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RESONANCE STRUCTURE IN THE FISSION OF (^{235}U + n)

ROUGH DRAFT

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A new multilevel reduced R-matrix analysis of the neutron-induced resonance cross sections of ^{235}U has been carried out. We used as a constraint in the analysis the angular anisotropy measurements of Pattenden and Postma, obtaining a Bohr-channel (or J, K channel) representation of the resonances in a two-fission vector space for each spin state. Hambsch *et al.*, have reported definitive measurements of the mass- and kinetic-energy distributions of fission fragments of (^{235}U + n) in the resonance region and analyzed their results according to the fission-channel representation of Brosa *et al.*, extracting relative contributions of the two asymmetric and one symmetric Brosa fission channels. We have explored the connection between Bohr-channel and asymmetric Brosa-channel representations. The results suggest that a simple rotation of coordinates in channel space may be the only transformation required; the multilevel fit to the total and partial cross sections is invariant to such a transformation.

1. INTRODUCTION: AN HISTORICAL PERSPECTIVE

The first low-energy neutron resonance total and fission cross-section measurements were made by E. E. Anderson *et al.*, as part of the Los Alamos Manhattan Project, and were reported in March 1947, informally, by B. McDaniel as part of the Los Alamos Technical Series[1]. Resonances were observed at 0.3, 1.1, 2.0, 3.3, 4.8, 5.7, 9, 12, 20, and 40 eV, and there was a follow-on experiment done to make sure that the 2.0 and 4.8 eV resonances were not due to a ^{234}U contaminant, as they were not seen in the fission cross section but only in the transmission measurements. It has been said that the very existence of resonance structure in fissile isotopes came as a surprise to many: If one assumes that $\Gamma_f \approx ND/2\pi$ (following Bohr and Wheeler[2], where Γ_f is the average fission width, D the spacing, and N the number of fission channels), and counts as a fission channel each of the possible pairs of fission products, one can conclude that $\Gamma_f \gg D$, such that there should be no observable resonance structure. Bohr and Wheeler were careful to avoid misleading their readers;

in their paper they defined N as the "number of levels in the transition state available with the given excitation".

When one refers to Bohr transition states or Bohr fission channels, however, the reference is not to the Bohr-Wheeler paper, but to the paper A. Bohr presented at the first Geneva Conference in 1955[3]. Here the idea was that the transition states are saddle-point states, where the nuclear angular momentum should be concentrated on simple collective modes of motion, associated with characteristic angular distributions of the fragments. Bohr also suggested that differences in the fission threshold for states of different spin and parity in the compound nucleus could lead to peculiar selection rules in the fission process. Following these ideas, Wheeler[4] suggested that symmetric fission for ^{235}U in the 3^- state might be anomalously low if the transition from saddle to scission were to preserve the wave-function symmetry properties.

Bohr's and Wheeler's suggestions sparked a large experimental effort. Early attempts to measure the angular distribution of the fragments from fission of aligned targets of ^{235}U by low-energy neutrons and to infer the K quantum numbers of the Bohr channels were undertaken by Dabbs, Roberts *et al.*,[5-7] and led to the definitive measurements by Pattenden and Postma[8]. Radiochemical measurements of the peak-to-valley distribution of fission fragment masses as a function of neutron energy in the resonance region led to the definitive work reported by Cowan *et al.*[8] The determination of resonance spins by measuring the interaction of polarized neutrons or polarized targets was first undertaken by Sailor, Schermer *et al.*,[10,11] and led to the definitive measurements by Keyworth *et al.*[12] Early fragment kinetic-energy measurements by Melkonian, Felvinci *et al.*[13,14] were forerunners of the recent measurements by Hambsch *et al.*[15]

The earliest measurements[1] of resonance fission in (^{235}U + n) showed that the fission widths are widely varying, from which it could be inferred that in this region fission is a few-channel process. Detailed measurements of the resonance shapes, e.g., those of Shore and Sailor[16], showed asymmetries due to level interference, and led to the recognized need for a multilevel multichannel approach in the analysis[17-19].

While it has always been obvious that a two-channel multilevel representation is not unique, in that the calculated fission cross section is invariant to rotation or mirror reflection of the fission-vector set, the full extent of the problem was not realized. In 1971, Auchampaugh carried out a study[20] showing that equivalent solutions can be obtained under reflection of certain fission-vector subsets about two orthogonal axes. The number of such

non-trivial solutions was estimated by Adler and Adler[21] as $\geq 1+(N-1)(N-2)/2$, where N is the number of resonances considered.

