

^{146}Gd and ^{144}Sm excited by the (p,t) reaction on radioactive targets

E. R. Flynn, J. van der Plicht,* and J. B. Wilhelmy

Los Alamos National Laboratory, Los Alamos, New Mexico 87545

L. G. Mann, G. L. Struble, and R. G. Lanier

Lawrence Livermore National Laboratory, Livermore, California 94550

(Received 19 July 1982)

The (p,t) reaction has been used to study the closed-shell nuclei ^{146}Gd and ^{144}Sm , the former exhibiting some characteristics of a doubly closed shell. Exotic radioactive targets of ^{148}Gd ($t_{1/2}=75$ yr) and ^{146}Sm ($t_{1/2}=7\times 10^7$ yr) obtained from chemical and isotope separation of irradiated beam-stop material were employed. The ground-state mass excess of ^{146}Gd was measured as being $\Delta u = -76.083(15)$ MeV and the first excited state is confirmed as being a 3^- state at 1.580-MeV excitation energy. Thirteen states were observed in ^{146}Gd and 23 in ^{144}Sm . The pairing-monopole and pairing-quadrupole states in both nuclei are observed for the first time. These levels are higher than systematics would predict, confirming a proton-subshell closure in ^{146}Gd , and are split in ^{144}Sm , indicating a strong interaction between nuclear modes.

NUCLEAR REACTIONS $^{148}\text{Gd}(p,t)^{146}\text{Gd}$ and $^{146}\text{Sm}(p,t)^{144}\text{Sm}$ with isotopically enriched targets. $E_p=25$ MeV; measured E_t and $\sigma(\theta)$ with Q3D spectrometer; DWBA analysis; deduced levels J^π .

NUCLEAR STRUCTURE Identified pairing monopole and quadrupole vibrations in ^{146}Gd and ^{144}Sm . Observed deviations from systematics of $N=82$ nuclei and mixing of both the monopole and quadrupole pairing vibrations in ^{144}Sm .

I. INTRODUCTION

There has been an extensive investigation of the nuclear region surrounding ^{146}Gd in recent years because of the apparent doubly closed-shell behavior of this nucleus.¹⁻³ In addition, there has been an attempt to understand this region theoretically.⁴ Unfortunately, it has been impossible to study ^{146}Gd by direct nuclear reactions except by complex, heavy-ion, transfer reactions^{5,6} or by poor-resolution experiments involving the detection of neutrons.⁷ The availability of radioactive targets prepared from materials irradiated at the Los Alamos Meson Physics Facility (LAMPF) circumvents these difficulties.⁸ In particular, the isotope ^{148}Gd ($t_{1/2}=75$ yr) (Ref. 9) permits a high-resolution study of ^{146}Gd via the $^{148}\text{Gd}(p,t)$ reaction. Since the mass of ^{148}Gd is known,¹⁰ the ground-state (g.s.) mass of ^{146}Gd may also be better determined. A variety of other long-lived isotopes also exist in this nuclear region. In particular, ^{146}Sm ($t_{1/2}=7\times 10^7$ yr) also may be used to study the neutron-shell closure at $N=82$ by the (p,t) reaction.

The (p,t) reaction is particularly interesting since pairing vibrations associated with neutron closed shells are strongly excited. The systematic behavior of these nuclear excitations¹¹ is indicative of the onset of certain nuclear characteristics, such as a proton-shell closure. Moreover, the position of pairing-vibration (PV) states is strongly influenced by the location of single-particle neutron states, and thus the energies of the PV states provide additional

constraints on calculations of single-particle levels in nuclei adjacent to shell closure.

In addition to the PV states, the (p,t) reaction also excites most of the low-lying normal-parity states. Thus, we expect to confirm and add many new level assignments in ^{146}Gd and ^{144}Sm . Furthermore, the interpretation of the first excited state in ^{146}Gd as the 3^- octupole state should also be confirmed.

II. EXPERIMENTAL PROCEDURE AND ANALYSIS

The ^{148}Gd and ^{146}Sm target material was produced by spallation reactions with a Ta target at the LAMPF accelerator.⁸ The material was dissolved after an irradiation of $\sim 5.6\times 10^4$ $\mu\text{A h}$ and the rare earths were separated by standard cation-exchange chromatography. Isotopic separation was then performed at the Lawrence Livermore National Laboratory (LLNL) and the target material was deposited on a $40\text{-}\mu\text{g}/\text{cm}^2$ C foil with a spot size of $\sim 2\times 6$ mm². The target material was not uniform; however, the effective thickness of the ^{148}Gd target during irradiation was ~ 5 $\mu\text{g}/\text{cm}^2$ and the thickness of the ^{146}Sm was 18 $\mu\text{g}/\text{cm}^2$.

