Cross Sections and Isomer Ratios for the $K^{41}(\alpha,n)Sc^{44m,44g}$ Reaction*

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(Received 20 April 1964)

The absolute cross sections for Sc^{44m} and Sc^{44g} have been determined at 1-MeV intervals from 8 to 19 MeV. The experimentally determined isomer-ratio function was compared with the theoretical predictions of a Huizenga-and-Vandenbosch-type calculation. A value of 1.7–1.8 for σ, the spin cutoff parameter, was found to give a good fit of the experimental isomer ratios below the $(\alpha,2n)$ threshold. Deviations of the calculated values from the experimental values above this threshold were interpreted to be the result of considerable spin fractionation.

I. INTRODUCTION

WITHIN the past several years, a number of studies have been made of nuclear reactions in which nuclear isomers are produced.[1–6] Such studies are of interest, for they bring into focus the law of conservation of angular momentum as it affects the course of a nuclear reaction. A scheme designed to predict quantitatively isomer ratios in compound-nucleus reactions was formulated by Huizenga and Vandenbosch.[3] The method of Huizenga and Vandenbosch is strictly valid only for a compound-nucleus reaction without any consideration given to direct interaction and to the possibility of particle emission competing with gamma decay in the final step of de-excitation. (We shall refer to this competition as spin fractionation.) A generalization of the Huizenga and Vandenbosch theory was given by Need.[5] The Need formalism is designed to cope with reactions which are only a small part of the total reaction, but again it does not consider spin fractionation as it has been defined above. Grover has considered the problem of spin fractionation in nuclear reactions, but this method is not feasible for application to isomer ratio calculations in its present state.[7]

The theoretical analysis of isomer ratios at sufficiently high energies is often marred by the presence of spin fractionation and direct interaction. In this work, we determined the cross sections and isomer ratios for the $K^{41}(\alpha,n)Sc^{44m,44g}$ reaction in the energy range from 8 to 19 MeV. Interference from direct interaction is thought to be of minor importance in the energy range studied as is evidenced by the shape of the isomer ratio curve between 20 and 40 MeV of Matsuo and Sugihara.[8] Furthermore, the Coulomb barrier for this reaction is low enough to permit accurate measurements of the cross sections several MeV below the $(\alpha,2n)$ threshold where spin fractionation sets in. This reaction was also studied below 19 MeV by Matsuo and Sugihara.[8] However, their data below 19 MeV contain only three isolated points and were not substantial enough to permit an unequivocal determination of the necessary isomer ratio parameters.

II. EXPERIMENTAL

The K^{41} targets were prepared by evaporating natural KCl upon aluminum backings with the thickness of KCl varying from approximately 1 to 2 mg/cm². The targets were then individually bombarded with 8–19-MeV alpha particles from the Florida State University Tandem van de Graaff accelerator. The incident-alpha-particle energies were known very accurately, and the energy loss of the particles in traversing the KCl was negligibly small.

After bombardment, each target underwent chemical separation to remove undesirable activities. The potassium chloride targets and aluminum backing were dissolved in 6*N* HCl. Standard scandium carrier was then added to the solution. The preliminary purification of the active scandium from active and nonactive contaminants such as aluminum, phosphorous, chloride, and potassium was accomplished by neutralizing the above acid solution with ammonium hydroxide and then precipitating scandium hydroxide from a 1*N* NaOH solution. The scandium hydroxide was then dissolved in 6*N* HCl and the preceding step repeated. The scandium hydroxide was again dissolved in hydrochloric acid, lanthanum carrier was added, and lanthanum precipitated as a fluoride from a solution of ammonium hydroxide and ammonium flouride, with scandium remaining in solution presumably as the strongly complexed ScF_4^- ion.[9] To the remaining solution, a small amount of $[Co(NH_3)_6]Cl_3$ reagent[10] was added with the scandium precipitating as $[Co(NH_3)_6]ScF_6$.[11] The precipitate was filtered, washed with water, alcohol, and ether, dried at 110°C for 10 min, and weighed. The

