FIG. 1. Proposed decay scheme of $^{180}Hf^m$. The well-known electromagnetic decay proceeds through highly K-forbidden γ transitions to the 8^+ and 6^+ members of the ground state rotational band. Intense internal conversion electrons from the 57.5, 93.3, and 215.2-keV transitions preclude direct observation of the weak K-allowed β branches. The 100.7-keV line observed in this experiment can be placed in ^{180}Ta on the basis of its low γ multiplicity and half-life. The numbers in italics are the logft values of the deduced β transitions.

II. EXPERIMENT

The normal electromagnetic (EM) decay of the 5.5 h ^{180}Hf isomer yields on the average 3.0 γ's and 1.5 internal conversion electrons as the nucleus spins down through the ground state rotational band. In a Ge(Li) spectrum a weak γ line at 100 keV will be overwhelmed by Compton scattered events from the 215.2, 332.3, 443.2, and 500.7-keV lines. Such scattered events have the Compton γ ray and on the average two more γ's in coincidence with it, while the 100 keV γ ray is only accompanied by a low energy β ray. An anticoincidence scheme can therefore be used to separate or veto multiple from single γ events.

A 2 mg sample of HfO_2 isotopically enriched to 89% ^{179}Hf with 8% ^{180}Hf was activated with thermal neutrons at the University of Washington reactor to produce ^{180}Hf^m. The source was mounted 2.7 cm from the front face of a 45 cm^3 Ge(Li) detector. To stop conversion electrons and β's from the decay of ^{181}Hf, 0.55 cm of plastic was inserted between the source and Ge(Li). The source and Ge(Li) were then enclosed in a NaI scintillator array consisting of an annular detector 30.0 cm long $\times$21.5 cm outer diameter with an 8.9 cm inner diameter, and a 7.6 cm $\times$7.6 cm NaI to close one end of the annulus. A schematic of the apparatus is shown in Fig. 2. Ge(Li) events were acquired in singles routing events according to whether there was an accompanying NaI veto or not. The source was counted for 20 h in 1 h time bins, beginning 2.5 h after the activation. The initial counting rate in the NaI array was 17 kHz and run by run deadtime corrections were made for the accidental vetoing of true singles events out of the NOVETO Route. The VETO and NOVETO spectra accumulated during the first ^{180}Hf^m half-life are shown in Fig. 3. The NaI array proved to be quite efficient at vetoing the high-multiplicity ^{180}Hf^m events where photopeak suppression factors of close to 100 were obtained for the principle γ rays. The ^{181}Hf 133.0 keV line with only the 482.0 keV line in coincidence was vetoed by a factor of 6.6 and the lone line at 343.4 keV from the decay of ^{175}Hf appeared only in the NOVETO Route. The remaining high energy peaks were due to the summing of the coincident ^{180}Hf^m γ rays or were well-known lines from other radioactive Hf isotopes. More importantly, the Compton background

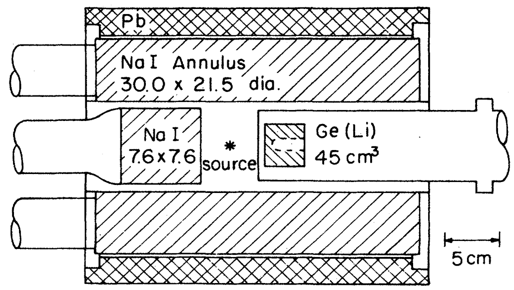

FIG. 2. Experimental setup. The ^{180}Hf^m source and Ge(Li) are enclosed by a nearly 4π NaI array to provide a gamma multiplicity filter. The Pb shielding assists in reducing cosmic ray background events in the annular NaI.