The proton beam was obtained from the Los Alamos National Laboratory's (LANL) three-stage Van de Graaff facility with an energy of 25 MeV and an intensity of 0.5 μA . The reaction tritons were analyzed in a Q3D spectrometer using a helical detector in the focal plane.¹² The

^{146}Sm target thickness was determined by measuring the elastically scattered 25-MeV protons with the Q3D spectrometer. The ^{148}Gd target thickness was not measured, but it can be estimated by assuming that the lowest 3^- states have equal cross sections for both nuclei. These collective octupole states should satisfy this condition approximately.¹³ The cross sections for the ^{144}Sm levels have 15% errors but, because of the aforementioned assumption, we feel that 25% errors are reasonable for the ^{146}Gd levels. The energy resolution was 8 keV.

Several different values of the mass excess (Δu) for ^{146}Gd appear in the literature based on the ($^{12}\text{C},^{10}\text{Be}$) and ($^3\text{He},n$) reactions^{6,7} and on a theoretical estimate based on a shell-model analysis.¹⁴ These values are, respectively, $\Delta u = -76.081 \pm 0.30$ MeV ($^3\text{He},n$), $\Delta u = -76.096 \pm 0.025$ MeV ($^{12}\text{C},^{10}\text{Be}$), and from theory $\Delta u = -75.591$ MeV. The present (p,t) experiment yields a value of $\Delta u = -76.083 \pm 0.015$ MeV using the measured (p,t) Q value of -7.844 MeV and the ^{148}Gd mass of $\Delta u = -76.268 \pm 0.006$ MeV.¹⁰ This measurement is based on a calibration using the $^{96}\text{Mo}(p,t)^{94}\text{Mo}$ reaction, which has a very similar Q value. Several calibration readings were used and found to be internally consistent to 5 keV. Our value for Δu agrees within errors with the results of

Refs. 6 and 7, and not with the theoretical prediction given in the table of Ref. 14.

Spectra of the $^{146}\text{Sm}(p,t)^{144}\text{Sm}$ and the $^{148}\text{Gd}(p,t)^{146}\text{Gd}$ reactions are shown in Figs. 1 and 2, respectively. The energies, yields, and L values for the reactions measured with both targets are given in Table I. With the thicker ^{146}Sm target, we observed more weakly populated states than with the ^{148}Gd target. However, it is clear that the (p,t) reaction in both cases populates most of the low-lying states known to exist in ^{144}Sm and ^{146}Gd (cf. Table I). The g.s. yields were measured only at two angles, but angular distributions from 10° to 50° were obtained for excited states using the 15% energy range of the Q3D detector system, or up to ~ 3.7 MeV of excitation. A detailed search for levels between the g.s. and the 3^- state of ^{146}Gd revealed none. The L -transfer assignments given in Table I are based on comparisons with DWBA calculations, systematics, and/or previously determined values of the spins and parities.

Figures 3 and 4 show the angular distributions obtained in this experiment for ^{144}Sm and ^{146}Gd , respectively. The solid lines correspond to the DWBA calculations using standard¹⁷ optical-model parameters for the p and t channels. These parameters fit the first excited 2^+ states well.

