ARGONNE NATIONAL LABORATORY
9700 South Cass Avenue
Argonne, Illinois

PHYSICS DIVISION
SUMMARY REPORT

February-March 1962

Morton Hamermesh, Division Director

Preceding Summary Reports:

ANL-6432, September-October 1961
ANL-6455, November-December 1961
ANL-6488, January 1962

Operated by The University of Chicago

under

Contract W-31-109-eng-38

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

FOREWORD

The Summary Report of the Physics Division of the Argonne National Laboratory is issued monthly for the information of the members of the Division and a limited number of other persons interested in the progress of the work. Each active project reports about once in 3 months, on the average. Those not reported in a particular issue are listed separately in the Table of Contents with a reference to the last issue in which each appeared.

This is merely an informal progress report. The results and data therefore must be understood to be preliminary and tentative.

The issuance of these reports is not intended to constitute publication in any sense of the word. Final results either will be submitted for publication in regular professional journals or, in special cases, will be presented in ANL Topical Reports.

TABLE OF CONTENTS

The date of the last preceding report is indicated after the title of each project below. Projects that are not reported in this issue are listed on subsequent pages.

I. EXPERIMENTAL NUCLEAR PHYSICS

		<u>PAGE</u>
I-2-19	NEUTRON DETECTORS (ANL-6072, October-November 1959) L. M. Bollinger, R. T. Carpenter, and J. P. Marion	1
	A Monte Carlo calculation was performed to determine the distribution of lifetimes of neutrons in glass scintillation neutron detectors in cases where multiple scattering cannot be ignored. The probability of capture is also determined. The results are characterized by two dimensionless ratios so that the distributions and capture probabilities may be applied to a large class of capturing media.	
I-18-14	NEUTRON POLARIZATION AND DIFFERENTIAL CROSS SECTIONS (ANL-6455, November- December 1961) R. O. Lane and W. F. Miller	5
	The probabilities for single, double, and triple scattering of neutrons in thin plates is calculated by an efficient Monte Carlo technique which takes into account energy loss of the neutrons following elastic scattering. The method is applied to several nuclei including C ¹² , Sn, and Pb, and also to resonance scattering from Li ⁶ .	
I-60-22	7.7-METER BENT-CRYSTAL SPECTROMETER (ANL-6235, November 1960) R. K. Smither	8
	The gamma-ray spectrum resulting from thermal-neutron capture in Hf ¹⁷⁷ was	

investigated with the Argonne 7.7-m bent-crystal spectrometer. The observed spectrum consisted of 250 gamma rays with energies between 50 kev and 1.6 Mev. These precision measurements of gamma-ray energies and intensities were used to modify and extend the level scheme of Hf^{178} . The extended level scheme contained 20 levels. These levels suggest the presence of three $K = 0$ rotational bands (besides the ground state) and two $K = 2$ rotational bands in the level scheme of Hf^{178} .

I-80-30 MOLECULAR BEAM STUDIES (ANL-6455,
November-December 1961)

W. J. Childs, J. Dalman, D. von
Ehrenstein, and L. S. Goodman . . .

36

Modifications and operating characteristics of the Mark II atomic-beam machine are discussed.

I-144-15 INVESTIGATION OF SCINTILLATORS (ANL-
6432, September-October 1961)

L. J. Basile and W. L. Buck

43

Fluorescence spectra and lifetimes have been studied for polystyrene samples having different average molecular weights and different concentrations of residual styrene monomer. The residual styrene monomer is fluorescent and acts as a typical fluorescent solute in a rigid medium. The fluorescence properties of polystyrene were found to depend on both the concentration of residual styrene monomer and, to a lesser extent, the average molecular weight.

PAGEII. MASS SPECTROSCOPY

- II-28-5 KINETICS OF CHEMICAL REACTIONS IN
THE GAS PHASE (ANL-5818, October-
December 1957)

J. Berkowitz and S. Wexler 55

An attempt was made to prepare CH_2 radicals and measure their appearance potential in a mass spectrometer. No evidence for the presence of CH_2 radicals was obtained, although the apparatus was easily able to detect CH_3 radicals. The result casts doubt on an earlier mass spectrometric experiment.

V. THEORETICAL PHYSICS, GENERAL

- V-3-9 DYNAMICS OF NUCLEAR COLLECTIVE
MOTION (ANL-6376, June 1961)

D. R. Inglis 63

The last two nucleons in the ground state of N^{14} have an angular momentum which can be acquired in the reaction $\text{C}^{12}(\text{Li}^6, \alpha)\text{N}^{14}$ only if the product α comes from the target C^{12} and this causes the α to go predominantly in the backward direction as observed.

- V-12-2 ACCURACY OF THE SUPERCONDUCTIVITY
APPROXIMATION FOR PAIRING FORCES
IN NUCLEI (ANL-6358, April-May 1961)

A. K. Kerman, R. D. Lawson, and
M. H. Macfarlane 64

This project has been terminated with publication of the report.

V-14-2	PROPERTIES OF NUCLEI WITH NEUTRONS AND PROTONS IN THE $1f_{7/2}$ SHELL (ANL- 6391, July-August 1961)	
	R. D. Lawson	64
	This project has been terminated with pub- lication of the report.	
V-25-1	SCATTERING OF ALPHA PARTICLES BY A VIBRATIONAL NUCLEUS (New project)	
	L. J. Tassie	65
	Both elastic and inelastic scattering are calculated by use of the plane-wave Born approximation. The treatment includes both single-phonon and two-phonon states and the effect of the diffuseness of the nuclear surface. The result for elastic scattering and the excitation of single- phonon 2^+ and 3^- states agrees well with experiments on Ni^{60} .	
V-33-4	FLUX QUANTIZATION AND TIME-REVERSAL DEGENERACY (ANL-6488, January 1962)	
	M. Peshkin and W. Tobocman	66
	It is proved that quantization of the magnetic flux through a superconducting ring, in units of $\pi \hbar c/e$, implies zero current at thermal equilibrium. The only approximation is the neglect of the penetration of the magnetic field into the body of the superconductor. The only characteristics of the Hamiltonian used are time-reversal invariance and gauge invariance.	

	<u>PAGE</u>
V-46-1 ELASTIC NUCLEON-NUCLEON SCATTER - ING AT HIGH ENERGIES AND SMALL ANGLES (New Project)	
K. Hiida	67
<p>The characteristic features of the elastic nucleon-nucleon scattering at high energies and small angles are explained on the basis of double diffraction scattering of a virtual boson by the two nucleons.</p>	
PUBLICATIONS	71
PERSONNEL CHANGES IN THE ANL PHYSICS DIVISION . . .	75

PROJECTS NOT REPORTED IN THIS ISSUE

A reference to the last preceding report is given in parentheses for each project.

I. EXPERIMENTAL NUCLEAR PHYSICS

- I-3. Cross Section Measurements with the Fast Neutron Velocity Selector (ANL-6072, October-November 1959), L. M. Bollinger and R. E. Cote¹.
- I-7. Gamma-Ray Spectra from Capture in Neutron Resonances (ANL-6146, April-May 1960), L. M. Bollinger, R. T. Carpenter, and H. E. Jackson.
- I-10. Tandem Van de Graaff Accelerator (ANL-6262, December 1960), F. P. Mooring and J. R. Wallace.
- I-11. Installation and Operation of the Van de Graaff Generator (ANL-6488, January 1962), J. R. Wallace.
- I-14. Pulsed Beams for the Van de Graaff Machine (ANL-6391, July-August 1961), R. E. Holland and F. J. Lynch.
- I-19. Nuclear Resonance Absorption of Gamma Rays (ANL-6455, November-December 1961), J. Heberle, P. N. Parks, and J. P. Schiffer.
- I-22. Scattering of Charged Particles (ANL-6455, November-December 1961), H. W. Broek, T. H. Braid, J. L. Yntema, and B. Zeidman.
- I-28. Angular Correlations in Charged-Particle Reactions (ANL-6358, April-May 1961), T. H. Braid.
- I-34. Decay of $_{68}\text{Er}^{172}$ (ANL-6432, September-October 1961), S. B. Burson and R. G. Helmer.
- I-35. Decay of $_{57}\text{La}^{135}$ (19.5 hr) (ANL-6391, July-August 1961), S. B. Burson and H. A. Grench.
- I-55. Capture Gamma-Ray Spectra for Neutrons with Energies from 0.1 to 10 ev (ANL-6052, September 1959), S. Raboy and C. C. Trail.

- I-98. Unbound Nuclear Energy Levels in the Kev Region (ANL-6432, September-October 1961), C. T. Hibdon.
- I-102. Neutron Cross Sections by Self-Detection (ANL-6376, June 1961), J. E. Monahan and F. P. Mooring.
- I-111. Semiconductor Detectors (ANL-6455, November-December 1961), H. M. Mann and J. W. Haslett.

II. MASS SPECTROSCOPY

- II-23. Sputtering Experiments in the Rutherford Collision Region (ANL-6488, January 1962), M. S. Kaminsky.
- II-29. Gaseous Species in Equilibrium at High Temperatures (ANL-6455, November-December 1961), J. Berkowitz.
- II-39. Fragmentation of Cyanogen (ANL-6488, January 1962), H. E. Stanton.
- II-40. Fragmentation of Hydrocarbons (ANL-6376, June 1961), H. E. Stanton.
- II-41. Consecutive Ion-Molecule Reactions (ANL-6455, November-December 1961), S. Wexler and N. Jesse.

III. CRYSTALLOGRAPHY

- III-4. Crystal Structure Studies of Compounds of Elements Ac-Am (ANL-6038, July-August 1959), W. H. Zachariasen.

IV. PLASMA PHYSICS

- IV-10. High-Frequency Plasmas (ANL-6455, November-December 1961), A. J. Hatch.

V. THEORETICAL PHYSICS, GENERAL

- V-1. The Deformation Energy of a Charged Liquid Drop (ANL-6432, September-October 1961), S. Cohen and W. J. Swiatecki.
- V-4. Relative β -Decay Probabilities for $_{19}\text{K}^{40}$ (ANL-6455, November-December 1961), D. Kurath.
- V-8. Relationships of Collective Effects and the Shell Model (ANL-6358, April-May 1961), D. Kurath.
- V-9. Interpretation of Experiments Involving Excitation of the 15.1-Mev Level of C^{12} (ANL-6391, July-August 1961), D. Kurath.
- V-15. Statistical Properties of Nuclear Energy States (ANL-6488, January 1962), N. Rosenzweig.
- V-42. Time Reversal and Superselection (ANL-6488, January 1962), H. Ekstein.
- V-45. Meson-Nucleon Interaction (ANL-6488, January 1962), K. Tanaka, K. Hiida, and M. Soga.

I. EXPERIMENTAL NUCLEAR PHYSICS

I-2-19

Neutron Detectors

(51210-01)

L. M. Bollinger, R. T. Carpenter, and J. P. Marion
Reported by J. P. Marion

In order that a neutron detector be useful for time-of-flight experiments, it is necessary that fluctuations in the neutron detection time, defined as the interval between the incidence of a neutron on the detector and its absorption in the detector, be small. For situations where the mean free path for scattering, λ_s , is either much less than the mean free path for absorption, λ_a , or much greater than the transverse dimension of the detector, w , the uncertainty in the detection time depends in a simple way on λ_a and w and may be calculated by elementary means. Also, in this case the efficiency of the detector is given simply by

$$\epsilon_0 = 1 - \exp(-w/\lambda_a) . \quad (1)$$

However, one may wish to detect neutrons with energies such that the first condition is not satisfied, and/or obtain higher efficiencies by using values of w such that the second condition is not satisfied. In this case a significant fraction of the absorptions may be preceded by one or more scatterings in the detecting mediums with a resulting increase in the detection time uncertainty and the efficiency.

In order to obtain quantitative information on the distribution of neutron detection times in boron-and lithium-loaded glass scintillators in cases where such multiple scattering cannot be neglected, a series of Monte Carlo calculations has been performed. The calculations were carried out on the GEORGE computer in a way similar to that described by Kennett and Bollinger.¹

¹T. J. Kennett and L. M. Bollinger, Nuclear Phys. 12, 249-260 (1959).

Briefly, the approach is as follows. A parallel beam of monoenergetic neutrons is incident normal to the front face of a plate of the capturing medium. It is assumed that the medium is composed of a single fictitious nuclide of nucleon number $A = 18$, that the scattering cross section is independent of neutron energy, and that the absorption cross section varies as $1/v$. The magnitudes of these quantities are chosen such that the probabilities for scattering and capture in the fictitious medium are the same at all neutron energies as in the real absorbing medium being investigated. The course of each neutron is followed

through the medium in the way described in reference 1. If the neutron is captured in the medium, the total distance which it traveled is recorded, and the event is inserted in the appropriate block of a histogram of distance increments. Since the materials included in this investigation moderate the neutron energies only slightly before the neutrons escape or are captured, the distribution of lifetimes is closely approximated by the distribution of path lengths of the neutrons.

An example of the results is shown in Fig. 1. For the case illustrated, the medium was a slab with infinite lateral dimensions and a thickness of 2.28 cm. The absorption and scattering cross sections were chosen so as to duplicate the absorption and scattering

Fig. 1. Results of the Monte Carlo calculation of the distribution of neutron path lengths before capture for lithium glass GL-304, 2.28 cm thick.

probabilities that actually exist for the lithium glass GL-304.² The histograms show, as expected, that the magnitude of the tail increases as neutron energy increases, or, in other words, as the probability of multiple scattering increases.

Although the conditions used in the Monte Carlo calculations were specified in terms of the characteristics of particular scintillators, the results actually may be applied in a more general way. The distribution of the ratio d/w , where d is the path length, is completely determined by the mean free paths for scattering and absorption, and by the thickness w . Thus, if all of these parameters are multiplied by the same constant, the distribution of d/w will not be affected. We choose to use the dimensionless ratios w/λ_s and λ_a/λ_s to characterize the distribution. The calculations were performed for a broad range of these ratios so that the distribution of lifetimes for glass scintillators with a large range of compositions could be obtained by interpolation. Any set of values used for w/λ_s and λ_a/λ_s corresponds to some set of values of thickness, density, and neutron energy for all absorbing slabs for which the absorption cross section varies as $1/v$ and the moderation is like that for a nucleus with mass number 18. Hence, these Monte Carlo calculations are applicable to all of the glass scintillators and for many other kinds of detectors as well.

