configurations of the other states to the values shown in Table I. An alternative approach, independent of any normalization, would have created greater uncertainties by its dependence on both the absolute values of $\Gamma_{mod}{}^{j}$ and the extraction of the elastic resonance widths (the latter being subject to the difficulty mentioned above).

Results from the most recent shell-model calculation by True[3] are compared in Table I with the experimentally determined wave functions. The theoretical column gives the energies and wave functions for the first seven predicted states in Pb^{206}, and the experimental column shows that at least five are in excellent agreement with the measurements. If the known 4^+ state at 1.68 MeV is similar to the predicted 4^+ state at 1.60 MeV, then resonances for this state would not have been observed. This is consistent with our assigning the resonances in the cross section at the bottom of Fig. 1 entirely to the 1^+ state at 1.71 MeV which, as Table I shows, is then in accord with the predicted 1^+ state.

The conclusions drawn from the results presented here may be summarized as follows:

(1) The low-energy structure of Pb^{206} appears to be well described in terms of two-neutron holes moving in the shell-model potential of the Pb^{208} core.

(2) It is possible to extract quantitative spectroscopic information from inelastic scattering at isobaric analog resonances, in a relatively easy manner, at least in those regions of the Periodic Table where the simple shell model provides a good description.

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Neutron Groups in the Spectrum of a PuF_4 Source*

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The neutron spectrum of a 1-kg PuF_4 laboratory neutron source was measured by use of nuclear track emulsion and a differentiation technique. The spectrum consists of a minor peak at 0.91 MeV and a major one at 1.55 MeV. The mean energy was found to be 1.35 MeV and the maximum energy 2.8 MeV. Special analysis of the track distribution revealed peaks at 0.91, 1.26, 1.44, 1.64, 1.8, and 2.1 MeV, and weaker ones at 0.5 and 2.5 MeV. The measured spectrum does not differ in principal features from an analytical spectrum obtained by a graphico-numerical study of published thin-target measurements. The analytical study predicts the effects on $F(\alpha,n)$-source spectra when α emitters of different energy from Pu^{239} are employed, and permits an interpretation of some previously unexplained observations by Chadwick and Constable.

1. INTRODUCTION

FLUORINE is one of the few elements that produce fast neutrons in good yield when bombarded with polonium-210 α rays. Because of this, the $F(\alpha,n)$ reaction was the subject of many early studies. Ion- and cloud-chamber measurements in the 1930's established the existence of several groups of neutrons, in the energy range 0.4 to 2.5 MeV, that were associated with α-particle resonances to levels in the resulting Na^{22} nucleus.[1,2] More recent studies of the $F^{19}(\alpha,n)Na^{22}$ reaction, by use of α-particle beams and CaF_2 targets, have provided detailed information on the neutron[3–6] and γ-ray[6–8] yields associated with resonances through the closely spaced 12.5- to 14.5-MeV levels in the compound Na^{23} nucleus. There is current interest in this reaction because it is a means of studying excited states in Na^{22} and Na^{23}, and because it is the basis for important laboratory neutron sources.[9–12]

Neutron spectral measurements have been reported for a small PoF source,[11] and one for the same 1-kg PuF_4 source used in this study.[12] The agreement between these spectra, however, was poor and the

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[1] J. Chadwick and J. E. R. Constable, Proc. Roy. Soc. (London) **A135**, 48 (1932).
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[12] E. Tochilin, in *Neutron Dosimetry* (International Atomic Energy Agency, Vienna, 1963).

PuF_4 spectrum did not match up particularly well with spectral parameters obtained by use of a precision-calibrated long counter.[13] For these reasons, and also because there have been recent improvements in the measurement and interpretation of fast-neutron spectra,[14,15] it seemed worthwhile to remeasure the neutron spectrum of the laboratory PuF_4 source, and to make an analytical attempt to reconcile the measured spectrum with published beam-target data.