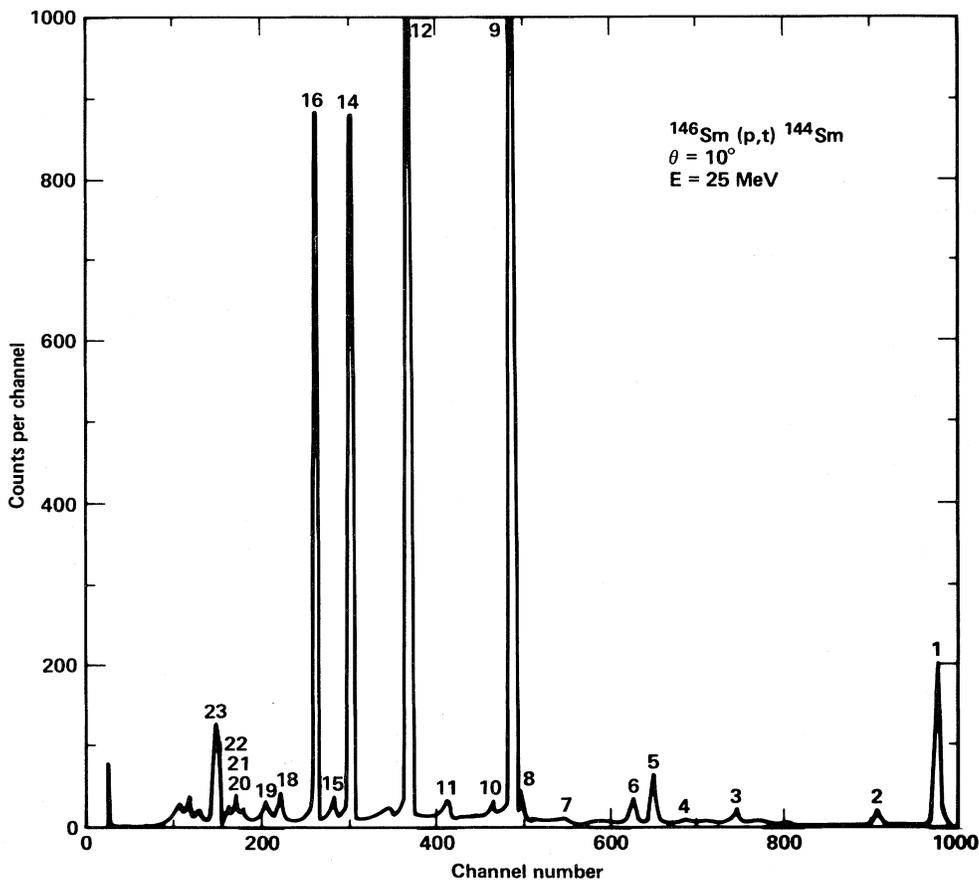


FIG. 1. Triton spectrum from the $^{146}\text{Sm}(p,t)^{144}\text{Sm}$ reaction at 10° .

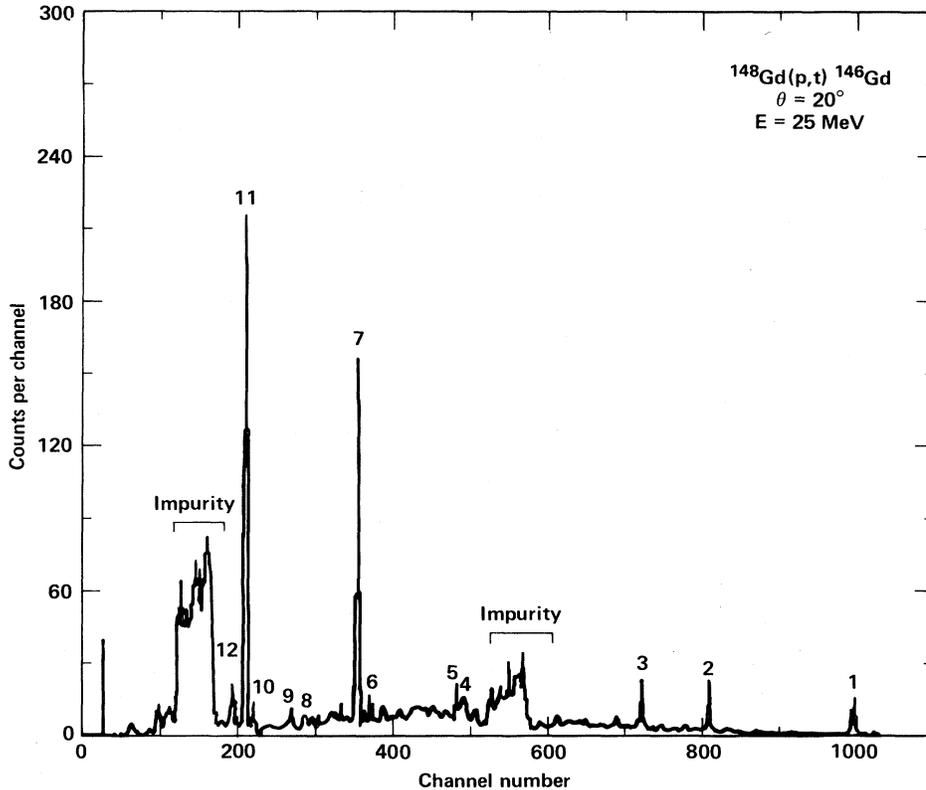


FIG. 2. Triton spectrum from the $^{148}\text{Gd}(p,t)^{146}\text{Gd}$ reaction at 20° .

They also fit the strong $L=0$ and 2 transitions near 3-MeV excitation energy. The lowest 3^- states show a distinctive deviation from these calculations at forward angles with both 3^- states having identical shapes. These states are rather weak, (about $14 \mu\text{b}/\text{sr}$ at maximum) in the (p,t) reaction, but should be strongly collective in inelastic scattering. Thus, the discrepancy may be due to coupled-channel effects through the inelastic-scattering channel. There are other indications that coupled-channel effects may be important. The known 4^+ and 6^+ states of ^{144}Sm have apparent $L=2$ angular distributions instead of $L=4$ and 6, respectively. Again, a multistep process is possible for these weakly populated excited states. An anomalous shape for the angular distribution of the lowest excited 0^+ state in ^{144}Sm is harder to understand, since the corresponding state in ^{146}Gd has a definitive $L=0$ shape. However, the ^{146}Gd state has six times the cross section.

III. THE PAIRING-VIBRATIONAL STATES

The PV monopole and quadrupole states are clearly observed in both nuclei. They are at a considerably higher

energy than the systematic trend of these states for lower Z , $N=82$ nuclei suggests (~ 2.3 MeV in ^{146}Gd). The transition strengths to the monopole PV states, relative to the g.s., are similar to those observed by (p,t) reactions in the Ce-Nd region,^{17,18} i.e., $\sim 50\%$.

