285
7-14-65

MASTER
IDO-17087
May 1965

A MACHINE PROGRAM FOR CALCULATING SLOW NEUTRON SCATTERING CROSS SECTIONS WITH EXACT AVERAGING OVER MOLECULAR ORIENTATION

L. J. Gannon and H. L. McMurry

PATENT CLEARANCE OBTAINED. RELEASE TO
THE PUBLIC IS APPROVED. PROCEDURES
ARE ON FILE IN THE RECEIVING SECTION.

**PHILLIPS
PETROLEUM
COMPANY**

ATOMIC ENERGY DIVISION

**NATIONAL REACTOR TESTING STATION
US ATOMIC ENERGY COMMISSION**

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.

PRINTED IN USA. PRICE \$1.00. AVAILABLE FROM THE CLEARINGHOUSE FOR FEDERAL
SCIENTIFIC AND TECHNICAL INFORMATION, NATIONAL BUREAU OF STANDARDS,
U. S. DEPARTMENT OF COMMERCE, SPRINGFIELD, VIRGINIA

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, express or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

IDO-17087
AEC Research and Development Report
Physics
TID-4500 (40th Ed.)
Issued: May, 1965

A MACHINE PROGRAM FOR CALCULATING SLOW NEUTRON SCATTERING
CROSS SECTIONS WITH EXACT AVERAGING OVER MOLECULAR ORIENTATION

L. J. Gannon
H. L. McMurry

PHILLIPS
PETROLEUM
COMPANY

Atomic Energy Division
Contract AT(10-1)-205
Idaho Operations Office
U. S. ATOMIC ENERGY COMMISSION

THIS PAGE
WAS INTENTIONALLY
LEFT BLANK

Abstract

This report describes a program for the IBM 7040 which calculates the scattering of slow neutrons by semi-rigid molecules in such a way that the correct average is obtained for scattering from all molecular orientations.

The program is a modification of an earlier one which treats rotations classically, but treats vibrational energy transfers quantum mechanically. The input data are explained and illustrative applications to CH_4 and C_2H_6 are presented.

A MACHINE PROGRAM FOR CALCULATING SLOW NEUTRON SCATTERING
CROSS SECTIONS WITH EXACT AVERAGING OVER MOLECULAR ORIENTATION

TABLE OF CONTENTS

	PAGE
I. INTRODUCTION	1
II. THEORETICAL	3
III. INPUT DATA	6
IV. ILLUSTRATIVE APPLICATIONS	9

TABLES

I. SIGMA CH ₄ EXACT AVERAGE	10
II. C ₂ H ₆ GAS EXACT AVG.	11
III. COMPARISON OF E.A. AND K.N. VALUES OF S(κ^2, β) FOR C ₂ H ₆	12

FIGURES

I. $\sigma(E_0, E, \mu)$ Vs. E For CH ₄	13
LIST OF REFERENCES	14

I. Introduction

When neutrons are scattered by an atom which is part of a molecule or crystal the cross section for scattering with specified energy change and scattering angle depends on how the molecule is oriented relative to the incident and scattered directions [1]. A previously reported program for the IBM 7040 [2], hereafter referred to as I or Macs 21* (PPCO 40.0005), calculates an average result for all possible molecular orientations by inserting average values of functions of the Eulerian angles (which define the orientation of the scattering system) wherever they appear in the scattering equations. This is called Krieger-Nelkin (K.N.) averaging [3].

Recently, calculations of the scattering by ethane and n butane using the K.N. averaging showed significant disagreement with experimental data under circumstances where agreement was expected. The most questionable approximation appeared to be the assumption of K.N. averaging. Therefore, a second program, now called Macs 22 (PPCO 40.0417), has been constructed which is identical with the K.N. program except for substitution of a routine which provides for calculating the scattering in many orientations, and then performing a numerical integration to get an exact average (E.A.) of the contributions from all orientations.

Except for the modifications described below, the new program is like the old one. It assumes that rotation-vibration coupling can be neglected, that rotations can be treated classically, and vibrations are harmonic. Criteria are incorporated which restrict computations of vibrational excitations and de-excitations to those which can significantly

*The origin of the name Macs 21 is a bit obscure. It is related to a long history of program alterations which began about 1959 or 1960.

affect the scattering. Other criteria insure a classical treatment of a vibrational mode whenever the energy transfer exceeds the vibrational level spacing by a sufficient factor. This greatly simplifies the computations.

II. Theoretical

The detailed discussion of the theory is given elsewhere [1]. The details of the program are described in I and the present discussion concerns only the modifications required for introducing the exact averaging procedure.

