STUDIES OF NUCLEAR FISSION, LOW-ENERGY NUCLEAR
REACTIONS AND TRANSURANIC NUCLEI

John R. Huizenga
Principal Investigator

June 1, 1972 - June 1, 1973

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Reactions and Transuranic Nuclei

Supported by the U. S. Atomic Energy Commission
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I. Introduction

The research activity of the group supported under the United States Atomic Energy Commission contract AT(11-1)3496 is carried out in the Nuclear Structure Research Laboratory and in Hutchison Hall, the new Chemistry-Biology building of the University of Rochester. Our research program includes studies of heavy ion reactions and fission, statistical nuclear decay including both experiment and theory, direct reaction studies on heavy deformed targets including inelastic scattering and transfer reactions (such as the (d,t) , $(^3\text{He},\alpha)$, $(^3\text{He},d)$, (α,t) and (p,t) reactions) and nuclear methods in chemical structure.

The major fraction of our experimental program is carried out with the Emperor tandem Van de Graaff in the Nuclear Structure Research Laboratory which is supported by a grant from the National Science Foundation. During the next year we plan to participate in collaborative programs to investigate heavy-ion reaction mechanisms at the Berkeley Super-Hilac and negative meson induced fission in the actinide elements at the Los Alamos Meson Physics Facility.

We wish to acknowledge the direct support of our research programs by the U. S. Atomic Energy Commission and the National Science Foundation for a nuclear equipment grant GP-28140 to the Chemistry Department. In addition, we are grateful to the National Science Foundation for providing a large amount of indirect support through the use of the facilities at the Nuclear Structure Research Laboratory.

II. RESEARCH PROGRAM

A) Heavy Ion Reactions and Fission

1) ✓ Elastic Scattering of Lithium on ^{232}Th and ^{238}U

H. Freiesleben, G. T. Rizzo, J. R. Huizenga and A. Prince*

Elastic scattering angular distributions were taken at energies of 24, 32 and 37.2 MeV incident ^6Li and ^7Li projectiles on ^{232}Th and ^{238}U . The data were taken with surface barrier detectors, the resolution was not good enough to discriminate inelastic scattered particles to the first 2^+ state. In order to see whether the inelastic processes are due to nuclear scattering or Coulomb excitation, the high resolution data reported earlier¹⁾ were analyzed using a Coulomb excitation code. Very good agreement was found between the high resolution data and calculations with the assumption that the nuclear inelastic scattering is negligible. Consequently, the unresolved groups of inelastic scattered projectiles are treated as part of the elastic group, since the optical model code JIB III²⁾ which we used does not include Coulomb excitation. Extensive calculations were performed to fit the scattering data and the total reaction cross section simultaneously (see paragraph on fission cross section). The "best" parameter set found for the 37.2 MeV data was a Saxon-Woods potential ($V_0=18.45$ MeV, $r_0=1.57\text{fm}$, $a_0=0.85\text{fm}$) with volume absorption ($W_s=14.93$ MeV, $r_{oi}=1.59\text{fm}$, $a_{oi}=0.60\text{fm}$, $r_{oc}=1.7\text{fm}$). No spin-orbit coupling was used.

* Brookhaven National Laboratory

Since we expected the static target deformation to be of influence on the differential cross section, coupled channel calculations were performed at Brookhaven National Laboratory using the code JUPITER-1 of Tamura³⁾. Only the coupling to the first excited 2+ state in ^{238}U was included by using the nonadiabatic approximation and a complex form factor. The calculated elastic and inelastic scattering cross sections were added for comparison with the experimental data. Again, an attempt was made to fit the scattering data at 37.2 MeV and the reaction cross section simultaneously.

The elastic scattering data of 37.2 MeV ^7Li on ^{238}U are shown in fig. 1 along with the predictions of the coupled channel calculation (Solid Line, $V_0=16.65$ MeV, $r_0=1.60$ fm, $a_0=0.85$ fm, $W_S=10.0$ MeV, $W_D=0$ MeV, $r_{oi}=1.59$ fm, $a_{oi}=0.60$ fm, $V_{so}=0$ MeV $r_{oc}=1.7$ fm, $\beta_2=.22$, $\beta_4=.05$) and the spherical optical model calculation (dashed line) with the same parameters except for an adjustment in the radius parameters to give volume conservation. The introduction of the deformed potential results in a better reproduction of the experimental data. However, due to the large amount of computer time needed for the coupled channel calculation, no attempt was made to improve the fit to the experimental data.

The same parameter set was used to calculate the scattering cross section at 32 MeV. The data and the calculations are shown in fig. 2. Again, the deformed potential gives a better description of the elastic scattering data, but the agreement between experiment and theory is not good. Little, if anything, is known about the energy dependence of the potential parameters,

Fig. 1 Plot of the cross section ratio of elastic scattering to Rutherford scattering of 37.2 MeV ${}^7\text{Li}$ on ${}^{238}\text{U}$.

Figure 1

Fig. 2 Plot of the cross section ratio of elastic scattering to Rutherford scattering of 32 MeV ${}^7\text{Li}$ on ${}^{238}\text{U}$.

Figure 2

and since the bombarding energies are below the Coulomb barrier one does not expect this effect to play an important role. The effect of projectile breakup may be important.

- 1) G. T. Rizzo, C. C. Lu and J. R. Huizenga, Annual Report (1971).
- 2) F. G. Perey, JIB III, Local Optical Potential Program with Automatic Parameter Search, 1965.
- 3) T. Tamura, Rev. Mod. Phys. 37, 679 (1965) and ORNL-4152 (1967).

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2) ✓ Fragment Correlation Angle Measurements of ${}^6\text{Li}$ and ${}^7\text{Li}$
Induced Fission of ${}^{232}\text{Th}$ and ${}^{238}\text{U}$ —

H. Freiesleben and J. R. Huizenga

The fragments of a binary fission process are emitted in opposite directions in the center-of-mass system. Due to the center-of-mass motion, the correlation angle measured in the laboratory system is less than 180° . For a special geometry where one fragment is detected at 90° , the most probable correlation angle for the second fragment is given by

$$\sin^2 \psi_{\text{mp}}(2) = (1 - x_2^2) / (1 + 2x_1x_2 + x_1^2)$$

where $x_i^2 = M_{\text{fi}} M_p E_p / E_{\text{fi}} M_{\text{CN}}^2$, $\psi_{\text{mp}}(2)$ is the most probable angle of fragment 2, M_p and E_p are the projectile mass and energy and M_{fi} and E_{fi} are the mass and energy of fission fragment i . A measurement of ψ_{mp} shows a distribution around ψ_{mp} , due to asymmetric mass division and neutron emission prior to or after fission. In addition, very asymmetric mass divisions give rise to smaller values of ψ_{mp} , making the correlation function slightly asymmetric. Since ψ_{mp} depends on the momentum transferred by the projectile to the target, the correlation angle measurement has often been used to distinguish between fission following compound nucleus formation [referred to as full momentum transfer (FMT)] or following direct interactions [referred to as incomplete momentum transfer¹⁾ (IMT)] at energies above the Coulomb barrier. We applied this method to ${}^6, {}^7\text{Li}$ induced fission on ${}^{232}\text{Th}$ and ${}^{238}\text{U}$ in order to investigate the percentage of the fission cross section

which is due to FMT. Measurements were performed at 30 and 38.4-MeV Li energies. Fission fragments were detected with surface barrier detectors. In order to have a large solid angle without losing angular resolution, rectangular apertures of $2 \times 15 \text{ mm}^2$ were used, subtending $\pm 0.8^\circ$ in the reaction plane ($\pm 6.5^\circ$ perpendicular to the reaction plane). The angular position of the detectors was checked by means of a telescope, and found to be accurate to $\pm 0.1^\circ$ with respect to the beam axis, which was defined by three aligned apertures. Standard electronics including a time-to-pulse height converter were used to detect and to discriminate accidental and real coincident events. By moving one detector, coincident counts were obtained as a function of angle. Special care was taken to establish the target position, because large errors in the correlation angle can be introduced by a target displacement along the beam axis. We checked the target position by Li-Li scattering. In any elastic scattering process of identical particles, the angle between scattered and recoiled particles is 90° if one particle is emitted at 45° . A typical result from a correlation experiment is shown in figure 3. The solid line is a fitted Gaussian distribution, following the experimental points very well. From the almost symmetric shape we conclude that less than 4% of the fission cross section follows IMT, at least at this energy.

A symmetric distribution at low incident energies (subcoulomb) does not prove that a compound nucleus has been formed. If one assumes that the Li-projectile disintegrates in the Coulomb field of the target nucleus, either of the breakup particles has

Fig. 3 Correlation angle for fission fragments
from ${}^6\text{Li}$ induced fission of ${}^{238}\text{Th}$.

Figure 3

a certain probability of being absorbed and inducing fission. If the second particle is scattered backwards, the fissioning system gets more than full momentum transfer. This can result in a broadening of the distribution and in a shift to smaller correlation angles, depending on the angular distribution of the outgoing particle. Our measured values of Ψ_{mp} are slightly smaller than the calculated values. However, a definite explanation of these results has not as yet been obtained.

- 1) W.J. Nicholson and I. Halpern, Phys. Rev. 116(1959)175;
T. Sikkeland, E. L. Haines and V. E. Viola, Jr., Phys. Rev. 125 (1962) 1350.

3) Angular Distributions for ${}^6\text{Li}$ and ${}^7\text{Li}$ Induced Fission
of ${}^{232}\text{Th}$ and ${}^{238}\text{U}$ —

H. Freiesleben, G. T. Rizzo and J. R. Huizenga

The differential fission cross section ratio $W(165^\circ)/W(90^\circ)$ for ${}^6\text{Li}$ and ${}^7\text{Li}$ induced fission of ${}^{232}\text{Th}$ and ${}^{238}\text{U}$ was measured between 23 and 38 MeV. An ${}^{238}\text{UF}_4$ target of $(118 \pm 4) \mu\text{g}/\text{cm}^2$ on $40 \mu\text{g}/\text{cm}^2$ C backing was used. The angle between the two surface barrier detectors, located at 90° and 165° and having the same geometry, was bisected by the target. Hence, any change in the beam position did not influence the cross section ratio since the solid angles cancel out.

The results are shown in fig. 4., converted to the CM-system. In addition, angular distributions have been measured at 37.2 MeV, using five detectors simultaneously. Fig. 5 shows the results, including a least square fit of Legendre polynomials to the data, using the equation

$$W(\theta_{\text{CM}}) = W(90^\circ) \sum_{L=0}^{L_{\text{max}}} a_{2L} P_{2L}(\cos \theta_{\text{CM}})$$

The coefficients are listed in table 1. Inclusion of a term $L=3$ did not improve the fit, showing that only angular momenta $L \leq 2$ between the two fragments are relevant.

Comparing the data for the same target in figs. 4 and 5, one observes that the ${}^7\text{Li}$ induced fission shows a larger anisotropy. This is expected from the fact that Γ_n/Γ_f is larger for the initial compound nucleus with one more neutron. Since the anisotropy for a fixed angular momentum distribution increases with decreasing excitation energy, more later chance fission (at lower excitation energy in the respective nuclei) increases the anisotropy. The

Fig. 4 Fission fragment anisotropy as a function of bombarding energy for ${}^6\text{Li}$ and ${}^7\text{Li}$ induced fission of ${}^{238}\text{U}$.

Figure 4

Fig. 5 Fission Fragment Angular distributions
for ${}^6\text{Li}$ and ${}^7\text{Li}$ Induced Fission of ${}^{238}\text{U}$
at bombarding energy of 37.2 MeV.