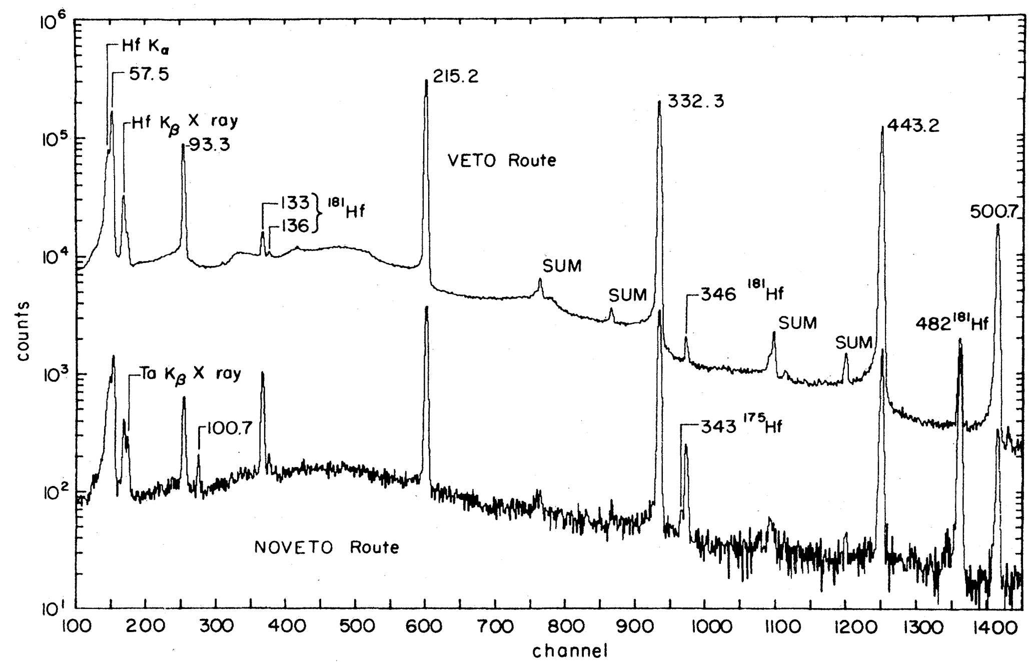

FIG. 3. Ge(Li) γ-ray spectrum of ^{180}Hf^m between 40 and 515 keV accumulated during the first 5 h of the decay. Ge(Li) events were routed according to whether there was an accompanying NaI event (VETO Route) or not (NOVETO Route). High multiplicity transitions have large VETO/NOVETO ratios. Multiplicity = 1 γ rays appear only in the NOVETO Route.

was suppressed by a factor of 80 in the vicinity of 100 keV. Here a new line at 100.70±0.05 keV appeared distinctly in the NOVETO Route with an area of 390±40 counts. A limit of 220 counts could be placed on a VETO Route component implying that the 100.7 keV line truly had no accompanying γ's. The line decayed away with a 4.7±0.6 h half-life. In Fig. 4 a fixed 5.5 h half-life decay curve has been fit run by run with a χ_ν^2 of 1.16. It is therefore concluded that the 100.7 keV γ transition follows the β decay of ^{180}Hf^{m} to an excited state in ^{180}Ta. The nearby 93.3 keV line has a total (VETO+NOVETO) area of 3.29×10^5 counts. When relative efficiencies and summing corrections (24%) are made for the 93.3 keV line, an absolute intensity of $(1.7\pm0.2)\times10^{-4}$ is obtained for the new transition.

Between the 93.3 and 133.0 keV peaks there is no evidence of a second new line in the NOVETO Route to an absolute intensity of 4.0×10^{-5}. The Hf 93.3 and 133.0 keV lines show no centroid shifts or broadening in the NOVETO Route which might hint at the existence of a nearly degenerate second line. There is a significant enhancement of the Hf $K_{\beta 2}$ x ray line at 65.0 keV in the NOVETO Route. Most of this strength is long-lived and can be attributed to the Ta $K_{\beta 1}$ x ray at 65.2 keV following the decay of ^{181}Hf. A limit of 220 counts was set on the excess 5.5 h component of this line and a limit of 70 counts was placed on the neighboring Ta $K_{\beta 2}$ x ray at 67.0 keV. This in turn limits the size of the internal conversion coefficient for the new 100.7 keV line.

III. DISCUSSION

Astrophysically important branching ratios for the production of ^{180}Ta^{m} rely heavily on nuclear structure details and warrant a review of the Nilsson orbitals in a deformed nucleus. In the $A=180$ mass region, the $\frac{9}{2}^+[514]$ and $\frac{7}{2}^-[514]$ orbitals lie near the Fermi surfaces of the proton and neutron quasiparticle seas, respectively. Allowed β^- or electron capture transitions between these two orbitals are designated "unhindered" because of their large spatial overlap. In odd rare-earth nuclei they are seen to occur with logft's of 4.5 to 5.0. Other allowed Gamow-Teller transitions between initial and final states lacking matching Nilsson quantum numbers are designated "hindered" and occur with logft values between 6.0 and 7.5.[11]