One of the early studies that pointed the way for the present paper may be of interest. In 1965, Moore and Miller[22] measured the relative cold fission yield as a function of neutron energy about the 0.28 eV resonance in ($^{235}U + n$) and showed that the fit to these data corresponds to a unique fission-vector orientation in a two-channel space. Some years later, Pattenden and Postma[23] inferred the relative distribution of fission through different K states in this same energy region, and observed almost exactly the same curve. We looked for this same characteristic curve in the fragment kinetic-energy distribution measured by Hambsch *et al.*[15] No significant variation is to be seen.

2. A NEW EVALUATION OF THE RESONANCE CROSS SECTIONS

In 1981, at an IAEA consultants meeting on uranium and plutonium resonance parameters, Moore *et al.*[24] described the resonance evaluation currently in use in ENDF/B-V, and sketched how one might proceed in obtaining a new evaluation with physically meaningful partial fission-width parameters. At that same meeting, it was recommended that a new evaluation of the ^{235}U resonance region be undertaken.

This new evaluation is now virtually completed[25]. It covers the energy range below 2250 eV, with most attention given to the well-resolved structure below 110 eV. In this region, four sets of data were included in the final fitting: the 1984 fission measurements of Gwin *et al.*[26] below 20 eV, the 1984 fission measurements of Weston and Todd[27] above 15 eV, the 1987 transmission measurements of Spencer *et al.*[28] below a few eV, and the 1988 transmission measurements of Harvey *et al.*[29] above that range. Spin assignments were made with the spin-separated fission data of Moore *et al.*,[12,30] but no attempt was made to utilize the K-distribution data of Pattenden and Postma.[8] That is the subject of the present paper.

The method of analysis used the resonance parameter code SAMMY[31] to perform consistent R-matrix multilevel analyses of the selected neutron cross section and transmission measurements. The fitting procedure is based on Bayes' method and allows the successive incorporation of new data in a consistent manner. The option to search not only for resonance parameters but also for experimental parameters such as sample thickness, sample effective temperature, backgrounds, normalizations, and the parameters of the instrumental resolution, all consistent with predetermined uncertainty limits, leads to realistic uncertainties and covariance matrices. A special feature of SAMMY programmed for this evaluation allows the fitting of the separate spin contributions to the

fission cross section. Another special feature programmed for the present paper allows one to search only on the fission-vector phases, and to calculate the separate channel contributions in each spin state.

3. TOWARD A UNIQUE MULTILEVEL DESCRIPTION.

Having obtained a fit to the fission, capture, and total cross sections of ^{235}U, we expect[21] that below 30 eV there are at least 381 + 931 = 1312 non-trivially different parameter sets characterized by different fission vector orientations. For only one of these parameter sets, the fission-vector orientations correspond to the correct K-quantum number distribution for each resonance. This is the solution that we should like to find, because we have assumed that this is the one that has physical content: it is the one reflected in the measurements by Pattenden and Postma[8] of the angular anisotropy of the fragments emitted in the resonance fission of aligned ^{235}U target nuclei.

There are three problems in finding this solution. First, the Pattenden-Postma data are incomplete, in that the anisotropies are reported only for a limited number of resonances. Secondly, the measured anisotropy for a given resonance does not uniquely determine the vector orientation in a two-channel space. It gives only the absolute value of the sine or cosine of the angle. Thus, even if we knew the anisotropies perfectly for each resonance, there would still be four possible vector orientations: $\pm\theta$, and $180°\pm\theta$, only one of which is the correct one. Thirdly, for spin 3^- resonances, a two-channel representation for the vector space may not be adequate, K = 0, 1, and 2 being the lowest expected open channels. We used it anyway, because the fitting code is restricted to a two-fission-channel representation, and because a detailed examination of the Pattenden-Postma results shows that the K = 0 contribution is anomalously low, for J = 3 as well as for J = 4.

The Bohr-channel phase constraint was calculated from Pattenden and Postma's A_2 values as follows: For J = 3, we ignored the K = 0 channel, with A_2 = -2.92, and used that for K = 1, A_2 = -2.19. Since A_2 = 0 for K = 2, and there is also no evidence for any K = 3 fission, the equation becomes

$$-A_2 = (0.90)(2.19)\ \Gamma_{f1}/\Gamma_f = (0.90)(2.19)\ \sin^2\theta \quad , \qquad (1)$$

where the factor 0.90 takes into account the Pattenden-Postma crystal misalignment (see note added in proof to Ref.8), and θ is the angle the fission-width vector makes with the K = 2 channel. We note that if there is a K = 0 component (such that $-A_2$ lies between 2.19 and 2.92 for an effective K = 0 plus K = 1 channel), the effect on our derived value of θ relative to the K = 2 axis can be represented as a slight rotation of the vector. For J = 4, the equation is

$$-A_2 = 0.90\ (2.48\ \sin^2\theta + 1.17\ \cos^2\theta)\ , \qquad (2)$$

where θ is again the angle relative to the K = 2 axis. We ignore K = 0, 3, 4.