Figure 5

same effect is reflected in the sudden increase of the anisotropy of ${}^7\text{Li}$ induced fission, if a new (${}^7\text{Li}, xn$) channel opens. This occurs at ${}^7\text{Li}$ energies of about 24 MeV (${}^7\text{Li}, 3n$) and 31 MeV (${}^7\text{Li}, 4n$) as can be seen in fig. 4. The threshold effect is less obvious in ${}^6\text{Li}$ induced fission. A careful comparison of both anisotropies is expected to shed some light on the initial compound nucleus properties, especially on the first chance Γ_n/Γ_f ratio. Similar results were obtained for the target ${}^{232}\text{Th}$.

Table 1. Legendre Coefficients

	a_0	a_1	a_2
${}^{232}\text{Th} + {}^6\text{Li}$	1.079	0.173	0.020
$+ {}^7\text{Li}$	1.107	0.259	0.059
${}^{238}\text{U} + {}^6\text{Li}$	1.060	0.136	0.020
$+ {}^7\text{Li}$	1.091	0.204	0.030

4) Total Reaction Cross Sections of ${}^6\text{Li}$ and ${}^7\text{Li}$ on ${}^{232}\text{Th}$ and ${}^{238}\text{U}$ —

H. Freiesleben, G. T. Rizzo, J. R. Huizenga, A. Prince*

The total fission cross sections were obtained by integrating the measured differential cross sections making use of the measured anisotropy $W(165^\circ)/W(90^\circ)$ ratios. The results are shown in Figs. 6 and 7. In addition, Fig. 7 shows the results of theoretical calculations for the ${}^{238}\text{U} + {}^7\text{Li}$ total reaction cross sections using different models.

The fission cross sections for these systems are to a good approximation equal to the total reaction cross section. For example, with 38-MeV ${}^7\text{Li}$ on ${}^{238}\text{U}$, the initial compound nucleus ${}^{245}\text{Am}$ is formed with 37.1 MeV of excitation energy. Since the fission barriers of such heavy nuclei are approximately 6 MeV, three or more chance fission occurs over the whole range of bombarding energy used in these experiments. Since previous estimates of Γ_f/Γ_n for each of these nuclei formed by neutron evaporation are approximately unity or larger, the sum of the fission cross sections (the quantity measured) for the multichance fission process approaches the compound nucleus cross section, although in a strict sense remains a lower limit to the reaction cross section. In Fig. 7, curve I represents the results obtained using the parabolic approximation of a spherical potential barrier with a standard set of parameters given by Thomas¹⁾. Curve II results from a spherical optical model calculation, using the code JIB III (see paragraph on elastic scattering). Curve III is calculated

* Brookhaven National Laboratory

Fig. 6 Fission cross sections as a function of
projectile energy for ${}^6\text{Li}$ and ${}^7\text{Li}$ Induced
Fission of ${}^{232}\text{Th}$.

Figure 6

Fig. 7. Comparison of experimental fission cross sections for ${}^6\text{Li}$ and ${}^7\text{Li}$ induced fission of ${}^{238}\text{U}$ with various models described in the text.

Figure 7

with a parabolic approximation of a deformed barrier, using a method similar to that of Rasmussen et al²⁾. From a comparison of curves II and III one observes a close similarity between these curves. This suggests that the imaginary potential in the spherical optical model and the deformation in the parabolic approximation affect the absorption in a similar way. Consequently a deformed optical model with imaginary potential is expected to further improve the description of the experimental data. The calculations were carried out using the coupled channel code JUPITER-1 of Tamura³⁾. The results are displayed in fig. 8 where the full line represents the coupled channel calculation. The same set of potential parameters has been used as described previously (see paragraph on elastic scattering). Again, the agreement between theory and experiment is improved.

A direct reaction, followed by fission like (${}^7\text{Li}, \alpha f$) is taken into account by the imaginary potential and has been shown to be negligible at high bombarding energies (see paragraph on correlation angle measurements). However, a Coulomb disintegration of the projectile followed by absorption of either of the break-up particles, which may in turn induce fission is not included in any model we used. The remaining discrepancy between data and calculations may be due to these processes. Presently, we cannot establish whether target deformation or projectile break-up or both of these effects are responsible for the enhancement of the fission cross section at subcoulomb bombarding energies.

1) D. T. Thomas, Phys. Rev. 116, 703 (1959).

2) J. O. Rasmussen and K. Sugarawa-Tanabe, Nucl. Phys. A171, 497 (1971).

3) T. Tamura, Rev. Mod. Phys. 37, 679 (1965) and ORNL-4152 (1967).

Fig. 8. Comparison of experimental fission cross sections for ${}^6\text{Li}$ and ${}^7\text{Li}$ induced fission of ${}^{238}\text{U}$ with a spherical optical potential (dashed line) and a coupled channel calculation (solid line).

Figure 8

5) Study of the ${}^7\text{Li} + {}^{209}\text{Bi}$ Reaction

H. C. Britt*, H. Freiesleben and J. R. Huizenga

An experimental program has been initiated to study various aspects of the ${}^7\text{Li} + {}^{209}\text{Bi}$ reaction at bombarding energies in the range 24-38 MeV. Cross sections for the $({}^7\text{Li},f)$ reaction are determined by detecting fission fragments in a semiconductor detector. Cross sections for the production of the various Rn, At and Po isotopes are determined by observing known energy alpha particle groups from the short lifetime decays of these isotopes. The background due to prompt reaction particles is eliminated by using a pulsed ${}^7\text{Li}$ beam and detecting the decay alpha particles between beam pulses.

This program is designed to investigate three semi-independent aspects of the ${}^7\text{Li} + {}^{209}\text{Bi}$ reaction.

1. Measure the total reaction cross section as a function of bombarding energy. It is hoped that a comparison of results from ${}^{209}\text{Bi}$ (a spherical nucleus) with ${}^7\text{Li}$ and ${}^4\text{He}$ measurements on Th and U isotopes (deformed nuclei) will give a direct measurement of the effect of deformation on the total reaction cross section at energies well below the Coulomb barrier.
2. Study the relative magnitudes of $({}^7\text{Li},xn)$, $({}^7\text{Li},txn)$, and $({}^7\text{Li},\alpha xn)$ reactions. From the relative cross sections for the production of Rn, At, and Po isotopes, information can be obtained on the relative cross sections for fusion and transfer reactions. In addition, the intensity as a function

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of the number of neutrons emitted for transfer reactions gives information on the average excitation energy following the alpha particle or triton transfers.

3. Direct measure of $\langle \Gamma_n / \Gamma_f \rangle$. From the measurements, the value for $\langle \Gamma_n / \Gamma_f \rangle$ for the compound nucleus ^{216}Rn is obtained directly. The experimental results can be compared directly with theoretical potential energy calculations to give a test of the current models in the transition region above Pb where shell effects are changing very rapidly with Z and A. This experiment will provide the first direct measurement of $\langle \Gamma_n / \Gamma_f \rangle$ in this region. Previous experiments have estimated this quantity by comparing a measured fission cross section with a calculated total reaction cross section.

A preliminary run on the accelerator indicated that the experiment is feasible. At 34 MeV bombarding energy all of the expected alpha particle groups were observed and preliminary values for relative cross sections for the production of various product nuclei are listed in table 2. At lower bombarding energies the ($^7\text{Li}, 2n$) reaction becomes more important as expected but it also appeared that the ($^7\text{Li}, n$) reaction accounted for a significant fraction of the total reaction cross section.

- 6) Fragment Kinetic Energies from ^{18}O Induced Fission of ^{246}Cm
H. Freiesleben and R. L. Ferguson*, F. Plasil*, C. Bemis, Jr.,*
and H. W. Schmitt*

Recently¹⁾ it was predicted on the basis of a static scission model, that total fragment kinetic energies in binary fission are

¹ H. W. Schmitt and U. Mosel, Nucl. Phys. A186 (72) 1.

* Oak Ridge National Laboratory

TABLE 2

Results from 34 MeV ^7Li + ^{209}Bi Reaction

Reaction	Product Nucleus	Relative Intensity
$(^7\text{Li}, \text{Capture})$	^{216}Rn	<.006
$(^7\text{Li}, n)$	^{215}Rn	.002
$(^7\text{Li}, 2n)$	^{214}Rn	~.02
$(^7\text{Li}, 3n)$	^{213}Rn	1.0
$(^7\text{Li}, 4n)$	^{212}Rn	.5
$(^7\text{Li}, t)$	^{213}At	~.01
$(^7\text{Li}, tn)$	^{212}At	.09
$(^7\text{Li}, t2n)$	^{211}At	a
$(^7\text{Li}, \alpha)$	^{212}Po	<.006
$(^7\text{Li}, \alpha n)$	^{211}Po	.22
$(^7\text{Li}, \alpha 2n)$	^{210}Po	a

^a These decays were observed but relative cross sections have not yet been obtained.

larger than those predicted with the liquid drop model in the mass region $250 \leq A \leq 290$. The largest kinetic energies were calculated¹⁾ for the cases where two fragments close to the double magic ^{132}Sn are emitted. To test these predictions, we measured the total kinetic energy of fission fragments produced by bombarding ^{232}Th and ^{246}Cm targets with ^{18}O . The liquid drop model (the static fission model with symmetric mass division) predicts $\langle E_k \rangle = 185 \text{ MeV}$ (185 MeV) for the $^{232}\text{Th} + ^{18}\text{O}$ system and 203 MeV (258 MeV) for the $^{246}\text{Cm} + ^{18}\text{O}$ system.

The experiments were performed at ORIC using $^{18}\text{O}^{5+}$ beams of 93.7, 105.2 and 125.9 MeV. Coincident fission fragments were detected with two solid state detectors. One detector subtending a small solid angle was set at forward angles, while the other subtending a large solid angle was properly set to detect complementary fragments. Pulse heights of coincident events were written on magnetic tape. The system was calibrated with a spontaneously fissioning ^{252}Cf source. Off line analysis was performed to calculate the CM-energies and masses of fragment pairs to get the total kinetic energy and mass distributions.

The experimental results for the $^{232}\text{Th} + ^{18}\text{O}$ system are in good agreement with the LD prediction. A preliminary analysis of the $^{246}\text{Cm} + ^{18}\text{O}$ data shows an even smaller $\langle E_k \rangle$ than the LDM predicts. This result does not necessarily prove the static scission model to be wrong, since the shell effects, leading to large $\langle E_k \rangle$ may be washed out at excitation energies above 50 MeV, where the compound nucleus $^{264}104$ is formed in our experiment. In addition,

the possibility of incomplete momentum transfer leading to fission in nuclei of lower Z has not been ruled out. The data analysis is still in progress.

7) ✓ Threshold Photofission: Theory and Experiment

J. R. Huizenga and H. C. Britt

Information on the properties of the fission channels in the highly deformed transition nucleus (or saddle point nucleus) is obtained from a study of the energy dependence of the fission fragment cross section and angular distribution. The photofission process is especially valuable in such studies due to the restricted angular momenta in the entrance channel.

Experimental measurements of near barrier photofission cross sections and angular distributions induced with bremsstrahlung and monoenergetic gamma rays have been theoretically analyzed in order to characterize barrier transition states. From the experimental data for heavy nuclei, we conclude that the $K=0^-$ and 0^+ barriers are nearly degenerate at the second or outer barrier, while the $K=0^-$ barrier is approximately 0.7 MeV higher at the first or inner barrier.