2. MATERIALS AND METHODS

Unmounted 1×3-in. pellicles of Ilford L.4 emulsion, 600 μ thick, were wrapped in two layers of black paper and exposed edge-normal 50 cm from a 1-kg-PuF_4 laboratory source[16] for a 3-day period under conditions designed to minimize scattering. The pellicles were subsequently developed and scanned by use of a random-drift sampling technique, and the tracks were analyzed by use of a differentiation procedure. The track sampling was carried out in two steps in order to equalize the number of short and long tracks in the channels of the track distribution.[14,15]

Approximately 9200 tracks, biased against those of shorter length, were selected and measured in the first sample. The second bias-free sample contained measurements of an additional 3000 tracks. A separate computer analysis was performed on each sample in order to find the number and fraction of tracks in each length channel that lay within a forward cone of half-angle

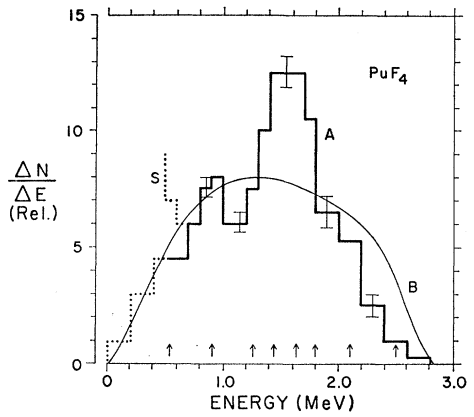

FIG. 1. Neutron spectrum of a PuF_4 source. A: measured by differentiation of 12 000-track recoil-proton distribution. (Region below 0.5 MeV is estimated.) Arrows indicate peaks revealed by special analysis of track distribution. B: theoretical spectrum from Ref. 15. S: spurious peak from plural scattering.

30° with respect to the source-emulsion axis. This fraction has been shown to be a sensitive indicator of neutron peaks hidden under the short-track scattering peak, and a useful means of identifying and verifying the presence of peaks elsewhere in the spectrum.[15]

3. RESULTS

A. Experimental. The PuF_4-source neutron spectrum derived from 12 200 measured tracks is presented in Fig. 1. The error bars represent satistical uncertainty only, and were obtained by allowing the points in the corrected[17] proton-recoil track-energy spectrum to vary randomly within their statistical limits, and by redrawing the smooth curve several times for differentiation.

In order to indicate roughly the size of the proton track-energy channels from which it was derived, the spectrum is depicted in 0.1-MeV steps below 1.8 MeV and 0.2-MeV steps above. These channel widths were chosen to be consistent with the inherent limitation in emulsion spectroscopy, arising from proton-range straggling that also varies from 0.1 to 0.2 MeV in these regions.[18]

Curve S indicates a spurious peak due to high-angle protons set in motion by low-energy neutrons that suffered plural scattering external to the source. This low-energy neutron scattering is a well-known phenomenon that is impossible to avoid experimentally. Although curve S does not represent the *emitted* source spectrum, it does correctly give the exposing neutron spectrum, a feature of emulsion spectroscopy important in neutron dosimetry.[19]

Below 0.5 MeV, where nuclear emulsion ceases to provide reliable spectral information, the curve follows a spectrum calculated by use of a graphico-numerical method, described below in Sec. 3B, that is based on beam-target data. Including this estimate of the 0- to 0.5-MeV region and excluding S, the mean energy of the measured spectrum (Fig. 1) is 1.35 MeV. This may be compared with 1.4 MeV obtained by direct measurement of a PuF_4 source.[13] A theoretical PoF spectrum calculated by Hess[20] is shown for comparison. The spectra are not strictly comparable but the agreement in the high-energy region is better than it appears, because the 5.3-MeV Po^{210} α rays are expected to produce more 2- to 2.9-MeV neutrons than the 5.10- to 5.15-MeV Pu^{239} rays.

B. Analytical. Recent measurements have established

[13] J. De Pangher, Nucl. Instr. Methods **5**, 61 (1959).
[14] R. L. Lehman, O. M. Fekula, and J. R. Wayland, Jr., Nucl. Instr. Methods **60**, 205 (1968).
[15] R. L. Lehman, Nucl. Instr. Methods **60**, 253 (1968).
[16] PuF_4-3, fabricated at the Hanford Laboratories (now Batelle Northwest) Richland, Wash. The source was in the shape of a 10-cm right circular cylinder and consisted of PuF_4 powder in a double aluminum welded container. Exposures were made about 6 months after its manufacture. The emission rate was 6×10^6 neutrons/sec (see Ref. 12).
[17] The raw track-energy spectrum was corrected for track loss depending upon track length, the dimensions of the emulsion, and the exposure geometry (see Ref. 14).
[18] See, for instance, J. R. Bird and R. H. Spear, Australian J. Phys. 8, 567 (1955); J. H. Roberts, Rev. Sci. Instr. **28**, 677 (1957).
[19] See, for instance, R. L. Lehman and O. M. Fekula, Nucleonics **24**, 35 (1964); in *Personnel Dosimetry for Radiation Accidents* (International Atomic Energy Agency, Vienna, 1965), p. 297.
[20] W. N. Hess, University of California Radiation Laboratory Report No. UCRL-3829, 1957 (unpublished); see also the PoF spectrum published on p. 70 of Ref. 13.

