Virtual Electron Capture in Ni^{59}†

MARCEL SCHMORAK
Brookhaven National Laboratory, Upton, New York
(Received 21 September 1962)

A careful measurement of the gamma-ray spectrum of Ni^{59} has been performed. The analysis of the data indicates that the inner bremsstrahlung spectrum is slightly distorted near the end point. This distortion is ascribed to destructive interference with a virtual e^- capture through the firxt excited state in Co^{59}. The probability of the virtual transition is found to be in the range of 0.0005 to 0.006 relative to the direct, second forbidden, inner bremsstrahlung to the ground state of Co^{59}. The product of the width of the first excited state in Co^{59} and the log(ft) value for the virtual e^- capture is determined.

1. INTRODUCTION

THE possibility of observing a combined beta-gamma transition through a virtual intermediate nuclear state, in competition with a direct forbidden β decay, has been investigated by Rose *et al.*[1] The decay of Ni^{59} affords a specially favorable case for the study of virtual e^- capture transitions; the relevant details of the decay scheme are shown in Fig. 1. The maximum energy available for inner bremsstrahlung photons associated with the second forbidden K-capture decay of Ni^{59} is (1076±2) keV −8 keV=1068±2 keV (the K binding energy of Ni is 8 keV).

The inner bremsstrahlung spectrum is expected to be distorted due to interference with the virtual transition via the first excited state 5/2− in Co^{59}. The degree of distortion depends on the relative probability of these two modes of decay, and on the sign of the interference. A rough estimate[1] indicates that the virtual transition might be of the order of a few percent of the direct inner bremsstrahlung. There is a large uncertainty in this estimate, because the $M1$ 5/2− to 7/2− transition probability in Co^{59}, and also the log ft value for the allowed 3/2− to 5/2− e^- capture, are not known. A measurement of the rate of virtual capture will determine, assuming the correctness of the theory, the product of the above-mentioned two unknown transition probabilities.

The photon spectrum associated with the virtual e^- capture is expected to be[1] of the form $K^3(1-K)^2/(1.03-K)^2$, K being the energy in units of the end-point energy (1068 keV). 1.03 is the ratio of the energy of the first excited state in Co^{58} to the energy available for K capture. The closeness of this ratio to unity makes Ni^{59} a specially favorable case for investigating virtual transitions.

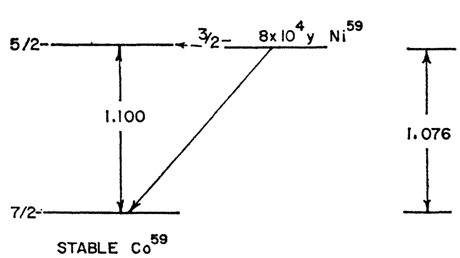

FIG. 1. Ni^{59} decay scheme. The virtual K capture through the 5/2- state is investigated.

Before this experiment was started, one measurement of the Ni^{59} inner bremsstrahlung spectrum was reported in the literature, that of Saraf.[2] This measurement was performed with a single-channel analyzer and a rather small (3.5 cm×3.5 cm) NaI crystal. The quality of the results near the endpoint was not sufficient to exclude a rather large virtual capture contribution (the possibility of such an effect was not considered in the above-mentioned paper). Therefore, it was decided to perform a careful measurement of the photon spectrum of Ni^{59}.

2. SOURCE PREPARATION

500 mg of Ni enriched to 99.95% Ni^{58} were irradiated for 4 weeks in an Oak Ridge reactor at a flux of 2×10^{14} n/sec cm^2. Since the capture cross section is 4 b, the source strength should be 4×10^6 disintegrations per sec. Several activities were found in the irradiated target, mainly Co^{58} produced by (n,p) reaction and also activities produced by neutron capture in chemical impurities in the targets, most of them not having been reported in the spectrographic analysis. Chemical separations were made (mainly using precipitation of Ni with di-methyl glyoxin). They were repeated several times after the last traces of contaminant γ-ray peaks were undetectable in a scintillation multichannel analyzer.

The source was finally obtained as NiO powder containing 455 mg of Ni^{58}. The powder was spread in a disk of 4 cm in diameter in order to reduce the self-absorption of the 8-keV K x-rays. For some of the singles measurements the source was concentrated to a "point" geometry.

3. MEASUREMENTS

In order to obtain a precise measurement of the γ spectrum, the following conditions have to be satisfied:

† Work performed under the auspices of the U. S. Atomic Energy Commission.