The observed probabilities for the fission of ^{232}Th and ^{238}U induced by photons and the (t, pf) reaction have been shown to be similar (see fig. 9). Since the decreases in the fission probabilities for the (t, pf) and the (γ, f) reactions above the neutron binding energy resemble each other closely, we feel that the previous assignment of photofission resonances at these energies is unwarranted. At energies below the neutron binding energy, one

Fig. 9. Comparison of the fission probability as a function of excitation energy for fission induced with photons and the (t,pf) reaction.

Figure 9

observes one or two small bumps or inflections in the ^{232}Th and ^{238}U photofission probabilities at energies which match similar structure in charged-particle induced fission. There is some evidence that the mass distribution is slightly altered for resonance fission.

From an analysis of the photofission results for ^{232}Th , where the outer barrier is the higher one, the quadrupole to dipole absorption cross section ratio for heavy elements is estimated to be 0.04 ± 0.02 .

8) A New Two-Center Shell Model for Nuclear Fission[†]

Klaus Albrecht

A two-center single-particle potential is proposed which has the Lawrence family of liquid drop shapes as its equipotentials. An improved parameterization of these shapes is given that facilitates their use. Only two deformation parameters are required to describe all relevant nuclear shapes for symmetric fission. These encompass Nilsson ground state shapes with ϵ - and (approximately) ϵ_2 -deformations, dumbbells and spheroidal fragments at large separation. The corresponding potential evolves from a deformed harmonic oscillator for the ground state of the fissioning nucleus to two deformed harmonic oscillators for well separated fragments.

In the single-particle calculation, a Thomas-type spin-orbit potential is used together with an ℓ^2 correction term. The nuclear deformation energy is calculated by Strutinsky's prescription, including pairing energy. Results are given as

[†] The major part of this work was done at Institute für Theoretische Physik der Universität Frankfurt.

deformation energy maps for the symmetric fission mode of various actinide nuclei. The asymmetric generalization of the model is outlined.

9) Negative Muon Induced Fission in the Actinide Elements

J. R. Huizenga, C. Kalbach-Cline, Z. Sujkowski^{*} and J. Unik[†]

A proposal has been submitted to study fission induced in the actinide elements by negative muons. We plan to perform this work at the stopped muon facilities of LAMPF. Initial work will probably include studies of fission induced by μ^- capture in the nucleus and by non-radiative muonic atom transitions in order to determine the relative yields from these two processes and the half-life for muon capture from the 1s atomic orbital. It is also hoped that additional measurements (by us or by other groups) of the yields of the different muonic x-rays per mu-stop event will provide information on the different radiationless transitions which can excite the nucleus above the fission threshold. This would permit the determination of the partial fission width following a radiationless transition. The systematics of all these quantities (prompt-to-delayed fission yield, capture half-life and post-radiationless-transition fission branching ratio) will be studied as functions of both the neutron and proton number of the system. Where possible, they will be related to theoretical expectations or to the results of fission studies using more conventional nuclear techniques.

^{*} Instytut Barden Jadrowych, Warsaw, Poland

[†] Argonne National Laboratory

Eventually these investigations may also be extended to include the study of neutrons or protons emitted following the stopping of a muon in the target.

B. Statistical Nuclear Decay

1) ✓ Dependence of Nuclear Level Width on Isospin From Ericson Fluctuation Measurements

M. Kildir and J. R. Huizenga

The statistical theory of fluctuations¹⁾ has been used successfully to describe fluctuations in the excitation functions of nuclear reactions which proceed through a statistical compound nucleus process. The average level width of the compound nucleus is extracted from a statistical analysis of excitation functions.

In the present study we report on the level widths of the compound nucleus ^{32}S determined from fluctuation analyses of the $^{31}\text{P}(p,p')$ and $^{31}\text{P}(p,\alpha)$ excitation functions. These level widths are used to calculate relative level densities of the $T=0$ and $T=1$ isospin states in the compound nucleus ^{32}S at an excitation energy 17.8 MeV. In addition, we have also calculated relative level densities of the $T=5/2$ and $T=7/2$ isospin states of the compound nucleus ^{55}Mn at an excitation energy of 17.6 MeV, using the level widths obtained from the $^{54}\text{Cr}(p,p')$ and $^{54}\text{Cr}(p,\alpha)$ excitation functions²⁾. The ^{32}S nucleus has been studied³⁾ previously in order to determine the effect of excitation energy and angular momentum on the level width.

Excitation functions of the $^{31}\text{P}(p,p')$ and $^{31}\text{P}(p,\alpha)$ reactions

were measured simultaneously in the proton energy interval of 8.51 to 10.01 MeV in 10-keV steps. A phosphorus target of approximately $40 \mu\text{g}/\text{cm}^2$ thickness was prepared on $20 \mu\text{g}/\text{cm}^2$ carbon backing by a vacuum evaporation method and was set at 30° to the beam direction in a 51-cm scattering chamber. The alpha particles and protons were detected by surface barrier solid-state detectors. Collimators of 0.45-cm diameter were placed in front of all detectors, and the proton detectors, except for the one at 90° were covered with a sufficient thickness of aluminum absorber to just stop the alpha particles.

The excitation functions show a decrease in cross section with increasing energy in the energy range studied. This energy dependence was removed from each excitation function before the fluctuation analysis was done. In the case of the $^{31}\text{P}(\text{p},\alpha)$ excitation functions, Hauser-Feshbach type calculations were performed to estimate the energy dependence of the cross section.

In figure 10, two of the excitation functions are shown before and after the energy dependence is removed. Level widths are determined from autocorrelation calculations of excitation functions resulting after the energy dependence is removed. The only correction applied to the level widths is the correction for the finite energy range of the data. The average level widths $\bar{\Gamma}$ extracted from $^{31}\text{P}(\text{p},\text{p}')$ and $^{31}\text{P}(\text{p},\alpha)$ excitation functions are 30.0 ± 1.4 and 38.7 ± 2.7 keV, respectively, where the errors on the average level widths are based on the mean square deviations of individual Γ values from $\bar{\Gamma}$. The average level widths for the $^{54}\text{Cr}(\text{p},\text{p}')$ and

Figure 10. (a) The experimental and Hauser-Feshbach corrected excitation functions for the reaction $^{31}\text{P}(p, \alpha_0)^{28}\text{Si}$ at $\theta_{\text{lab}} = 165^\circ$.
(b) The experimental and corrected excitation functions for the reaction $^{31}\text{P}(p, p_2)^{31}\text{P}$ at $\theta_{\text{lab}} = 150^\circ$.

Figure 10

$^{54}\text{Cr}(p,\alpha)$ reactions are 14.6 ± 0.6 and 10.8 ± 0.4 keV, respectively.

In order to compare experimental results with the theoretical predictions we performed calculations of level widths with the statistical model of nuclear reactions. The calculational procedure is described elsewhere³⁾. However, in the present calculations σ_J is defined as the partial reaction cross section for a particular reaction channel proceeding through the compound nucleus with a specific spin J . The values of σ_J are estimated from a Hauser-Feshbach calculation. We used standard Fermi Gas and Back-Shifted Fermi Gas level densities for residual and compound nuclei. The standard optical model parameters are used to calculate transmission coefficients.

The total width is given by

$$\frac{1}{\Gamma} = \sum \frac{\sigma_J}{\sigma} \cdot \frac{1}{\Gamma_J}$$

where $\sigma = \sum_J \sigma_J$. From the calculated widths for different reaction channels we conclude that the effect of angular momentum on the level width is small. The dependence of level width on the reaction angle was investigated also and found to be small.

One possible explanation for the experimental level width of the (p,α) reaction channels being different from that of the (p,p') reaction channels is due to partial or complete isospin conservation. It has already been shown⁴⁾ that isospin plays an important role in statistical compound nuclear reactions. If isospin is a good quantum number one sees that the (p,α_0) , (p,α_1) and (p,α_2) reactions proceed through only the T_z isospin states. The width extracted from the

(p, p_0) , (p, p_1) and (p, p_2) excitation functions on the other hand is a weighted average of the widths of the T_z and T_y isospin states. From the experimental values of average level widths we determined the level widths of the $T_z=0$ and $T_y=1$ isospin states of the compound nucleus ^{32}S to be 38.7 ± 2.7 keV and 26.2 ± 3.5 keV, respectively. In the case of the compound nucleus ^{55}Mn , the determined level widths of the $T_z=5/2$ and $T_y=7/2$ isospin states are 10.8 ± 0.5 keV and 16.1 ± 1.5 keV, respectively. These level widths are used to calculate the level densities of the T_z and T_y isospin states in the compound nucleus. In table 3, the ratios of densities of the two isospins are given for different sets of level density parameters of the residual nuclei. One sees that this ratio is only weakly dependent on the parameters of the residual nuclei.

Energy differences between T_y and T_z isospin states in ^{32}S and ^{55}Mn nuclei are about 7 and 10 MeV, respectively. Experimental result for ^{32}S indicates that the two isospin states have about the same density. This result is partially accounted for by nuclear pairing. Whereas, the $T=0$ states originate from an even-even nucleus with a large pairing energy, the $T=1$ states of ^{32}S are analog states of ^{32}P which is an odd-odd nucleus. Hence, the effective energy difference between the two isospin states is reduced by the pairing energy. In the case of odd-mass ^{55}Mn nucleus, the pairing force doesn't alter the energy difference between T_y and T_z isospin states. In the future we plan to confirm this explanation of the relative densities of the levels of different isospin with realistic level density calculations including isospin.

TABLE 3

Relative level densities of $T_<$ and $T_>$ isospin states of the compound nuclei ^{32}S and ^{55}Mn for different sets of level density parameters of the residual nuclei.

The Compound Nucleus ^{32}S , $U_c=17.8$ MeV

a	$\Delta^{31}\text{P}$	$\Delta^{31}\text{S}$	$\Delta^{28}\text{Si}$	$\frac{\rho(U_c, J=0, T=1)}{\rho(U_c, J=0, T=0)}$
A/9	0.0	0.0	3.0	1.230
A/9	1.0	1.0	3.0	1.008
A/9	0.0	0.0	4.0	1.428
A/8	0.0	0.0	3.0	1.263
A/10	2.0	2.0	4.0	1.038

The Compound Nucleus ^{55}Mn , $U_c=17.6$ MeV

a	$\Delta^{54}\text{Cr}$	$\Delta^{54}\text{Mn}$	$\Delta^{51}\text{V}$	$\frac{\rho(U_c, J=1/2, T=7/2)}{\rho(U_c, J=1/2, T=5/2)}$
A/9	1.5	-2.5	-0.5	3.613×10^{-3}
A/9	2.5	-2.5	-0.5	2.686×10^{-3}
A/9	1.5	-1.5	-0.5	4.495×10^{-3}
A/8	1.5	-2.5	-0.5	2.625×10^{-3}
A/10	2.0	0.0	1.0	7.344×10^{-3}

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2) Calculation of the Level Density and Spin Cut-off Factor with
the Inclusion of Nuclear Pairing

A. N. Behkami[†], G. Chan and J. R. Huizenga

We are continuing our program to compare theoretical spin
dependent nuclear level densities with similar experimental results.
We employ a statistical method for computation of state densities
which is based on the grand partition function. These microscopic
calculations make use of realistic sets of single particle energy
levels and nuclear pairing is included in the grand partition
function.

As sample results we show the comparison of the experimental
and theoretical level densities and spin cutoff factors for ⁵⁶Fe
in Figs. 11 and 12. These results are based on Seeger's single
particle energy levels and nuclear pairing parameters of

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Fig. 11 Comparison of the experimental and
theoretical level densities for ^{56}Fe .