Table I is a tabular representation of the distribution of detection times. The area under the histogram was arbitrarily divided into three sections. Area A_0 contains all the neutrons with path lengths $d < w$, A_1 contains all neutrons with path lengths in the range $w \leq d \leq 3w$, and area A_2 contains all those with path lengths $d > 3w$. Each histogram was determined from the capture of 2000 neutrons. Here again the table shows the expected results. That is, there is an increase in the area under the tail of the histogram when either the energy of the neutron or the thickness of the absorber increases.

It is also of interest to see how the magnitude of the tail

² L. M. Bollinger, G. E. Thomas, and R. J. Ginther, (to be published in Nuclear Instr. and Methods).

TABLE I. Summary of results of Monte Carlo calculations for slabs with infinite lateral dimensions and thickness w . The conditions specified in columns 1, 2, and 3 are for a lithium glass with characteristics similar to those of the lithium glass GL-304.

Conditions				Results			
w/λ_s	λ_a/λ_s	E_n (ev)	w/v (nsec)	$\frac{A_1}{A_0}$	$\frac{A_2}{A_0}$	ϵ (%)	ϵ/ϵ_0
0.518	0.429	10	457	0.12	0.001	74.5	1.06
0.518	1.35	10^2	145	0.31	0.034	39.0	1.22
0.518	4.29	10^3	46	0.40	0.088	16.7	1.46
0.518	13.5	10^4	14.5	0.43	0.16	5.7	1.52
0.518	42.9	10^5	4.6	0.42	0.17	2.0	1.67
0.130	1.35	10^2	36.2	0.22	0.052	10.6	1.17
0.130	13.5	10^4	3.6	0.24	0.084	1.10	1.15
0.052	1.35	10^2	14.5	0.13	0.051	4.14	1.10
0.052	13.5	10^4	1.5	0.19	0.046	0.41	1.07

depends upon the lateral dimensions of the absorber. Therefore, calculations were performed for an absorbing medium in the form of a right circular cylinder with diameter D as a variable parameter. All other conditions were the same as for the case of the infinite slab. The values for the ratios A_1/A_0 and A_2/A_0 are given in Table II. These data have meaning for a large class of detectors if, for any given detector, the diameter is adjusted to give the same value of the ratio D/λ_s . Note that the tail of the histogram increases as the diameter of the detector increases.

In Tables I and II, the true (Monte Carlo) capture probability ϵ is listed. The probability ϵ_0 is the result calculated from Eq. (1), which neglects multiple scattering. From these data, it is apparent

TABLE II. Results of Monte Carlo calculations for cylinders of absorber. When the absorber has the density and composition of lithium glass GL-304, the conditions of the calculation correspond to a neutron energy of 11 400 ev, a thickness of 2.28 cm, and the diameter D.

D (cm)	$\frac{A_1}{A_0}$	$\frac{A_2}{A_0}$	ϵ (%)
0	0	0	3.77
2.28	0.14	0	3.93
5.70	0.26	0.005	4.57
11.4	0.33	0.040	5.01
22.8	0.37	0.074	5.36
∞	0.43	0.16	5.73

that ϵ_0 is a good approximation when the energy is low or when the detector is thin, i.e., when either $\lambda_a/\lambda_s < 1$ or $w/\lambda < 1$, where λ is the total mean free path for neutrons in the scintillator. If both of these conditions fail, the error introduced by neglecting multiple scattering may be quite large. Finally, it can be shown from the tables that ϵ_0 is a good approximation ($\pm 10\%$) to the probability of capture in the region A_0 for all conditions studied.

I-18-14 Neutron Polarization and Differential Cross Sections (51210-01)

R. O. Lane and W. F. Miller (AMD)

Reported by R. O. Lane

ENERGY-DEPENDENT MULTIPLE SCATTERING OF
NEUTRONS IN PLATES

Measurements of the angular distribution of neutrons scattered from nuclei often contain serious errors produced by the contribution of neutrons multiply scattered within the sample before emerging from the

sample and being detected at a given angle. A technique was developed for the GEORGE computer which calculated the intensities for single, double, and triple scattering for the case of a scattering sample in the shape of a plate. The most unusual feature of this technique is that, unlike previous treatments, it takes account of the dependence of the differential cross sections on energy following each successive elastic-scattering event. Such a feature is essential to any calculation dealing with light nuclei whose cross sections exhibit resonance structure. The energy-dependence as well as the angle-dependence of the differential cross section $\sigma(\theta, E)$ are included in the input data for the calculation.

Since these data are actually the results sought in measurements of this type, one usually begins by giving as initial input data the measured values of $\sigma(\theta, E)$. Then by comparison of the consequent calculated intensities with the measured ones, corrected values of $\sigma(\theta, E)$ may be supplied as input to the calculation, and the whole calculation may be repeated. Iteration of this procedure converges rather quickly to a set of $\sigma(\theta, E)$ as a limit if the initial data did not contain too much multiple scattering. In most experiments dealt with to date, one calculation using the measured $\sigma(\theta, E)$ as input data was sufficient to give results with an error within the statistical uncertainty of the experiment.

The single-scattering intensities can be calculated analytically from the data. The double- and triple-scattering intensities are calculated by a highly efficient Monte Carlo technique which takes into account the variations of cross sections at each step of the process. For a given set of input $\sigma(\theta, E)$ the double- and triple-scattering intensities can be calculated quite accurately without excessive use of computer time.

Figure 2 shows an example of a polar plot of the calculated relative intensities for the case of neutrons of 0.32-Mev incident energy scattered from a plate of Li^6 . The scattering of the neutrons, which are incident in the direction of the arrow, is measured in the plane of the

Fig. 2. Scattering of 0.32-Mev neutrons by Li^6 . Curve A represents the calculated single scattering, B the double scattering, E the sum of single and double scattering, and T the single scattering without attenuation by subsequent collisions.

figure. The plate is perpendicular to the plane of the figure. Curves A and B represent the single- and double-scattering intensities, respectively, of the emergent neutrons. Curve E is the sum of A and B. Triple scattering in this case is too small to be shown. Curve T represents the single-scattering intensity without the effects of the attenuation of the scattered beam on the way out of the sample. The energy lost by some of the neutrons after elastic scattering reduces their energy to coincide with a large resonance at 0.26 Mev. Noticeable dips in curve A near 130° and 320° are produced by attenuation effects of this resonance. Neutrons removed from curve A at these points contribute to the double scattering in Curve B.

Multiple-scattering probabilities have also been calculated for other thicknesses and energies for Li^6 and also for C^{12} , Sn, and Pb. A detailed description of both the method and the results is given in an article to be published in Nuclear Instruments and Methods.

Calculations of this type are useful not only in understanding the detailed processes of multiple scattering of neutrons, but also in the extension of these relatively simple cases to the more complicated problems of transport and shielding calculations in reactor design.

I-60-22 7.7-Meter Bent-Crystal Spectrometer (51210-01)

R. K. Smither

THE THERMAL-NEUTRON-CAPTURE GAMMA-RAY
SPECTRUM OF $\text{Hf}^{177}(\text{n}, \gamma)\text{Hf}^{178}$ AND THE ASSOCIATED
ENERGY LEVELS IN Hf^{178}

The gamma-ray spectrum resulting from thermal-neutron capture in Hf has been investigated with the Argonne 7.7-m bent-crystal spectrometer.¹ Two samples of Hf were investigated, one of natural abundance and one enriched to 93% Hf^{178} . The observed gamma-ray

¹B. Hamermesh, D. Rose, and H. Ostrander, Rev. Sci. Instr. 28, 233 (1957).

spectrum consisted of 350 gamma rays with energies between 40 kev and 1.6 Mev. By comparing the relative intensity of each gamma ray in the enriched and unenriched sample, 217 of the observed gamma rays were identified with neutron capture in Hf^{177} . A list of these gamma-ray energies, with their errors and intensities, appears in Table III. The spectrum is

TABLE III. A list of the energies, errors, and intensities of the thermal-neutron capture-gamma-ray spectrum resulting from $\text{Hf}^{177}(\text{n}, \gamma)\text{Hf}^{178}$ as measured with the Argonne 7.7-m bent-crystal spectrometer. The energies and errors (kev) are given in columns 1 and 2 while the intensities (in photons per 100 neutron captures in Hf^{177}) appear in column 3. The errors are meant to be probable errors and to reflect both the statistical and systematic errors of the measurement. The probable error in the intensities is 10 - 20%, with the latter prevailing at the extremes of the spectrum. The gamma-ray intensities have not been corrected for internal conversion. Column 4 indicates if the gamma ray appears in the final scheme of Hf^{178} (Fig. 6).

(1) Energy (kev)	(2) Error (kev)	(3) γ 's/100 neutron captures in Hf^{177}	(4) In scheme (Fig. 6)?	(5) Remarks
1552.1	± 2.8	0.40		
1500.4	± 2.7	0.76		} Doublet structure
1473.9	± 2.7	0.32	Yes	
1420.9	± 2.0	2.14	Yes	} Doublet structure with possible line at ≈ 1419 kev (Int. ≈ 0.8)
1402.9	± 2.6	0.81	Yes	
1381.8	± 3.0	0.27	Yes	
1350.5	± 3.0	0.44		
1338.2	± 2.0	4.65	Yes	} Doublet structure with possible line at ≈ 1322 kev (Int. ≈ 0.8)
1327.5	± 2.2	2.05	Yes	
1310.3	± 2.4	1.24	Yes	
1291.9	± 2.3	0.57	Yes	
1269.5	± 1.6	1.80	Yes	
1228.8	± 1.4	7.30	Yes	{ Poss. sym. dbt. (Sep. $\approx 1-2$ kev)
1206.4	± 0.6	5.98	Yes	

(1) Energy (kev)	(2) Error (kev)	(3) γ 's/100 neutron captures in Hf ¹⁷⁷	(4) In scheme (Fig. 6)?	(5) Remarks
1176.3	± 1.2	9.08	Yes	} Doublet structure
1167.4	± 1.2	4.71	Yes	
1143.7	± 0.9	1.82	Yes	
1125.4	± 1.2	0.19	?	
1103.1	± 0.5	3.56	Yes	
1081.8	± 1.0	2.80	Yes	} Doublet structure
1077.8	± 1.0	2.90		
1057.7	± 1.2	0.24		
1032.3	± 2.0	0.27		
1016.5	± 0.7	0.56	Yes	
1006.9	± 1.0	0.38	←	} Troublesome impurity line at 1003.5 kev
983.5	± 1.0	0.27		
969.5	± 0.8	0.40	Yes	} Doublet structure
962.1	± 0.7	1.09		
949.09 ^a	± 1.6	0.24		
937.97 ^a	± 1.6	0.21		
921.26	± 0.4	1.09		
901.30	± 0.4	0.36		
881.27	± 1.2	0.15	Yes	} Overlapping structure
870.11 ^b	± 1.4	0.11		
861.84	± 1.2	0.28		
855.87 ^b	± 1.5	0.15		
847.71	± 1.5	0.12		
841.11	± 0.9	0.17	Yes	
831.65 ^a	± 1.2	0.08		
822.05	± 1.2	0.09		} Doublet structure
817.50	± 1.2	0.11		

(1) Energy (kev)	(2) Error (kev)	(3) γ 's/100 neutron captures in Hf ¹⁷⁷	(4) In scheme (Fig. 6)?	(5) Remarks
792.54	± 1.0	0.10		
780.58	± 1.0	0.10		} Doublet structure
775.68	± 1.0	0.22		
763.11	± 0.9	0.13		} Triplet structure
758.01	± 0.9	0.31		
752.35	± 0.4	0.36	Yes	
744.15	± 0.6	0.19		
736.90	± 0.4	0.19		
716.33	± 0.4	0.21		
704.24	± 0.4	0.14		
698.17	± 0.6	0.11		} Doublet structure
694.82	± 1.2	0.07		
687.44	± 0.6	0.10		
682.37	± 0.6	0.16		
672.35	± 0.8	0.08		} Doublet structure
668.69	± 0.5	0.10		
662.40	± 0.5	0.10		} Doublet structure with troublesome impurity at 654.8 kev
658.29	± 0.6	0.14		
649.43	± 0.4	0.13		} Doublet structure
645.37	± 0.2	0.33		
632.93 ^b	± 0.3	0.13		
628.32	± 0.3	0.23		
617.81	± 0.3	0.23		
611.10	± 0.4	0.11		
606.84	± 0.3	0.39		
597.64	± 0.3	0.41		} Doublet structure
596.33	± 0.3	0.31		

(1) Energy (kev)	(2) Error (kev)	(3) γ 's/100 neutron captures in Hf ¹⁷⁷	(4) In scheme (Fig. 6)?	(5) Remarks
590.33	± 0.4	0.09		} Doublet structure
587.20	± 0.4	0.14		
583.07	± 0.4	0.12		} Doublet structure
580.45	± 0.4	0.08		
572.23	± 0.3	0.11		
567.78	± 0.5	0.03		
563.08 ^a	± 0.5	0.017		
559.38	± 0.5	0.035		
550.10 ^a	± 0.5	0.016		
547.36	± 0.2	0.51		
542.99	± 0.3	0.12		
534.66	± 0.2	0.21		
522.08	± 0.1	0.36		} Annihilation rad. at 510.94 kev
503.90	± 0.2	0.10		
497.62	± 0.1	1.46		} Troublesome impurity at 507.87 kev
494.22	± 0.1	0.17		
488.70	± 0.3	0.09		} Troublesome impurity at 486.2 kev
483.92	± 0.3	0.046		
481.40	± 0.2	0.09		
478.31	± 0.3	0.047		} Doublet structure
477.31	± 0.3	0.039		
472.91	± 0.1	0.23		
466.83	± 0.4	0.28		
459.91	± 0.1	0.12		
453.99	± 0.1	0.16		
449.79	± 0.2	0.042		