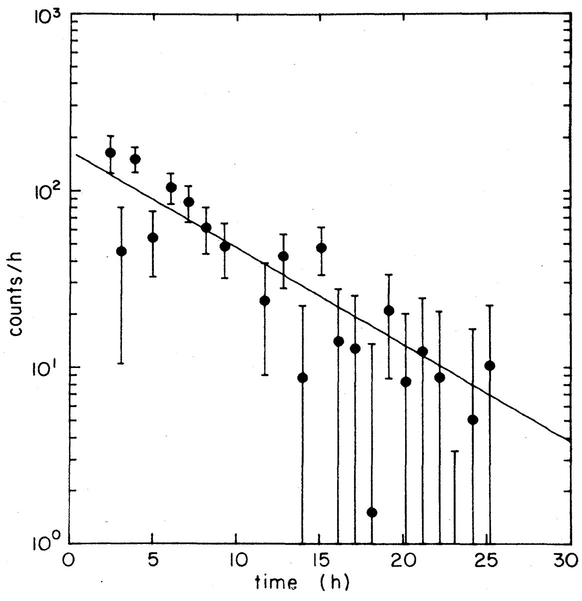

FIG. 4. Decay of the NOVETO Route 100.7 keV line. The curve is fit to the data with a fixed 5.5 h half-life.

The 9^- isomer in odd-odd ^{180}Ta is formed by a coupling of the $\frac{9}{2}^-[514]_\pi$ with the $\frac{9}{2}^+[624]_\nu$. The 8^+ level is the spin antialigned coupling of the $\frac{7}{2}^+[404]_\pi$ with the $\frac{9}{2}^+[624]_\nu$ and forms the Gallagher-Moszkowski doublet with the 1^+ ground state. The 7^- level is formed by coupling the $\frac{7}{2}^-[404]_\pi$ with the $\frac{7}{2}^+[514]_\nu$.[12]

The 8^- isomer in even-even ^{180}Hf can be formed by either breaking a proton pair and coupling the proton quasiparticles $\frac{7}{2}^+[404]_\pi$ and $\frac{9}{2}^-[514]_\pi$, or by breaking a neutron pair and coupling $\frac{9}{2}^+[624]_\nu$ and $\frac{7}{2}^-[514]_\nu$. There are two observed 8^- levels in ^{178}Hf, at 1147.4 keV (a 4.0 sec isomer) and at 1479.0 keV, which have been shown to be substantial mixtures of just these two states.[13–15] By the time two more neutron orbitals have been filled in ^{182}Hf, the low-lying 8^- level (a 61.5 min isomer) is in nearly a pure broken proton pair configuration.[16,17] On the basis of its large magnetic moment $\mu=8.6\pm1.0\mu_N$,[18,19] the isomer in ^{180}Hf might be expected to be a pure broken proton pair state like ^{182}Hf^{m}. However, the neutron transfer reaction ^{179}Hf(d,p) successfully produces the isomer suggesting that there must be a significant admixture of the broken neutron pair state.[20] The higher-lying, predominantly two-neutron 8^- level has not been identified.

The hindered transitions of importance to the present study are the β decays of the ^{180}Hf $8^-_{(2\pi)}$ to the 9^- isomer and 8^+ excited state in ^{180}Ta. These involve the $\frac{9}{2}^+[624]_\nu\rightarrow\frac{7}{2}^+[404]_\pi$ and $\frac{9}{2}^+[624]_\nu\rightarrow\frac{9}{2}^-[514]_\pi$ transitions, respectively. The first case is seen with logft values of 6.5 and 6.7 in the β^- decay of ^{177}Yb and in the electron capture of ^{181}W. A logft of 6.4 is observed in the analogous β^- decay of ^{182}Hf^{m}. The latter case is actually a first-forbidden, nonunique transition seen with consistent logft values of 6.7±0.2 in the decays of the nearby odd nuclei ^{177}Yb, ^{181}W, and ^{183}Os.

The β decay of the $8^-_{(2\nu)}$ to the 9^- isomer and the decay of the $8^-_{(2\pi)}$ to the 7^- excited state in ^{180}Ta involve the $\frac{7}{2}^-[514]_\nu\rightarrow\frac{9}{2}^-[514]_\pi$ unhindered quasiparticle transition. logft values of 4.7 and 4.4 are seen in the decays of the odd mass nuclei ^{175}Yb and ^{181}Os. Neighboring ^{182}Hf^{m} is observed to decay to the same 7^- level in ^{182}Ta with a logft of 4.8. Original interest in the isomer to isomer β decay centered on the possibility of state mixing in the structure of ^{180}Hf^{m}.[5] The β-decay strength is sensitive to a small admixture of the broken-neutron state into the broken-proton state. A limit on the direct isomer to isomer β-decay branch of 1.4% corresponding to a log$ft>5.82$ argues that the mixing is less than 10%.[6]

Table I displays the logft values and x ray relative intensities derived by assuming the new 100.7-keV transition is $E1$ or $E2$. A straightforward determination of the multipolarity on the basis of the factor of 3 difference in

TABLE I. β branches and $\log ft$ values derived for the new 100.7 keV γ transition. These are compared with $\log ft$ values systematically derived from the decays of nearby nuclei. Present limits on the 214-keV K-allowed direct isomer to isomer β decay are included for reference.