From Auchampaugh's study[20], we know that the non-uniqueness of the fit involves only the phase angle describing the vector orientations. Our fitting procedure, then, was to solve for the absolute phase angles for those resonances for which it had been determined by Pattenden and Postma, and then use the multilevel fit to establish the correct value of $\pm\theta$ or $180°\pm\theta$ using the one that most nearly resembled the solution found. We next allowed the other vectors (those for resonances that were unreported by Pattenden and Postma) to vary, in order to find a solution for the phases of these vectors that would be most compatible with those that were constrained. When convergence stopped, we then allowed all the vector orientations to vary, the Pattenden-Postma data uncertainties being large enough that the angles are known only to perhaps ±10 to 20°.

For J = 3, we found that the results of this procedure were quite satisfactory. After a mirror transformation of the trial set[25], we found that only a small rotation was required to bring essentially all the vectors below 30 eV into agreement with the Pattenden-Postma data, as shown in Fig. 1. For J = 4, we encountered some inconsistencies. For example, the multilevel fit to the 8.8-9.3 eV doublet requires that the phase angle between these two fission vectors be ±80°, while the Pattenden-Postma data seem to require them to be nearly parallel. Preliminary results of this fitting are also shown in Fig. 1.

4. THE BROSA-CHANNEL DESCRIPTION

Hambsch *et al.*[15] reported fluctuations in the fission-fragment mass distribution, which are strongly correlated with fluctuations in $\bar{\nu}$ and total kinetic energy. But the calculated correlation of the Hambsch results with the Bohr-channel distribution of K values (i.e., Pattenden and Postma A_2 variations) was reported to be quite weak: r = (-0.37 ±0.17). Hambsch *et al.* analyzed their data by fitting their mass distribution to five Gaussians, two for each of the two asymmetric mass peaks and one for symmetric fission, which described the mass distribution as partial mass distributions for each of the three exit fission channels in the multimode model of Brosa *et al.*[32] Their data are reported as a relative value of (W_1/W_2) where W_1 and W_2 are the weights of the "Standard-1" and "Standard-2" characteristic mass distributions arising out of the Brosa *et al.* study.

In order to interpret these results as fluctuating vectors in a two-channel fission-vector space, we simply took the largest and smallest values of $(W_1/W_2)_R$ reported by Hambsch *et al.* for the J = 4 resonances in (^{235}U + n):

0

Fig. 4 Comparison of fission-width vector orientations. In the top curves are shown the vectors corresponding to a possible multilevel solution for J = 3 and J = 4 for those resonances for which Pattenden and Postma[8] reported data. The two middle curves show the Bohr channel vectors we derived as a starting point from the Pattenden-Postma results, from which these multilevel solutions was obtained. In the lower curves are shown the Brosa-channel vectors derived from the data of Hambsch et al.[15] The tilt of the Brosa-channel space relative to that of the Bohr-channel space is the same for both spins.

0.941 for the resonance at 4.85 eV, and 1.206 for the resonance at 19.3 eV, and assumed that these represent fission vectors oriented at 90° in a two-channel vector space, along the two axes. According to this assumption, the phase relation is then given for any spin-4 resonance as

$$(W_1/W_2)_R - 0.941 = 0.265 \sin^2 \theta' \quad , \tag{3}$$

where θ' is the angle between the fission vector of the resonance being considered and the Brosa "Standard-2" axis.

In principle, we could have at this stage carried out the same kind of phase optimization that we did in using the Pattenden-Postma data as a constraint on the multilevel description. In practice, we found by inspection that a rotation of the axes of 18° appears to be sufficient to transform the Bohr-channel description determined by Pattenden and Postma into the Brosa-channel description of Hambsch *et al.*

For J = 3 resonances, we arbitrarily assumed the prescription

$$(W_1/W_2)_R - 0.941 = 0.299 \sin^2\theta' \quad , \tag{4}$$

and found again that the same rotation of 18° appears to obtain; these results are also shown in Fig. 1. Perhaps it is not surprising that the same transformation applies for both spin states, i.e., it seems to be independent of J.

When we began this study, we thought we might be able to show that each of the Bohr channels is spread into the two asymmetric Brosa channels with slightly different weights. If this were the case, the fluctuations in mass and kinetic energy observed by Hambsch *et al.* would follow the Bohr-channel description, which would be superposed on a constant background. The rotation we seem to have seen suggests that the feeding of Bohr channels into Brosa channels takes place coherently, so that the reduced-width amplitudes are summed in the transformation.