that the neutron emissions associated with reactions in a PuF_4 source occur as a series of many sharp (10- to 40-keV) α-particle resonances through closely spaced (40-keV) levels in the compound nucleus.[6] Other studies have directly shown that neutrons are emitted in resonance reactions leading to five states in the final Na^{22} nucleus.[4,5] This complex situation is depicted in Fig. 2 by use of a Lauritsen plot,[21] and the limits in neutron energy determined by the 2-body Q equation for the transitions to the five final states[22] are shown in Fig. 3. Because of this complexity, and the fact that presently there is very little information available on the

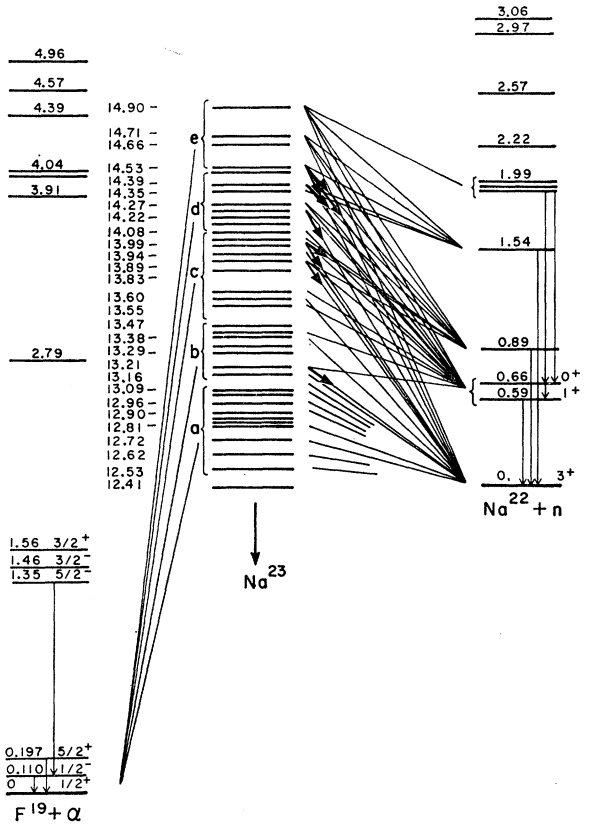

FIG. 2. Energy-level scheme used in analysis of neutron groups. Assembled from data in Ref. 20. γ rays associated with reaction are also shown.

angular distribution of neutrons to any of these states, it seems hopeless to attempt an exact theoretical calculation of a neutron spectrum for a PuF_4 source. However, in order to compare the measured source

[21] T. Lauritsen and F. Ajzenberg-Selove, Rev. Mod. Phys. **27**, 77 (1955); W. Kunz and J. Schintlmeister, *Nuclear Tables* (Pergamon Press, Frankfurt, 1965), Vol. I, Part II; C. M. Lederer, J. M. Hollander, and I. Perlman, *Table of Isotopes* (John Wiley and Sons, Inc., New York, 1967), 6th ed., p. 162; C. Maples, G. W. Goth, and J. Cerny, University of California Radiation Laboratory Report No. UCRL-16964, 1966 (unpublished).

[22] The 0.588 and 0.662 states are considered as one state in this analysis.