β endpoint	113 keV		214 keV
Absolute γ intensity	$1.7(2)\times 10^{-4}$		
Measured α_K (Ta)	<5.3		
β Transition	$8^- \to 8^+$	$8^- \to 7^-$	$8^- \to 9^-$
γ Multipolarity	E1	E2	
α_T[a]	0.369	3.77	
α_K[a]	0.301	0.862	
f_β	2.3×10^{-4}	8.1×10^{-4}	$<1.4\times 10^{-2}$[b]
$\log ft$ (experiment)	6.74	6.20	>5.82
$\log ft$ (systematic)	6.7	4.6	6.4[c]

[a]Internal conversion coefficients were taken from Ref. 25.
[b]Reference 6.
[c]Assumes a pure broken-proton pair configuration.

K conversion rates was not possible. The presence of the 57.5-keV line masks the existence of weak Ta K_α x rays. Poor statistics in the Ta K_β x rays allow for only an upper limit on the 5.5 h component. However the factor of 100 difference in expected β-decay strengths between the two branches makes a conclusive assignment possible. The $8^-_{(2\pi)} \to 7^-$ case is far too weak to be an allowed-unhindered transition, while the $8^-_{(2\pi)} \to 8^+$ $\log ft$ value is perfectly consistent with other $\frac{9}{2}^+[624]_\nu \to \frac{9}{2}^-[514]_\pi$ transitions. It is therefore concluded that the observed 100.7 keV γ line is the $E1$ transition from the 8^+ level. If an upper limit of $\log ft = 5.0$ is placed on the β-decay strength to the 7^- state, then the 7^- level can be excluded below 262 keV of excitation in ^{180}Ta.

IV. ASTROPHYSICAL IMPLICATIONS

Beer and Ward have shown that $^{180}\mathrm{Hf}^m$ is produced during the slow (s) neutron capture process.[4] The observed abundance of $^{180}\mathrm{Ta}^m$ is reproduced for a total β branch from $^{180}\mathrm{Hf}^m$ of $f^*_\beta = 6.5\%$, where the ($*$) denotes s-process stellar conditions. The s process is thought to occur at temperatures hot enough ($kT = 15-35$ keV) that even $Z = 72$ atoms are practically fully ionized. Beer and Macklin have pointed out that two effects due to the high degree of ionization in an s-process environment enhance f_β, the laboratory β branches of $^{180}\mathrm{Hf}^m$.[24] First, the empty L shell blocks the internal conversion of the 57.5-keV transition. This effect alone increases the half-life of $^{180}\mathrm{Hf}^m$ against EM decay by a factor of 1.4. Second, the nearly empty K shell opens the door to bound-state β decay which significantly supplements β-decay strength into the continuum:

$$\lambda^*_\beta = \lambda_{\beta C}\left[1 + \frac{\lambda_{\beta B}}{\lambda_{\beta C}}\right]. \quad (1)$$

Beer and Macklin used the early formalism of Bahcall[22] to obtain $\lambda_{\beta B}/\lambda_{\beta C} = 0.5$ for the 214-keV isomer to isomer decay. But recently, Takahashi[23] has applied a procedure developed by Takahashi and Yokoi[24] which takes into account depression of the effective electron continuum in hot, dense stellar matter to show that $\lambda_{\beta B}/\lambda_{\beta C}$ for the 214-keV decay ranges between 0.5 and 3.0 for plausible s-process conditions. A similar calculation for the 113-keV transition results in a factor of between 1.5 and 11.0.

Combining these two effects, nominal ($kT = 25$ keV, $\rho = 1000$ g/cm^3) s-process enhancement factors of 14.4 and 4.9 are obtained for the 113- and 214-keV β decays, respectively. Therefore the laboratory measured $f_\beta(8^- \to 8^+) = 2.3\times 10^{-4}$ corresponds to $f^*_\beta(8^- \to 8^+) \approx 0.33\%$, which might account for 5% of the observed abundance of $^{180}\mathrm{Ta}^m$. The laboratory limit of $f_\beta(8^- \to 9^-) \le 1.4\%$ for the direct isomer to isomer transition corresponds to an upper limit on the production of $^{180}\mathrm{Ta}^m$ of 105%. As discussed in Sec. III, this limit requires a 10% mixing of the $8^-_{(2\pi)}$ and $8^-_{(2\nu)}$ states. A pure 2π state would result in $f_\beta(8^- \to 9^-) = 0.37\%$ and thus a contribution, which, together with the branch through the 8^+ excited state, constitutes less than one-third of the observed $^{180}\mathrm{Ta}^m$ abundance. A more careful measurement of the direct isomer to isomer β decay is clearly necessary. The identification of the second 8^- level in ^{180}Hf between 1.5 and 2.0 MeV of excitation through neutron transfer reactions would be another helpful step in determining the extent of state mixing in $^{180}\mathrm{Hf}^m$.