(1) Energy (kev)	(2) Error (kev)	(3) γ 's/100 neutron captures in Hf ¹⁷⁷	(4) In scheme (Fig. 6)?	(5) Remarks
438.84	± 0.2	0.07		
435.57	± 0.14	0.21		
430.81	± 0.30	0.19		
427.71	± 0.20	0.07	Yes	{ Triplet structure ground-state band (8 ⁺ $\rightarrow$ 6 ⁺) Possible doublet
426.21	± 0.14	0.58		
424.39	± 0.17	0.23		
420.87	± 0.20	0.08		
417.00	± 0.30	0.021		{ Troublesome impurity at 408.07 kev
406.70	± 0.20	0.024		
403.64	± 0.09	0.16		
399.84	± 0.09	0.17		
397.48	± 0.20	0.025		
395.65	$\pm 0.14^b$	0.040		Possible doublet
390.90	± 0.20	0.015		
388.04	± 0.20	0.015		
383.26	± 0.07	0.26		
379.44	± 0.07	0.11	Yes	
376.45	± 0.10	0.016		
374.62	± 0.10	0.027		
371.74	± 0.10	0.012		
364.33	± 0.14	0.011		{ Doublet structure
363.24	± 0.15	0.06		
356.83	± 0.05	0.38		
354.59	± 0.10	0.025		
350.98	± 0.10	0.046		
348.26	± 0.05	0.50		

(1) Energy (kev)	(2) Errors (kev)	(3) γ 's/100 neutron captures in Hf ¹⁷⁷	(4) In scheme (Fig. 6)?	(5) Remarks
345.43	± 0.15	0.040		
341.98 ^b	± 0.05	0.020		
339.08	± 0.04	1.14		
337.51	± 0.05	0.05		
331.60	± 0.06	0.049		
328.59	± 0.08	0.042		
325.51	± 0.04	6.82	Yes	{ Ground-state band (6 ⁺ $\rightarrow$ 4 ⁺)
321.46	± 0.08	0.015		
318.89	± 0.08	0.017		
318.29	± 0.08	0.017	Yes	{ Triplet structure
317.28	± 0.03	0.12		
313.38	± 0.05	0.15		
312.42	± 0.05	0.026		{ Doublet structure with possible impurity at 312.5 kev
311.16	± 0.07	0.042		
308.38	± 0.10	0.009		
306.37	± 0.04	0.10	Yes	
296.72	± 0.03	0.06		
289.47	± 0.03	0.60		
280.63	± 0.03	0.10	Yes	
279.31	± 0.06	0.036		
278.58	± 0.06	0.021		{ Doublet structure
277.28	± 0.06	0.22	Yes	
276.88	± 0.06	0.22	Yes	{ Doublet structure
274.69	± 0.04	0.047		
273.09	± 0.03	0.25		

(1) Energy (kev)	(2) Error (kev)	(3) γ 's/100 neutron captures in Hf ¹⁷⁷	(4) In scheme (Fig. 6)?	(5) Remarks
270.52	± 0.08	0.06		} Doublet structure with troublesome impurity at 269.85 kev
270.06	± 0.08	0.027		
268.69	± 0.04	0.012		
264.59	± 0.02	0.025		
259.99	± 0.02	0.05		
256.56	± 0.05	0.34		} Doublet structure
256.01	± 0.05	0.28		
249.37	± 0.03	0.07		
247.54	± 0.04	0.015		
246.88	± 0.04	0.024	Yes	
245.23	± 0.02	0.51	Yes	
244.23	± 0.02	0.52		
242.55	± 0.03	0.029		
241.95	± 0.04	0.08		
234.49	± 0.03	0.10		
230.74 ^a	± 0.07	0.010		
230.22	± 0.04	0.045		
229.31	± 0.02	0.15		
227.20	± 0.02	0.19	Yes	
224.37 ^b	± 0.06	0.006	Yes	
223.96	± 0.06	0.027		
223.32	± 0.14	0.006		
218.93	± 0.02	0.09		
217.76	± 0.03	0.044		
216.63	± 0.09	0.014		} Doublet structure
216.29	± 0.02	0.15		
213.42	± 0.02	41.30	Yes	{ Ground-state band (4 ⁺ $\rightarrow$ 2 ⁺)
208.26	± 0.03	0.012		

(1) Energy (kev)	(2) Error (kev)	(3) γ 's/100 neutron captures in Hf ¹⁷⁷	(4) In scheme (Fig. 6)?	(5) Remarks
204.05	± 0.04	0.13	Yes	} Doublet structure
203.75	± 0.04	0.20	Yes	
201.04 ^b	± 0.03	0.009		
190.07	± 0.01	0.083		
187.59	± 0.01	0.071		
185.31 ^b	± 0.06	0.007	Yes	
180.39	± 0.02	0.021		
176.19	± 0.02	0.051		} Doublet structure
173.63	± 0.03	0.014		
173.31	± 0.03	0.038		
169.47	± 0.02	0.031		
166.63	± 0.01	0.014		
161.92	± 0.02	0.037	Yes	
157.86	± 0.02	0.023		
151.29	± 0.01	0.063	Yes	
149.14	± 0.01	0.10	Yes	
148.07	± 0.03	0.042		
147.78	± 0.01	0.16	Yes	
144.49	± 0.01	0.27	Yes	
140.86	± 0.01	0.19		
137.67	± 0.01	0.05		
129.33	± 0.01	0.044	?	
128.09	± 0.03	0.018		
126.58	± 0.01	0.25	Yes	} Troublesome impurity at 125.94 kev
125.84	± 0.04	0.013		
124.09	± 0.01	0.022		

(1) Energy (kev)	(2) Error (kev)	(3) γ 's/100 neutron captures in Hf ¹⁷⁷	(4) In scheme (Fig. 6)	(5) Remarks
122.85	± 0.01	0.39		
116.58	± 0.01	0.082		
112.63	± 0.01	0.043		
111.66	± 0.01	0.092		
107.31	± 0.01	0.14		
105.14	± 0.01	0.12		
102.58	± 0.01	0.017	Yes	
100.94 ^b	± 0.02	0.023		
97.91	± 0.01	0.066		
93.17	± 0.005	18.09	Yes	{ Ground-state band (2 ⁺ $\rightarrow$ 0 ⁺)
88.84	± 0.01	0.35	Yes	
86.97	± 0.02	0.023		
85.59	± 0.01	0.43	Yes	
82.70	± 0.01	0.40		
73.07	± 0.01	0.077	Yes	
71.05	± 0.01	0.030	Yes	
53.90 ^b	± 0.01	0.012		
52.74 ^b	± 0.01	0.018		

^aQuestionable line (poor statistics or complicated structure).

^bQuestionable isotopic assignment (usually due to the low intensity of the gamma when observed with the natural-abundance sample).

quite complicated, especially in the region from 100 kev to 300 kev. The quoted errors in Table III are a combination of the statistical error (rms) and an estimated systematic error. Many of the large errors assigned to the gamma rays above 1 Mev are due to the complicated nature of the gamma spectrum rather than to lack of precision in the instrument. The observed line width (full width at half maximum) was 1% (10 kev) at 1 Mev and 0.1% (0.1 kev) at 100 kev during this experiment. The precision of the instrument was one part in 2500 at 1 Mev and one part in 20 000 at 100 kev.

The observed gamma-ray spectrum is shown in the form of a line graph in Figs. 3, 4, and 5. The intensities of the γ rays shown in these figures are those listed in Table III. They have been

Fig. 3. Line graph of the $\text{Hf}^{177}(\text{n}, \gamma) \text{Hf}^{178}$ gamma-ray spectrum below 400 kev, observed with the Argonne 7.7-m bent-crystal spectrometer. The intensities of the gamma rays are given in photons per 100 neutron captures in Hf^{177} .

Fig. 4. Line graph of the $\text{Hf}^{177}(n,\gamma)\text{Hf}^{178}$ gamma-ray spectrum between 400 and 800 keV. The intensities of the gamma rays are given in photons per 100 neutron captures in Hf^{177} .

Fig. 5. Line graph of the $\text{Hf}^{177}(n,\gamma)\text{Hf}^{178}$ gamma-ray spectrum between 800 and 1600 keV. The intensities of the gamma rays are given in photons per 100 neutron captures in Hf^{177} .

corrected for the self-absorption in the sample, the absorption of the material (He, air, Al, Li, paraffin, quartz) between the sample and the detector, the reflectivity of the quartz crystal, the transmission of the collimator, and the efficiency of the NaI detector. All of these quantities, except the self-absorption correction, were determined by experiment. No correction is made for the internal conversion of the gamma rays. The observed relative intensities of the gamma rays originating from the natural-abundance sample (Hf metal) and the gamma rays originating from the enriched sample (HfO_2) form a good check on the self-absorption calculations. The predicted intensity ratios (enriched/unenriched) agreed with the observed ratios to within 10% throughout the energy range.

A number of interesting features of the spectrum are visible in these line graphs. The three gamma-ray transitions between the lowest four members of the ground-state rotational band (0^+ , 2^+ , 4^+ , 6^+) are evident as strong lines at 93, 213, and 325 keV in Fig. 3. The strength of the 213-keV γ -ray transition (41%)² from the 4^+ to the 2^+ member of the ground-state band reflects the previously mentioned fact that high-spin states are well populated in the decay scheme. A second prominent group of strong transitions occurs between 1.0 and 1.5 MeV in Fig. 5. The concentration of appreciable transition strength in a single gamma ray implies that although the level in question is strongly populated from levels of higher excitation, there are relatively few competing exit channels available for the depopulation of this level. A level with these characteristics is most likely to occur at low excitation energies. In the proposed decay scheme, these strong gamma rays are interpreted as transitions between a group of excited-state rotational bands (1.1 to 1.5 MeV) and the ground-state band. A third group of gamma rays stands out in the line graph (Fig. 3) between 70 and 140 keV. Their

² The gamma intensities given in percent are equivalent to intensities given in photons per 100 neutron captures in Hf^{177} .

proximity to the 93-kev gamma ray suggests that they may be the transitions between the lower members of the excited-state rotational bands. This appears to be the case in the proposed decay scheme. If these low-energy gamma rays are M1 or E2 transitions, then their total intensities (γ + conversion electron) are four or five times the γ intensity shown. They then become 1-2% transitions which compare favorably with the intensities observed for the γ transitions between 1.0 and 1.5 Mev (Fig. 5).

The precision measurements of the energies and intensities of the capture-gamma-ray spectrum, performed with the bent crystal spectrometer, were used to correct and extend the level scheme of Hf^{178} . This new level scheme appears in Fig. 6. In order to develop a decay scheme of this type, it is very convenient to start with a set of established levels on which one can build. In this case, the first four levels of the ground-state rotational band (0^+ , 2^+ , 4^+ , 6^+) were used. The energies of these states were obtained by comparing the capture-gamma-ray measurements (this paper) with the conversion electron work of F. F. Felber, F. S. Stephens, Jr., and F. Asaro³ and the Coulomb excitation work of E. Chupp, J. DuMond, F. Gordan, R. Sopson, and H. Mark.⁴

The excitation energy at which new intrinsic states⁵ appear in the level scheme of Hf^{178} is of considerable theoretical interest. For this reason it was felt worthwhile to attempt to generate the level scheme of Hf^{178} with a relatively limited use of the conversion-electron data. If this new approach reproduced most of the levels found in the conversion

³ F. F. Felber, Jr., (Master's thesis) University of California UCRL-3618, January, 1956; F. F. Felber, Jr., F. S. Stephens, Jr., and F. Asaro, J. Inorg. Nuclear Chem. 7, 153 (1958).

⁴ E. Chupp, J. DuMond, F. Gordan, R. Sopson, and H. Mark, Phys. Rev. 112, 518 (1958).

⁵ The term "new intrinsic states" refers to states that have a different configuration, and thus a different nuclear shape, than the ground state.

Fig. 6. Level scheme of Hf^{178} as deduced from the gamma-ray measurements made with the Argonne 7.7-m bent-crystal spectrometer. The energies of the levels and gamma rays are given in keV. The width of a line in the level scheme is meant to reflect the relative intensity of the gamma ray. The width scale used for the low-energy gammas (lying between 1195 and 1575 keV) is four times that used for the gamma rays with energies between 1 and 2 MeV and for the ground-state band. No corrections have been made for internal conversion. An asterisk following the energy of a level indicates that that level has not been observed previously.

electron work,³ then some confidence could be put in the new levels found by the same method. The new level scheme of Hf^{178} appears in Fig. 6.

The basic assumption in the method used to develop this level scheme of Hf^{178} was that the energies and spin assignments of the ground-state rotational band were a 0^+ ground state, a 2^+ level at 93.17 keV, a 4^+ level at 306.59 keV, and a 6^+ level at 632.10 keV. The energies of these levels are derived from the sum of the energies (as measured with the Argonne bent-crystal spectrometer) of the cascade gammas that depopulate the level. The spin assignments, however, depend heavily on the previous conversion-electron work³ and on the Coulomb-excitation experiments.⁴ The upper levels (which appear in Fig. 6) were then built on the ground-state band in a straightforward manner. The energy differences between all strong gamma-ray transitions above 700 keV were calculated and compared with the three cascade gammas (93, 213, and 325 keV) in the ground-state band (Fig. 6). About 16 such energy differences matched the energies of the three gamma rays. Combinations of these differences were compared until a consistent level scheme was obtained. In this manner, 8 levels were suggested at energies of approximately 1269, 1323, 1384, 1402, 1421, 1431, 1473, and 1513 keV. The low-energy part of the gamma-ray spectrum was then investigated for possible interconnecting transitions. Ten such interconnections were found.

Although the above method appears to be effective for states with spins of 1, 2, 3, and 4, it will not generate 0^+ states. A spin-0 state will have only one gamma transition to the ground-state band ($0^+ \rightarrow 2^+$). All other transitions are highly forbidden.

³C. J. Gallagher, H. L. Nielsen, and O. B. Nielsen, Phys. Rev. 122, 1590 (1961).

In the conversion-electron work of Gallagher, Nielsen, and Nielsen,⁶ a strong conversion-electron line was observed at 1197 ± 12 keV following the 9.3-min. electron-capture decay of Ta^{178} to Hf^{178} . No appreciable gamma-ray intensity was observed at this energy so Gallagher, Nielsen, and Nielsen conclude that the 1197-keV transition has either an E0, or an M2, and/or higher multipole assignment. This, coupled with the presence of a strong c. e. transition⁷ at 1105 ± 11 keV with a conversion coefficient close to the theoretical value for an E2 transition, lead to the establishment of a 0^+ level at 1197 ± 10 keV.