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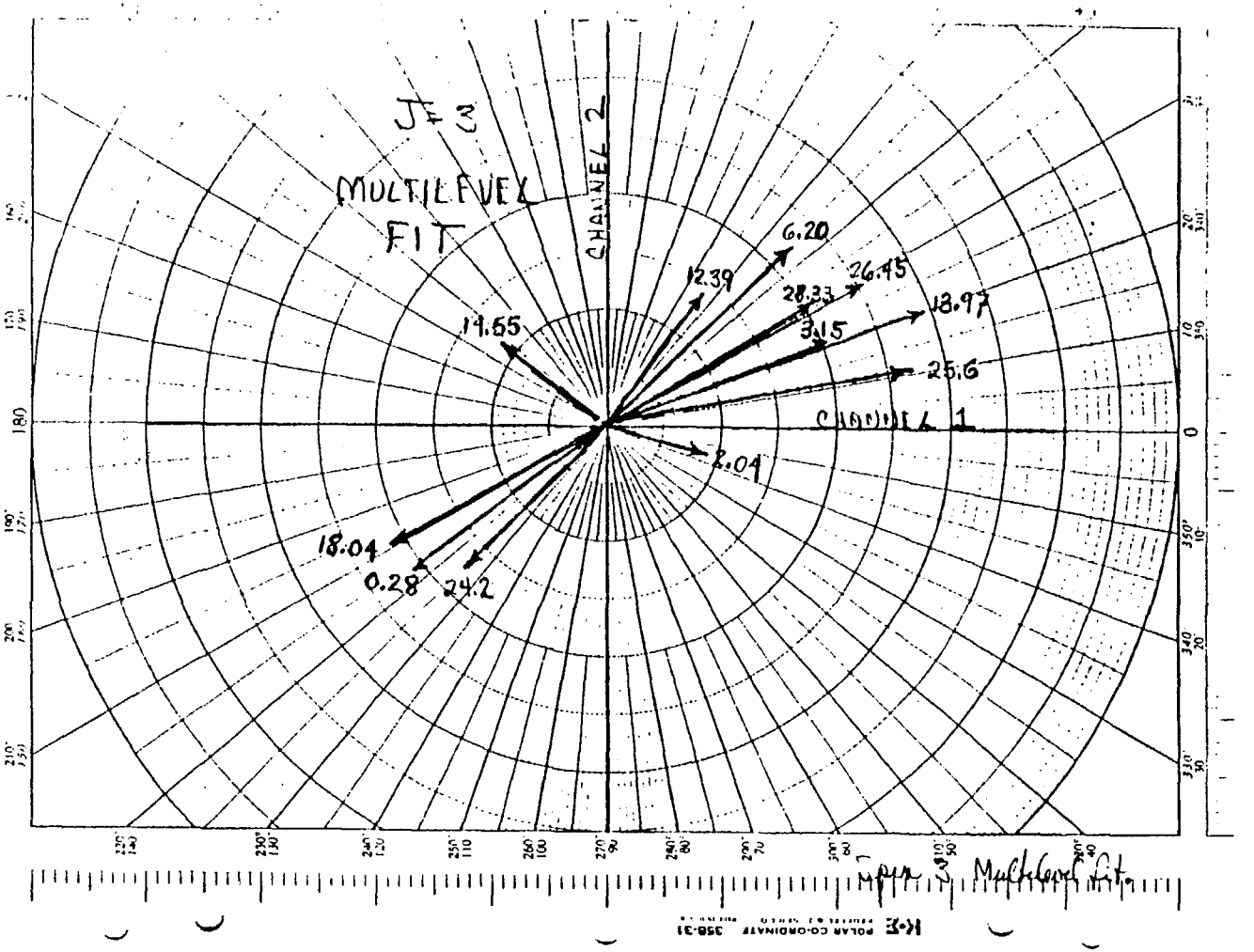

J#3
MULTILEVEL
FIT
CHANNEL 2
CHANNEL 1
6.20
12.39
26.45
28.33
18.97
3.15
25.6
14.55
2.04
18.04
0.28
24.2
180
0
Open 3 Multilevel Fit.
K+E POLAR CO-ORDINATE 358-31