In ^{146}Gd , a 0^+ state near the expected energy of the PV state is seen^{15,20} at 2.165 MeV, but it contains only 13% of the intensity of the 0^+ state at 3.016 MeV. The quadrupole PV strength seems primarily to reside in the state at 3.378 MeV, although it is possible that additional strength is in levels above 3.5 MeV. The 2^+ state at 3.378 MeV contains about 55% of the transition strength of the 0^+ state at 3.016 MeV, a ratio quite similar to that found in Ce.¹⁸ The monopole PV state at 3.016 MeV has 58% of the g.s. transition strength exactly as in ^{142}Nd .¹⁹ The systematics of these states and the low-lying states are shown in Fig. 5. The striking feature of these systematics in the case of ^{146}Gd is the rise of E_x above the trend of the lighter nuclei.¹¹

^{144}Sm is interesting because both the monopole and quadrupole PV states are fractionated into two large components. Such a splitting is not observed in any of the other $N=82$ nuclei and indicates a substantial interaction between two nuclear modes. To assure that levels from

TABLE I. Properties of energy levels in ^{144}Sm and ^{146}Gd .

Level No.	^{144}Sm					Level no.	^{146}Gd				
	Known ^a E_x (MeV)	J^π	E_x^b (MeV)	Present L	$d\sigma/d\Omega$ ($\mu\text{b}/\text{sr}$) at 10°		Known ^c E_x (MeV)	J^π	E_x (MeV)	Present L	$d\sigma/d\Omega^g$ at 25°
0	0	0 ⁺	0	0	460.0	0	0	0 ⁺	0	0	350.0
1	1.6600	2 ⁺	1.660	2	14.0	1	1.5795	3 ⁻	1.580	3 ^c	14.0
2	1.8101	3 ⁻	1.810	3 ^c	14.0	2	1.972 ^f	2 ⁺	1.971	2	10.0
	2.167					3	2.165 ^f	0 ⁺	2.162	0	26.0
3	2.1906	4 ⁺	2.190	(2) ^d	2.5	4	2.612	≤ 4	2.615		7.5
4	2.3232	6 ⁺	2.324	(2) ^d	0.8	5	2.658	5 ⁻	2.658		7.0
5	2.424	2 ⁺	2.422	2	7.0	6	2.982	7 ⁻	2.985		5.0
6	2.479	0 ⁺	2.480	(4) ^d	4.5		2.996	4 ⁻			
	2.588	4 ⁺				7			3.016	0	200.0
	2.645						3.099	6 ⁻			
	2.661					8	3.183	8 ⁻	3.181		8.0
	2.687					9			3.231		6.0
	2.704						3.294	8 ⁻			
7			2.729	(6)	1.4		3.313				
8	2.800	2 ⁺	2.804	2	4.3	10			3.354		6.0
9	2.824		2.827	0	190.0	11			3.378	2	110.0
10	2.884	4 ⁺	2.887	(4)	3.8	12	3.423	3 ⁻	3.417		14.0
11	3.021		3.020	2	3.5		3.428	9 ⁻			
	3.080					13			3.442		10.0
	3.1238										
	3.136	7									
12			3.142	0	78.0						
13	3.198	(4 ⁻)	3.205		wk.						
	3.228	1 ⁻									
	3.266										
14	3.310	(3,4 ⁻)	3.318	2	130						
	3.362	(4,3 ⁻)									
15	3.3761	8	3.375	3	11.0						
	3.393	(3,2 ⁻)									
	3.405	(3 ⁻)									
16			3.426	2	130.0						
	3.4605	9									
17			3.481		wk.						
	3.5183										
18	3.530	(3 ⁻)	3.542	(3,4)	10.0						
	3.564										
19			3.579	(4,3)	4.3						
	3.629										
	3.6051										
20			3.661	(4,3)	3.3						
	3.671	(5 ⁻)									
21			3.683	5	12.0						
22			3.708	(3)	3.5						
	3.724										
23	3.734	(3 ⁻)	3.741	(3)	30.0						

^aFrom Ref. 15.^bThe error on these values is ± 5 keV.^cThe assignment is based on $L=3$ systematic shapes. See Ref. 13.^dThese L transfers are clearly due to two step mechanisms (see text).^eFrom Refs. 1–3, 16, and 19.^fThere is substantial disagreement in the literature on these energies.^gThe thickness of the ^{146}Gd target was not measured directly. If we assume that the cross sections for populating the first 3^- state in ^{144}Sm and ^{146}Gd are equal, then this column gives cross sections in $\mu\text{b}/\text{sr}$.