A strong gamma transition at 1103.1 ± 0.5 keV was observed in the capture-gamma-ray spectrum obtained with the Argonne bent-crystal spectrometer. Fortunately this gamma ray was well resolved from its nearest neighbors (1125 and 1079 keV) and could be uniquely assigned to the same transition as the c. e. line at 1105 keV. The (1103.1 ± 0.5) -keV gamma ray was used to establish a 0^+ level at 1196.27 ± 0.5 keV. This value agreed very well with the previously generated scheme (see three paragraphs back) since there were four low-energy gamma rays in the observed gamma spectrum that would interconnect this level with the rest of the level scheme.

The value obtained by Gallagher, Nielsen, and Nielsen⁶ for the conversion coefficient of the 1197-keV c. e. line ($0^+ \rightarrow 0^+$) was ≥ 0.017 . The major problem in obtaining a good value for this number is the relatively poor resolution of the NaI crystal used to observe the gamma rays.⁶ By using the bent-crystal spectrometer to compare the gamma-ray intensity of the 1103.1-keV transition with the upper limit of the 1195-keV transition and combining this with the relative c. e. intensities,⁶ a new lower limit is obtained for the conversion coefficient of the 1195-keV

⁷ The phrase "conversion electron" is abbreviated as "c. e." throughout this paper.

transition. If one assumes that the 1103.1-keV transition is pure E2, the new limit is $a_k \geq 0.04$. The alternative to the E0 assignment is now E5 or M4, which are out of the question if the 1103.1-keV gamma is assumed to be the transition to the 2^+ state at 93.17-keV. The argument here is that an M4 transition would be so very much slower than the competing M2 or E3 transition that it would not be observed as a strong c.e. line. The 0^+ level at 1237 keV was added after the 1323-keV level had been given an assignment of $K = 0$, spin = 2^+ .

Four additional levels were established above this upper group (between 1194 and 1473 keV) through a detailed analysis of the low-energy gamma rays.⁸ If this process is repeated, more levels are suggested at higher energies and the level spacings become smaller. Unfortunately, the likelihood of making an error (placing a gamma ray in the wrong position) in the decay scheme increases with the square of the number of levels in the scheme. For this reason it was felt advisable to limit the level schemes at this point. This limited extension of the level scheme is very interesting, however, in that the added levels can be interpreted as constituting the second members of the rotational bands started in the better-established levels below it.

Much information about the spins and parities of the levels in Fig. 6 can be obtained from the detailed consideration of the gamma-ray transitions that connect it to the rest of the scheme. For transitions between levels of the same parity, only M1 and E2 transitions are expected to have observable intensities. The competing M3 and E4 transitions are expected to be slower by at least a factor of 10^5 . The assignment of most of the levels in Fig. 6 to positive parity depends strongly on the previously mentioned conversion-electron work.⁶ Negative-parity assignments cannot be ruled out for the levels not seen in the electron-capture decay of Ta^{178} ,^{3,6} because no conversion

⁸The IBM-704 computer was used for this analysis.

coefficients are available for the gamma-ray transitions that connect them to the ground-state band. Tentative parity assignments were made for some of these levels by considering the relative intensities of the gamma rays that connect the levels to the rest of the scheme.

In the development of the spin assignments of the levels in Fig. 6, it has been assumed that observed gamma rays correspond to $\Delta(\text{spin}) \leq 2$. The spin assignments for positive-parity levels were made in the following manner. A 1^+ state will have two transitions to the ground-state band, $1^+ \rightarrow 2^+$ and $1^+ \rightarrow 0^+$; a 2^+ state will have three transitions to the ground-state band, $2^+ \rightarrow 4^+$, $2^+ \rightarrow 2^+$, and $2^+ \rightarrow 0^+$; a 3^+ state will have two transitions, $3^+ \rightarrow 4^+$ and $3^+ \rightarrow 2^+$; a 4^+ state will have three transitions, $4^+ \rightarrow 6^+$, $4^+ \rightarrow 4^+$, and $4^+ \rightarrow 2^+$; etc. A similar set of rules can be made for negative-parity states. A method of determining the value of K^θ for the band with which an individual level is associated is still needed.

The collective model predicts that relative intensities of gamma-ray transitions (of the same multipolarity) originating at one level and proceeding to different levels of another band are simple functions of their energies and Clebsch-Gordan coefficients. These Clebsch-Gordan coefficients are functions of the initial and final K values as well as the spins of the levels and the multipolarity of the transitions. If the spins of the levels, multipolarity of the transitions, and the K value of the final states are known, then it is possible to determine K_i , the K of the initial state, from the relative gamma-ray intensities. This is the principal method used to establish the K values that appear in Fig. 6.

Before discussing the levels in Fig. 6, it is instructive to consider in detail what the collective model predicts for the relative

⁹ The quantum number K refers to the projection of the spin or angular momentum of the state on the axis of rotational symmetry of the nucleus.

intensities of gamma transitions leading to the ground-state rotational band. The ratio of the gamma transition probabilities for two transitions originating at the same level and proceeding to two lower levels of another rotational band is given by

$$\frac{T}{T'} = \frac{(I_i \lambda I_f K_f | I_i K_i \lambda K_f - K_i)^2 E^{2\lambda + 1}}{(I_i \lambda I'_f K'_f | I_i K_i \lambda K_f - K_i)(E')^{2\lambda + 1}},$$

where the primed and unprimed quantities correspond to the two different final states. Compilation of the relative intensities of gamma transitions that can proceed to the ground-state band ($K_f = 0$) appears in Table IV.

TABLE IV. Relative gamma-ray intensities for transitions from levels of excited-state bands with $K = 0, 1$, and 2 to the members of the $K = 0$ ground-state band. The relative intensities are actually the squares of the appropriate Clebsch-Gordan coefficients $(I_i \lambda I_f K_f | I_i K_i \lambda K_f - K_i)$, where I_i and I_f are the spins of the initial and final states, respectively, K_i and K_f are their K values, and λ is the multipolarity of the radiation. The omissions that result from the selection rule $\Delta K \leq \lambda$ are indicated by "X". The omissions that result when the Clebsch-Gordan coefficient equals zero are indicated by "O". The observed ratios of gamma-ray intensities must be corrected by the appropriate energy factor $(E'/E)^{2\lambda + 1}$ before they can be compared with this table.

		Relative Intensity ^a ($K_f = 0$)					
$I_f \longrightarrow$		0	2	4	6	8	10^b
$K_i \ I_i \ B\lambda$							
0 0 E2			1.00				
0 0 M1							
1 1 E2			0.50				
1 1 M1		0.33 γ	0.17 γ				

TABLE IV. (Contd.)

		Relative Intensity ^a ($K_f = 0$)					
$I_f \rightarrow$		0	2	4	6	8	10^b
K_i	$I_i \ B\lambda$						
0	2 E2	0.20	0.29	0.51			
0	2 M1		O				
1	2 E2	0.20	0.07				
1	2 M1		0.50 γ				
2	2 E2	0.20	0.29	0.014			
2	2 M1		X				
1	3 E2		0.14	0.36			
1	3 M1		0.29 γ	0.21 γ			
2	3 E2		0.36	0.14			
2	3 M1		X	X			
0	4 E2		0.29	0.26	0.46		
0	4 M1			O			
1	4 E2		0.24	0.02	0.24		
1	4 M1			0.50 γ			
2	4 E2		0.12	0.35	0.03		
2	4 M1			X			
1	5 E2			0.18	0.32		
1	5 M1			0.27 γ	0.23 γ		
2	5 E2			0.32	0.18		
2	5 M1			X			
0	6 E2			0.31	0.25	0.43	
0	6 M1				O		

TABLE IV. (Contd.)

$I_f \longrightarrow$	Relative Intensity ^a ($K_f = 0$)					
	0	2	4	6	8	10^b
$K_i \ I_i \ B\lambda$						
1 6 E2			0.24	0.01	0.25	
1 6 M1				0.50 γ		
2 6 E2			0.10	0.36	0.04	
2 6 M1				X		
1 7 E2				0.20	0.30	
1 7 M1				0.27 γ	0.23 γ	
2 7 E2				0.30	0.33	
2 7 M1				X	X	
0 8 E2				0.33	0.25	0.43
0 8 M1					O	
1 8 E2				0.25	0.005	0.26
1 8 M1					0.50 γ	
2 8 E2				0.09	0.37	0.045
2 8 M1					X	

^aThe energy factor has been removed from the relative intensity or intensity ratio as discussed in the text.

^bExtrapolated values.

The relative intensities listed in Table IV are given by the square of the Clebsch-Gordan coefficient, $(I_i \lambda I_f K_f | I_i K_i \lambda K_f - K_i)^2$, where I_i and I_f are the relative spins of the initial and final states, K_i and K_f are their

K values, and λ is the multipolarity of the radiation.¹⁰ The energy factor (E^5 for E2 and E^3 for M1) must be removed from the observed gamma-ray intensities before they can be compared with Table IV.

The relative intensities of many transitions that would be strong in a single-particle-model estimate of gamma transition rates, do not appear in Table IV. Some of these omissions result from the selection rule $\Delta K \leq \lambda$, and are indicated by an "X." The others occur when the Clebsch-Gordan coefficient is zero and are indicated by an "O." The quantity γ appearing with all M1 values is a proportionality factor that reflects the ratio of the nuclear matrix elements (M1/E2) and thus the relative speeds of the M1 and E2 transitions. The single-particle estimate of γ in the relevant energy range (700-1400 kev) is $\gamma = 200-800$. Thus the M1 multipole can easily dominate a 1-Mev transition, even if its speed is only a tenth of the single-particle estimate. The factor γ is expected to increase as the energy of the transition decreases. The simple collective model does not predict any enhancement of the E2 transitions between levels of different bands. Therefore if this simple picture were to hold, E2 transitions would not be observed in competition with M1 transitions unless the latter were in some way hindered, e. g., through K forbiddenness or because of an accidental cancellation of terms (as indicated by O's in Table IV). In these cases, the main M1 contribution experimentally observed may come from a small admixture of a $K_i \pm 1$ band into the K_i band in question. A small (2%) admixture of $K = 1$ in the initial state is sufficient to explain the observed M1 strengths in Fig. 6. Table V lists the ratios of the relative intensities of transitions (see

¹⁰ The values of the Clebsch-Gordan coefficients were calculated with the use of the coefficients in the compilation of M. Rotenberg, R. Bivins, N. Metropolis, and J. Wooten, Sr., The 3j and 6j Symbols, (The Technology Press, Massachusetts Institute of Technology, Cambridge, Massachusetts, 1959).

TABLE V. Ratios of the relative intensities of transitions from levels of excited-state bands with $K = 0, 1$, and 2 to the members of the $K = 0$ ground-state band. The intensity ratios are actually the ratios of the squares of the appropriate Clebsch-Gordan coefficients, $(I_i \lambda I_f K_i | I_i K_i \lambda K_f - K_i)$, where I_i and I_f are the spins of the initial and final states, respectively, K_i and K_f are their K values, and λ is the multipolarity of the radiation. The notation I_f/I_i above the ratio columns refers to the spins of the final states of the transitions being compared. The ratios in parenthesis assume an admixture of 2% of $K = 1$ into the excited-state band and are meant only as rough estimates of the effect. The observed ratios of gamma-ray intensities must be corrected by the appropriate energy factor $(E'/E)^{2\lambda+1}$ before they can be compared with this table.

Pure E2 Intensity Ratios ^a					[M1 + E2] Intensity Ratios ^a ($\gamma = 150$)				
$\frac{I_f}{I_i} \rightarrow$	4/0	6/2	8/4	10/6 ^b	2/0	4/2	6/4	8/6	10/8 ^b
$K_i I_i$									
0 0					∞				
1 1					0.5				
0 2	2.6				1.4(9)	1.8(0.3)			
1 2	1.1				370	0.003			
2 2	0.07				1.4(9)	0.05(0.001)			
1 3						0.75			
2 3						0.4(0.6)			
0 4		1.6				0.9(6)	1.8(0.3)		
1 4		1.0				320	0.003		
2 4		0.25				2.9(15)	0.08(0.02)		
1 5							0.85		
2 5							0.6(0.8)		
0 6			1.4				0.8(5)	1.7(0.4)	
1 6			1.0				310	0.003	
2 6			0.4				3.6(19)	0.1(0.02)	
1 7								0.89	
2 7								1.1(1.05)	
0 8				1.3				0.8(5)	1.7(0.2)
1 8				1.0				300	0.003
2 8				0.5				4.2(21)	0.1(0.02)

^aThe energy factor has been removed from the relative intensity or intensity ratio as discussed in the text.

^bExtrapolated values.

Table IV) from levels of excited-state bands with $K = 0, 1$, and 2 to the members of the $K = 0$ ground-state band. The notations at the heads of these columns refer to the spins of the final states (I_f/I_f'). Two values are listed for some cases. The first number assumes that the upper band is pure. The second number (in parentheses) assumes a 2% admixture of $K = 1$ in the upper band and $\gamma = 150$. This second ratio (in parentheses) is not meant to be an upper or lower limit but rather an estimate of the effect of a small admixture of a $K = 1$ band with the even K_1 bands.

With the assumption that all observed gamma-ray transitions correspond to a spin change of 2 or less, and with the theoretical intensity ratios listed in Table V, it is possible to assign K values and spins to the levels in Fig. 6. These quantities appear at the left of each level in Fig. 6, in the order indicated above. If an excited state has negative parity, then the most likely multipoles for gamma transitions are $E1$ and $M2$. In this case, one need only change $M1$ to $E1$ and $E2$ to $M2$ in Tables IV and V to convert Tables IV and V for use with negative-parity states. The value of γ will of course be different.

A list of the energies, K values, spins, and parities of the levels in Fig. 6 appears in Table VI. All energies are given in kev. The errors in the level spacings are of the order of 0.5 kev when the members of the ground-state band are compared with the upper levels (above 1100 kev) while the errors in the levels spacings between these upper levels are about 0.03 kev. The parentheses around an assignment indicate uncertainty or conflict in the assignment.