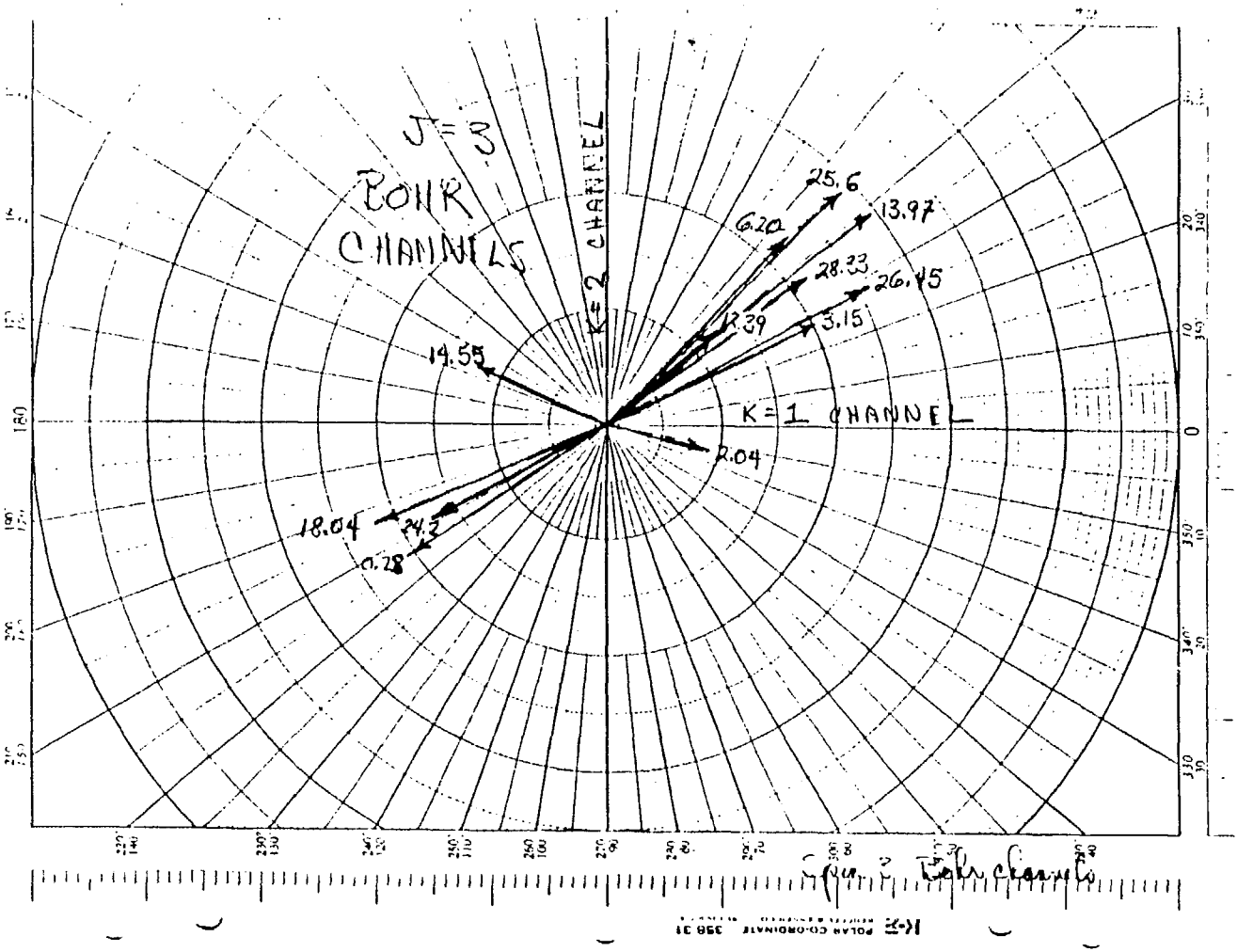
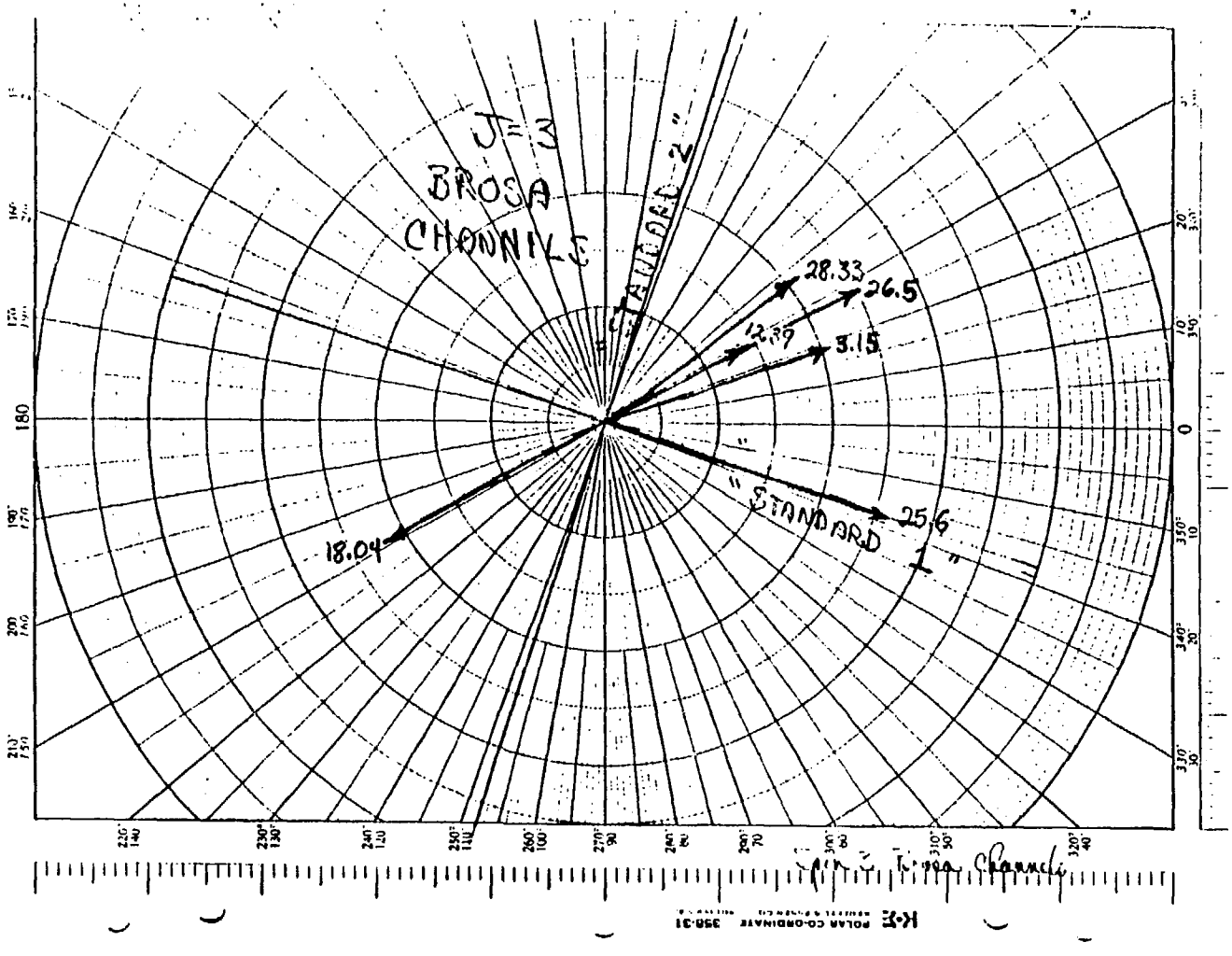