* Supported by the U. S. Atomic Energy Commission under contract No. AT-(40-1)-2628.
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[1] J. W. Meadows, R. M. Diamond, and R. A. Sharp, Phys. Rev. **102**, 190 (1956).
[2] B. Linder and R. A. James, Phys. Rev. **114**, 322 (1959).
[3] J. R. Huizenga and R. Vandenbosch, Phys. Rev. **120**, 1305, 1313 (1960).
[4] J. L. Need and B. Linder, Phys. Rev. **129**, 1298 (1963).
[5] J. L. Need, Phys. Rev. **129**, 1302 (1963).
[6] C. Riley and B. Linder, Phys. Rev. **134**, B559 (1964).
[7] J. R. Grover, Phys. Rev. **123**, 267 (1961).
[8] T. Matsuo and T. Sugihara, Can. J. Chem. **39**, 697 (1961). This seems reasonable because we are below 20 MeV and also the total cross sections are still of considerable magnitude so that any small direct interaction cross section would have a negligible effect upon the experimental cross-section ratios.
[9] J. Kleinberg, Atomic Energy Commission Report No. LA-1721, 1958 (unpublished).
[10] W. Blitz, Z. Anorg. Chem. **83**, 177 (1914).
[11] Y. Takashima, University of California, Lawrence Radiation Laboratory Report UCRL-Trans. 339 (L), 1960 (unpublished).

TABLE I. Experimental results for $K^{41}(\alpha,n)Sc^{44m,44g}$ reaction.

Alpha energy (MeV)	σ_m (mb)	σ_g (mb)	$\sigma_m+\sigma_g$ (mb)	$\frac{\sigma_m}{\sigma_m+\sigma_g}$	$\frac{\sigma_m}{\sigma_g}$
8	17	151	168	0.10	0.12
9	53	309	362	0.15	0.17
10	79	338	417	0.19	0.23
11	124	358	482	0.26	0.35
12	138	360	498	0.28	0.38
13	171	329	500	0.34	0.52
14	189	296	485	0.39	0.64
15	199	221	420	0.47	0.90
16	209	175	384	0.54	1.19
17	205	152	357	0.57	1.35
18	190	120	310	0.61	1.58
19	140	84	224	0.62	1.67

total time required for purification of each sample was about 75 min and the chemical yields were of the order of 80–90%.

A 3-×3-in. NaI(Tl) crystal scintillator coupled with a 256-channel analyzer was used for gamma counting. The photopeak efficiencies used in the calculation of the absolute disintegration rates were taken from Heath[12] and Vegors, Marsden, and Heath.[13] As a check upon the efficiency of our geometry, the disintegration rate of a standard Cs^{137} 0.662-MeV photon was determined using Heath's calculated data.[12] The determined disintegration rate agreed with the calibrated standard to within 2%.

From the decay scheme of Fig. 1, it is seen that in principle both isomers of Sc^{44} can be determined by following the decay of the 1.16-MeV gamma.[14,15] (A small amount of 1.12 interference from Ca^{44} necessitates

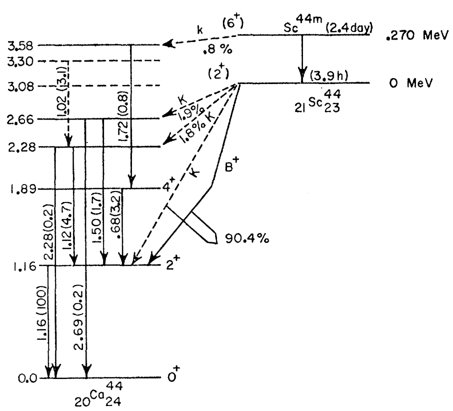

FIG. 1. Decay scheme of $_{21}Sc_{23}^{44}$. Taken from J. D. McCullen and J. J. Kraushaar, Phys. Rev. **122**, 555 (1961). Spins of Sc^{44m} and Sc^{44} taken from D. L. Harris and J. D. McCullen, Phys. Rev. **132**, 310 (1963).

[12] R. L. Heath, Atomic Energy Commission Report No. IDO-16408, 1957 (unpublished).

[13] S. H. Vegors, L. L. Marsden, and R. L. Heath, Atomic Energy Commission Report No-16370 1958 (unpublished).

[14] J. D. McCullen and J. J. Kranshaar, Phys. Rev. **122**, 555 (1961).

[15] D. L. Harris and J. D. McCullen, Phys. Rev. **132**, 310 (1963).