Figure 11

Fig. 12 Comparison of the experimental and
theoretical spin cutoff factors for
 ^{56}Fe .

Figure 12

$\Delta_p = 1.43$ MeV and $\Delta_n = 1.08$ MeV. The inclusion of pairing has an important effect on both the theoretical level density and its spin dependence. Excellent agreement is obtained between theory and experiment for both the total level density and its spin dependence as shown in Figs. 11 and 12.

Comparisons are being made presently between theory and the neutron resonance data.

3) A Note on Statistical State Density Calculations

K. Albrecht, A. N. Behkami[†] and J. R. Huizenga

Nuclear state densities derived from single-particle plus pairing Hamiltonians have been calculated by various authors^{1,2)}. The same Hamiltonian has been treated by Jensen and Damgaard³⁾ in a computationally slightly different way. As these authors claim that their calculations yield results which are apparently different from the earlier ones, we would like to clarify the situation with this note.

In the framework of statistical mechanics, average state densities are usually obtained from the well-known relation (see e.g., ref.⁴⁾)

$$\omega(E, N) = \frac{1}{(2\pi i)^2} \int_{-i\infty}^{+i\infty} d\beta \int_{-i\infty}^{+i\infty} d\alpha Z(\beta, \alpha) \exp(\beta E - \alpha N), \quad (1)$$

which expresses the state density as the inverse Laplace transform of the grand partition function $Z(\beta, \alpha)$. For our purpose, it suffices to consider only one kind of fermion, e.g., neutrons. Evaluating the above integrals with the saddle-point approximation

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yields the following state density

$$\omega(E, N) = \frac{z^{\beta, \alpha}(\beta_0, \alpha_0) \exp(\beta_0 E - \alpha_0 N)}{2\pi |D^{\beta, \alpha}(\beta_0, \alpha_0)|^{1/2}} \quad (2)$$

The stationary point $\beta_0(E, N)$, $\alpha_0(E, N)$ of the integrand in (1) is found from

$$\left(\frac{\partial \ln z^{\beta, \alpha}}{\partial \beta} \right)_{\beta_0, \alpha_0} + E = 0; \quad \left(\frac{\partial \ln z^{\beta, \alpha}}{\partial \alpha} \right)_{\beta_0, \alpha_0} - N = 0 \quad (3)$$

and the determinant $D^{\beta, \alpha}$ is given by

$$D^{\beta, \alpha}(\beta_0, \alpha_0) = \begin{vmatrix} \frac{\partial^2 \ln z^{\beta, \alpha}}{\partial \beta^2} & \frac{\partial^2 \ln z^{\beta, \alpha}}{\partial \beta \partial \alpha} \\ \frac{\partial^2 \ln z^{\beta, \alpha}}{\partial \alpha \partial \beta} & \frac{\partial^2 \ln z^{\beta, \alpha}}{\partial \alpha^2} \end{vmatrix}_{(\beta_0, \alpha_0)} \quad (4)$$

The superscript β, α has been included here as an explicit reminder that β and α are the independent variables in $z^{\beta, \alpha}$. Most applications of (2) have been made in terms of β and α . It may, however, be more convenient to use another set of variables in order to simplify the explicit expressions of D which arise from a specific model. Thus β and λ were used in ref.³⁾ as independent variables, where $\lambda = \alpha/\beta$. These authors find an apparent disagreement between their $D^{\beta, \lambda}$ and the corresponding $D^{\beta, \alpha}$ obtained earlier^{1,2)}. We will show now that such disagreement is spurious.

If any function $F^{\beta, \alpha}$ is expressed in terms of β and λ , the following identities are seen to hold

$$F^{\beta, \alpha}(\beta, \alpha) = F^{\beta, \lambda}(\beta, \lambda(\beta, \alpha))$$

$$\frac{\partial F^{\beta, \alpha}}{\partial \beta} = \left(\frac{\partial}{\partial \beta} - \frac{\lambda}{\beta} \frac{\partial}{\partial \lambda} \right) F^{\beta, \lambda}; \quad \frac{\partial F^{\beta, \alpha}}{\partial \alpha} = \frac{1}{\beta} \frac{\partial F^{\beta, \lambda}}{\partial \lambda} \quad (5)$$

Using these transformations, one can establish D in terms of the new variables. We find

$$D^{\beta, \alpha}(\beta_0, \alpha_0) = \begin{vmatrix} \frac{\partial^2 \ln Z^{\beta, \lambda}}{\partial \beta^2} & \frac{\partial}{\partial \beta} \left(\frac{1}{\beta} \frac{\partial \ln Z^{\beta, \lambda}}{\partial \lambda} \right) \\ \frac{\partial}{\partial \beta} \left(\frac{1}{\beta} \frac{\partial \ln Z^{\beta, \lambda}}{\partial \lambda} \right) & \frac{1}{\beta^2} \frac{\partial^2 \ln Z^{\beta, \lambda}}{\partial \lambda^2} \end{vmatrix}_{\beta_0, \lambda_0 = \frac{\alpha_0}{\beta_0}} = D^{\beta, \lambda}(\beta_0, \lambda_0) \quad (6)$$

Since the right-hand side of this equation is also obtained by Jensen and Damgaard³⁾, we have proved that there cannot be any discrepancy between their $D^{\beta, \lambda}$ and the $D^{\beta, \alpha}$ given, e.g., by Moretto¹⁾ as well as by eq. (4). It is important to note, however, that this holds only for the value of D, whereas the corresponding elements of $D^{\beta, \lambda}$ and $D^{\beta, \alpha}$ need not be the same. In fact they are all different except for the (2,2) element. It seems that this difference in the elements has led the authors of ref.³⁾ to their erroneous conclusion that the D values must be different, too. When the elements of D are written explicitly for the Hamiltonian under discussion, it can indeed be seen that those obtained with β and λ are simpler than those obtained with β and α .

We further note that an alternative and faster proof of (6) can be given which makes use of the transformation properties of the second-order derivative matrix at a stationary point. Let $F(x_i)$, $i = 1, \dots, n$ be stationary at $\{x_i^0\}$ and consider the coordinate transformation $u_k = u_k(x_i)$. If the transformed function is called f , we have $F(x_i) = f(u_k(x_i))$ and it can readily be proved that the determinant transforms with the square of the Jacobian:

$$\left| \frac{\partial^2 F}{\partial x_i \partial x_k} \right|_{(x_i^0)} = \left| \frac{\partial^2 F}{\partial u_i \partial u_k} \right|_{(u_i^0)} \cdot \left| \frac{\partial u_i}{\partial x_k} \right|_{(x_i^0)}^2 \quad (7)$$

Application of this result for $n = 2$ to the function of $\ln Z + \beta E - \alpha N$ yields (6).

Finally, we note that another related discrepancy mentioned in ref. 3) is also spurious. These authors claim that Moretto's expression (1) for the derivative $\partial \Delta / \partial \beta$ of the gap parameter seems incorrect because it differs from their $\partial \Delta / \partial \beta$. It is true that $\partial \Delta^{\beta, \alpha} / \partial \beta \neq \partial \Delta^{\beta, \lambda} / \partial \beta$, but this does not mean that one expression is incorrect. Realizing again that two different sets of variables are used, we see from (5) that $\partial \Delta^{\beta, \lambda} / \partial \beta$ has to be compared to $\partial \Delta^{\beta, \lambda} / \partial \beta - (\lambda / \beta) \partial \Delta^{\beta, \lambda} / \partial \lambda$. If the explicit expressions for these quantities are inserted, agreement is found again. We thus conclude that the (β, α) approach and the (β, λ) approach are equally valid for the problem under discussion, provided the mathematics is handled properly.

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4) Nuclear Level Densities - A Review

J. R. Huizenga and L. G. Moretto[†]

A review was recently completed on nuclear level densities for the Annual Review of Nuclear Science. The following topics are discussed in this review.

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I. THEORETICAL OUTLINE OF THE METHODS AND MODELS EMPLOYED IN THE EVALUATION OF LEVEL DENSITIES

A. Methods of Calculation

1. The combinatorial method
2. The partition function method

B. Nuclear Models

1. System of noninteracting fermions
2. The equidistant model
3. Level densities for more complex sets of single particle levels
4. Level densities from the shell model
5. System of interacting fermions
6. Inclusion of collective degrees of freedom

II. EXPERIMENTAL SOURCES OF INFORMATION ON LEVEL DENSITIES

A. Neutron Resonances

B. Charged-Particle Resonances

C. Inelastic Scattering and Nuclear Reactions to Resolved Levels

D. Spectrum of Evaporated Particles

E. Excitation Functions of Isolated Levels

F. Ericson Fluctuation Widths

G. Compound Nucleus Lifetime and Level Density from the Blocking Effect

H. EXcitation Functions

I. Spin Distribution of the Nuclear Level Density

J. Yrast Levels

5) Relative Yields of (p,p') and (p,n) Reactions

C. Kalbach-Cline, J. R. Huizenga, S. M. Grimes[†], J. D. Anderson[†], C. Wong[†] and J. T. Davis[†]

For about six years, people have been using various modifications of Griffin's preequilibrium model to analyze the energy spectra of particles emitted in medium energy nuclear reactions. One crucial feature of the model that has remained untested through the lack of good data is the predicted relative intensities of preequilibrium neutrons and protons. The present work is designed to provide a test of these model predictions.

The purpose of these experiments was to measure both the proton and neutron spectra for reactions involving the same type and energy projectile incident on the same target. Since neither shell nor pairing effects are included, at present, in the extended Griffin model, the systems for study were chosen to minimize these effects. Thus they are all far removed from shell closures and involve protons incident on odd Z targets. This latter condition means that the composite system will be even-even, producing residual nuclei which are both odd A. The target materials chosen were ^{103}Rh , ^{159}Tb , ^{169}Tm and possibly ^{181}Ta . Initial measurements are being carried out with an 18-MeV beam but later work with higher energy protons is being considered. The (p,n) measurements are being done at the Livermore Cyclograff using a fixed neutron array in a low background room. Spectra have been obtained for the $^{103}\text{Rh}(p,n)$ reaction at 3.5° , 9.3° , 17° , 32° , 54° , 69° , 84° , 113° , 129° , and 144° . These have been analyzed, and an angle integrated

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spectrum has been obtained. The (p,p') runs are taking place at the Rochester MP Van de Graaff using a 2-element counter telescope. Data has recently been taken on the $^{103}\text{Rh}(p,p')$, $^{159}\text{Tb}(p,p')$ and $^{169}\text{Tm}(p,p')$ reactions at angles of 32° , 54° , 69° , 84° , 113° , 129° and 144° . No analysis of this data has yet been performed.

Freeequilibrium calculations are being carried out using the extended Griffin model code PRECOM. This program includes Pauli principle effects and uses empirical estimates of the 2-body matrix element to calculate absolute cross sections. It runs on the NSRL-PDP-6 computer.

6) ✓ Mechanisms of (p,p') Reactions on the Heavy Tin Isotopes

C. Kalbach-Cline, J. R. Huizenga and H. K. Vonach[†]

The measurement of energy spectra from (p,p') reactions induced by 14.0- and 17.8-MeV protons incident on targets of ^{118}Sn , ^{120}Sn and ^{124}Sn was reported in the 1971 Annual Report from this laboratory. The measured proton spectra are expected to contain only negligibly small contributions due to equilibrium proton evaporation from the T_γ composite states. The observed protons should result primarily from T_γ state equilibrium decay plus decay of any T_γ states which have not undergone isospin mixing. For purposes of comparison, and in order to obtain an empirical shape for a proton evaporation peak, additional measurements have been made on the more neutron deficient target ^{112}Sn . Simultaneously some of the measurements from the heavy isotope targets were repeated as a consistency check.