Three requirements were imposed on the levels in Fig. 6. First, each level was required to have at least three connections (observed transitions) with the rest of the level scheme. The average number of such connections for the levels in Fig. 6 is five. The one exception to this rule is the level at 1058 kev. This level is believed to have an 8^+ spin assignment and is therefore expected to make only one connection ($8^+ \rightarrow 6^+$) to the rest of the level scheme. The

TABLE VI. A list of the energies, K values, spins, and parities of the levels in Hf^{178} (Fig. 6) as deduced from the capture gamma-ray measurements and their comparison with previous data. The parentheses indicate uncertainties in the number or assignment as discussed in the text. The errors in the spacings between the upper levels is 0.01-0.04 keV, while the error in the spacings between the upper levels and the ground-state band is ≈ 0.5 keV. The following abbreviations are used in the remarks column: G.S.R.B. $\rightarrow$ Ground state rotational band; A $\rightarrow$ also observed following Coulomb excitation (reference 4); B $\rightarrow$ also observed following the 2.2-hr decay of Ta^{178} (reference 3); and C $\rightarrow$ also observed following the 9.3-min decay of Ta^{178} (reference 6).

Energy (keV)	Assignment (K, spin, π)	Remarks
0.0	0, 0^+	Ground state
93.17	0, 2^+	G.S.R.B. A, B, C
306.59	0, 4^+	G.S.R.B. B, C
632.10	0, 6^+	G.S.R.B. B
1058.31	(0, 8^+)	(G.S.R.B.) B
1196.27	0, 0^+	C
1237.26	(0, 0^+)	Tentative
1269.34	0, 2^+	C
1322.85	0, 2^+	
1384.48	(0, 4)	
1402.25	2, $2^{(+)}$	
1420.64	2, $2^{(+)}$	C
1431.25	0, 0^+ ; (0, 2^+)	C
1473.13	(1, 1^+); (0, 2^+)	(C)
1473.32	0, 4^+	
1512.92	0, 4^+	
1514.56	((, 2^+))	Tentative
1550.02	((2, 3^+))	Tentative
1569.78	((2, 3^+))	Tentative
1575.73	((0, 2^+))	Tentative

second requirement of each level was that all gamma-ray transition energies should agree with the level spacings to within 1.5 probable errors. The average energy difference between the level spacings and the gamma rays is 0.4 times the probable error. This better-than-expected agreement is believed to be due to the cancellation of systematic errors. The third requirement was that the sum of the intensities (γ + an estimated c. e.¹¹) of the transitions feeding a level must be equal to or less than the sum of the intensities of the transitions which deplete the level, to within the errors of the measurements.

Although the level scheme in Fig. 6 may still be incomplete, considerable progress has been made in its development. Two low-lying excited-state $K = 0$ bands have been identified with base states at 1196.3 keV and 1237.3 keV. The moments of inertia of these bands are 5% and 10% larger than that of the ground-state rotational band. This suggests slightly larger nuclear deformations for these bands. After a short energy interval (≈ 200 keV) many new levels appear. Another $K = 0$ band with a base (0^+) state at 1431.3 keV and two $K = 2$ bands with base (2^+) states at 1402.2 keV and 1420.6 keV are suggested by this group of levels. The nuclear deformations of these three upper excited-state bands, as derived from their level spacings, are very similar (within 4%) to each other and slightly smaller than the deformation of the ground-state band. The collective nature of these excited states is evident in the strong enhancement of the low-energy transitions between the lower members of these bands. A good example of this collective enhancement is the 73.07-keV transition from the 1269.3-keV level with $K = 0$, spin = 2^+ to the 1196.3-keV level with $K = 0$, spin = 0^+ . This E2

¹¹ L. A. Sliv and I. M. Band, Leningrad Physic-Technical Institute Reports (1956 and 1958); reproduction: Reports ICC 57 K 1, and ICC 58 L 1, issued by the Physics Department of the University of Illinois, Urbana, Illinois (unpublished).

transition competes with an E2 transition (1269.5 keV) to the ground state ($K = 0$, spin = 0^+). The observed gamma intensity of the 73.07-keV transition is only 1/20 that of the 1269.5-keV one. After the removal of the E^5 energy factor, the ratio of the reduced transition probabilities (73.07 keV to 1269.5 keV) is 6.7×10^4 . In the light of present theory it is difficult to even account for the presence of so many relatively strong low-energy transitions in the gamma-ray spectrum.

The success experienced in determining K values for levels through the investigation of relative gamma-ray intensities is very encouraging. In all cases the $(I \rightarrow I + 2)/(I \rightarrow I - 2)$ ratios of E2 gamma intensities lead to clear-cut assignments. The further fact that the observed $(2^+ \rightarrow 2^+)/(2^+ \rightarrow 0^+)$ ratios for transitions from the $K = 2$ bands to the $K = 0$ ground-state band were quite close to the ratio predicted for E2 radiation alone (experimental ratios = 2.2 and 1.4; theoretical ratio = 1.4) demonstrates that the $\lambda \geq \Delta K$ selection rule is effective for these low-lying transitions. This result can be interpreted to mean that these $K = 2$ bands are relatively pure, with only small $K = 1$ admixtures in their wave functions.

A number of coincidence experiments have been performed on the $\text{Hf}^{177}(n, \gamma)\text{Hf}^{178}$ reaction with thermal and epi-thermal neutrons.¹² The 3-dimensional recording and analyzing system of the Argonne chopper group was used for this work.¹³ Only preliminary results are available to date but a number of the important features of the level scheme of Hf^{178} (in Fig. 6) have been confirmed. In particular, the strong coincidence of the 93-keV and 213-keV gammas with the group of gammas lying between 1.0 and 1.3 MeV is quite evident. Further, there is some evidence for coincidences between the high-energy gamma rays (6-7 MeV)

¹² Unpublished.

¹³ Argonne National Laboratory Report ANL-6275.

and the low-energy gamma rays which connect the upper levels in Fig. 6. The high-energy coincidence rate with a 140-150 keV gamma is particularly striking. This gamma-gamma coincidence can be associated with the level at 1575.29 keV ($K = 0$, spin = 2^+) and/or the level at 1569.34 keV ($K = 2$, spin = 3^+) and/or the level at 1549.59 keV ($K = 2$, spin = 3^+).

In the level scheme of Hf^{178} , as presented in Fig. 6, there are 121 transitions that correspond to a change of spin of 2 units or less. Of these, 93 are within the energy range of the bent-crystal spectrometer. The observation of 54 (60%) of the observable transitions is considered to be quite satisfactory since additional selection rules may exist for these gamma transitions.¹⁴ As the levels in Fig. 6 are either confirmed or rejected by future experiments, it may be possible to employ the unused members of the gamma spectrum to extend the level scheme of Hf^{178} to higher energies.

A more detailed discussion of this work will appear in a paper being prepared for publication in the Physical Review.

¹⁴ G. Alaga, Nuclear Phys. 4, 625 (1957).

I-80-30 Molecular Beam Studies

(51210-01)

William Childs, John Dalman, Dieter von Ehrenstein,
and Leonard Goodman
Reported by William Childs

1. IMPROVEMENTS AND MODIFICATIONS OF THE MARK II ATOMIC-BEAM MACHINE

Nearly all of the design work for modification of the universal detector has been completed, and shop work is in progress on several components. In the new system, the radius of the ion trajectory in the mass-spectrometer magnet is increased from 4 in. to 12.5 in.

This will permit the ion accelerating voltage to be raised from the present 1-2 kv to 10 kv for all masses, so that the resolution will be less impaired by an energy spread originating in the extraction of ions from the ionization region. A further consequence of the new design will be to remove the detector (an electron multiplier) from the fringing field of the spectrometer magnet.

A new and completely different ion box has been designed to eliminate many of the problems associated with the first model. Ions will be formed by electron bombardment, as previously, but will be extracted in the direction of the atomic beam instead of perpendicular to it. Ionization can thus be carried out along at least as great a beam length as before, but the ions will be extracted through a thin slit instead of through a very wide aperture. After extraction, the ions will be accelerated and then deflected through 90° by means of a small electrostatic condenser. After further acceleration, the ions will present a very small, energetically homogeneous source for the mass analyzer. Since angular spread in the ion beam can greatly reduce resolution, however, it is planned at this point to introduce a pair of small quadrupole lenses to render the beam sufficiently parallel. The new ion box will be in contact with a liquid nitrogen trap, and will have provision for baking. Shielding will also be introduced to reduce the stray magnetic field in the ion box to an acceptable value.

In most electron-bombardment detectors for atomic beams, nearly all the ions formed and detected (even when using a mass analyzer) originate in the residual gas instead of the atomic beam. Although various synchronous detection schemes are in use elsewhere, a different approach appeared desirable to us. A "forward-backward" scaling system is being constructed which, in conjunction with beam-modulation techniques, will perform the same function with greater discrimination. Square-wave modulation can be introduced either by chopping the beam mechanically or by modulating the rf signal used to induce transitions.

During the half-cycles when the beam is "on", the scaler will add all ion pulses received from the multiplier, and during the half-cycles when the beam is "off", the scaler will subtract all (background) pulses received. The counting rate due to the beam alone may then be examined after any integral number of cycles, and the statistics will, of course, improve as the counting time is increased. The maximum counting rate of the scaler will be 10^6 sec^{-1} , and the system should give good statistics quickly, even for noise-to-signal ratios of 100. At the maximum counting rate of 10^6 sec^{-1} , even a signal of 10^3 sec^{-1} can be measured to within about $\pm 10\%$ within 2 min. For higher counting rates, a narrow-band ac detector can be used.

Because of the large number of ions produced (in the universal detector) from the "rest gas", it is clear that a very clean vacuum is desirable in the detection region. It has been found that a 400-liter/sec titanium getter-ion pump does a far better job on our detector chamber than did the Hg diffusion pump previously used. In addition, no refrigeration or trapping is required. A vacuum of 2×10^{-7} mm is attainable in a few hours, and 5×10^{-8} mm can be reached in 2-3 days without baking.

2. OPERATION OF THE MARK II MACHINE

A considerable amount of operating experience has been gained with the new machine with beams of K^{39} . One feature of the surface-ionization detector used is unusual and of interest. In conventional atomic-beam machines, the detector is located immediately behind the detector slit so that the two beams of atoms (corresponding to the two possible beam paths in the inhomogeneous "B" magnet) are refocused through the slit, and are immediately detected. In the new machine, however, the hot-wire detector is located some distance behind the detector slit, with the result that the two beams, after being refocused

through the slit, again diverge and are well separated in space by the time they reach the detector. The nonresonant part of the beam, often referred to as the "noise," does not, in general, have the same angular divergence as the signal when it passes through the slit, and thus one may achieve unusually high signal-to-noise ratios by judicious placement of the detector. Resonances in K^{39} are often seen with a signal-to-noise ratio of 200 in the new machine, while 25 is a good figure for the Mark I. This new technique cannot increase the signal itself, however.

The measured absolute size of the flop-in signal indicates that about 50% of all atoms that can be flipped and observed do indeed contribute to the resonance. It may be mentioned that the present, temporary oven mount does not have provision for adjustment of its tilt (a feature which has been found critical on the Mark I). Progress is being made on the permanent oven mount which does include this feature, and it is not unreasonable to expect still further improvement in signal-to-noise ratio when the oven can be tilted.

Comparison of the resonance frequency as a function of position in the C magnet (where resonance is induced) was obtained by the use of the four rf loops available. Such studies show that even when the inhomogeneous deflecting magnets are fully energized (20 kilogauss), the central half of the C field has a homogeneity of about 1 part in 10^4 , and it appears to be better when the deflecting fields are reduced.

After completion of the tests using K^{39} , we switched to atoms in the $P_{1/2}$ atomic ground state of the stable indium isotopes. These atoms, which can also be detected with a surface-ionization detector, have deflections of only about 1/5 that of K^{39} because of the smaller electronic g factor and the higher oven temperature required.

Resonances were quickly observed and, after optimizing the position of the detector wire as described above, signal-to-noise ratios about an order of magnitude better than those seen in the Mark I were observed.

3. THE "ANOMALOUS" RESONANCE IN INDIUM

In preparing to make measurements on the radioactive nucleus In^{110} in the old machine, it was decided to check out the machine with nonradioactive In^{115} first. One reason for this was that no observations had been made on indium since the great improvement in homogeneity of the C field several years ago. All observations showed not only the normal transition, but also a second transition a little higher in frequency for any given field. Subsequent quantitative investigations showed that (1) the "anomalous" resonance always appeared when using the Mark I machine, (2) its separation from the normal transition was proportional to the field, and (3) the "anomalous" transition could not be observed on the Mark II machine, although the normal transition was very sharp and clear.

The explanation of the anomaly, though not arrived at immediately, is entirely satisfactory. Figure 7 shows schematically the hyperfine structure of the atomic $P_{1/2}$ ground state of In^{115} as a function of magnetic field strength. For most atoms, a field intensity of 5000 gauss

Fig. 7. Hyperfine-structure diagram for the $P_{1/2}$ atomic ground state of In^{115} .

would be represented by a line at least as far to the right as that labelled "II" in the figure. For In^{115} , however, the hyperfine splitting $h\Delta\nu$ is so large that line II represents about 20 000 gauss (the intensity of the inhomogeneous fields in the Mark II machine), and line I about 6000 gauss (the intensity of the inhomogeneous fields in the Mark I machine). Line "c" represents a weak field such as that used in the C fields of both machines for the indium transitions. Figure 8 shows the situation at

weak field in more detail. Of the transitions indicated, only that labeled "1" is of the usual single-quantum type. All of the multiple-quantum transitions at this weak field have very nearly the same frequency as the single-quantum transition, and thus it is not always possible to be sure which transition is being observed.

In order for an atom which has undergone a transition to be refocused to the detector, the effective magnetic moments in the two inhomogeneous fields must be equal and opposite (for the design in both the Argonne machines). The force on an atom in these fields may be expressed as

$$F = - \text{grad } W = - \frac{\partial W}{\partial H} \text{ grad } H .$$

From this, it is clear that the effective moment for a particular state is just the negative of the slope shown in Fig. 7. If the inhomogeneous fields are operated at the field "II" in Fig. 7 (as is the case on the Mark II machine), it is seen that the only transitions which can be refocused are those labeled 1, 2, 3, . . . , 10. This is in complete agreement with what is observed on the new machine when the inhomogeneous fields are at 20 000 gauss. In the Mark I machine, however, the maximum inhomogeneous fields (about 6000 gauss) correspond to position "I", where the only transitions normally expected are of the type 1, 2, . . . , 7, 8, 9, 10. It can be seen, however, that transitions 11-19 can also be seen under these conditions. Calculations show that transitions 16-19 lie exactly $2g_I \mu_0 H$ above transitions 11-14, respectively. The spectrum actually observed on the Mark I machine shows two peaks, separated by $2g_I \mu_0 H$ in accord with the theory. Transitions 16-19 were also observed on the Mark II machine, but only after reducing the strengths of the inhomogeneous fields to the value given by "I".