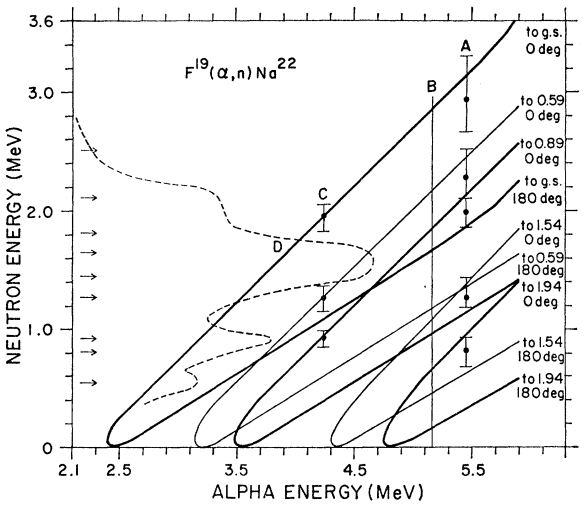

FIG. 3. Energy limits of the neutron groups. A: 30° data from Ref. 5. B: maximum energy of Pu^{239} α rays. C: 0° data from Ref. 5. D: measured source spectrum. Arrows indicate peaks in neutron energy, as in Fig. 1.

spectrum with the beam-target data, a spectrum was calculated by use of the following numerical method.

Relative excitation functions to the first four relevant states in Na^{22} were obtained from data in Refs. 4 and 6, and sketched out as curves A through D in Fig. 4. Since the experimental curves consisted of many sharp resonances, it was necessary, in order to facilitate the present analysis, to smooth them very strongly. Moreover, because the important excitation to the ground state (curve A) has not been systematically measured beyond 3.5 MeV, it was necessary to estimate this portion of curve from isolated observarions.

In order to find the number of neutrons in each group, the areas under the broad resonances in the partial cross sections were obtained graphically and weighted by the relative number of "steady-state" α particles

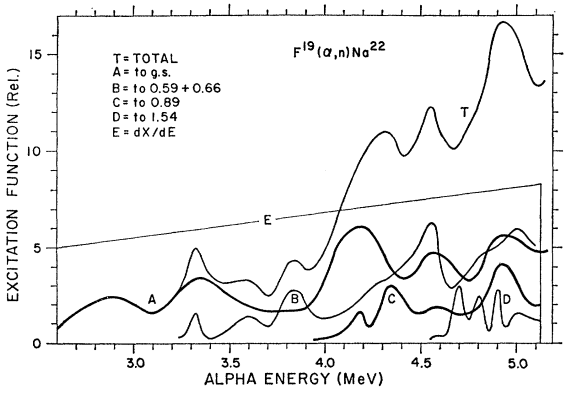

FIG. 4. Excitation functions to levels in Na^{22}. Relative values, deduced from measurements in Refs. 4–8. The curves have been very strongly smoothed. Curve E is relative steady-state α-particle energy distribution in the source.

TABLE I. Neutron groups in the PuF_4 spectrum.

Transitions to	Group No.	E_α (MeV)	Principal Na^{23} resonance levels	Principal peaks E_n (MeV)	Group size
g. s.	a_0	2.4–3.1	12.72, 12.81	0.5	5.1
	b_0	3.1–3.6	13.16, 13.21, 13.38	0.8–1.0	5.8
	c_0	3.6–4.0	13.60	1.2	3.9
	d_0	4.0–4.4	13.89, 13.94, 13.99	1.4–1.7	13.6
	e_0	4.4–4.8	14.22	1.8–2.0	8.5
	f_0	4.8–5.1	14.53, 14.71, 14.90	2.1–2.8	10.3
0.590 & 0.660	b_1	3.3–3.6	13.21, 13.40	0.5	1.0
	c_1	3.6–4.0	13.55, 13.65	0.5–0.6	2.2
	d_1	4.0–4.4	13.89, 13.99, 14.08	0.8–1.0	3.9
	e_1	4.4–4.8	14.22, 14.27, 14.39	1.2–1.4	7.7
	f_1	4.8–5.1	14.53, 14.71, 14.90	1.5	17.4
0.89	d_2	4.0–4.4	13.89, 13.99, 14.03	0.4–0.8	4.8
	e_2	4.4–4.8	14.27, 14.39	0.9–1.1	5.8
	f_2	4.8–5.1	14.53, 14.71, 14.90	1.2–1.5	4.2
1.54	e_3	4.6–4.8	14.35, 14.39	0.5	2.2
	e_3	4.8–5.1	14.53, 14.71, 14.90	0.6–0.90	2.9
1.94	f_4	4.8–5.1	14.90	0.5	0.7
					99.8

having the given energy inside the PuF_4 source, as determined by a dx/dE curve.[23] In the relevant energy region the slope of the dx/dE curve does not vary appreciably from element to element, so it was not necessary to compute partial contributions from F and from Pu.