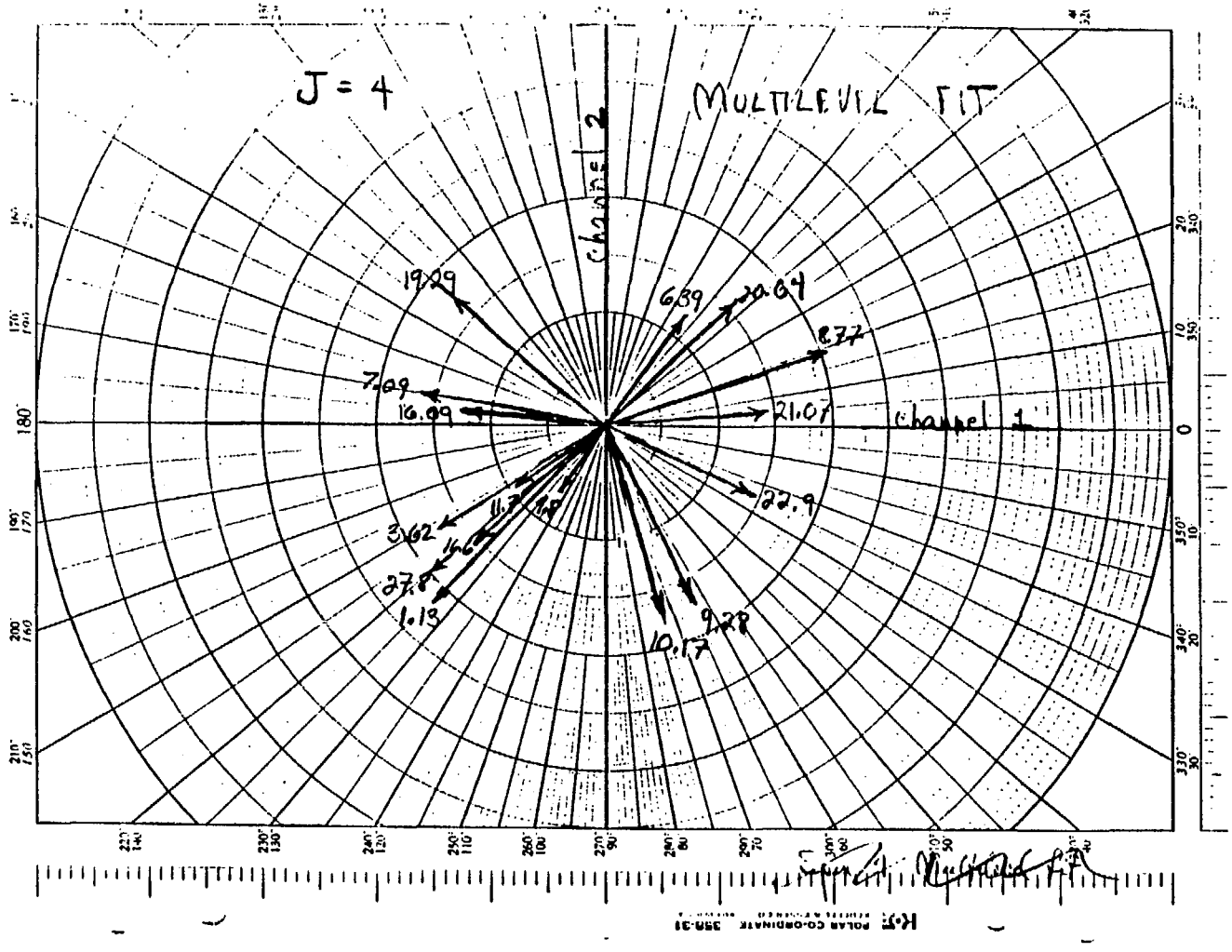