It would appear that the technique of measuring the separation of the group 16-19 from the group 11-15 would be a good method for measuring the nuclear g factor for several radioactive indium nuclei, and considerable thought is being given to this possibility. A likely candidate for early work will be 54-min $\text{In}^{115\text{m}}$, for which the nuclear moment and spin have been measured¹ to be $+(4.4 \text{ nm} \pm 10\%)$ and 5, respectively.

4. OPERATION OF THE MARK I MACHINE

The Mark I machine, in addition to extensive use in connection with understanding the "anomalous" resonance in the $P_{1/2}$ state of In^{115} , was used for preliminary work on two radioactive nuclei — 85-day Sc^{45} and 66-min In^{110} . For various reasons, it has

¹L. S. Goodman and S. Wexler, Phys. Rev. 108, 1524 (1957).

been decided to concentrate on the latter isotope, which is produced by the $\text{Ag}^{107}(\alpha, n)\text{In}^{110}$ reaction in the Argonne cyclotron. Initial runs indicate that adequate production of the In^{110} can be achieved. As mentioned above, 54-min $\text{In}^{115\text{m}}$ may be reinvestigated in an attempt to measure the nuclear dipole moment more precisely.

5. NEW EQUIPMENT

A high-precision signal generator has been purchased from Solartron Elektronik, GMBH, Munich. The maximum rf output level is 1 v from 50 kc/sec to 50 Mc/sec, and 2 v from 50 Mc/sec to 500 Mc/sec, and may be attenuated with a built-in precision attenuator calibrated in 1 db steps to 99 db. The frequency is locked to a crystal (stable within 2 parts in 10^8) every 10 kc/sec to 50 Mc/sec and every 100 kc/sec to 500 Mc/sec. Between crystal checkpoints it is resettable to ± 10 cps below 50 Mc/sec and ± 100 cps above 50 Mc/sec. Provision is made for either am or fm modulation.

I-144-15 Investigation of Scintillators

(51300-01)

L. J. Basile and W. L. Buck
Reported by L. J. Basile

EFFECT OF STYRENE MONOMER ON THE FLUORESCENCE PROPERTIES OF POLYSTYRENE

I. Introduction

This report presents results obtained in a detailed investigation of some luminescence properties of polystyrene, a typical plastic solvent used in preparing a plastic scintillator. An attempt was made to see if any correlation exists between the luminescence properties of polystyrene and the average molecular weight of the

polymer. Particular emphasis has been placed on measurements of the decay times and fluorescence spectra for polystyrene samples having different average molecular weights.

The fluorescence spectrum of polystyrene has been measured by excitation with uv radiation,¹⁻⁴ x rays,^{5,6} and β particles.⁴ There are some differences in the shapes of the reported spectra, which may well be due to differences in experimental conditions. All of the reported spectra show one characteristic feature, namely, a maximum fluorescence intensity in the region of 310 m μ .

In all previously reported investigations (both here and elsewhere) on the use of polystyrene in plastic scintillators, it was assumed that the polystyrene sample was completely polymerized. It is well known that bulk thermal polymerization of styrene monomer leaves some residual styrene monomer within the polystyrene, but heretofore it tended to be ignored on the tacit assumption that its concentration would be too low to have any appreciable effect on the results. The data obtained in this investigation show, however, that residual styrene monomer does have a significant effect on the luminescence properties of polystyrene.

The samples were thermally polymerized in a constant-temperature oil bath. The molecular weight of the polystyrene was varied

¹R. K. Swank and W. L. Buck, Phys. Rev. 91, 927 (1953).

²L. Pichat, P. Pesteil, and J. Clement, J. Chim. Phys. 50, 26 (1955).

³G. W. King, AWRE Report No. -0-12/54 (1954).

⁴E. A. Andreyeshchev and I. M. Rozman, Optika i Spectroscopia 2, 488 (1957).

⁵W. S. Koski, Phys. Rev. 82, 230 (1951).

⁶F. H. Krenz, Trans. Faraday Soc. 51, 1 (1955).

by changing the temperature of polymerization. The length of time the samples were kept in the oil bath was made shorter for higher temperatures. Samples polymerized at 85°C were held at temperature for 18 days; at 110°C, 125°C, and 145°C for a period of 10 days; at 170°C for a period of 150 hours. Polystyrene of high molecular weight was obtained by adding varying amounts of a cross-linking agent to the styrene monomer. The cross-linking agent was 80-90% pure divinylbenzene, freshly distilled before use. Samples containing the cross-linking agent were polymerized at 145°C for 10 days.

Samples of cast polystyrene were prepared in the following manner. Polystyrene, previously polymerized under the above conditions, was put into a vial which was then attached to the vacuum line and kept evacuated. The vial was then carefully heated so that the sample was fairly fluid. This temperature was maintained for a 2-3 hour period with occasional pumping to remove any volatile components. When the sample was free of bubbles, it was slowly cooled and then removed from the vacuum line.

The amount of residual styrene monomer was determined by using the extinction coefficient of styrene monomer in chloroform to measure the absorbance at 291 mμ of a 5-gm/liter chloroform solution of thermally polymerized polystyrene.

II. Results and Discussion

a. Absorption Studies

From absorption studies, the concentration of residual styrene monomer was found to range from 0.5% for the samples polymerized at 170°C to at least 7% for the samples polymerized at 85°C. There was approximately the same amount of styrene monomer in the 125°C, 145°C, and 170°C samples. Samples polymerized at 110°C contained about 1% styrene monomer. The 85°C samples were badly

crazed after standing in air for two weeks. Absorption studies on cast polystyrene showed that less than 0.05% of monomer was left in samples prepared by this method.

b. Fluorescence Spectra

Fig. 9. Changes in the fluorescence spectra of polystyrene in cyclohexane upon addition of styrene monomer. Concentration of polystyrene was 4 g/liter. The concentration of the styrene monomer (ml/liter solution) is indicated on each curve. Excitation was by 2650-Å Hg radiation.

Figure 9 shows the fluorescence spectrum of a cyclohexane solution of polystyrene which was previously polymerized at 145°C. The shape of the spectrum is quite symmetrical and does not show any peaks in the region around 310 mμ. Addition of trace quantities of styrene monomer to the solution enhances the fluorescence intensity. A rather sharp band of fluorescence, due to the presence of styrene monomer, appears around 305-308 mμ. The fluorescence spectrum (not shown) of dilute solutions of styrene monomer in cyclohexane

are very similar in shape to the spectra shown in Fig. 9.

The fluorescence spectra of the solid polystyrene samples are shown in Figs. 10 and 11. The spectra shown in Fig. 10 are for samples which were polymerized at the indicated temperatures. The fluorescence spectrum of the solid samples polymerized at the lower temperatures (85°C, 110°C, and 125°C) are quite similar to the spectrum for the dilute styrene solutions shown in Fig. 9. The intensity peak at about 305-310 mμ is due to the presence of residual styrene monomer in the bulk-polymerized samples. As the temperature of polymerization is increased, there is a decrease in the fluorescence

Fig. 10. Fluorescence spectra of cast polystyrene and polystyrene samples that had been bulk polymerized at the indicated temperatures. Excitation was by 2650-A Hg radiation.

Fig. 11. Fluorescence spectra of cross-linked polystyrene. The cross-linking agent was divinylbenzene in the concentration indicated (ml/liter styrene). Excitation was by 2650-A Hg radiation.

intensity in the region from 300-330 mμ and the spectrum shifts to slightly longer wavelengths. The spectrum for cast polystyrene, which has been shown to be free of styrene monomer, is symmetrical and similar to that observed for polystyrene in cyclohexane. The shape of the fluorescence spectrum of cast polystyrene is independent of the average molecular weight. Although there may be slight differences in intensity, the general shapes of the fluorescence spectra for different cast polystyrene samples are quite similar to the one shown.

For all practical purposes the absorption spectra of the samples polymerized at 125°C, 145°C, and 170°C were identical to each other. It was also shown that the concentration of residual styrene was the same ($\approx 0.5\%$) for the samples polymerized at these three different temperatures. Therefore, the decrease in the styrene fluorescence in going from the polystyrene samples polymerized at 125°C to those polymerized at 170°C must be due to changes in the polystyrene molecule.

The average molecular weight of polystyrene decreases as the temperature of polymerization increases (Table VII). The observed

TABLE VII. Intrinsic viscosity, measured at 25°C, for various polystyrene samples. Units are deciliters per gram.

Polymerization temperature				145°C cast polystyrene
110°C	125°C	145°C	170°C	
1.47	1.21	0.82	0.37	0.80
1.47	1.22	0.81	0.37	0.81

decrease in fluorescence intensity between 125° and the 170° samples could be related to a corresponding increase in the number of shorter polymer chains. That is to say, for some unknown reason the quenching probability may be higher for shorter polymer chains than for longer ones. Evidence supporting this conclusion has also been obtained from our studies by use of β excitation (Sec. IIc).

The most conspicuous effect of adding crosslinking agent (Fig. 11) is to shift the emission spectrum to longer wavelengths. This change is probably due to a combination of several effects. One is that adding crosslinking agent (divinylbenzene) results in more complete polymerization, i. e., more complete removal of styrene monomer. Another effect is a change in the structure of the polymer as the polystyrene chains are increasingly interconnected by phenyl groups⁷. This changes both the optical and physical properties of the polymer. Crosslinked polymer is much harder, more brittle, and very difficult to dissolve in typical organic solvents.

⁷ R. H. Boundy and R. F. Boyer, Styrene: Its Polymers, Copolymers, and Derivatives (Reinhold Publishing Corporation, New York, 1952).

c. Intensity Measurements under β -Particle Excitation

The results of the light-output measurements are listed in Table VIII. Among the different polystyrene samples in the series

TABLE VIII. Relative light outputs for thick polystyrene samples polymerized at different temperatures T_p . The samples were excited with β particles from a Sr^{90} source^p and the light outputs were measured by an RCA-6903 photomultiplier. Values are expressed relative to a standard anthracene crystal having the same approximate dimensions, uncorrected for the spectral response of the phototube. Each value listed represents a different sample.

T_p ($^{\circ}C$)	85 $^{\circ}$	110 $^{\circ}$	125 $^{\circ}$	145 $^{\circ}$	170 $^{\circ}$
	1.5	4.4	3.9	3.2	2.8
	1.5	4.5	4.4	3.2	
	1.4	4.4	3.8	3.2	
Cast polystyrene samples					
		1.4	1.6	1.2	0.9
		1.4	1.2	1.2	
		0.9	1.2	1.1	

polymerized at each particular temperature, the bulk-polymerized sample (which contains residual styrene monomer) has a much higher light output than the corresponding cast polystyrene sample. These results suggest that residual styrene monomer is partly responsible for the relatively high light output observed in polystyrene.

When bulk-polymerized polystyrene is excited by β rays, the amount of direct excitation of residual styrene monomer is negligible. However, the intensity with styrene present is almost three times that

without it. Therefore, it is inferred that excited polystyrene is transferring energy to the residual styrene monomer.

The light output for the 85°C sample is quite low, while the residual styrene monomer concentration is quite high ($\approx 7\%$). A similar decrease in intensity was observed under uv excitation (Fig. 10). The decrease in intensity is probably due to some type of self-quenching action by the styrene monomer itself. Studies in liquid solution have established that self quenching of styrene monomer is rather high (Fig. 9). Unfortunately we were not successful in our attempts to cast 85°C bulk-polymerized polystyrene.

d. Lifetime Studies

The decay times of scintillation pulses from polystyrene samples are tabulated in Table IX, which also lists values obtained for aerated cyclohexane solutions of styrene monomer and of polystyrene.

For styrene monomer in cyclohexane, the decrease in decay time with an increase in concentration is probably due to self quenching. The decay time of styrene monomer is relatively short compared to that of the polymerized material. The decay time of polystyrene in solution is also shorter than in the rigid medium. It must be remembered that the results are for an aerated solution in one case and a rigid medium in the other. Oxygen quenching, self-quenching, and viscosity effects are important factors in the study of liquid solutions.^{8,9} As a rule, oxygen quenching tends to shorten the average lifetime of the excited molecules. Because of the conditions used, one should look upon the above results for the liquid medium as lower limits for the decay time of these molecules in cyclohexane.

⁸ I. B. Berlman, J. Chem. Phys. 33, 1124 (1960).

⁹ A. Weinreb, J. Chem. Phys. 35, 91 (1961).

TABLE IX. Decay times of polystyrene samples. Thick samples measured 12 mm thick by 2 cm in diameter; thin samples were 0.5 mm thick by 5 mm in diameter. For cross-linked polystyrene, the values in parentheses are the concentration of cross-linking agent per 1000 ml of styrene. Also shown are the decay times of styrene and of polystyrene in cyclohexane at the concentrations indicated. Units are nanoseconds. Each value listed is for a separate sample.

Polymerization temperature					Cross-linked polystyrene	
85°	110°	125°	145°	170°		
Thick polystyrene samples						
8.7	13.3	14.7	14.1	13.3	14.9	(0.04)
8.4	13.0	14.4	14.2	13.5	15.0	(1.0)
8.9	13.0	14.4	14.0	13.8	15.0	(2.0)
8.6	13.2	14.7	14.8	13.8	14.8	(5.0)
	13.2				15.1	(7.0)
Thin polystyrene samples						
7.2	12.6	13.5	13.7	13.3		
7.6	12.8	14.0	13.5	13.2		
8.0	12.7	13.7	13.5	13.3		
8.2	12.8	14.0	13.2			
Cast polystyrene samples						
	20.0	20.3	19.5	19.4		
	19.9	19.8	19.2	18.9		
	19.2	20.0	19.0			
Liquid solutions						
Styrene in cyclohexane				Polystyrene in cyclohexane		
0.1 ml/liter	6.5			(4 g/liter)	12.7	
	6.5				12.6	
0.2 ml/liter	5.1					
	5.1					

Let us consider the results of the decay-time measurements for polystyrene samples which were polymerized at different temperatures. Absorption and emission studies have shown that these samples are two-component systems. The bulk material is polystyrene and can be considered as the solvent. Imbedded in the polystyrene is residual monomer in relatively dilute concentrations. It has been shown that this dilute monomer is highly fluorescent.