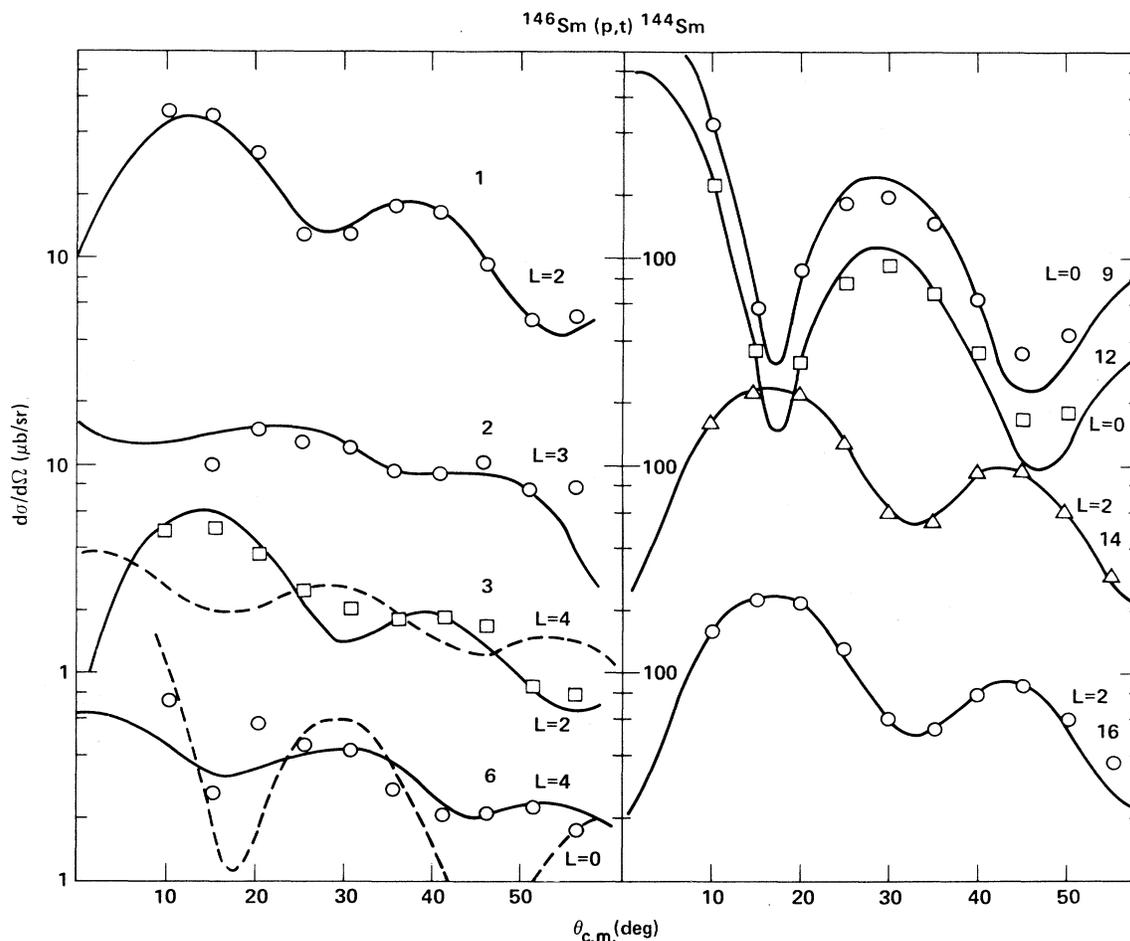


FIG. 3. Triton angular distributions and distorted-wave calculations for selected levels in the $^{146}\text{Sm}(p,t)^{144}\text{Sm}$ reaction.

impurities could not account for the second states, an examination of all the Q values for relevant reactions was made. No possibilities exist which can explain either splitting. The monopole strength in ^{144}Sm is divided between two states at 2.827 and 3.318 MeV with a strength ratio of 2.4 to 1. Their centroid is thus at 2.919 MeV, only 97 keV lower than in ^{146}Gd . The quadrupole PV strength is split equally between two levels at 3.318 and 3.426 MeV with a centroid at 3.372 MeV or 0.453 MeV above the monopole centroid. The ^{146}Gd separation of monopole and quadrupole PV states is 0.362 MeV. The ratio of PV monopole to g.s. transition strength is 58%, exactly as in the Gd case. The total quadrupole PV strength ratio to monopole PV is 97%, or almost double the ^{146}Gd result. This indicates there is missing 2^+ strength in Gd above the energy region examined here. This result for ^{144}Sm would correspond to the ^{140}Ce result where all high-energy, $L=2$ transitions were summed and gave a ratio of 94%. This indicates that in ^{144}Sm the quadrupole PV strength is concentrated in just two levels.