Fig. 3 Bohr channels


J=3
BROSA
CHANNILS
"STANDARD 2"
"STANDARD 1"
28.33
26.5
12.39
3.15
25.6
18.04
180
0
K+E POLAR CO-ORDINATE 359-31

[redacted] [redacted]



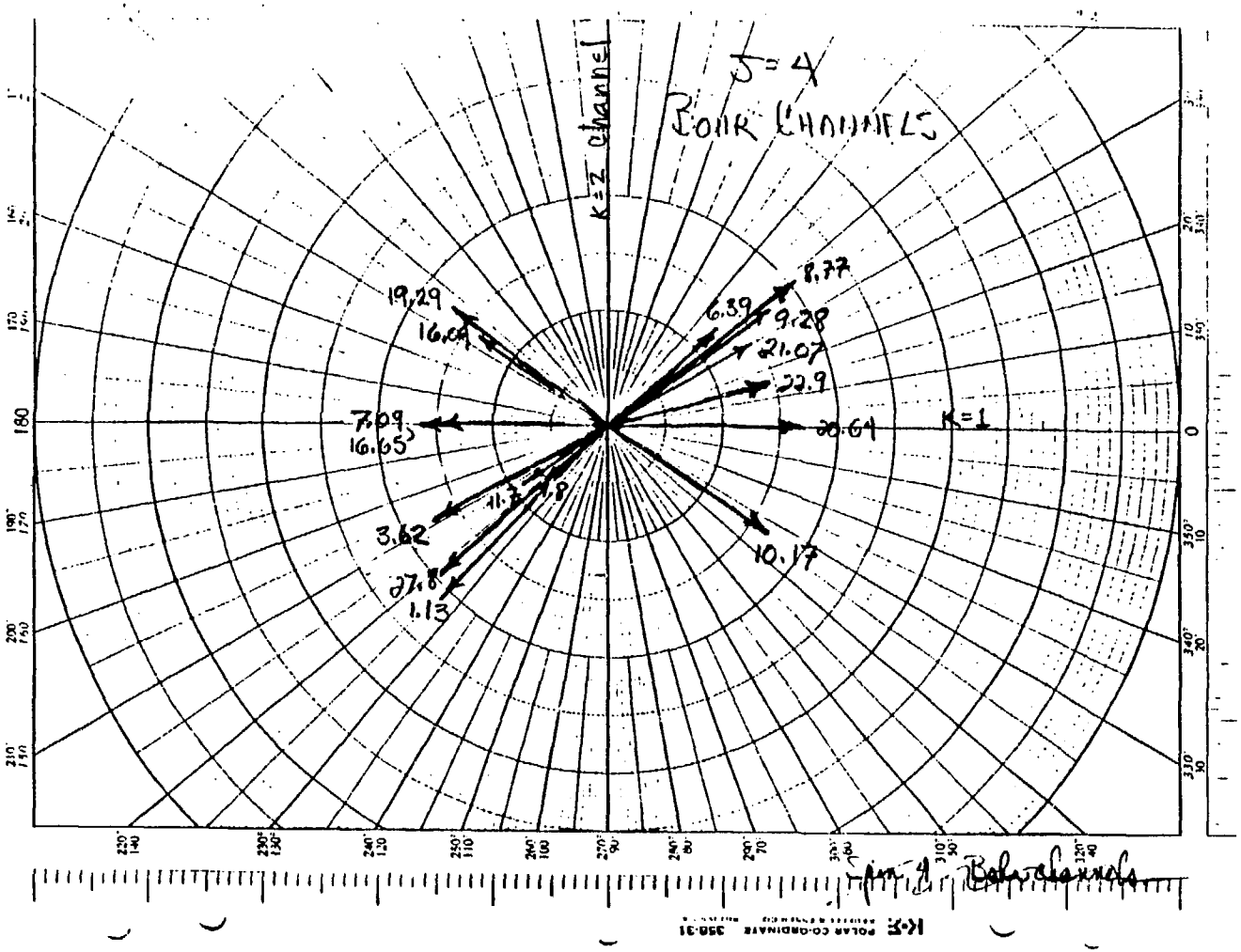