The residual styrene monomer affects the decay time of the scintillation pulse from the polystyrene samples. Cast polystyrene, which has been shown to be free of styrene monomer, has a relatively long lifetime. Samples that were polymerized at 125°C , 145°C , and 170°C contain approximately the same amount of residual styrene monomer (0.5%). The measured decay times of these samples are approximately the same and are at least 25% shorter than for cast polystyrene. This shows how a small amount of styrene monomer affects the decay time of polystyrene. As the concentration of styrene monomer is increased (in going successively to the 110°C and 85°C samples), the decay time continues to decrease quite markedly.

These results are also interpreted to mean that polystyrene is transferring energy to the residual styrene monomer, which acts as if it were a typical fluorescent solute incorporated in a plastic medium. The measured decay time is quite dependent on the styrene concentration, i. e., the decay time becomes shorter as the concentration of styrene is increased. Finally the concentration of styrene becomes so high (7% for the 85°C sample) that what is being measured is the decay time of styrene in a rigid medium. This value is quite close to the value for the decay time of styrene in cyclohexane.

There is a slight suggestion that the decay time depends somewhat on the average molecular weight. The decay times for the 170°C samples are consistently shorter than those for the 125°C samples.

Since the styrene concentration is the same, the shorter polystyrene chains must have a higher probability of quenching than the longer chains. This also supports the conclusions reached from the studies of the emission spectra.

The decay times of the various cast polystyrene samples are relatively long. Once again there is a suggestion that the longer chains have a slightly longer lifetime than the shorter chains. However, the reduced accuracy of the measurements of the relatively long decay times makes it difficult to decide whether the observed effect is real or due to experimental error.

The decay time measured for cross-linked polystyrene is approximately 10% higher than for polystyrene polymerized at 145°C. It is apparent that the amount of cross-linking agent has little effect on the lifetime; increasing the concentration of cross-linking agent almost 20-fold makes no measurable change in the decay time. If divinylbenzene acted as a quencher, one would expect the lifetime to decrease with an increase in concentration of the cross-linking agent. The lengthening of the decay time for cross-linked polystyrene could be due to such effects as more complete removal of the residual styrene monomer. Perhaps the various effects tend to counteract each other to produce the observed constant decay time for cross-linked polystyrene.

It must be emphasized that the measured decay times for the samples reported were obtained by fitting the best straight line to the semilogarithmic plots of the decaying parts of the pulse. This assumes that the decaying part of the pulse can be adequately described by the simple exponential relation $I = Ae^{-t/\tau}$.

These results are part of a paper which has been accepted for publication in the Journal of Chemical Physics.

II. MASS SPECTROSCOPY

II-28-5 Kinetics of Chemical Reactions in the Gas Phase (51300-01)

J. Berkowitz and S. Wexler

After more than a generation of experimentation, the methylene radical (CH_2) has finally been identified as a chemical entity. The far ultraviolet spectrum of this species was obtained by Herzberg¹ who used flash photolysis to decompose diazomethane (CH_2N_2), and an auxiliary light source to detect absorption in the dissociated gas. The conditions for this experiment were critical; it was necessary to use an excess of inert "buffer" gas and to search for the absorption spectrum within 50 μsec of the flash.

The assignment of the observed absorption was checked by isotopic substitution of both deuterium and C^{13} in the CH_2N_2 . Further analysis showed that the absorption spectrum was part of a Rydberg series, whose terms fit the formula

$$\nu = 83\,851 - \frac{R}{(n - 0.12)^2} .$$

Therefore, the limit of the series ($83\,851\text{ cm}^{-1}$ or 10.396 eV) represents an ionization potential of CH_2 . Herzberg gives some strong arguments to prove that this is the ionization potential of the ground state of CH_2 . An earlier mass spectrometric study² arrived at a

¹ G. Herzberg, Can. J. Phys. 39, 1511 (1961); Proc. Roy. Soc. (London) A262, 291 (1961).

² A. Langer, J. A. Hipple, and D. P. Stevenson, J. Chem. Phys. 22, 1836 (1954).

significantly different value, $11.7_{\text{S}} \pm 0.1$ ev, for the appearance potential of CH_2^+ . In this latter experiment, diazomethane was allowed to pass through a quartz tube which could be heated to any desired temperature up to 1000°C . The end of the quartz tube had a pinhole leak, through which the gas flowed into a conventional mass spectrometer.

The appearance potential of CH_2 from cold diazomethane, represented by the reaction

was first measured. The authors then set the energy of their ionizing electron beam slightly below this appearance potential and began to heat the quartz tube. At a temperature of $\approx 400^\circ\text{C}$, the ion signal at mass 14 began to rise and appeared to reach a peak at ca. 900°C . The investigators concluded from this evidence that the sequence of events was

The appearance potential for this latter process was reported as $11.7_{\text{S}} \pm 0.1$ ev.

The discrepancy between the recent spectroscopic analysis and the older mass spectrometric results prompted us to reinvestigate the mass spectral experiment. Accordingly, we fashioned a quartz reactor, shown in Fig. 12. The design permits fairly rapid pumping of the pyrolysis products from the quartz tube. The assembly was tested by pyrolyzing dimethyl mercury $(\text{CH}_3)_2\text{Hg}$ and searching for the appearance of methyl radical CH_3 .

The rate of flow of dimethyl mercury from the quartz reactor was determined by observing the intensity of mass 232, the

Fig. 12. Quartz reactor for pyrolysis of $\text{Hg}(\text{CH}_3)_2$ and CH_2N_2 .

$[(\text{CH}_3)_2\text{Hg}^{202}]^+$ ion, in the mass spectrometer with the movable slit alternately permitting and then blocking the flow to the ionization chamber. This "shutter effect," which distinguishes between the mass -232 ions formed from $(\text{CH}_3)_2\text{Hg}$ molecules in direct flight from the reactor and the mass -232 ions formed from the ambient gas, had a magnitude of 3680 arbitrary intensity units when the quartz reactor was at room temperature and 260 units when at a measured temperature of 900°C . This diminution was evidently due to pyrolysis of the dimethyl mercury. Simultaneously, the CH_3^+ intensity from CH_3 radicals increases. In Fig. 13 (a) the ratio of the intensity at, say, 13 ev to that at 20 ev is observed to increase with the temperature of the quartz reactor. At the lower temperature, virtually the only source of CH_3^+ is the reaction

At the higher temperatures, pyrolysis produces free methyl radicals which contribute CH_3^+ ions by simple ionization, i.e.,

The importance of the latter reaction in comparison with the former becomes most pronounced below 14 ev. The appearance potential of CH_3^+ (10.0 ± 0.3 ev) from methyl radical, as measured in this experiment, is in good agreement with previous measurements by both spectroscopic³ (9.843 ev) and mass spectrometric^{2,4} methods. A further verification of the source of the methyl radicals was obtained by placing the movable slit between the quartz reactor and the ionization chamber.

³ G. Herzberg and J. Shoosmith, Can. J. Phys. 34, 523 (1956).

⁴ F. P. Lossing, K. W. Ingold, and I. H. S. Henderson, J. Chem. Phys. 22, 621 (1954); O. Osberghaus and R. Taubert, Z. phys. Chem. 4, 264 (1955).

Fig. 13. Change in the ionization efficiency curves of mass 15 $[\text{CH}_3]^+$ from $\text{Hg}(\text{CH}_3)_2$ with the temperature of the quartz reactor. The intensities have been normalized at 20 ev electron energy for all three heating conditions. The intensity at 13 ev relative to that at 20 ev is seen to rise with temperature.

Figure 14 demonstrates that the intensity of CH_3^+ from CH_3 radicals (when the energy of the ionizing electrons was 11.5 ev) decreases rapidly as the temperature of the reactor decreases. This figure also clearly shows the effect of alternately blocking the passage of gas from quartz tube to ionization chamber, then permitting gas to pass through a slit.

The experiment was then repeated with diazomethane. The latter was prepared by the reaction of KOH on N-nitroso methyl urea. The procedure adopted was similar to the one described by Arndt,⁵ except that n-amyl ether was substituted for the ethyl ether used by Arndt and the CH_2N_2 was isolated free of solvent. The appearance potential of the

⁵

F. Arndt, Organic Synthesis, Collective Volume II, edited by A. H. Blatt (John Wiley and Sons, New York, 1943), p. 165.

Fig. 14. Shutter effect on mass 15 (CH_3^+ from CH_3 radicals). The energy of the ionizing electrons is 11.5 ev, which is below the threshold for dissociative ionization of $\text{Hg}(\text{CH}_3)_2$. The intensity of the peak is seen to decrease rapidly as the quartz reactor cools, and pyrolysis of $\text{Hg}(\text{CH}_3)_2$ ceases to be important.

parent peak (CH_2N_2^+ was measured to be 9.2 ± 0.3 ev, in good agreement with earlier work by Lossing.^{1(b)} This corroborated that CH_2N_2 had indeed been prepared. The sample was then heated. In Fig. 15 (a), the decay of CH_2N_2^+ as a function of temperature is clearly evident. Figure 15 (b) shows the effect of this heating on mass 14 [$\text{CH}_2^+(?)$] at an ionizing energy of 19 ev. Figure 15 (c) shows the effect of heating when the ionizing energy used is 12.5 ev. This latter energy should be sufficient to directly ionize CH_2 radicals if any are present, but be below the appearance potential of CH_2^+ and or N^+ from CH_2N_2 . The same temperature range is covered in all three experiments. No evidence could be obtained for the formation of CH_2^+ , even after 90% of the CH_2N_2^+ peak had decayed as a result of pyrolysis.

The current result is thus unable to confirm the earlier mass spectrometric study. Our experiment has been repeated several times with different modes of preparation of CH_2N_2 , but never were we able to detect any signal of CH_2^+ from CH_2 . Comparison of our experimental

(a)

(b)

(c)

Fig. 15. (a) Diminution in intensity of mass 42 (CH_2N_2^+) as the temperature of the quartz reactor is increased. About 90% of the diazomethane apparently is destroyed by pyrolysis. (b) Effect on the intensity of mass 14 [CH_2^+ (?)] when the temperature of the quartz reactor is increased. The Energy of the ionizing electrons is 19 ev. (c) Effect on the intensity of mass 14 when the temperature of the quartz reactor is increased and the ionizing energy is 12.5 ev. There is no evidence for the formation of CH_2^+ from methylene radical, whose appearance potential should be less than 12.5 ev. The ionizing voltage used is below the appearance potential of CH_2^+ or N^+ from CH_2N_2 .

arrangement with that of Herzberg suggests that the CH_2 radicals probably have ample time to recombine on the walls of the reaction vessel before they can be detected. Virtually any collision is likely to lead to loss of CH_2 . Hence the present work, while not necessarily confirming Herzberg's result, casts considerable doubt upon the only competing value for the ionization potential of CH_2 .

V. THEORETICAL PHYSICS, GENERAL

V-3-9 Dynamics of Nuclear Collective Motion

(51210-01)

D. R. Inglis

HEAVY PARTICLE STRIPPING AND THE STRUCTURE OF Li^6

A qualitative explanation can be given for the interesting contrast observed between the angular distributions of the groups going to the ground state and the second excited state in the reaction $\text{C}^{12}(\text{Li}^6, \alpha)\text{N}^{14}$. The ground-state group of alphas has a peak only in the backward direction because only the mechanism of "heavy-particle stripping" can satisfy the requirements on the orbital angular momentum for the ground state of N^{14} (primarily a ^3D). The "deuteron" of the incident Li^6 initially has insufficient angular momentum about the final core but acquires more (Fig. 16)

because the force attracting it toward the target nucleus is not directed toward the center of mass of the twelve nucleons (eight from the target C^{12} and four from Li^6) which form the core of the final N^{14} . The other four nucleons from the target form the product alpha. The second excited state of N^{14} is approximately a ^3S and there is no such angular momentum limitation so both light- and heavy-particle stripping occur and give rise to forward and backward peaks. The validity of the cluster model for Li^6 is also discussed in a paper to appear in the Physical Review.

Fig. 16. Schematic representation of the proposed mechanism of heavy-particle stripping. The attraction F acting on the "deuteron" (the pair of small circles) of the incident Li^6 gives the "deuteron" additional angular momentum about the center of mass of the core of the final N^{14} nucleus (dashed triangle). The other alpha (medium sized circle with cross marks) is emitted backward in the reaction.

V-12-2 Accuracy of the Superconductivity Approximation for Pairing
 Forces in Nuclei (51210-01)

A. K. Kerman, R. D. Lawson, and M. H. Macfarlane
Reported by R. D. Lawson

This project has been completed and the results have been published in a report entitled "Accuracy of the Superconductivity Approximation for Pairing Forces in Nuclei," A. K. Kerman, R. D. Lawson, and M. H. Macfarlane, Phys. Rev. 124, 162-167 (October 1, 1961).

V-14-2 Properties of Nuclei with Neutrons and Protons in the
 $1f_{7/2}$ Shell (51210-01)

R. D. Lawson

This project has been completed and the results have been published in a report entitled "Properties of Nuclei with Neutrons and Protons in the $1f_{7/2}$ Shell," R. D. Lawson, Phys. Rev. 124, 1500-1505 (December 1, 1961).

V-25-1 Scattering of Alpha Particles by a Vibrational Nucleus
(51210-01)

L. J. Tassie^{*}

The elastic and inelastic scattering of α particles by a vibrational nucleus is calculated by use of the plane-wave Born approximation. The excitation of both single-phonon and two-phonon states is considered. The effect of the diffuseness of the nuclear surface is included. The result for elastic scattering and the excitation of the single-phonon 2^+ and 3^- states is in good agreement with experiment for Ni^{60} . The approximations used are discussed, and it is suggested that excitation of 0^+ , 1^- , and 5^- states should provide the best experimental test of the theory of two-phonon excitation of nuclei. The energies of the vibrational states are also considered.

A report on this work has been prepared for publication.

* Now returned to the Research School of Physical Sciences, Australian National University, Canberra, Australia.