[1] M. E. Rose, R. Perrin, and L. L. Foldy, Phys. Rev. **128**, 1776 (1962).

[2] B. Saraf, Phys. Rev. **102**, 466 (1956).

(1) The electronics has to be stable.

(2) The background radiation should be small and constant and the statistics good.

(3) The efficiency of the detector has to be well known.

(4) Radioactive impurities in the source have to be eliminated.

The electronic system used consisted of Franklin amplifiers and TMC 256-channel transistorized pulse-height analyzer. For final analysis, 3 runs of 300 min each with a 3-in.×3-in. NaI crystal and 3 runs of 240 min each with a 5-in.×5-in. NaI crystal were made. Energy calibration was repeated before every run and the maximum variations in the calibration were less than $\frac{1}{3}\%$. The amplifier and multichannel analyzer were linear to better than $\frac{1}{2}\%$ in the energy region of interest.

The background was reduced by placing the NaI detector in an iron shield lined with bismuth bricks (in order to reduce backscattering). The statistical errors (including the effect of background) are negligible except near the end point, where resolution correction uncertainties are even larger, as described in Sec. 5. For example, the statistical error is 5% at an energy of 0.98 of the end point and it is 1.4% at 0.90 of the end point. The background is 3% of the source strength at 0.5 MeV.

The accurate knowledge of the efficiency of the detector and its variation with energy is an important aspect of the problem. Three main corrections to the measured spectrum have to be made: (1) the change of photoefficiency with energy; (2) contribution of the Compton scattered electrons; and (3) the finite resolution of the NaI detector. The Compton correction is smaller in bigger crystals while the resolution correction is usually smaller in smaller crystals. As a check on the reliability of the corrections, the measurements were made with two crystals of different dimensions, 3 in.×3 in. and 5 in.×5 in.

In addition to these three corrections, there are two minor ones: There are some excess counts around 200 keV due to backscattering from the shield, and there is a distortion of the spectrum below about 300 keV due to the nonlinear response of the NaI detector in this region. This nonlinearity was found to be approximately the same as reported by Iredale.[3] These corrections were not taken into account because the data were analyzed above 300 keV only.

The elimination of radioactive impurities was accomplished by a careful chemical separation. As a double check, the γ spectrum was measured in coincidence with K x rays of ~8 keV. The statistics in this measurement were poorer than in the singles measurement; however, within the errors the singles and coincidence spectra were the same above 300 keV. At low energies there were less counts in the coincidence spectrum because the inner bremsstrahlung associated with L capture was eliminated (see, for example, Biavati *et al.*[4]).

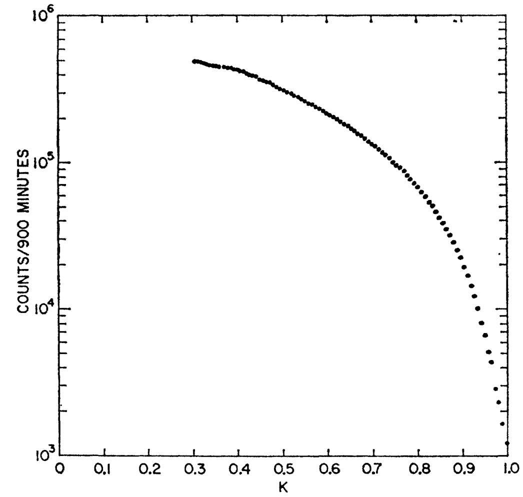

FIG. 2. Gamma spectrum of Ni^{59}. K is the energy in fractions of the end-point energy. The inner bremsstrahlung spectrum is slightly distorted around $K=0.9$ due to interference with the virtual transition through the first excited state in Co^{59}.

4. RESULTS AND INSTRUMENTAL CORRECTIONS

The results obtained with the 3-in.×3-in. NaI crystal are shown on Fig. 2. The background is subtracted.

The correction for finite resolution was made by measuring the resolution of the detector at several energies (it was 8.4% at 660 keV and 7% at 1 MeV). It was assumed that the photopeak shape is Gaussian and this Gaussian was folded with the theoretical inner bremsstrahlung spectrum for $\lambda=\frac{1}{2}$ (see Sec. 5). The fact that the real spectrum is in fact slightly different makes only a second-order error on this correction. As could be expected, the resolution correction is significant only near the end point.