IV. DISCUSSION AND SUMMARY

There are several unexpected results in these data. The primary ones are the increased excitation energy of the neutron PV monopole state in ^{146}Gd over previous estimates based on systematics¹¹ and the splitting of both the monopole and quadrupole PV states in ^{144}Sm . The increased excitation energy of the monopole PV in ^{146}Gd implies simpler shell structure than in surrounding $N=82$ nuclei. In all cases where one shell is not closed, e.g., in $N=82$ nuclei,^{19,20} below Sm, and in $N=50$ nuclei,²¹ the PV state is lowered in energy compared to the harmonic prediction,²² and only part of the harmonic strength is seen. Thus, the fact that the PV state is closer to the harmonic energy (which in this case would be ~ 3.9 MeV based on binding-energy systematics) than in the lighter isotones indicates smaller interactions with proton degrees of freedom and, therefore, a better-closed proton shell. The intensity of this state relative to the g.s. does not support this argument since this ratio is in agreement with

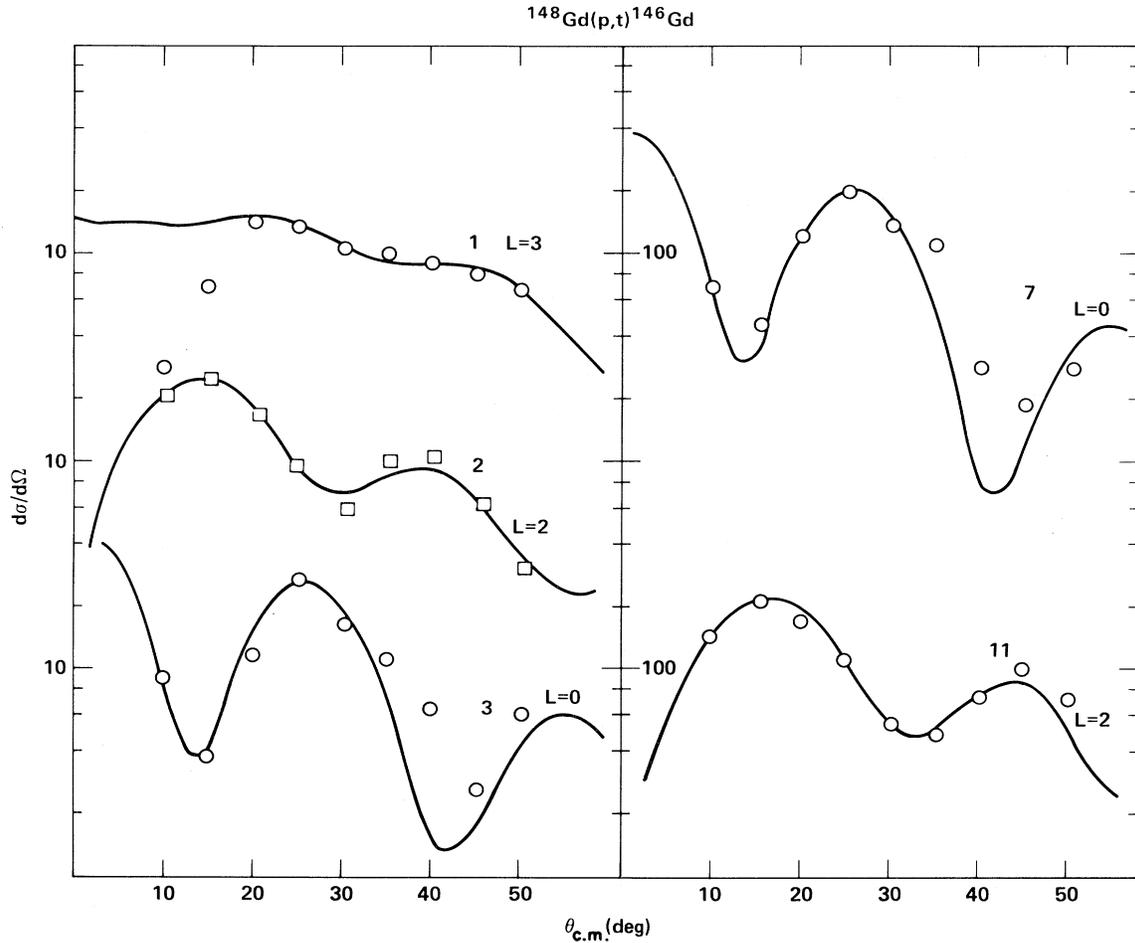


FIG. 4. Triton angular distributions and distorted-wave calculations for selected levels in the $^{148}\text{Gd}(p,t)^{146}\text{Gd}$ reaction. The ordinate scale is normalized to the ^{146}Sm data under the assumption that the first 3^- states are populated with the same cross section in ^{144}Sm and ^{146}Gd (see text).

lower- Z cases. However, the proper test of the harmonic model is the ratio of the cross section for the monopole state to the cross section for the g.s. of the $^{146}\text{Gd}(p,t)$ reaction. No one yet has fabricated a ^{146}Gd target for this measurement. Thus, the increased excitation energy of the PV supports the view that ^{146}Gd represents a neutron and proton shell and subshell closure, respectively. One therefore expects a proton PV in ^{146}Gd . Although there is some evidence⁷ for $L=0$ strength above 2.0 MeV in the ($^3\text{He},n$) reaction, there is no evidence that two-proton transfer reactions with heavy ions^{5,6} excite 0^+ states in ^{146}Gd .