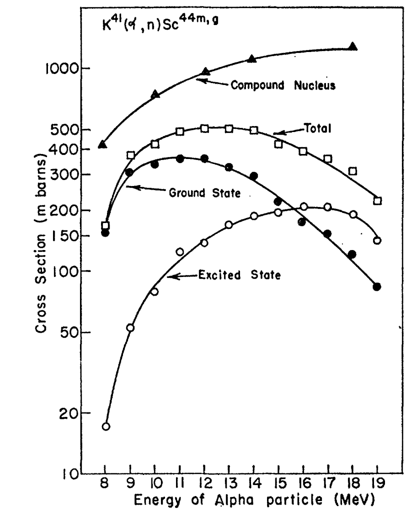

FIG. 2. Excitation functions for $K^{41}(\alpha,n)Sc^{44m,44g}$ reaction. Open circles represent $K^{41}(\alpha,n)Sc^{44m}$ excitation function. Solid circles represent $K^{41}(\alpha,n)Sc^{44g}$ excitation function. Squares represent total $K^{41}(\alpha,n)Sc^{44m,44g}$ excitation function. Solid triangles represent calculated compound nucleus excitation function taken from J. R. Huizenga and G. J. Igo, Atomic Energy Commission Report No. ANL-6373, 1961 (unpublished).

multiplication of the experimentally determined 1.16 activity by a 0.96 correction factor. Also, any interference from the 1.05-MeV photon of Sc^{43}, which should be small, is neglected.[16]) However, the necessity of counting at 8.35-cm distance from the crystal to minimize summing of the 1.16- and 0.511-MeV photons reduced the counting rate of the 1.16-MeV photon to such an extent that statistics were too poor for an accurate determination of the 58.8-h excited-state activity with our mμA alpha beam, without very long bombardments. To resolve this difficulty, the ground-state 3.9-h activity was determined by counting the 1.16-MeV gamma at 8.35 cm distance immediately after the sample had been chemically prepared for counting. Then approximately 40 h were allowed to pass so that the initial ground-state activity of Sc^{44} would minimize to the equilibrium stage, and any interfering Sc^{43} would decay away (Sc^{43} has 0.38- and 0.511-MeV photons which decay with a 3.9-h half-life).[16] The samples were then counted flat against the crystal with the 0.270-MeV photon being used to determine the 58.8-h excited-state activity. The low-energy contribution from the small amount of the 0.511-MeV decay that still existed from the equilibrium decay of the 3.9-h ground state was subtracted out with the aid of a Na^{22} standard.

Calculations of the absolute cross sections were performed using the standard decay and bombardment equations.[6] The conversion coefficient of the 0.270-MeV

[16] K. Way, R. W. King, C. L. McGinnis, and R. Van Lieshout, Atomic Energy Commission Report No. TID-5300, 1955 (unpublished).

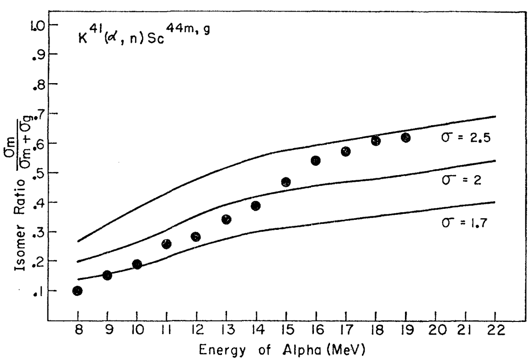

FIG. 3. Isomer ratios for $Sc^{44g,44m}$ pair produced by $K^{41}(\alpha,n)$ $Sc^{44g,44m}$. Experimental points are represented by dots and calculated values as solid lines for indicated spin cutoff parameters.

gamma was taken to be 0.14 and that of the 1.16-MeV photon was negligible.[17]

III. EXPERIMENTAL RESULTS

The experimental results are tabulated in Table I. The error in the absolute cross sections for the separate states is estimated to be about ±15%. Excitation functions for the excited and ground states are displayed in Fig. 2. Also shown are the total absolute cross sections for the $K^{41}(\alpha,n)Sc^{44}$ reaction from 8 to 19 MeV. Calculated compound-nucleus cross sections are displayed in Fig. 2 for comparison.[18] Figure 3 shows the experimental isomer-ratio function. The error in the isomer ratios is estimated to be ±10%. The solid lines (of Fig. 3) represent the theoretical isomer ratios calculated for spin cutoff parameters of 1.7, 2, and 2.5, respectively, about which comment is reserved for the discussion.