The photoefficiency variation with energy was taken from Heath.[5] To check the effect of the nonstandard source dimensions, the geometry of the source was changed to that of a "point" source on the axis, and it was found that the relative photoefficiency did not change to any significant degree.

The Compton correction was performed numerically as follows: A theoretical spectrum with $\lambda=\frac{1}{2}$ (see Sec. 5) which represents the data to a first approximation was assumed. Compton to total ratios were taken from Miller *et al.*[6] The spectrum was divided into strips and the Compton contribution of each strip assumed to be of rectangular shape up to the Compton edge; then all

[3] P. Iredale, Nucl. Instr. Methods 11, 340 (1961).

[4] M. Biavati, S. Nassiff, and C. S. Wu, Phys. Rev. 125, 1364 (1962).

[5] R. L. Heath, Atomic Energy Commission Report IDO-16408 (TID-4500) (unpublished).

[6] W. F. Miller and W. J. Snow, Argonne National Laboratory Report ANL-6318 (unpublished).

TABLE I. Experimental spectrum and instrumental corrections. K is the energy in units of the end-point energy (1068 keV); φ is the experimental spectrum for the 3-in.×3-in. NaI detector in arbitrary units; σ is the error (in percents) used for the least-square fitting, the statistical error, in brackets, is negligible below $K=0.8$; C is the calculated Compton contribution; r is the resolution; μ is the corrected spectrum (see Sec. 4).

K	φ	σ	C	r	μ
0.3	240.6	3	63.0		0.2388
0.35	218.9	3	48.5		0.2570
0.4	204.5	3	36.5		0.2832
0.45	178.0	2	26.5		0.2841
0.5	151.6	2	18.2		0.2766
0.55	126.5	2	11.7		0.2611
0.6	103.5	2	6.7		0.2400
0.65	81.8	2	3.0		0.2107
0.7	64.0	1	1.1		0.1817
0.75	46.7		0.2		0.1436
0.8	32.7	(0.3)	0.0		0.1061
0.85	19.97	1	0.0		0.0688
0.9	9.86	2.3(1.4)		1.05	0.03386
0.93	5.29	1.7		1.09	0.01825
0.95	3.08	5.5(2.8)		1.26	0.00932
0.97	1.68	11 (4)		1.76	0.00366

the contributions were summed up. As a check, a more realistic shape for the Compton spectrum was assumed; the maximum change compared to the preceding calculation was $\frac{1}{3}\%$. The Compton contribution at 0.5 MeV is ~14% for the 3-in.×3-in. NaI and ~4% for the 5-in.×5-in. NaI.

To check the effect of backscattered gammas, the measurements were repeated outside the shield. The change in the shape of the spectrum above 300 keV was negligible. The results with the 5-in.×5-in. NaI agree very well with the 3-in.×3-in. results.

Table I shows the experimental results and the various corrections for the 3-in.×3-in. NaI detector; the assigned errors σ are discussed in Sec. 5. $\mu(K)$ is the experimental spectrum corrected for finite resolution of the detector, for the Compton contribution, and for the change of photoefficiency with energy.

5. ANALYSIS OF THE RESULTS

The results were compared with several theoretical predictions. Two different forms of the inner bremsstrahlung spectrum and two possibilities of interference with the virtual capture process (constructive and destructive interference) were investigated. A least-square fit was calculated for different probabilities of virtual capture, keeping one free parameter λ in the shape of the inner bremsstrahlung spectrum.

The inner bremsstrahlung spectrum for second forbidden transitions is not precisely known due to the presence of several unknown matrix elements. No such spectrum of similar energy to that of Ni^{59} is measured at present. This fact complicates the analysis of the results.

The first form of the second forbidden inner bremsstrahlung spectrum used was[7] $I_{ib}=K(1-K)^2[K^2+\lambda(1-K)^2]$, K being the energy as a fraction of the endpoint energy, λ essentially a ratio of matrix elements and treated as an unknown parameter.[8]

The second form for the inner bremsstrahlung spectrum used for fitting the data included relativistic and Coulomb corrections.[9] The results did not differ appreciably from those obtained with the first form, because these corrections are small above $K=0.3$, which was the energy region analyzed.