A variety of statistical model calculations have been done in an effort to interpret the heavy isotope data. The size of the

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T_z evaporation peak was estimated in an isospin and angular momentum dependent calculation of partial level widths. Three common sets of level density parameters were used in an effort to estimate the uncertainty in the results.

The extended Griffin model was modified to include the effects of isospin conservation in an approximate way, and T_z preequilibrium components were calculated using a recent matrix element prescription¹⁾ as well as the usual values of $[p_0=2, h_0=1$ and $g=(A/13) \text{ MeV}^{-1}]$ particle level density. These preequilibrium components are almost identical to those resulting from non-isospin dependent calculations and can account reasonably well for the high energy emitted protons not populating the strong 2^+ and 3^- collective states in the target nucleus. However, even with the inclusion of the largest possible T_z evaporation component, a large number of the low energy protons remain unaccounted for. These protons are attributed to decay of the T_z composite nucleus states prior to isospin mixing. This interpretation is supported by work of Cohen et al²⁾ in Pittsburgh who have compared (p,p') and (d,d') spectra for reactions on ^{124}Sn .

The preequilibrium model calculations for the T_z states in the composite system predict that, in the absence of isospin mixing, virtually all the available cross section will go into proton emission before statistical equilibrium is achieved. They lead to T_z preequilibrium component spectra which are dominated by low energy protons emitted in the middle and late stages of the equilibration process. These components have many of the features of an evaporation spectrum, so it is not surprising that one can

account for the data almost as well by assuming pure equilibrium as pure preequilibrium emission from the T_2 states. Zero free parameter preequilibrium model fits to the 14-MeV, 60° data are shown in Fig. 13. There are indications, however, that neither set of T_2 calculations predict accurate spectral shapes. The problem arises because as equilibrium is approached the rate for emitting low energy protons increases and eventually can become comparable to or greater than the rate for the internal two-body interactions. Thus, the internal transitions cannot occur quickly enough to maintain the assumed equal population of all n-exciton states. The calculated spectral shapes depend on this assumption and will become of doubtful significance for low energy particles emitted from the more complex states.

Unfortunately to get around this problem or indeed even to estimate its severity, it is necessary to have more detailed knowledge of the number and structure of n-exciton states with $T=T_2$ as well as the fraction of such states which are unbound toward emission of a particular energy of proton. This problem is currently under investigation but appears to be of greater complexity than was first imagined.

In summary, it appears that isospin selection rules play an important role in (p,p') reactions on the heavy tin isotopes. The observed high energy protons appear to be the result of strong collective excitations and of statistical preequilibrium emission from the T_2 composite states. The low energy particles are apparently due primarily to decay of the T_2 states prior to isospin mixing and there is some evidence that this decay, too, may be

Fig. 13 Experimental and calculated proton spectra from the $^{118, 120, 124}\text{Sn}$ (p,p') reactions. The calculated spectra include preequilibrium components from both $T_{<}$ and $T_{>}$ states.

Figure 13

largely preequilibrium. If this is true, it raises serious questions about the validity of existing statistical model calculations for emission of low energy protons from fairly complex T_0 states. These questions can only be answered when better knowledge of the T_0 states is obtained.

This experimental work is currently being written up for publication.

1) See Section B8 in this report ("Effective Matrix Elements for Use in the Extended Griffin Model", C. Kalbach-Cline).

2) B. L. Cohen, G. R. Ras, J. H. Degan and K. C. Chan, University of Pittsburgh preprint (1972) and B. L. Cohen, private communication.

7) Charged Particle Spectra from Reactions Induced by 59-MeV

α -Particles

[†] F. E. Bertrand, [†] R. W. Peele and C. Kalbach-Cline

The energy spectra for protons, deuterons, tritons, and alpha particles emitted in reactions induced by 59-MeV α particles incident on targets of ^{12}C , ^{16}O and ^{54}Fe have been measured using the Oak Ridge Isochronous cyclotron. The experimental technique and the measured spectra have already been reported by the first two authors¹⁾. This data is currently being written up for publication and to this end has been submitted to statistical model analysis.

Angle integrated spectra have been constructed for the ^{12}C and ^{54}Fe data. These have been analyzed by a method similar to the one used for studying complex particle emission in proton induced reactions²⁾ except that the preequilibrium part of the calculations could be done on an absolute basis by using the recent prescription³⁾ for M^2 . A 4p-0h initial configuration was assumed.

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The Fe results are of comparable quality to results for proton induced reactions at similar energies. That is, most of the particles are ascribed to preequilibrium emission; the relative intensities of the different particle types are reasonably well accounted for, the spectral shapes less well so; and the number of high energy alpha particles is grossly underestimated. This last point is almost certainly due to the experimental importance of knock-on α clusters. Further evidence for this can be seen by comparing the (p,p') and (α , α') spectra for the ^{54}Fe target.

The results for ^{12}C are quite a bit different. Nuclei as light as ^{12}C (or ^{16}O , the composite nucleus,) are not usually amenable to statistical analysis, so it is hardly surprising that the present calculations fail to reproduce the relative intensities of the different types of emitted particles. In the figure, the calculated preequilibrium components have been multiplied by the factors indicated next to the particle-type designation. What is surprising is the relatively good agreement in spectral shapes. This may result from the fact that much of the preequilibrium strength comes from the simplest possible states (4p-0h for p,d, and t; 5p-1h for ^4He). The weighting of contributions from different exciton numbers is less important here than for heavier nuclei. Again, it seems probable that most of the observed cross section involves preequilibrium emission.

1) F. E. Bertrand and R. W. Peele, Oak Ridge National Laboratory Report ORNL-4694 (1971).

- 2) C. K. Cline, Nucl. Phys. A193 (1972) 417.
- 3) See section B8 in this report ("Effective Matrix Elements for use in the Extended Griffin Model," C. Kalbach-Cline).
- 8) Effective Matrix Elements for use in the Extended Griffin Model
C. Kalbach-Cline

In the Griffin model, the rate expressions for the internal transitions involving exciton-exciton scattering ($\Delta h = \Delta p = 0$) or creation or destruction of a particle-hole pair ($\Delta p = \Delta h = \pm 1$) are taken from time dependent perturbation theory. They have the form

$$\lambda_{\Delta n}(p, h, E) = \frac{2}{\pi} M^2 \omega_{\Delta n}(p, h, E)$$

where $\omega_{\Delta n}$ is the density of states accessible in the interaction. Thus it is assumed that the average matrix element M is related to only a small residual part of the two-body interaction, with the rest of the interaction having gone into the potential well.

Without some knowledge of the value for M^2 , the model cannot be used to calculate absolute cross sections. However, until recently M^2 has generally been left as an unknown quantity in performing model calculations. The apparent reason is that M^2 is an effective as well as a residual matrix element. It depends on the details of the model formulation, and it is thus difficult to evaluate from first principles.

In the present work empirical values for M^2 have been obtained as a function of mass number and excitation energy. This has been done by using experimental particle energy spectra to obtain

empirical estimates of the branching ratio for preequilibrium nucleon emission in the first step of the reaction. The empirical branching ratios were then equated with theoretical ones containing the unknown quantity M^2 . The results of this analysis lead to systematic trends for the square of the average matrix element which can be summarized as

$$M^2 = K_a A^{-3} E^{-1}$$

$$K_p = 95 \text{ MeV}^3$$

$$K_\alpha = 725 \text{ MeV}^3$$

where the subscript a on the proportionality constant refers to the projectile, and A and E are the mass number and excitation energy of the composite system.

The observed mass number and energy dependence are consistent with expectations based on nuclear matter calculations. However, the M^2 values lead to interaction rates which are consistently smaller than the nuclear matter rates of Blann and Mignerey^{1,2)} by a factor of about 15 for incident protons and 2-3 for incident alpha particles. The present values of M^2 are consistent with those found by Braga-Marcazzan et al³⁾ for (n,p) reactions at about 20 MeV of excitation when it is assumed that $K_p \approx K_n$. Braga-Marcazzan et al are also using a modified version of Griffin's model while Blann and Mignerey are using the Hybrid Model. This may account for some of the discrepancy.

If the matrix elements from the present work are extrapolated to 1 MeV of excitation, they are found to represent about 2 to 5% of the full two-body interaction. This last quantity has been estimated from the centroid energies of two-particle weak-coupling multiplets of states in nuclei near doubly closed shells. This

observation is consistent with the use of perturbation theory in the extended Griffin model.

C) Direct Reaction Studies on Heavy Deformed Nuclei

1) Collective States Excited in the $^{234}\text{U}(d,d')$ and $^{236}\text{U}(d,d')$

Reactions

J. S. Boyno*, Th. W. Elze**, J. R. Huizenga and C. E. Bemis, Jr.***

Inelastic scattering of 16-MeV deuterons has been used to excite collective states in ^{234}U and ^{236}U . As reported previously¹⁾ the (d,d') reaction is especially well suited for the study of such states in the actinide nuclei. Spectra of high resolution, which allow identification of collective levels below 2 MeV of excitation are readily obtained. In addition, relative reduced transition probabilities, B(E2) and B(E3), may be extracted for strongly excited states since there is a direct proportionality between the B(E λ) value and the differential cross section.

In this work, thin targets of highly enriched ^{234}U and ^{236}U were bombarded with 16-MeV deuterons obtained from the Rochester tandem. The spectra were analyzed at 90° and 125° in the Enge spectrograph and recorded on emulsion plates. Three exposures, each of different length, were obtained at each angle in order to excite smaller peaks to a measurable intensity. Our lower limit of detection was $\approx 2\mu\text{b}$. Intensity and position matching insured proper energy and intensity relationships. Absolute normalization was accomplished by matching the ground state 0⁺ with

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an elastic scattering cross section calculated with a DWBA code. A typical spectrum, that of the $^{236}\text{U}(d,d')$ reaction at $\theta_L=125^\circ$ is shown in fig. 14. The $^{234}\text{U}(d,d')$ spectra were contaminated with a significant amount of W impurity; however, the energies of the impurity peaks are well known, and these lines are easily removed from consideration.

The spin-parity assignments for ^{234}U shown in table 4 are based primarily on a comparison of energy and intensity patterns found in this nucleus with those obtained in inelastic deuteron work on other actinide nuclei¹⁾. In addition, the large body of previous work on this nucleus²⁾ has yielded many strong assignments in ^{234}U . These previous assignments are a good test of our empirical spin-parity assignments. In addition, they often allow us to adopt K-values for bands in cases where this cannot be done on the basis of inelastic scattering data alone.

An interesting aspect of the ^{234}U spectroscopy is the large number of negative parity states excited. As seen in table 4, we assign, at least tentatively, members of five different negative parity bands, including two $K^\pi=0^-$ bands. Theoretical predictions³⁾ are that there should be just four negative parity bands in this region, namely, the octupole quadruplet. The existence of a second, strongly collective $K^\pi=0^-$ band is an unexplained anomaly.

The levels in ^{236}U , while less well established by previous work⁴⁾, again are identified by comparison with the other nuclei studied¹⁾, especially the comparison with levels in ^{234}U and ^{238}U . The spectroscopic results are in table 5.

Figure 14 Spectrum of deuterons scattered from ^{236}U . The spin and parity assignments are given. The first number is the K assignment.