IV. DISCUSSION

The cross sections displayed in Fig. 2. may be compared at two energies with those reported by Matsuo and Sugihara.[8] The agreement is good at 11 MeV, but poor at 16 MeV. Our excitation curve peaks at 13 MeV and has a value of 500 mb at its maximum while the excitation function of Matsuo and Sugihara peaks at 16 MeV and has a value larger than 700 mb. The measurements of Matsuo and Sugihara differed in two respects from ours. Their reaction was obtained by cyclotron bombardment using foil-degradation techniques to determine the incident-alpha-particle energy, whereas our cross sections were obtained by individual bombardments using a Tandem van de Graaff. This factor coupled with the difference in counting technique may possibly account for these discrepancies in the cross sections.

Our experimentally determined isomer ratios displayed in Fig. 3. (solid circles) appear to have excellent internal agreement resulting in the formation of a smooth, well-defined isomer-ratio function. The isomer ratio curve of Matsuo and Sugihara appears to extend our curve beyond 19 MeV.[19] Tabulated in Table II are the predicted isomer-ratio values calculated in the manner of Huizenga and Vandenbosch.[3] For the calculations the level density parameter a for Sc^{44} was taken to be $0.094A$ MeV^{-1} (4.1 MeV^{-1}), where A is the nuclear mass.[20] The average energy of the evaporated neutrons was taken to be $2T$, where T is the nuclear temperature given by[21]

$$T=(E_r/a)^{1/2}, \tag{1}$$

with E_r being the energy of the residual nucleus. The average number of gamma rays emitted at each calculated point $\bar{N}_\gamma$ was taken to be[22]

$$\bar{N}_\gamma=\tfrac{1}{2}(aE_r)^{1/2}. \tag{2}$$

When fraction values of $\bar{N}_\gamma$ were calculated with (2), a linear interpolation between the integers spanning the fraction was used. The alpha-particle transmission coefficients were taken from Huizenga and Igo,[23] and the

TABLE II. Calculated isomer ratios.

Alpha energy (MeV)	Spin cutoff parameter σ	Number of gammas emitted $\bar{N}_\gamma$	$\frac{\sigma_m}{\sigma_m+\sigma_g}$	$\frac{\sigma_m}{\sigma_g}$
8	1.7	1.5	0.14	0.17
	2	1.5	0.20	0.25
	2.5	1.5	0.27	0.37
	3	1.5	0.32	0.46
10	1.7	2	0.18	0.22
	2	2	0.27	0.37
	2.5	2	0.38	0.63
	3	2	0.46	0.83
12	1.7	2.3	0.25	0.33
	2	2.3	0.34	0.56
	2.5	2.3	0.48	0.95
	3	2.3	0.57	1.29
14	1.7	2.6	0.30	0.44
	2	2.6	0.42	0.73
	2.5	2.6	0.55	1.26
	3	2.6	0.64	1.74
18	1.7	3.2	0.35	0.53
	2	3.2	0.48	0.94
	2.5	3.2	0.63	1.75
	3	3.2	0.71	2.53
22	1.7	3.6	0.40	0.69
	2	3.6	0.54	1.18
	2.5	3.6	0.69	2.21
	3	3.6	0.76	3.27

[17] D. Strominger, J. M. Hollander, and G. T. Seaborg, Rev. Mod. Phys. **30**, 621 (1958).
[18] J. R. Huizenga and G. J. Igo, Atomic Energy Commission Report No. ANL-6373, 1961 (unpublished).

[19] The results are published in Ref. 4 in graphic form. Exact results were obtained from Taku Matsuo (private communication).
[20] R. L. Bramblett and F. W. Bonner, Nucl. Phys. **20**, 395 (1960).
[21] J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley & Sons, Inc., New York, 1952).
[22] V. M. Strutinky, L. V. Groshev, and M. K. Akimova, Nucl. Phys. **16**, 657 (1960).
[23] J. R. Huizenga and G. J. Igo, Atomic Energy Commission Report No. ANL-6373, 1961 (unpublished).

neutron coefficients were taken from Feld *et al.*[24] The calculations were performed on the Florida State University IBM-709 computer using a program written by Hafner, Huizenga, and Vandenbosch.[25]

Referring to Fig. 3, we notice several distinct results. First, the experimental ratios are described quite well from about 8 to 14 MeV by a nearly constant spin cutoff parameter, but require a continuously varying parameter after that. Second, the value of the spin cutoff parameter is smaller than ordinarily found in this mass region.