The data were fitted with a function of the form

$$F(K,X)=I_{ib}+XI_{vc}\pm(2I_{ib}XI_{vc})^{1/2}.$$

I_{ib}, the inner bremsstrahlung spectrum, was described above. I_{vc} is the spectrum of the virtual capture γ rays.[1] $I_{vc}=cK^3(1-K)^2/(\epsilon-K)^2$, $\epsilon=1.03$, is the ratio of the energy of the first excited state in Co^{59} to the energy available for the K capture decay of Ni^{59}; c is a normalization constant chosen so that

$$\int_0^1 I_{ib}dK=\int_0^1 I_{vc}dK.$$

The positive sign represents a case of constructive interference, while the negative sign corresponds to destructive interference. (No other phase is possible due to time reversal invariance.)

The factor $\sqrt{2}$ is due to the fact that only 50% interference is possible. The inner bremsstrahlung spectrum is circularly polarized, while the virtual capture gammas are not polarized.

The least-squares fit was made on an IBM 7090 computer for many different values of λ and X. Table II shows some of the results. K is the energy of the photons in units of the endpoint energy. The region below $K=0.3$ was not included in the analysis, because of the larger uncertainties in the spectrum, due to the Compton contribution and the distortions caused by backscattered gammas. The uncertainties in the theoretical inner bremsstrahlung shape are also larger in the low-energy region due to P-wave capture process and due to the larger Coulomb corrections. In the analysis more values of K were included near the endpoint, because the effect of the virtual capture is biggest around $K=0.9$.

$\mu(K)$ is the experimental spectrum corrected for instrumental effects as described in Sec. 4 and in Table I.

$\sigma(K)$ are the errors assigned. The larger errors near the endpoint are due mainly to the statistical errors

[7] R. Cutkowsky, Phys. Rev. **95**, 1222 (1954).

[8] See, for example, C. S. Wu, *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (North-Holland Publishing Company, Amsterdam, 1955).

[9] R. Marr, Harvard thesis, 1958 (unpublished); and private communication.

TABLE II. Some of the results of the least-squares fit (see Sec. 5). The best fit was obtained for $X=-0.002$ and $\lambda=0.49$. dev/$\sigma(K)$ is the deviation between the fitted curve $F(K,X)$ and the corrected experimental spectrum $\mu(K)$, divided by the experimental error $\sigma(K)$. K is the energy of the photons in units of the end-point energy.

K	$\mu(K)$	$\sigma(K)$	$F(K,-0.002)$ $\lambda_0=0.49$	dev/$\sigma(K)$	$F(K,-0.041)$ $\lambda_0=0.29$	dev/σ	$F(K,0.0001)$ $\lambda_0=0.66$	dev/σ
0.97	0.0036	0.0004	0.0031	1.289	0.0055	−4.840	0.0044	−2.084
0.95	0.0093	0.0005	0.0089	0.826	0.0098	−0.919	0.0112	−3.898
0.93	0.0182	0.0003	0.0175	2.355	0.0160	7.340	0.0205	−7.506
0.9	0.0338	0.0007	0.0345	−0.991	0.0301	5.217	0.0374	−5.180
0.85	0.0688	0.0007	0.0699	−1.536	0.0648	5.778	0.0708	−2.856
0.8	0.1061	0.0011	0.1087	−2.338	0.1072	−0.975	0.1062	−0.085
0.75	0.1436	0.0015	0.1463	−1.799	0.1507	−4.753	0.1402	2.282
0.7	0.1817	0.0019	0.1799	0.973	0.1904	−4.583	0.1708	5.756
0.65	0.2107	0.0042	0.2079	0.667	0.2232	−2.987	0.1971	3.234
0.6	0.2400	0.0048	0.2299	2.097	0.2471	−1.482	0.2193	4.314
0.55	0.2611	0.0052	0.2465	2.807	0.2624	−0.254	0.2378	4.481
0.5	0.2766	0.0054	0.2584	3.364	0.2697	1.272	0.2534	4.288
0.45	0.2841	0.0056	0.2668	3.094	0.2705	2.436	0.2670	3.058
0.4	0.2832	0.0075	0.2724	1.444	0.2662	2.264	0.2788	0.591
0.35	0.2570	0.0075	0.2756	−2.476	0.2585	−0.195	0.2885	−4.206
0.3	0.2388	0.0075	0.2759	−4.942	0.2481	−1.235	0.2951	−7.501

(see Table I) and to an estimated uncertainty in the resolution correction.