Table 4
Levels Excited in ^{234}U

Energy (keV)	$\left(\frac{d\sigma}{d\Omega}\right)^{90^\circ}$ μbsr^{-1}	$\left(\frac{d\sigma}{d\Omega}\right)^{125^\circ}$ μbsr^{-1}	$R = \frac{d\sigma^{90^\circ}}{d\Omega^{125^\circ}}$	Assignment $K, I^{\pi a)}$	
				This work	Previous
0	44336	9310	4.76	$0, 0^+$	$0, 0^+$
43	6171	2434	2.54	$0, 2^+$	$0, 2^+$
144	482	324	1.49	$0, 4^+$	$0, 4^+$
295	40	50	0.8	$0, 6^+$	$0, 6^+$
504	---	6	---	$0, 8^+$	$0, 8^+$
787	9	28	0.3	$0, 1^-$	$0, 1^-$
810	12	6	2	$0, 0^+\beta$	$0, 0^+\beta$
850	178	104	1.71	$0, 3^-$	$(0, 3^-) +$ $(0, 2^+\beta)$
927	67	31	2.2	$2, 2^+\gamma$	$2, 2^+\gamma$
964	10	15	0.7	$0, 5^-$	$(0, 5^-)$
1023	109	71	1.5	$2, 3^-$	$(2, 3^-) +$ $(2, 4^+)$
1126	10	14	0.7	$2, 5^-$	$(2, 5^-) +$ $(2, 2^+)$
1150	---	3	---	---	---
1174	---	3	---	---	---
1218	(5)	(8)	(0.6)	---	---
1238	obscured	6	---	$(0), 1^-$	$(0, 1^-)$
1278	obscured	4	---	$(2, 7^-)$	---
1312	105	58	1.8	$(0), 3^-$	---
1339	---	4	---	---	---
1446	11	12	0.9	$(0), 5^-$	---
1486	10	9	1.1	$(1, 3^-)$	$(1, 3^-)$
1555	---	3	---	---	(1^+)
1586	5	7	0.7	$(1, 5^-)$	$(1, 5^-)$
1652	15	12	1.2	---	$(6, 9^-) +$ $(1, 6^-)$
1675	7	3	2	---	---

Table 4 - Continued

Energy (keV)	$(\frac{d\sigma}{d\Omega})_{90^\circ}$ μbsr^{-1}	$(\frac{d\sigma}{d\Omega})_{125^\circ}$ μbsr^{-1}	$R = \frac{d\sigma_{90^\circ}}{d\sigma_{125^\circ}}$	Assignment K, I^π ^{a)}	
				This work	Previous
1696	6	---	---	---	---
1721	12	9	1.3	(3, 3 ⁻)	(3, 3 ⁻)
1786	11	8	1.4	---	(4, 5 ⁺)
1863	17	17	1.0	---	---

Cross section errors are $\pm 10\%$ $d\sigma/d\Omega \leq 100 \mu\text{b}$, $\pm 5\%$ $d\sigma/d\Omega > 100 \mu\text{b}$.

Energies are ± 2 keV; quantities enclosed in parentheses are considered uncertain.

a) Certain K-values adopted, see text for details.

Table 5
Levels Excited in ^{236}U

Energy (keV)	90°		$R = \frac{d\sigma}{d\Omega} \frac{90^\circ}{125^\circ}$	Assignment K, I^π ^{a)}	
	$(\frac{d\sigma}{d\Omega})_{\mu\text{bsr}}^{-1}$	$(\frac{d\sigma}{d\Omega})_{\mu\text{bsr}}^{-1}$		This work	Previous
0	44489	9322	4.77	$0, 0^+$	$0, 0^+$
44	6434	2718	2.37	$0, 2^+$	$0, 2^+$
149	588	270	2.18	$0, 4^+$	$0, 4^+$
310	42	51	0.8	$0, 6^+$	$0, 6^+$
524	---	3	---	$0, 8^+$	$0, 8^+$
686	12	30	0.4	$0, 1^-$	$2, 2^-$
746	216	89	2.4	$0, 3^-$	---
848	18	23	0.8	$0, 5^-$	---
919	---	3	---	$0, 0^+ \beta$	$0, 0^+ \beta$
959	104	42	2.5	$2, 2^+ \gamma$	$(0, 2^+ \beta)$
1002	---	4	---	$(0, 7^-)$	---
1037	95	53	1.8	$(2), 3^-$	$1, 3^-$
1060	25	15	1.7	---	---
1150	66	44	1.5	$-, (3^-)$	---
1240	---	5	---	---	---
1262	---	13	---	---	---
1333	---	6	---	---	---

Cross section errors are $\pm 10\%$ $d\sigma/d\Omega \leq 100 \mu\text{b}$, $\pm 5\%$ $d\sigma/d\Omega > 100 \mu\text{b}$.
Energies are ± 2 keV; quantities enclosed in parentheses are considered uncertain.

a) Certain K-values adopted, see text for details.

Energy and intensity patterns of the groups at 686, 746, and 848 keV strongly resemble those of the 1^- , 3^- and 5^- members of the $K^\pi = 0^-$ octupole bands seen in other actinide nuclei. From rotational systematics, we also tentatively identify the $I^\pi = 7^-$ member of this band. Previous work⁵⁾ led to a $K, I^\pi = 2, 2^-$ assignment for a level at 688 keV in ^{236}U . Our evidence makes this assignment highly unlikely although a $0, 1^-$ assignment is seemingly incompatible with the multipolarities of various transitions seen in gamma decay. Our arguments in this case are explained in detail elsewhere.⁶⁾

There is a direct relationship between the differential cross section for inelastic scattering and the reduced transition probability for a given state. The cross section as calculated in a macroscopic DWBA theory may be written

$$\left(\frac{d\sigma}{d\Omega}\right)_{0^+ \rightarrow J=\lambda} = N B_\lambda^2 \sigma_\lambda(\Theta, Q) \quad (1)$$

The normalization constant is known and the σ_λ is calculated with the code DWUCK. Since the reduced transition probability $B(E\lambda)$ for an $E\lambda$ transition from the ground state may be written

$$B(E\lambda) = \left[\frac{3}{4} Z e R_0^\lambda A^{\lambda/3}\right]^2 \beta_\lambda^2, \quad (2)$$

the β_λ extracted from eq. 1 may be used to calculate the $B(E\lambda)$. In practice, the use of the DWBA limits the $B(E\lambda)$ values extracted from (d, d') cross sections to relative values, which require normalization before they are compared to Coulomb excitation values. The normalization factors obtained in a similar calculation for ^{238}U ¹⁾ have been used throughout the actinide series without

change, and they yield excellent agreement here as well, as seen in tables 6 and 7.

The spectroscopy of actinide nuclei from the (d,d') reaction is quite straightforward. Identifications based on the systematics previously developed in this region indicate that the behavior of most excitations is exceptionally stable. Coupled channels calculations would undoubtedly be of great use in the cross section estimates, perhaps making the extraction of absolute $B(E\lambda)$ from inelastic scattering data quite feasible.

- 1) Th. W. Elze and J. R. Huizenga, Nucl. Phys. A187, 545 (1972).
- 2) Nuclear Data B4 (Academic Press, New York), 581 (1970).
- 3) V. G. Soloviev and T. Siklos, Nucl. Phys. 59, 145 (1964);
K. Neergard and P. Vogel, Nucl. Phys. A149, 609 (1970).
- 4) Nuclear Data B4 (Academic Press, New York) 623 (1970).
- 5) C. M. Lederer, University of California, LRL Report UCRL-10028 (PhD thesis) 1963.
- 6) J. S. Boyno, Th. W. Elze, J. R. Huizenga and C. E. Bemis, Jr., to be published.

TABLE 6

Relative $B(E^2; 0^+ \rightarrow 2^+)/e^2 \cdot 10^{-48} \text{ cm}^4$

Nucleus	E (keV)	Band	$(d, d')_{\text{Av.}}$	Coul. Ex.
^{234}U	43	Rot	11.8	10.0 ^{a)}
^{234}U	927	Gamma	0.14	--
^{236}U	44	Rot	12.2	12.1 ^{a)}
^{236}U	959	Gamma	0.20	0.26 ^{b)}

a) Refs. 3,4

b) F. E. Durham, et al in Proc. Int. Conf. Nucl. Struct., ed. D. A. Bromley and E. W. Vogt (University of Toronto Press, Toronto, 1960), using

$$B(E2)_{\text{s.p.}} \approx 2.95 \cdot 10^{-53} A^{4/3} e^2 \text{ cm}^4$$

TABLE 7

Relative $B(E3; 0^+ \rightarrow 3^-) / e^2 \cdot 10^{-72} \text{cm}^6$

Nucleus	E(keV)	$(d, d')_{Av.}$	Theory ^{a)}
^{234}U	850	0.65	0.56
^{234}U	1023	0.42	0.29
^{234}U	1312	0.37	--
^{234}U	1486	0.04	0.002
^{234}U	1721	0.05	0.11
^{236}U	746	0.70	0.50
^{236}U	1037	0.34	0.29
^{236}U	1150	0.26	--

a) Ref. 3

-75-80 4249

2) Single Proton States in ^{249}Bk

J. S. Boyno, H. Freiesleben, J. R. Huizenga, C. Kalbach-Cline, C. E. Bemis* and R. W. Hoff†

A study of the $^{248}\text{Cm}(^3\text{He},d)$ and $^{248}\text{Cm}(^4\text{He},t)$ reactions has been instituted at Rochester. This data is being combined with the results of studies of the γ -decay of ^{253}Es and the β -decay of ^{249}Cm in order to obtain a more complete picture of the nucleus ^{249}Bk and in particular of its single proton states. Of special interest is the energy of the expected $[521\frac{1}{2}^+]$ proton states which should be of primarily $f_{5/2}$ character and should therefore, in the spherical limit, originate above the $2=114$ shell closure. Its identification would provide information on the size of the shell gap.

The $(^3\text{He},d)$ reaction has been studied at an energy of 28.5 MeV on two targets prepared in different ways at Oak Ridge and at Livermore and containing different impurities. Deuteron spectra for an emission angle of 60° were recorded in photographic emulsions using the Enge split pole spectrograph. The spectra are shown in figure 15. The poor statistics are due to the low cross sections and the limited time that photographic plates can be left in vacuum. The $(^4\text{He},t)$ reaction was performed at the Oak Ridge Isochronous Cyclotron at 40 MeV using emulsions in a Brown-Buchner spectrograph; and at Rochester at 29.4 MeV in the Enge split-pole. The $(^4\text{He},t)$ data clearly confirms many of the assignments based on the $(^3\text{He},d)$ results but is not yet completely analyzed.

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Figure 15. Two spectra from the $^{248}\text{Cm}(^3\text{He},d)^{249}\text{Bk}$ reaction.

Figure 15

States in ^{249}Bk have been identified by comparisons of observed energies with those of states known from α - and β -decay studies, and through predicted intensity and rotational energy spacing patterns within a band. DWBA calculations for the $(^3\text{He},d)$ reaction were performed both with and without finite range and spin orbit effects. The more complete calculations yielded better agreement with experiment. Theoretical spectroscopic factors were initially obtained from a Nilsson model program using the deformation parameters $\epsilon_2=0.25$ and $\epsilon_4=-0.02$. Later, in order to obtain reasonable agreement with experiment, it was found necessary to include pairing effects and multi-band Coriolis mixing. Finally, we have examined the predictions of Gareev et al.¹⁾ for the $[521+]$, the $[400+]$ and $[660+]$ bands. These predictions include the effects of the coupling between collective and single particle degrees of freedom.