It is to be noted that σ can change with energy as is shown by the classical form

$$\sigma^2 = IT/\hbar^2, \tag{3}$$

where I is the effective moment of inertia and T is again the nuclear temperature. Replacing T by (1) gives as an alternate form of (3)

$$\sigma^2 = I(E_r/a)^{1/2}/\hbar^2, \tag{4}$$

in which all terms have been previously defined. The energy dependence of σ is seen from (4) to be small, being directly proportional to the fourth root of the residual excitation energy, and the square root of the moment of inertia which also has some energy dependence. It seems unreasonable to attribute the variation in σ completely to energy when there is only a strong variation over one segment of the isomer-ratio function, but virtually no variation over another. Likewise, Bishop[26] could describe his experimental isomer ratios with a constant σ from 10 to 18 MeV for the $Ag^{107}(\alpha,n)In^{110m,g}$ reaction, but had σ increase from 3 to approximately 8 between 18 and 22 MeV.

As mentioned previously, there are several factors (none of which were taken into account in the calculations) which could make the theoretical values deviate from the experimental values. Among these are spin fractionation and direct interaction. Direct interaction is though to be negligible for this reaction in the energy range which is pertinent as stated in the introduction.

Corrections for spin fractionation to the Huizenga and Vandenbosch theory could in principle be made using the method of Grover's "average channel fraction."[7] However, Grover's method requires a knowledge of level spacings and radiation widths, information about which is almost totally lacking at the present time. In a later reference, Grover proposed an approximation which entails preliminary averaging.[27] Unfortunately, this leads to a cancellation of the effect of spin fractionation when the cross-section ratio is calculated. From a qualitative standpoint, it is possible to explain the high-spin enhancement of spin fractionation. This explanation depends upon the findings of Grover[7] that, for those energy states just above the binding energy of the next neutron, gamma-ray emission will compete with the emission of another neutron. Consider the distribution of states of Sc^{44*} in the neighborhood of the binding energy of the next neutron. If a particle state is neutron unstable, the neutron must be emitted with very little energy. As a consequence of this energy limitation, the centrifugal-barrier requirement would limit the emitted neutrons to those of low orbital angular momentum. Now, if the spin states of the product nucleus (Sc^{43}) accessible to neutron decay were limited to low-J values, then decay of the high-spin states of Sc^{44*} by neutron emission would be hindered because of the necessity of the neutrons for having high-orbital angular momentum. Bishop,[26] has proposed a possible explanation based on an energy-dependent σ which limits *all* of the spins states of the product nucleus to low-J values. His argument is that if σ is relatively small at low excitation energies, the distribution of states of the product nucleus (Sc^{43}) will peak at a low-J value, resulting in few high-spin states to which Sc^{44} can neutron decay. Another possible explanation for limiting the neutron accessible states to those of low J, which is not based on an energy-dependent σ, results from the necessity for decay of the product nucleus itself. If the product nucleus were left in a state of high angular momentum, and little excitation energy, decay to a low-spin ground state would then require that a large amount of angular momentum be carried off by an energy-limited gamma cascade. If only low-spin states of the product nucleus are open for decay, then all neutron emission of Sc^{44*} in the vicinity of the $(\alpha,2n)$ threshold must come from low-spin states of Sc^{44*}. Gamma decay of the total distribution of Sc^{44*} will not be hindered because each decay results in only a slight redistribution of the former spins and a small change in energy. Hence, in the vicinity of the $(\alpha,2n)$ threshold there will be an enhancement of the high-spin isomer, because of this necessity for gamma decay.

Attributing the apparent strong variation in σ demonstrated in Fig. 3 and Table II over the latter half of the isomer-ratio function to the effects of spin fractionation, the proper value of the spin cutoff parameter would then be determined by a comparison of the experimental isomer ratios with those calculated below the $(\alpha,2n)$ threshold.

The low value of σ (1.7–1.8) determined for Sc^{44} is perplexing. Although a low σ value has been reported for $Al^{28}(\sim 1.7)$,[28] most studies seem to indicate that nuclei in this mass region have much larger values (of the order of 3 or 4). It warrants mentioning that the σ determined by the Huizenga and Vandenbosch form-

[24] B. T. Feld, H. Feshbach, M. L. Goldberger, H. Goldstein, and V. F. Weisskopf, Atomic Energy Commission Report No. NYO-636, 1951 (unpublished).

[25] W. L. Hafner, Jr., J. R. Huizenga, and R. Vandenbosch, Atomic Energy Commission Report No. ANL-6662, 1962 (unpublished).

[26] C. T. Bishop, Atomic Energy Commission Report No. ANL-6405, 1961 (unpublished).