The errors (3%) in the low-energy region are due mainly to uncertainties in the Compton correction and possible distortions by backscattered gammas. The errors are smallest (1%) in the region $0.7\leq K\leq 0.85$, where mainly the uncertainties in the photoefficiency of the NaI crystal contribute. It is clear that this estimate of essentially systematical errors is somewhat arbitrary; however, the precautions taken (described in Sec. 3) and the agreement between the 3-in.×3-in. and 5-in.×5-in. NaI results support this estimate.

$F(K,X)$ is the least-squares fitted function of the form given above treating λ (and an arbitrary normalization factor) as adjustable parameters.

The column dev/$\sigma(K)$ is the ratio $[F(K,X)-\mu(K)]/\sigma(K)$ which gives an idea of the goodness of the fit.

Figure 3 summarizes the analysis. X is again the ratio of the probability for the virtual capture transition to the probability for inner bremsstrahlung decay. Negative X indicates destructive interference; positive X, constructive interference. $\lambda_0(X)$ is the value of λ giving the best least-squares fit with the data. n is the number of free parameters (14 in our case) and χ^2/n is the usual test for goodness of fit.

The best fit is obtained for $X=-0.002$ and $\lambda=0.49$. It is clear that the case of constructive interference is excluded completely. For destructive interference the fit gets rapidly worse for $X>0.06$. The worsening of the fit on both sides of the minimum at $X=-0.002$ is especially noticeable around $K=0.93$. This is to be expected, in view of the shape of the virtual capture photon spectrum.

Even the best fit cannot be considered good from a purely statistical point of view (for a good fit $\chi^2/n\approx 1$). Either the errors assigned are underestimated by about a factor of 2, or else the function $F(K,X)$ does not describe the situation perfectly. As already mentioned,

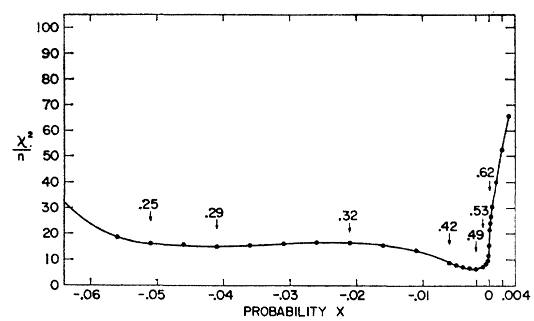

FIG. 3. Results of the least-square analysis. χ^2/n indicates the goodness of the fit. X is the probability of the virtual capture transition relative to the direct inner bremsstrahlung decay. Negative X refers to the case of destructive interference, positive X to constructive interference. The numbers above the arrows are the values of the inner bremsstrahlung parameter λ_0 which give the best fit; these values vary smoothly with X. The best fit with the data is obtained for $X=-0.002$ and $\lambda=0.49$.

no improvement in the least-square fit was obtained by including Coulomb and relativistic corrections. An attempt will be made to include some of the matrix elements which were neglected in Cutkowsky's formula and check their effect on the least-square fit.

It is difficult to state the error in the determination of X because it depends very sensitively on $\sigma(K)$; however, a reasonable estimate would be $X=-0.0020_{-0.004}^{+0.0015}$.

Saraf,[2] considering the Ni^{59} spectrum to be due to inner bremsstrahlung alone (i.e., $X=0$), obtained a best fit with $\lambda=0.33$. Hayashi[10] got a best fit with $\lambda=0.5$.

CONCLUSIONS

The best fit for the data is obtained by assuming that the probability for the virtual capture transition is 0.002 of the direct inner bremsstrahlung, and that the interference is destructive. $X=0.002$ corresponds to a

[10] T. Hayashi and J. R. Comerford, Atomic Energy Commission Report TID-6080, 1960 (unpublished).

half-life of 4×10^7 yr for the virtual capture transition. The preliminary estimate[1] for X was 0.025 and was based on the assumption that log $(ft)=5.7$ for the 3/2− to 5/2− transition, and $\Gamma_\gamma=10^{-9}mc^2$ for the width of the $M1$ transition. The experimental results indicate that the product of those two probabilities is in fact smaller by a factor of ∼10.

ACKNOWLEDGMENTS

The author is indebted to M. E. Rose and L. L. Foldy for proposing the experiment, to M. Kinsley for help with the chemical separations, to A. Schwarzschild for providing the least-square fitting program, and to J. Weneser, G. Scharff-Goldhaber, G. Emery, E. der Mateosian, and R. Marr for helpful discussions.