Figure 16 shows the known single proton bands in ^{249}Bk , with the heavy lines indicating states identified in this work. The $1/2^-$ band labeled $[521+]$ is predicted by Gareev et al to contain only 22% of the $[521+]$ single particle strength while the $1/2^+$ state at 379 keV is part of a band expected to contain a mixture of the $[400+]$ and $[660+]$ single particle configurations. Both predictions are in agreement with the present results. Our results are also consistent with the recent single proton transfer data of Erskine and Kyle²⁾.

Analysis and interpretation of the current results is proceeding.

Figure 16. Single proton bands in ^{249}Bk .
The heavy lines indicate levels
excited by the $^{248}\text{Cm}(^3\text{He},d)^{249}\text{Bk}$
reaction.

^{249}Bk

Figure 16

- 1) F. A. Gareev, S. P. Ivanova, L. A. Malov and V. G. Soloviev, Nucl. Phys. A171, 134 (1971).
- 2) J. R. Erskine and G. Kyle, Bull. Am. Phys. Soc. 17, 928 (1972).
- 3) Levels of ^{233}Pa Excited by the $^{232}\text{Th}(^3\text{He},d)$ and $^{232}\text{Th}(\alpha,t)$ Reactions
Th. W. Elze[†] and J. R. Huizenga

The $(^3\text{He},d)$ and (α,t) reactions on ^{232}Th targets had previously¹⁾ been studied. Based on the spectroscopic factors extracted from both reactions, levels belonging to the $1/2^- [530]$ and $3.2^- [521]$ rotational bands were identified. However, a considerable number of levels below 600 keV could not be interpreted. The major difficulties in the data analysis stemmed from both poor resolution of the $(^3\text{He},d)$ spectrum and considerable Coriolis mixing between low-lying states which strongly affect the spectroscopic factors. We have therefore rerun the $^{232}\text{Th}(^3\text{He},d)^{233}\text{Pa}$ reaction with somewhat improved resolution (≈ 20 keV FWHM) and better statistics. The spectrum of emergent deuterons as measured in an Enge type magnetic spectrograph is shown in fig. 17. At present, an attempt is made to interpret most of the levels below 600 keV as being members of the $1/2^+ [660]$, $3/2^+ [651]$ and $5/2^+ [642]$ rotational bands with strong Coriolis interaction between them. The possibility of $\Delta N=2$ mixing between the $1/2^+ [660]$ and $1/2^+ [400]$ configurations is also being considered.

- 1) Th. W. Elze and J. R. Huizenga, Contribution 8.5, International Conf. on Properties of Nuclear States, Montreal 1969.

[†] Present address: Institut für Kernphysik, Universität Frankfurt, Germany.

Figure 17. Spectrum of deuterons from the
 $^{232}\text{Th}(^3\text{He},d)^{233}\text{Pa}$ Reaction.

Figure 17

4) A Study of Levels in ^{239}Np via Proton Transfer Reactions

T. von Egidy[†], Th. W. Elze^{††} and J. R. Huizenga

The $^{238}\text{U}(^3\text{He},d)$ and $^{238}\text{U}(\alpha,t)$ reactions have been performed in order to study Nilsson proton levels in ^{239}Np . The $(^3\text{He},d)$ reaction was run in 1968 at angles of 20° , 35° , 60° and 90° with a ^3He beam energy of 30 MeV. This past year the $(^3\text{He},d)$ and (α,t) reactions were performed at 60° with beam energies of 28.5 MeV. In all cases the reaction products were detected on photographic emulsions in an Enge split-pole magnetic spectrograph. The deuteron and triton peak widths (FWHM) are 16 and 14 keV, respectively. Rotational bands built upon the following Nilsson proton configurations have been identified; $5/2+[642]$, $5/2-[523]$, $1/2-[530]$ and $3/2-[521]$. Further analysis of the data is in progress in order to identify additional strong lines in the spectra.

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5) Collective States of ^{248}Cm Excited by Inelastic Deuteron Scattering

Th. W. Elze[†] and J. R. Huizenga

The inelastic scattering of 16-MeV deuterons from a ^{248}Cm target has been studied at $\theta = 90^\circ$ and 125° . The spectra of scattered deuterons were measured in an Enge magnetic spectrograph with an overall energy resolution of approximately 10 keV

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Figure 18. Spectrum of deuterons scattered from
 ^{248}Cm .

Figure 18

(cf. fig.18). Excited by the reaction is the ground-state rotational band up to its 8^+ member, as well as many levels of a mainly collective nature above 1 MeV. Among the latter strongly excited states are probably the γ vibrational band and one or more octupole bands. We are presently attempting to identify some of them. The intensity pattern of the octupole bands in ^{248}Cm may be slightly different from that reported for other actinide nuclei^{1,2)}.

1)

Th. W. Elze and J. R. Huizenga, Nucl. Phys. A187, 545 (1972).

2)

J. S. Boyno, Th. W. Elze, J. R. Huizenga and C. E. Bemis, Jr., to be published

6) Study of the (p,t) Reactions on Targets of $^{188,190,192}\text{Os}$

Th. W. Elze[†], D. G. Burke^{††}, J. S. Boyno^{†††} and J. R. Huizenga

The (p,t) reactions have been studied with 18-MeV protons incident on isotope separated targets of ^{188}Os , ^{190}Os and ^{192}Os . Angular distributions of the emergent tritons were measured at 12 angles between 10° and 65° with emulsion plates located in the focal plane of an Enge split-pole magnetic spectrograph. The relative cross sections for the ground state transitions measured at 25° and 30° with a natural Os target are $1:0.98\pm0.02:0.72\pm0.05$ for the targets 192:190:188. The angular distributions for the g.s. transitions have a deep minimum at 40° . The relative σ for the $\ell=0$ transitions populating states in ^{190}Os are 1(g.s.), 0.04

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(911 keV), 0.01 (1540 keV) and 0.05 (1731 keV).

D. Nuclear Methods in Chemical Structure

1) Effect of Pressure on the Decay Constant of ${}^7\text{Be}^*$

W. K. Hensley, J. R. Huizenga and W. A. Bassett[†]

Twenty-five years have passed since the independent suggestions of Segre¹ and Daudel² that the decay constants of certain radioactive nuclides might be altered by varying the electron density in the vicinity of the nucleus. Since that time, several groups have placed nuclides that decay by electron capture and internal conversion in different chemical and physical environments in order to observe these effects.³ The two most frequently used nuclides for these studies have been ${}^7\text{Be}$ and ${}^{99\text{m}}\text{Tc}$.

The first attempts to observe the effect of high pressure on the half-life of a nuclide were carried out by comparing the measured activities before and after compression of a radioactive sample. Bainbridge⁴ observed that the difference in the decay rate between ${}^{99\text{m}}\text{Tc}$ compressed to 100 kbars and ${}^{99\text{m}}\text{Tc}$ at one bar was about 0.02%. A few years later Gogarty et al⁵ used a similar technique to measure the effect of pressure on the decay rates of ${}^7\text{Be}$ and ${}^{131}\text{Ba}$. The technique of measuring the effect while the sample remained at a high pressure was first used by Cooper⁶ to measure the effect of pressure on the decay of ${}^{90\text{m}}\text{Nb}$ and again recently by Mazaki et al⁷ who repeated the ${}^{99\text{m}}\text{Tc}$ measurement.

Recent advances⁸ in the field of high pressure technology have eliminated the need for the large hydraulic presses used

* This work also supported in part by the National Science Foundation.

[†]Department of Geological Sciences and Space Science Center, Univ. of Roch

in previous studies.⁴⁻⁷ In this communication we wish to report results on the change in the decay constant of ^7Be in BeO at pressures up to 270 kbars obtained on a small sample in a diamond anvil press. Diamond anvil presses have been used in increasing numbers principally in the fields of high pressure physics and geology for over a decade. One of the reasons for their gaining popularity as a research tool is that they have a number of distinct advantages over conventional hydraulic presses. Most of these advantages stem from the small overall size and the simplicity in the basic design. They require no external support to the system to sustain the pressure after it has been applied to a sample. Another feature is that the pressure of the sample can be determined internally without disturbing the sample or altering the pressure.

In fig. 19 is shown a schematic diagram of the type of press used in this work. It is a simple piston and screw device constructed from stainless steel. The body of the press is basically cylindrical, one inch in diameter, and two inches long. The anvils are 25 mg. (1/8 carat) brilliant cut single-crystal diamonds which have had their culet faces enlarged to 0.3 and 0.4 mm. diameter on the upper and lower anvils, respectively. In order to permit the alignment⁸ of the anvil faces parallel to each other, the diamonds are cemented to a pair of rocker joints that have their axes of rotation at right angles to each other. In addition, each rocker joint is able to move laterally in a trough in the piston. This allows

Figure 19. Schematic diagram of the
diamond anvil press.

Figure 19

complete translational and angular adjustment of the anvil faces. The pistons have a flat side ground onto them in order to prevent rotation of the anvil. Compression of the sample is accomplished by tightening the loading screws. The dashed lines in the loading screws, pistons, and rocker joints indicate holes through which the sample can be observed while under pressure.

Samples of BeO were prepared in the conventional radiochemical manner⁹ from the carrier-free isotope in 0.5N HCl¹⁰. The pressure samples were made by first compressing a layer of NaCl between the diamond faces. Then the piston with the smaller anvil face was removed from the press and a piece of the ⁷BeO in the shape of a thin platelet about 0.1 mm. diameter transferred to the center of the large anvil. The piston was then reinserted into the press, and the press loaded to its final pressure.

The purpose of including a layer of NaCl between the anvils of the press is two-fold. First, as a pressure-transmitting medium; that is, a substance which surrounds the sample and provides a uniform pressure on the entire sample. This results in part from the high compressibility and low shear strength of the NaCl. The second function that the NaCl serves is as an internal pressure standard. Because of the design of the press, it is possible to use standard X-ray diffraction techniques to measure the pressure between the anvils.

A slightly modified 114.59 mm. diameter Debye-Scherrer Powder camera was used for this work. The geometry of the press and camera is such that the X-rays passed through the

sample perpendicular to the long axis of the press and thus across the pressure axis. As a result, the X-ray record which is obtained contains information about the pressure over the entire sample area. The diffraction pattern produced by the part of the sample at the edge of the anvil, at one bar pressure, enables one to calculate V_0 , the unit cell volume of the uncompressed sample.

In fig. 20 is shown a portion of two films that were exposed in the manner described above. The patterns, which are both from NaCl, were obtained from samples which were loaded to pressures of 120 and 270 kbars in the upper and lower patterns, respectively. The strongest line in each pattern is from the 200 plane of the high pressure portion of the sample. The line from the 200 plane at 1 bar pressure is seen to be inside the corresponding 200 line at high pressure and clearly separated from it. This is because the pressure gradient is concentrated near the edges of the anvils where the sample is very thin and as a result, there is not enough material between the two pressure extremes to provide the intensity necessary to produce a broad band. The spots on the patterns are Laue reflections from the single crystal diamond anvils.

To calculate the pressure, the isothermal compressibility of NaCl given by Weaver et al¹¹ was used. The procedure was to measure the diffraction pattern and then with Bragg's Law to determine the lattice parameter of the NaCl unit cell. From this one calculates V/V_0 , the ratio of the compressed volume to the volume at 1 bar, and then one determines the sample pressure from the Weaver equation.