V-33-4 Flux Quantization and Time-Reversal Degeneracy (51300-01)
 (formerly "Time Reversal, Flux Quantization, and the
 Current-Carrying State")

M. Peshkin and W. Tobocman*
 Reported by M. Peshkin

The magnetic flux trapped by a ring superconductor has been found to be quantized in (Gaussian) units of

$$L = \pi \hbar c / e. \quad (1)$$

The previously reported idea¹ that time-reversal degeneracy relates this quantization to the Meissner effect has been developed further. It is now necessary to assume only that the penetration of the magnetic field into the body of the superconductor may be neglected. By using the time-reversal invariance and gauge invariance of electromagnetic forces, it is then proved that if the magnetic flux is an integral multiple of $\underline{L}$, then the states of the superconductor exhibit time-reversal degeneracy. The present proof of this assertion differs substantially from the earlier proof¹ only in that the motion of the lattice is included.

The time-reversal degeneracy implies that at thermal equilibrium, where states are weighted according to their energy alone, the expectation of the current density vanishes everywhere. This zero-current result is necessary for consistency with the Meissner effect.

Vanishing expectation of the current does not, however, require the quantization of flux in units of $\underline{L}$. An elementary

* Case Institute of Technology, Cleveland, Ohio. Supported in part by a grant from the Sloan Foundation.

¹ Murray Peshkin, Physics Division Summary Report ANL-6488 (January 1962), p. 26.

counter-example is provided by the "di-electron" model, where e in Eq. (1) must be replaced by $2e$.

A paper enlarging on these ideas has been submitted for publication.

V-46-1 Elastic Nucleon-Nucleon Scattering at High Energies and
Small Angles (51151-01)

K. Hiida

The elastic proton-proton scattering cross section for incident energies of 13-28 Bev and for scattering angles of 56.5 and 60.5 milliradians was determined at CERN.¹ The behavior of this cross section in the laboratory system is given by

$$d\sigma_{\text{lab}}/d\Omega \approx (4\pi)^{-2} E_i^2 \sigma_{N-N}^2(E_i) \exp[-a(-t)^{1/2}] , \quad (1)$$

where E_i is the energy of the incident proton in the laboratory system, $(-t)^{1/2}$ is the invariant momentum transfer, $\sigma_{N-N}(E_i)$ is the total cross section for nucleon-nucleon scattering at E_i , and a is nearly constant but depends weakly on E_i and t .

The mechanism to explain the behavior described by Eq. (1) is the double diffraction scattering of a virtual boson by the

¹ G. Cocconi, A. N. Diddens, E. Lillethun, G. Manning, A. E. Taylor, T. G. Walker, and A. M. Wetherell, Phys. Rev. Letters 7, 450 (1961).

Fig. 17. Feynman diagram for our process. The circles on nucleon lines denote the high-energy incident and observed nucleons. The momenta p_i , p_f , q_i , and q_f denote the momenta of the incident, observed, target, and recoil nucleons, respectively; and k and $(p_i - p_f + k)$ are the momenta of two virtual bosons exchanged between the two nucleons.

The cross section is given by

$$\begin{aligned} \frac{d\sigma_{\text{lab}}}{d\Omega} &\approx \left| \frac{1}{(2\pi)^5} \frac{1}{4m} \sum_j \int d^4k \theta(k) D^j(p_i, p_f, k) \right. \\ &\times \{(s_A - m^2) \sigma_{B-N,A}^j(s_A) [g_{B-N,A}^j(s_A, t)]^{1/2}\} \\ &\times \{(s_B - m^2) \sigma_{B-N,B}^j(s_B) [g_{B-N,B}^j(s_B, t)]^{1/2}\} \left. \right|^2, \end{aligned}$$

where m is the nucleon mass. The summation over j is over all possible intermediate boson states; D^j is given by

$$D^j(p_i, p_f, k) = f(k, p_i - p_f + k) [k^2 - \mu_j^2 + i\epsilon]^{-1} [(p_i - p_f + k)^2 - \mu_j^2 + i\epsilon]^{-1},$$

μ^j is the boson mass, and $f(k, p_i - p_f + k)$ is the form factor and is normalized to $f = 1$ when $k^2 = (p_i - p_f + k)^2 = \mu_j^2$; $s_A = (p_i + k)^2$, and $s_B = (q_f + k)^2$ are the squares of the energies of the boson-nucleon systems at (A) and (B); $\sigma_{B-N,A}^j$ and $\sigma_{B-N,B}^j$ are the total cross sections for the boson-nucleon scatterings at (A) and (B) and are nearly constant at high energies; and $g_{B-N,A}$ and $g_{B-N,B}$ describe the diffraction shapes for the boson-nucleon scatterings at (A) and (B), respectively, and are defined by

$$\frac{d\sigma_{B-N}(s, t)}{d\Omega} = \frac{d\sigma_{B-N}(s, 0)}{d\Omega} g_{B-N}(s, t) .$$

Experimentally, g_{B-N} depends strongly on t but weakly on s .

One can see that Eq. (2) can explain the characteristic feature of the nucleon-nucleon diffraction shape (1) in terms of boson-nucleon diffraction shapes: (i) Since $s_A, s_B \propto E_i^2$, the product $(s_A - m^2) \times (s_B - m^2)$ becomes important at high energies so that $d\sigma_{lab}/d\Omega \propto E_i^2$. (ii) It is obvious that $d\sigma_{lab}/d\Omega \propto g_{B-N,A}(t) \times g_{B-N,B}(t)$ because of the weak dependence of $g(s, t)$ on s . Another t -dependent factor $h(t)$ comes from $D(p_i, p_f, k)$. For example, even when $k^2 = \mu^2$, $(p_i - p_f + k)^2$ becomes large when $(-t)$ is large. Numerical calculations show

$$g_{B-N,A}(t) g_{B-N,B}(t) h(t) \propto \exp[-a(-t)^{\frac{1}{2}}] .$$

(iii) It is known experimentally that the form factors $g_{\pi-N}$ and g_{p-p} (the form factor of the proton-proton diffraction peak, which is defined in analogy with g_{B-N}) have a tendency to narrow with increasing s . Since $h(t)$ does not depend on E_i in the approximation retaining only terms proportional to E_i^2 , Eq. (2) can explain the narrowing of the nucleon-nucleon diffraction peak with increasing E_i in terms of $g_{B-N,A}(s_A, t)$ and $g_{B-N,B}(s_B, t)$.

This work was published in Phys. Rev. Letters 8, 148 (1962). There will be no further reports on this subject.

PUBLICATIONS SINCE THE LAST REPORT

PAPERS

ENERGY DISTRIBUTION AND FRAGMENTATION PROCESSES RESULTING FROM ELECTRON IMPACT ON PROPANE AND n-BUTANE

W. A. Chupka and M. S. Kaminsky (Project II-28)
J. Chem. Phys. 35, 1991-1998 (December 1961)

ISOBARIC-SPIN SPLITTING OF SINGLE-PARTICLE RESONANCES

J. B. French and M. H. Macfarlane (Unattached)
Nuclear Phys. 26, 168-176 (July 1961)

INTERFERENCE EFFECTS OF RADIOFREQUENCY FIELDS IN RESONANCE RADIATION

M. N. Hack (Project I-19)
Nuovo cimento 22, 168-177 (October 1, 1961)

ELASTIC NUCLEON-NUCLEON SCATTERING AT HIGH ENERGIES AND SMALL ANGLES

K. Hiida (Project V-46)
Phys. Rev. Letters 8, 148-150 (February 1, 1962)

TOTAL CROSS SECTIONS AT HIGH ENERGIES

K. Hiida, M. Soga, and K. Tanaka (Project V-45)
Phys. Rev. Letters 8, 187-190 (February 15, 1962)

PRECISION MEASUREMENTS OF THE NEUTRON TOTAL CROSS SECTION OF U^{235}

A. Saplakoglu (Project I-3)
Nuclear Sci. Eng. 11, 312 (November 1961)

SYMMETRY THEORY OF THE AHARONOV-BOHM EFFECT: QUANTUM MECHANICS IN A MULTIPLY CONNECTED REGION

L. J. Tassie and M. Peshkin (Project V-33)
Ann. Phys. 16, 177-184 (1961)

DISSOCIATION OF MOLECULES BY NUCLEAR DECAY OF CON- STITUENT ATOMS

S. Wexler (Project II-38)
Chemical Effects of Nuclear Transformations (International
Atomic Energy Agency, Vienna, 1961), pp. 115-145

ABSTRACTS

DOUBLE-OVEN EXPERIMENTS WITH LITHIUM HALIDES

J. Berkowitz, H. A. Tasman, and W. A. Chupka..(Project II-29)
 Abstracts of Scientific Papers Presented at the 18th
 International Congress of Pure and Applied Chemistry,
 Montreal, August 1961 (University of Toronto Press,
 Toronto, 1961), p. 109

REACTION OF GRAPHITE WITH HYDROGEN. HEAT OF FORMATION
OF THE METHYLENE RADICAL

W. A. Chupka, D. J. Meschi, and J. Berkowitz ..(Project II-29)
 Abstracts of Scientific Papers Presented at the 18th
 International Congress of Pure and Applied Chemistry,
 Montreal, August 1961 (University of Toronto Press,
 Toronto, 1961), p. 115

HIGH-FREQUENCY DIELECTRIC CONSTANT OF HIGH-FREQUENCY
PLASMOIDS

A. J. Hatch (Project IV-10)
 Bull. Am. Phys. Soc. 7, 133 (February 23, 1962)

ANALYSIS OF GAMMA-RAY SPECTRA OBTAINED WITH A NaI
CRYSTAL WITH AN ANTICOINCIDENT ANNULUS

J. E. Monahan, S. Raboy, and C. C. Trail . . . (Project I-55)
 Proceedings of the Total Absorption Gamma-Ray Spec-
 trometry Symposium, Gatlinburg, Tennessee, May 10-
 11, 1960, TID-7594, pp. 168-187

TIME-REVERSAL INVARIANCE AND FLUX QUANTIZATION

M. Peshkin (Project V-33)
 Bull. Am. Phys. Soc. 7, 117 (February 23, 1962)

CHARGE STATES OF MOLECULAR FRAGMENTS FOLLOWING
NUCLEAR DECAY

S. Wexler (Project II-38)
 Papers Presented at Mass Spectrometry Conference, June
 4-9, 1961, Chicago, Illinois, ASTM Committee E-14, p. 106

ACM STUDENT REPORTS

ANALYSIS OF THE BETA-RAY SPECTRUM OF ${}_{74}^{188}\text{W}$

Gerald C. Nelson (Project I-36)
ACM Student Report to St. Olaf College

SOME MASS SPECTROSCOPIC STUDIES ON THE IONIZATION OF
Kr, Xe, AND He

D. L. Spears (Project II-31)
ACM Student Report to Monmouth College

AN ATTEMPT AT OBSERVING THE MOSSBAUER EFFECT WITH
THE 6.3-KEV GAMMA RAY IN Ta^{181}

D. Thorstenson (Project I-19)
ACM Student Report to Monmouth College

ADDITIONAL PAPERS ACCEPTED FOR PUBLICATIONEFFECT OF STYRENE MONOMER ON THE FLUORESCENCE
PROPERTIES OF POLYSTYRENE

L. J. Basile (Project I-144)
J. Chem. Phys. (April 1962)

THE HEAT OF FORMATION OF THE CN RADICAL

J. Berkowitz (Project II-29)
J. Chem. Phys.

DOUBLE-OVEN EXPERIMENTS WITH LITHIUM-HALIDE VAPORS

J. Berkowitz, H. A. Tasman, and W. A. Chupka.. (Project II-29)
J. Chem. Phys. (April 1962)

THE DEFORMATION ENERGY OF A CHARGED DROP. IV. EVIDENCE
FOR A DISCONTINUITY IN THE CONVENTIONAL FAMILY OF SADDLE
POINT SHAPES

S. Cohen and W. J. Swiatecki (Project V-1)
Ann. Phys.

POLARIZATION AND DIFFERENTIAL CROSS SECTION FOR NEUTRON
SCATTERING FROM SILICON

R. O. Lane, A. J. Elwyn, and A. S. Langsdorf, Jr... (Project I-18)
Phys. Rev. (May 1962)

ENERGY-DEPENDENT MULTIPLE SCATTERING OF NEUTRONS
IN THIN PLATES

R. O. Lane and W. F. Miller (Project I-18)
Nuclear Instr. and Methods

ELECTRIC QUADRUPOLE MOMENT OF THE 14.4-KEV STATE OF
 Fe^{57}

C. E. Johnson, W. Marshall, and G. J. Perlow..(Project I-19)
Phys. Rev. (May 15, 1962)

FLUX QUANTIZATION AND THE CURRENT-CARRYING STATE IN
A SUPERCONDUCTING CYLINDER

H. J. Lipkin, M. Peshkin, and L. J. Tassie . . .(Project V-33)
Phys. Rev. (April 1, 1962)

CHARGE STATES OF MOLECULAR FRAGMENTS FROM CCl_3Br
FOLLOWING NUCLEAR DECAY OF $\text{Br}^{80\text{m}}$ AND Br^{82}

S. Wexler (Project II-38)
J. Chem. Phys. (April 1962)

PERSONNEL CHANGES IN THE ANL PHYSICS DIVISION

NEW MEMBERS OF THE DIVISION

Student Aides (ACM)

Mr. John Heil, Beloit College. Working with S. B. Burson on the gamma-ray spectrum and decay scheme of 65-day W^{188} . Came to ANL on January 29, 1962.

Mr. Russell Marshall, Monmouth College. Working with S. Wexler on mass spectrometric studies of the reactions between methane and hydrogen isotopes (H_2 , D_2 , and T_2) under electron bombardment. Came to ANL on January 29, 1962.

Mr. Richard W. Reno, Knox College. Working with S. S. Hanna on superconducting magnets for use in Mössbauer experiments. Came to ANL on January 29, 1962.

DEPARTURE

Dr. Relja Popić has been an Institute Affiliate in the ANL Physics Division since October 5, 1960. He has collaborated with S. S. Hanna on (1) excited states of B^{10} from the $B^{11}(He^3, \alpha)B^{10}$ reaction (Project I-21) and (2) the $Be^9(p, d)Be^8$ reaction for $2.97 \text{ Mev} \leq E_p \leq 3.5 \text{ Mev}$ (Project I-21); and with S. Raboy and C. C. Trail on

(3) neutron capture in He^3 (Project I-56) and (4) parity conservation in $\text{H}(n,\gamma)\text{D}$ (Project I-56). He terminated at ANL on March 23, 1962 to return to the Boris Kidric Institute of Nuclear Sciences, Belgrade, Yugoslavia.