[27] J. R. Grover, Phys. Rev. 127, 2142 (1962).

[28] T. Ericson, Nucl. Phys. 11, 481 (1959).

alism appears to be quite sensitive to assumptions made for the final gamma decay, in which spin states decay to the isomeric state for which the change in angular momentum is a minimum. As a consequence, the value for σ would depend strongly on the spin values of the final states. For example, if the spins of the Sc^{44} isomers had been established as 7 and 3 (instead of the now recognized values of 6 and 2), the σ value would have been closer to 3.

To summarize, we have seen that the Huizenga and Vandenbosch formalism can adequately describe the isomer ratios for a compound-nucleus (α,n) reaction when interference from the $(\alpha,2n)$ is not possible. When multiple-particle emission becomes possible, however, the cross sections are strongly governed by the channel fraction parameters. The indiscriminate application of the Huizenga and Vandenbosch theory to even the most simple compound-nucleus reactions can be erroneous and yield parameters which are not meaningful once multiple-particle emission becomes possible.

ACKNOWLEDGMENTS

We are grateful to the U. S. Atomic Energy Commission for providing financial support for this research and to the U. S. Air Force for support of the FSU Tandem van de Graaff Program. We also wish to thank Peter Stoycheff for helping with the operation of the accelerator.

PHYSICAL REVIEW VOLUME 135, NUMBER 6B 21 SEPTEMBER 1964

Effect of the Harwell $A(\theta)$ Data on the 50-MeV Proton-Proton Phase Shifts*

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The new $A(\theta)$ and $\sigma(90°)$ data at 50 MeV are found to decrease the probable range of the phase shifts. The $^3F_3$ phase, however, is strongly predicted to be far from the value expected on the basis of models; this reflects on the consistency of the data. The most significant result is the rejection of solution 2. Comparison is made to the results of Batty and Perring.

I. INTRODUCTION

IN a previous communication[1] energy-independent modified phase-shift analyses were made of 25 proton-proton scattering data measured at energies near 50 MeV; these included cross-section, polarization, depolarization, and correlation measurements. The analysis energy was chosen to be 51.8 MeV, the energy of the data subgroup with the smallest quoted errors. An interesting result was that the $^3P_0$ phase shift was found to probably lie between 13 and 19°, somewhat higher than the 10.7–12.0° given by current potential models.

There has recently become available from Ashmore *et al.*[2] a set of quite good $A(\theta)$ measurements at 47.5 MeV, and from Batty *et al.*[3] a much improved absolute cross-section measurement at 50 MeV. The principal effects of these measurements are (1) that the probable $^3P_0$ phase-shift range is halved and lowered, and (2) that solution 2 is eliminated, for all practical purposes, at this energy.

II. DATA SELECTION AND TREATMENT

Including the 25 data previously considered, 5 new $A(\theta)$ data, and a new absolute cross-section measurement at 90°, there are available a total of 31 data in the energy range 47.5–52.0 MeV. One has then to decide upon the energy at which to make the analysis.

The previous analysis[1] was performed at 51.8 MeV, the energy of the (then) most precise data. The new A measurements, however, are at 47.5 MeV, and there are no A data at nearby energies for use in interpolation. If the interpolation is not too large, one might consider using the results of an energy-dependent phase-shift analysis. The published phase-shift representation which appears to give the best fit to the moderate energy proton-proton data would seem to be that labeled "CR21" in a previous communication.[4] Using the CR21-predicted $A(\theta)$ at 47.5, 50, and 51.8 MeV, each experimental A datum was shifted by the difference of the predictions at the datum angle. For example, CR21 predicted $A(39°)=-0.051$ at 47.5 MeV and

TABLE I. Interpolated $A(\theta)$ minus experimental $A(\theta)$ values, as fractions of the experimental errors. The 50-MeV numbers can be constructed from Table II.

Energy (MeV)	c.m. angles (deg) 23.5	39.0	54.6	71.1	87.1
50	−0.37	−0.47	−0.68	−0.58	−0.38
51.8	−0.64	−0.82	−1.17	−1.00	−0.66

* Supported in part by the U. S. Atomic Energy Commission.
[1] P. Signell, N. R. Yoder, and N. M. Miskovsky, Phys. Rev. **133**, B149 (1964).
[2] Table II, Ref. a.
[3] Table II, Ref. c.
[4] P. Signell and N. R. Yoder, Phys. Rev. **134**, B100 (1964).