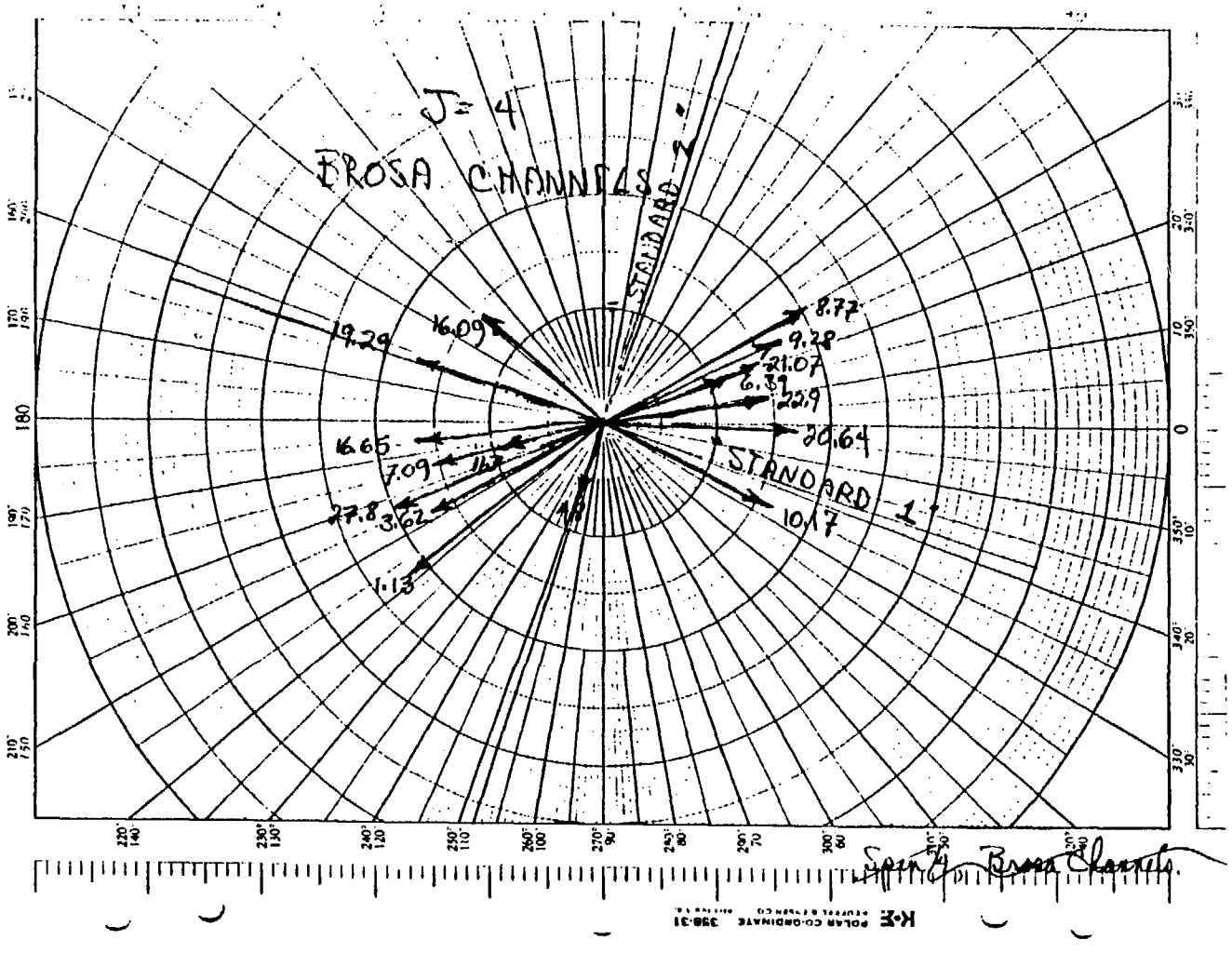


Fig. 4. Bohr channels


J= 4
BROSA CHANNELS
STANDARD 2
STANDARD 1
19.29
16.09
8.77
9.28
21.07
6.39
22.9
20.64
10.17
16.65
7.09
27.8
3.62
1.13
180
0
Spin 4 Brosa Channels.
K+E POLAR CO-ORDINATE 358-31