The splitting of both the monopole and quadrupole PV strength in ^{144}Sm is unusual. A splitting of the quadrupole PV in ^{208}Pb was noted previously.²³ This splitting is clearly due to the accidental degeneracies of the excitation energies of the first 2^+ states of ^{206}Pb and ^{210}Pb . We must assume a similar phenomenon occurs here because the

$^{142,146}\text{Sm} (2_1^+)$ states are also approximately degenerate in energy with excitation energies of 0.768 and 0.747 MeV, respectively. For the monopole PV an additional argument is required. Since the PV represents the lowest $2p-2h$ state for neutrons, a strong excitation of an additional 0^+ state implies a degeneracy with a proton 0^+ state. The heavy-ion two-proton transfer reactions^{5,6} excite neither of these states, but then these 0^+ states may have no particular coherence among the protons.

ACKNOWLEDGMENTS

The authors are grateful to P. Kleinheinz for useful discussions and for carefully reading the manuscript, to S. Orbeson for his assistance with the Q3D, to R. E. Brown for assistance with the experiments, to group P-9 of

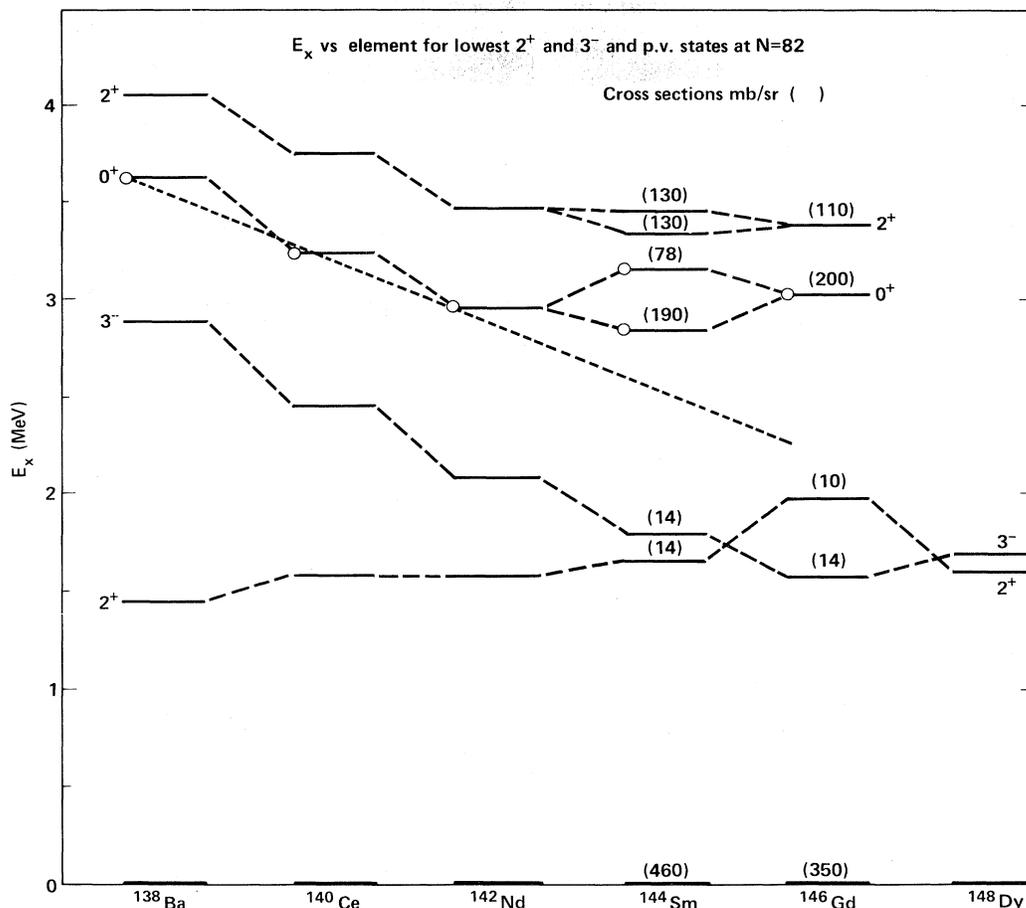


FIG. 5. Systematics of the lowest 2^+ and 3^- states in the $N=82$ nuclei. Also included are the dominant PV monopole and quadrupole states. For the present results on ^{144}Sm and ^{146}Gd , the cross sections for each level as seen in the (p,t) reaction are also given. The continuous dashed line is a linear extrapolation of the PV monopole energies from the lighter isotopes.