The neutrons in each group were then distributed in energy as allowed by the limits of the Q-equation curves (Fig. 3). The results of this analysis are given in Table I and Fig. 5. The center-of-mass angular distributions were assumed to be isotropic within each group, except in one or two cases where large resonances were known to occur at the extremes of the α-particle energy leading to a group. This assumption seems reasonable because

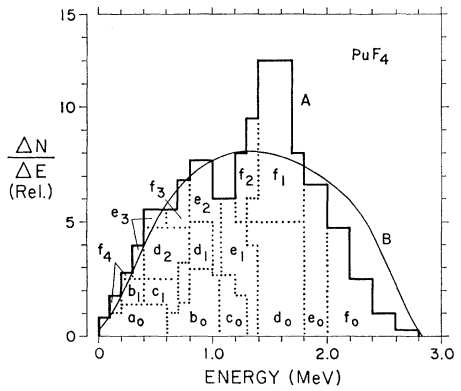

FIG. 5. Neutron groups in the PuF_4 source spectrum. A: composite graphico-numerical result (see Table I for identification of groups). B: theoretical spectrum from Ref. 19.

partial-wave analysis shows that only s- and p-wave neutron emission is probable in this energy region,[24] because one of the prominent resonance emissions (12.72 MeV in Na^{23}) is known to be isotropic,[4] and when the width of the groups and the resolution in the measured spectrum is considered, only extreme backward or forward scattering can make an appreciable difference in the shape of the total neutron spectrum. Nonetheless, the angular distributions are strongly peaked forwards and backwards in parts of the excitation spectrum,[5] indicating mixing and overlapping of adjacent levels, so the validity of assuming uniform laboratory neutron distributions for the groups may be questioned.

4. DISCUSSION

A. Measurements. When considering the neutron emission from a 10-cm source, it is important to examine the possible modification of the spectrum by internal scattering. By assuming atomic densities of 0.8 and 3×10^{22} cm^{-3} for Pu and F, respectively, and a mean path of 4 cm for 1.5-MeV neutrons, one can estimate that 6% of the emitted neutrons excite fast n,f and 5% n,n' reactions in Pu^{239}, and that 22% of the neutrons suffer elastic scattering with F within the source. About 12% of the emitted neutrons will suffer elastic scattering with Pu atoms, but neither this nor the elastic scattering by F or Al in the outer jackets will appreciably affect the mean neutron energy. Moreover, since the n,n' group is modest in relative size and is spread over the entire spectrum, it is not expected to modify the emitted spectrum significantly. Although the source material is poor at thermalizing 1.5-MeV neutrons, the thermal neutron fission of Pu^{239}, because of the 700-b cross section, adds an unknown contribution to the stray neutrons from fast fission. The occurrence in the recoil spectrum of occasional tracks extending to 5 MeV indicates that fission neutrons are present in the source spectrum, but their total contribution must be small ($<10\%$); otherwise the characteristic peaking in fission spectra at 0.75 MeV would be a prominent feature in the measured spectrum.[25]

The shape of the experimental spectrum generally agrees with that reported for a small PoF source,[11] but the position of the major peak was 0.25 MeV below the 1.55 MeV reported here. The same workers provided an analytical spectrum that was consistent with their measurements. They incorrectly assumed, however, that the stopping power of 3- to 5-MeV α rays in their source was flat; had they used a sloping dx/dE curve,[23] their analytical spectrum would peak at about 1.5 MeV. The fractions of neutrons emitted in transitions to the 0.59- and 0.66-MeV states, corresponding to groups b_1 through e_1, amount to 0.32 in our work. This may be

[23] L. C. Northcliffe, Ann. Rev. Nucl. Sci. **13**, 87 (1963); National Academy of Science—National Research Council Publication No. 1133, 1964, p. 374; G. W. Gobeli, Phys. Rev. **103**, 275 (1956).

[24] Robley D. Evans, *The Atomic Nucleus* (McGraw-Hill Book Co., New York, 1955), p. 410 ff and p. 872 ff.