ACKNOWLEDGMENTS

The authors gratefully acknowledge the assistance of Pat Miller and Deloss Fry for the reactor activations. They wish to thank R. Dewberry and R. Naumann for providing an early version of their paper, and K. Takahashi for performing the bound-state β-decay calculations. They would also like to thank R. Ward and P. Sood for helpful discussions, and W. Haxton for carefully reading the manuscript. This work was supported in part by the U.S. Department of Energy, Division of Nuclear Physics.

*Present address: Nuclear Science Division, Lawrence Berkeley Laboratory, Berkeley, CA 94720.

[1]E. B. Norman, Phys. Rev. C 24, 2334 (1981).
[2]T. Cousins, T. J. Kennett, and W. V. Prestwich, Phys. Rev. C 24, 911 (1981).
[3]A. H. Wapstra and G. Audi, Nucl. Phys. A 432, 1 (1985).
[4]H. Beer and R. A. Ward, Nature 291, 308 (1981).
[5]C. J. Gallagher, M. Jorgenson, and O. Skilbreid, Nucl. Phys. 33, 285 (1962).
[6]S. E. Kellogg and E. B. Norman, Bull. Am. Phys. Soc. 27, 705 (1982); and in Proceedings of the Second Workshop on Nuclear Astrophysics, Ringberg Castle, Tegernsee, West Germany, Max Planck Institut fur Astrophysik Report No. 90, 1983, p. 92.
[7]E. Warde, Ph.D. thesis, Universite Louis Pasteur Strasbourg, 1979 (unpublished).
[8]E. Warde, G. J. Costa, D. Magnac, R. Seltz, C. Gerardin, M.

Buenerd, Ph. Martin, W. Saathoff, and C. A. Wiedner, Phys. Rev. C **27**, 98 (1983).
[9]R. A. Dewberry and R. A. Naumann, Phys. Rev. C **28**, 2259 (1983).
[10]P. C. Sood and R. N. Singh, in *Proceedings of the International Conference on Nuclear Physics, Florence, Italy, 1983* (Tipografia Compositori, Bologna, Italy), Vol. 1, p. 284.
[11]C. J. Gallagher, Nucl. Phys. **16**, 215 (1960).
[12]E. Warde, R. Seltz, G. Costa, D. Magnac, and C. Gerardin, J. Phys. (Paris) **1**, L-1 (1978).
[13]C. J. Gallagher and H. L. Nielsen, Phys. Rev. **126**, 1520 (1962).
[14]H. Massmann, J. O. Rasmussen, T. E. Ward, P. E. Haustein, and F. M. Bernthal, Phys. Rev. C **9**, 2312 (1974).
[15]T. E. Ward and Y. Y. Chu, Phys. Rev. C **12**, 1632 (1975).
[16]T. E. Ward, P. E. Haustein, J. B. Cumming, and Y. Y. Chu, Phys. Rev. C **10**, 1983 (1974).
[17]T. E. Ward and P. E. Haustein, Phys. Rev. C **4**, 244 (1971).
[18]H. J. Korner, F. E. Wagner, and B. D. Dunlap, Phys. Rev. Lett. **27**, 1593 (1971).
[19]K. S. Krane, S. S. Rosenblum, and W. A. Steyert, Phys. Rev. C **14**, 656 (1976).
[20]J. I. Zaitz and R. K. Sheline, Phys. Rev. C **6**, 506 (1972).
[21]H. Beer and R. L. Macklin, Phys. Rev. C **26**, 1404 (1982).
[22]J. N. Bahcall, Phys. Rev. **124**, 495 (1961).
[23]K. Takahashi (private communication).
[24]K. Takahashi and K. Yokoi, Nucl. Phys. **A404**, 578 (1983).
[25]F. Rosel, H. M. Fries, K. Alder, and H. C. Pauli, At. Data Nucl. Data Tables **21**, No. 4-5 (1978).