LANL for their operation of the Van de Graaff, to group CNC-11 at LANL for preparation of the Sm and Gd fractions, to D. Sisson at LLNL for the chemical purification of the Sm and Gd oxides, and to R. J. Dupzyk and C. M.

Henderson at LLNL for the isotope separations. This work was performed under the auspices of the U.S. Department of Energy by the Lawrence Livermore National Laboratory under Contract No. W-7405-Eng-48.

*Present address: National Superconducting Cyclotron Laboratory, Michigan State University, East Lansing, MI 48824.

¹P. Kleinheinz, S. Lunardi, M. Ogawa, and M. R. Maier, Z. Phys. A **284**, 351 (1978).

²M. Ogawa, R. Broda, K. Zell, P. J. Daly, and P. Kleinheinz, Phys. Rev. Lett. **41**, 289 (1978).

³P. Kleinheinz, R. Broda, P. J. Daly, S. Lunardi, M. Ogawa, and J. Blomqvist, Z. Phys. **290**, 279 (1979).

⁴A. L. Goodman, Nucl. Phys. **A331**, 401 (1979); R. R. Chasman, Phys. Rev. C **21**, 456 (1980); P. Mukherjee, R. Bhattacharya, and I. Mukherjee, *ibid.* **24**, 1810 (1981).

⁵W. von Oertzen, H. Homeyer, B. G. Harvey, D. Hendrie, and D. Kovar, J. Phys. A **279**, 357 (1976).

⁶R. C. Pardo, S. Gales, R. M. Ronningen, and L. H. Harwood, Phys. Lett. **91B**, 41 (1980).

⁷W. P. Alford, R. E. Anderson, P. A. Batay-Csorba, R. A. Emigh, D. A. Lind, P. A. Smith, and C. D. Zafiratos, Nucl. Phys. **A321**, 45 (1979).

⁸J. Wilhelmy, G. E. Bentby, K. E. Thomas, R. E. Brown, E. R. Flynn, J. van der Plicht, L. G. Mann, and G. L. Struble, *Proceedings of the Fourth International Conference on Nuclei Far from Stability, Helsingør, Denmark, 1981*, edited by P. G.

- Hansen and G. B. Nielsen (CERN, Geneva, 1981), p. 684.
- ⁹R. J. Prestwood, D. B. Curtis, and J. H. Capps, *Phys. Rev. C* **24**, 1346 (1981).
- ¹⁰A. H. Wapstra and K. Bos, *At. Data Nucl. Data Tables* **19**, 185 (1977).
- ¹¹E. R. Flynn, J. A. Cizewski, R. E. Brown, and J. W. Sunier, *Phys. Lett.* **98B**, 166 (1981).
- ¹²E. R. Flynn, S. D. Orbesen, J. D. Sherman, J. W. Sunier, and R. Woods, *Nucl. Instrum. Methods* **128**, 35 (1975).
- ¹³E. R. Flynn, J. G. Beery, and A. G. Blair, *Nucl. Phys.* **A218**, 285 (1974).
- ¹⁴J. Blomqvist, P. Kleinheinz, R. Broda, and P. J. Daly, *Proceedings of the Fourth International Conference on Nuclei Far from Stability, Helsingør, Denmark, 1981*, edited by P. G. Hansen and G. B. Nielsen (CERN, Geneva, 1981), p. 545.
- ¹⁵*Table of Isotopes*, 7th ed., edited by C. M. Lederer and V. S. Shirley (Wiley, New York, 1978).
- ¹⁶J. Styczen, P. Kleinheinz, M. Piiparinen, and J. Blomqvist, *Proceedings of the Fourth International Conference on Nuclei Far from Stability, Helsingør, Denmark, 1981*, edited by P. G. Hansen and G. B. Nielsen (CERN, Geneva, 1981), p. 548.
- ¹⁷F. G. Perey, *Phys. Rev.* **131**, 745 (1963); E. R. Flynn, D. D. Armstrong, J. G. Beery, and A. G. Blair, *ibid.* **182**, 1113 (1969).
- ¹⁸T. J. Mulligan, E. R. Flynn, O. Hansen, R. F. Casten, and R. K. Sheline, *Phys. Rev. C* **6**, 1802 (1972).
- ¹⁹J. B. Ball, R. L. Auble, J. Rapaport, and C. B. Fulmer, *Phys. Lett.* **30B**, 533 (1969).
- ²⁰R. Julin, J. Kantele, M. Luontama, A. Passoja, P. Kleinheinz, and J. Blomqvist, *Phys. Lett.* **94B**, 123 (1980).
- ²¹J. B. Ball, R. L. Auble, and P. G. Roos, *Phys. Lett.* **29B**, 172 (1969).
- ²²A. Bohr and B. Mottelson, *Nuclear Structure* (Benjamin, Reading, Mass., 1975), Vol. II, p. 645.
- ²³G. Igo, P. Barnes, and E. Flynn, *Phys. Rev. Lett.* **24**, 470 (1970).