Figure 20. X-ray diffraction photographs obtained from ${}^7\text{BeO-NaCl}$ samples compressed to 120 kbar (A) and 270 kbar (B). Lines from the 200 and 220 planes of NaCl were produced using filtered copper radiation. Lines 1 and 3 are reflections from the portion of the sample at 1 bar; lines 2 and 4 were produced from the high pressure portion of the sample.

Figure 20

After preparation and preliminary pressure determination, the samples were fixed to special mounts for counting. The 477-keV gamma ray was used to monitor the decay of ^7Be . The samples were counted with Ge(Li) detectors¹², which were connected through an F.E.T. preamplifier and a linear amplifier to a 4096 channel analyzer with computer compatible output. After the raw data were acquired they were stored on magnetic tape, and the activities calculated later.

The method of integrating over a constant energy region bracketing the photopeak was used to determine the activity of the counted sample. We chose the energy region between 455 and 485 keV to represent the total activity. After the activities were calculated, they were used as input to a standard least-squares code which fits an exponential decay curve to the data to determine the value of the decay constant for each sample. Values of $(\lambda_c - \lambda)/\lambda$ were calculated directly from the fitted decay constants, where λ_c and λ are the decay constants that were measured for the compressed and uncompressed samples, respectively. This method of analysis was chosen because it is insensitive to efficiency changes in the gamma-ray detection system. In order to provide a set of reference standards for the system, samples of BeO (1 bar) were counted along with the pressurized samples. In addition, two of the experiments contained a pair of identical chemical samples which provided a null measurement to check the system's electronic and geometric stability. Activities were measured on the average every other day for a period of three half-lives, about six months.

There are several possible sources of error in the calculations of $\Delta\lambda/\lambda$. Statistical fluctuations in each of the measured activities of the sample are of the order of $\pm 0.1\%$. Another possible source of error is in the pressure measurements where the uncertainty is about ± 10 kbar.

In the two cases where identical chemical samples were counted, in addition to the compressed sample, the $\Delta\lambda/\lambda$ values were found to be less than 0.0004 ± 0.0004 . This value is considerably smaller than the values of $\Delta\lambda/\lambda$ measured for the compressed and uncompressed samples.

The experimental results obtained from measurements of $\Delta\lambda/\lambda$ at 120, 210, and 270 kbar are shown in fig. 21, where a least-squares curve fit to the equation $(\lambda_c - \lambda)/\lambda = K_p P$ is shown. The least squares value obtained for K_p is 0.000022 ± 0.00001 kbar⁻¹ when P is in units of kbars. The K_p value based on the data of Gogarty et al⁵ for the compression of ${}^7\text{BeCO}_3 \cdot \text{Be}(\text{OH})_2$ is calculated by us to be 0.00002 ± 0.00003 kbar⁻¹ and agrees within experimental error with our value of K_p given above. Diffraction patterns produced when an X-ray beam traverses the entire pressure range in a single sample of pure BeO provide evidence that no phase transition occurs. The maximum pressure in this sample calculated from the measured lattice parameters is 300 kbar or 240 kbar, based on the Birch equation with Anderson's elastic constants¹³ or the shock compression data of Cline and Stephens,¹⁴ respectively.

The increase in the decay constant of ${}^7\text{BeO}$ with pressure has been estimated with the electron wave functions of the free Be atom¹⁵. In our model calculation the fractional amount of electron density in the outermost portion of the Be atom (a

Figure 21. Fractional increase in the total decay constant of ^7Be in ^7BeO as a function of pressure where λ_C is the decay constant of the compressed sample. The error bars represent one standard deviation. The line is a least-squares fit of our data to the equation $(\lambda_C - \lambda) / \lambda = K P$. The data point of Gogarty et al⁵ is calculated from a least-squares fit to the above equation of some twenty experimental measurements taken near 100 kbars.

radius of $0.89\text{\AA}$ was used) containing 10% of the volume is redistributed inside the new volume after compression ($0.9 V_0$) with the original electron density distribution. This gives a 0.35% increase in the electron density at the nucleus for a free Be atom. We assume the same enhancement in λ of 0.0035 for BeO at a value of V/V_0 of 0.9, which corresponds to a pressure of 240 kbar when use is made of the equation of state of Cline and Stephens. The experimental values of $(\lambda_c - \lambda)/\lambda$ measured at 210 and 270 kbar are 0.0051 and 0.0059, respectively. By interpolation between these two points, one observes that the above calculation gives an enhancement in λ which is 2/3 of the experimental value. However, this calculation is based on several assumptions and more rigorous theoretical calculations need to be made. These are being pursued with more realistic models.

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III Publications and Activities

1. Articles

F. C. Williams, Jr., G. Chan and J. R. Huizenga, THE SIGNIFICANCE OF SHELL CORRECTIONS IN THE PARAMETERIZATION OF NUMERICAL STATE DENSITY CALCULATIONS, Nucl. Phys. A187, 225 (1972).

Th. W. Elze, J. S. Boyno and J. R. Huizenga, COLLECTIVE STATES OF Gd ISOTOPES FROM (p,t) REACTIONS, Nucl. Phys. A187, 473 (1972).

Th. W. Elze and J. R. Huizenga, COLLECTIVE STATES OF ^{232}Th , ^{238}U and ^{242}Pu , Nucl. Phys. A187, 545 (1972).

C. C. Lu, L. C. Vaz and J. R. Huizenga, COMPUTER PROGRAMS FOR STATISTICAL NUCLEAR DECAY, University of Rochester Report UR-NSRL-51, April (1972).

L. C. Vaz, C. C. Lu and J. R. Huizenga, STATISTICAL PROGRAM GAMAJ, University of Rochester Report UR-NSRL-54, May (1972).

C. C. Lu, L. C. Vaz and J. R. Huizenga, LEVEL DENSITIES FROM SPECTRA OF DIFFERENT REACTIONS POPULATING THE SAME NUCLEUS, Nucl. Phys. A190, 229 (1972).

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C. K. Cline, EXTENSIONS OF THE PREEQUILIBRIUM STATISTICAL MODEL AND A STUDY OF COMPLEX PARTICLE EMISSION, Nucl. Phys. A193, 417 (1972).

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C. K. Cline, Precom: PROGRAM FOR CALCULATING PREEQUILIBRIUM PARTICLE ENERGY SPECTRA, University of Rochester Report UR-NSRL-62 (1972).

J. R. Huizenga, H. Freiesleben, J. P. Unik and K. L. Wolf, INVESTIGATIONS OF HEAVY-ION REACTION MECHANISMS, Research Proposal for Use of the Super-Hilac.

J. R. Huizenga, C. Kalbach-Cline, Z. Sujkowski and J. R. Unik, NEGATIVE MUON INDUCED FISSION IN THE ACTINIDE ELEMENTS, Research proposal for use of Los Alamos Meson Physics Facility.

C. Kalbach-Cline, J. R. Huizenga and H. K. Vonach, ISOSPIN CONSERVATION AND PREEQUILIBRIUM DECAY IN HEAVY ISOTOPE $\text{Sn}(p,p')$ REACTIONS, Submitted to Nucl. Phys.

J. S. Boyno, J. R. Huizenga, Th. W. Elze and C. E. Bemis, Jr., LEVELS OF ^{234}U AND ^{236}U EXCITED BY THE (d,d') REACTION, Submitted to Nucl. Phys.

J. R. Huizenga and H. C. Britt, THRESHOLD PHOTOFISSION: THEORY AND EXPERIMENT, To be published in Proceedings of International Conference on Photonuclear Reactions and Applications held on March 26-30, 1973.

R. Vandenbosch and J. R. Huizenga, NUCLEAR FISSION, Textbook on the subject matter of nuclear fission in publication and to appear approximately June, 1973.

2. PhD Theses Completed

"The Significance of Shell Corrections in the Parameterization of Numerical State Density Calculations" by Grace Chan.

"The Effect of Pressure on the Radioactive Decay Rate of ^7Be " by W. K. Hensley.

3. Contributed Papers at Professional Meetings

J. S. Boyno, C. K. Cline, H. Freiesleben, J. R. Huizenga, C. E. Bemis, Jr., and R. W. Hoff, PROTON STATES IN ^{242}Bk , Bull. Am. Phys. Soc. 18, 38 (1973).

H. Freiesleben, G. T. Rizzo and J. R. Huizenga, ^6Li and ^7Li INDUCED REACTIONS ON ^{232}Th and ^{238}U , Bull. Am. Phys. Soc. 18, 97 (1973).

J. R. Huizenga, H. Freiesleben and G. T. Rizzo, EFFECT OF TARGET DEFORMATION ON HEAVY ION REACTION CROSS SECTIONS, Meeting of American Chemical Society Division of Nuclear Chemistry and Technology, Jan. 29-Feb. 1, (1973).

M. Kildir and J. R. Huizenga, DEPENDENCE OF THE NUCLEAR LEVEL WIDTH OF ^{32}S ON ISOSPIN, Bull. Am. Phys. Soc. 18, 668(1973).

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Th. W. Elze, D. G. Burke, J. R. Huizenga and J. S. Boyno, STUDY OF THE (p,t) REACTIONS ON TARGETS $^{188},^{190},^{192}\text{Os}$, Bull. Am. Phys. Soc. 18 700(1973).

W. K. Hensley, J. R. Huizenga and W. A. Bassett, EFFECT OF PRESSURE on the Radioactive Decay Constant of ^7Be , Meeting of American Geophysical Union, April 16-20, 1973.

4. Invited Lectures

"Dependence of the Nuclear Level Density on Single Particle Structure," J. R. Huizenga, Gordon Research Conference on Nuclear Chemistry, June 1972.

"Experimental and Theoretical Nuclear Level Densities," J. R. Huizenga, Democritus Institute Meeting on Physics, Athens, Greece, Nov. 1972.

"Effects of Pressure and Chemical Environment on the Radioactive Decay Constant of ^7Be ," W. K. Hensley, W. A. Bassett and J. R. Huizenga, 164th ACS Meeting, New York City, Aug.-Sept. 1972.

"Investigation of Isospin Conservation in Compound Reactions," C. Kalbach-Cline, Autumn School on Level Densities and Compound Processes, Schleching, Germany, Oct. 1972.

"Effect of Pressure on the Decay of ^7Be ," W. K. Hensley, Physical Chemistry Seminar, University of Rochester, October 30, 1972.

"Isospin Effects in Preequilibrium Emission and Evaporation from Compound Nucleus," C. Kalbach-Cline, Villars (Switzerland) Meeting on Nuclear Physics, Jan. 21-27 (1973).

" ^6Li and ^7Li Induced Reactions on ^{232}Th and ^{238}U ," H. Freiesleben, Brookhaven National Laboratory, Nov. 27, 1972.

"Threshold Photofission: Theory and Experiment," J. R. Huizenga, International Conference on Photonuclear Reactions and Applications, Asilomar Conference Grounds, Pacific Grove, Calif., March 26-30, 1973.

"Studies of Statistical and Direct Reactions," J. R. Huizenga, University of Maryland, April 27, 1973.

Professional Activities

Member of Editorial Board of the Journal of Physical Chemistry.

Member of Advisory Council of Princeton University's Chemistry Department.

Member of Review Committee of Argonne Physics Division.

Member of Executive Committee of Super-Hilac Users Association.

Member of Ad-Hoc Panel on Nuclear Data Compilation, NAS-NRC.

Chairman of Division of Nuclear Chemistry and Technology of American Chemical Society (1972).

Member of University of Rochester Senate.

IV. Personnel

Dr. J. R. Huizenga - Professor, Departments of Chemistry and Physics

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†† Supported in large part during Academic Year by a fellowship through the Department of Chemistry.