[25] N. Nereson, Phys. Rev. **88**, 823 (1952).

compared with 0.30, the lower limit for transitions to these levels based on n,γ coincidence measurements by these authors.

The spacing of the eight peaks found by special examination of the track sampling channels (Sec. 2 and arrows, Fig. 1) is just sufficient to suggest that future detailed emulsion or crystal spectrometry may be able to resolve them.[26] Above 1 MeV the positions of these peaks agree with those obtained for the neutron groups in cloud-chamber measurements,[2] if one assumes that these measurements did not resolve the three peaks centering at 1.44 MeV. The cloud-chamber peak at 1.08 MeV is well above ours at 0.91 MeV, but considering the uncertainty in early range-energy determinations, these may be the same peak. The detailed interpretation of these peaks must await differential cross-section measurements. Even then, the interpretation will be complicated in the region below 1.8 MeV because each peak is composed of at least two neutron groups (Fig. 5).

Figure 5 permits one to interpret some puzzling observations of Po-α-F neutrons by Chadwick and Constable.[1] By use of ion chambers they found six groups of recoil protons in the region above 1 MeV. Groups 5 and 6 (2.3 to 3 MeV) disappeared from the spectrum when the energy of the α rays reaching the thick CaF_2 target was reduced to 5 MeV. Moreover, when the α-ray energy was reduced to about 4.7 MeV, groups 2 and 4 disappeared; finally, when the energy was 4.3 MeV groups 1 and 3 disappeared. These observations may be explained in part as follows. When the α-particle energy is reduced from 5.3 to 5 MeV, the high-energy tail of group f_0 (corresponding to groups 5 and 6) disappears. As the impinging α-ray energy is further reduced the rest of f_0 (2.1 MeV) as well as f_1+f_2 (1.55+1.3 MeV, respectively), corresponding to groups 2 and 4, vanish. When the α energy drops still more to 4.3 MeV, e_0 (1.9 MeV) as well as e_1+e_2 (1.2+0.95 MeV, respectively), corresponding to groups 1 and 3, vanish, leaving only low-energy groups undetected by these authors.

B. Analytical spectrum. The high-energy part of the calculated spectrum can only be approximate because of the uncertainty associated with the 4- to 5-MeV region of curve A in Fig. 4, and therefore with the size of the neutron groups d_0, e_0, and f_0. The agreement between the calculated and measured spectra, however, may be interpreted as a *prediction* that the excitation function to the ground state is substantially as estimated. More beam-target measurements are needed to settle this point. The numerical calculation would also be strengthened by angular-distribution studies and by a total cross-section measurement for the $F^{19}(\alpha,n)$ reaction similar to that reported for the Be^9 (α,n) reaction.[27] The total neutron excitation curve in Fig. 4 was simply estimated by summing the curves A through D. No direct-reaction component was included in the figure because the classical potential barrier for $F^{19}(\alpha,x)$ reactions is 6.7 MeV, far in excess of the 5.3-MeV energy limit considered here. That the total cross section for neutron emission should begin a strong rise at 4 MeV, well below this barrier, seems reasonable not only because of progressive reduction in potential barrier width, but also because more reaction exit channels for neutron emission become available just above this energy.

The use of a more energetic α-emitter than Pu^{239} in laboratory $F^{19}(\alpha,n)$ neutron sources, such as Am^{241} (5.49 MeV), Cm^{244} (5.81 MeV), or Es^{254} (6.44 MeV), should increase the number of neutrons in the f groups and excite higher resonance (and possibly direct-interaction) groups g, h, etc., not considered here, while the groups a through e remain unchanged. On the other hand, if the 3.18-MeV α-emitter Gd^{148} were employed, only group a_0 would be excited.

ACKNOWLEDGMENTS

The author is indebted to Eugene Tochilin for exposing the nuclear emulsions to a PuF_4 neutron source at the U. S. Naval Radiological Defense Laboratory, Miss Olga M. Fekula for technical assistance in obtaining and analyzing the measurements, Professor Walter H. Barkas for valuable discussions, and Dr. Roger Wallace for supporting the development of the automatic coordinate-readout microscope that made these measurements possible.

[26] For instance, see the resolution in D. Haag and H. Fuchs, Z. Physik **174**, 227 (1963).

[27] J. H. Gibbons and R. L. Macklin, Phys. Rev. **137**, B1508 (1965).