PHYSICAL REVIEW VOLUME 129, NUMBER 4 15 FEBRUARY 1963

Slightly Inelastic Proton-Deuteron Scattering*

D. G. STAIRS,† RICHARD WILSON, AND P. F. COOPER, JR.
Cyclotron Laboratory, Harvard University, Cambridge, Massachusetts
(Received 24 September 1962)

Measurements of the inelastic p-d cross section, in the region of very small momentum transfer, have been made at laboratory scattering angles of 5°, 10°, 15°, and 20°. The elastic p-d cross section has also been measured at these angles and compared with Postma's data. These measurements have been performed with a high-resolution magnetic spectrometer designed especially for this experiment. The p-d cross sections have been obtained by normalizing the p-d spectra to p-p spectra obtained by filling the same target with liquid hydrogen. The shape of the p-p spectrum, at a particular angle, was used to effect the separation of the inelastic p-d from the elastic p-d spectrum. The over-all energy resolution at small angles was about 0.75%.

A comparison of the elastic p-d data with the impulse-approximation calculation of Kerman, McManus, and Thaler yielded a value of Σ_t, the triplet amplitude sum, at the four angles measured. The singlet amplitude sum, Σ_s, was obtained by fitting Cromer's theory to the inelastic p-d cross section. The experimental values of the parameters Σ_s and Σ_t are compared with the predictions of the most recent phase-shift analyses. Σ_s appears to be particularly sensitive to the values of the $T=0$ amplitudes and thus experimental values of Σ_s may be useful in future phase-shift analyses.

INTRODUCTION

THE cross section for slightly inelastic proton deuteron scattering has been measured at four angles between 5° and 20° for an incident proton energy of 158 MeV. This report is concerned mainly with the experimental measurements whereas the theory and analysis of the data are dealt with in a companion article by Cromer.[1]

By the adjective slightly inelastic we refer to those collisions in which the incident proton transfers barely enough momentum to distintegrate the deuteron. Characteristically the momentum spectrum of the outgoing high-energy proton exhibits a rather small peak immediately below the elastic p-d peak. The shape of this slightly inelastic peak is determined mainly by the strong attractive potential operating in the final state (principally ^1S) of the recoiling two-nucleon system. The interest in studying collisions of this type derives from the fact that the cross section[1–3] can be shown to be rather sensitively dependent on the $T=0$ nucleon-nucleon scattering amplitudes.[4,5]

Earlier experimental work providing an important basis for the investigations reported here has been carried out at Harwell,[6,7] Uppsala,[8] and Harvard.[9,10]

GENERAL METHOD

The 158-MeV unpolarized proton beam was allowed to strike a small target of liquid deuterium. Protons scattered from the target were momentum-analyzed by a high-resolution magnetic spectrometer and detected by a simple array of scintillation counters. At each particular scattering angle the momentum spectrum of

* Supported by joint program of the Office of Naval Research and U. S. Atomic Energy Commission.
† Supported by the National Research Council of Canada.

[1] A. H. Cromer, following paper [Phys. Rev. **129**, 1680 (1963)].
[2] L. Castillejo and L. S. Singh, *Proceedings of the International Conference on Nuclear Forces and the Few-Nucleon Problem* (Pergamon Press Ltd., New York, 1960).
[3] A. K. Kerman, H. McManus, and R. L. Thaler, Ann. Phys. (New York) **8**, 551 (1959).
[4] G. Breit, M. H. Hull, K. E. Lassila, and K. D. Pyatt, Phys. Rev. **120**, 2227 (1960).
[5] M. H. Hull, K. E. Lassila, H. M. Ruppel, F. A. McDonald, and G. Breit, Phys. Rev. **122**, 1606 (1961).
[6] J. P. Scanlon and J. J. Thresher, in *Proceedings of the International Conference on Nuclear Forces and the Few-Nucleon Problem*, edited by T. C. Griffith and E. A. Power (Pergamon Press Ltd., New York, 1960).
[7] M. J. Esten, T. C. Griffith, G. J. Lush, and R. J. Metheringham, *Proceedings of the International Conference on Nuclear Forces and the Few-Nucleon Problem* (Pergamon Press Ltd., New York, 1960).
[8] H. Tryen, G. Tibell, and Th. A. J. Maris, Nucl. Phys. **7**, 281 (1958).
[9] H. Postma and R. Wilson, Phys. Rev. **121**, 1229 (1961).
[10] A. Kuckes, R. Wilson, and P. F. Cooper, Jr., Ann. Phys. (N. Y.) **15**, 193 (1961).