All calculations are based on computing the scattering law functions $S_v(\kappa^2, u, X, \beta)$ for each atom v in the scattering system. The arguments in S_v show dependence on the square of the neutron momentum transfer κ , and on its energy transfer $\beta = (E - E_0)/kT$, and on variables u and X which define the molecular orientation. Here E_0 and E are the initial and final neutron energies, respectively. The function u is defined by $u = \cos \theta = \underline{K} \cdot \underline{k}$ where $\underline{K}$ is a unit vector along the Z axis of a space fixed system which is chosen to coincide with $\underline{K}$, and $\underline{k}$ is along the z axis of a system which rotates with the molecule. The angle X measures the rotation of the x, y axes in the molecule about $\underline{k}$ relative to the lines of nodes. This convention is the same as is used by Wilson, Decius, and Cross [4]. The function δ_v depends on u and X through parameters $\delta_{v\tau}$ which are obtained from the relations which tell how atom v moves during the τ normal vibration. In treating scattering by gases motions of translation and rotation are treated as normal modes of zero frequency. The quantities $\delta_{v\tau}$ are defined by the relation

$$\delta_{v\tau} = 3\{-(\sqrt{1-u^2} \cos X)T_{v\tau x} + (\sqrt{1-u^2} \sin X)T_{v\tau y} + uT_{v\tau z}\}^2 \quad (1)$$

When K.N. averaging is used the square of Eq. (1) is formed and average values of $1 - u^2 = \cos^2\theta$, $\cos^2\chi$ and $\sin^2\chi$ are inserted. This yields

$$\bar{\delta}_{v\tau} = T^2_{v\tau x} + T^2_{v\tau y} + T^2_{v\tau z} \quad (2)$$

The K.N. program described in I includes the $\bar{\delta}_{v\tau}$ values as input. The present program replaces these input values by values calculated from Eq. (1) for specified values u_i and χ_i and then calculates $S_v(\kappa^2, u_i, \chi_i, \beta)$. If the range $0 < u_i < 1$ is divided into N_u increments, and $0 < \chi < 2\pi$ into N_χ increments, the average of $S_v(\kappa^2, \beta)$ for molecules in all possible orientations can be approximated by the sum

$$\bar{S}(\kappa^2, \beta) = \frac{1}{N_u N_\chi} \sum_{i=1}^{N_u} \sum_{j=1}^{N_\chi} S_v(\kappa^2, u_i, \chi_j, \beta) \quad (3)$$

Eq. (2) is an approximation to

$$S_v(\kappa^2, \beta) = \frac{1}{4\pi} \int_{u=-1}^1 \int_{\chi=0}^{2\pi} S_v[\kappa^2, u, \chi, \beta] du d\chi \quad (4)$$

In equations (3,4) the bar is omitted over $\bar{S}_v(\kappa^2, \beta)$ since this notation has been used to denote K.N. averaging in which calculations are made using equation (2) [1,2].

The range needed for u in Eq. (1) is 0 to 1 instead of -1 to 1 because the range -1 to 0 merely reproduces the terms from 0 to 1. To see this, the quantity in brackets in Eq. (1) is examined for some u_i in the range $0 < u_i < 1$, and for χ values of $0 < \chi_j < \pi/2$; $\pi - \chi_j$; $\pi + \chi_j$ and $2\pi - \chi_j$. The four values of Eq. (1) which result are the same, except for sign, with what is found when u_i is replaced by $-u_i$.

Hence, all values of $\delta_{v\tau}$ which are needed to evaluate Eq. (3) are obtained using $0 < u_i < 1$.

Recipes for obtaining the $T_{v\tau x}$, $T_{v\tau y}$, $T_{v\tau z}$ from normal coordinate analyses are described elsewhere [5].

III. Input Data

The input data are the same as in Macs 21 except for minor changes and some additions.

It is now possible to insert comments in columns 71-78 on all card types except 1, and 7 through 15. Other changes are

Card Type 1

- a. Storage limitations restrict calculation to options 1, 2, 3. Columns 1, 2 are reserved for these and future option numbers.
- b. Further input through Column 70 is same as in I.
- c. N_u (denoted by NOU) in Columns 71, 72. Fixed point - this tells the number of mesh intervals for u in the range $0 < u < 1$.
- d. N_x (denoted by NCHI) in columns 73, 74. Fixed point -- this is the number of intervals for x in the range $0 < x < 2\pi$.
- e. A 1 in Column 77 will call for a detailed print out. The card number 1 must be in Column 80.

Note: The detailed print out is always that of option 1, even though another option is specified. The total values always correspond to the option specified.

Card Type 2

This is like Macs 21, but it is necessary to have at least one mode for the translations and three modes for the rotations. The form of the T_{vtx} , T_{vty} , T_{vtz} for the translations is such that δ_{vt} for translations along x , y , and z are all just $1/M$ where M is

the mass of the molecule [5]. In calculating scattering by gases it suffices to use a single mode of zero energy and to assign $\bar{\delta}_{vT} = 3/M$ for every atom v for this mode. The rotations must be averaged over, and energies must be assigned for the rotations about each axis. For scattering by gases these modes all have zero energy.

Card Type 6

These are the $\bar{\delta}_{vT}$ values as in Macs 21. For those modes for which an E.A. calculation is made the program replaces the $\bar{\delta}_{vT}$ values by the ones calculated from Eq. (1). However, since it is not necessary to treat all modes this way it has been convenient to retain the Macs 21 input on the six cards and to have the program replace these as the need arises. In treating gases on Macs 21, it suffices to treat all six modes of rotation and translation as one mode of zero energy. The $\bar{\delta}_{vT}$ for this mode is the sum of the quantities for the six degrees of freedom. In the present program it is necessary to give separate zero frequencies for each rotational degree of freedom and to assign $\bar{\delta}_{vT}$ values for each.

New Cards

Card Type 16

- a. A number NTK (Ni) which tells the number of modes to be averaged over using Eq. (1) when atom v is treated. Not all modes need to be treated by Eq. (1). Seven NTK numbers per card, one for each atom v , with numbers in the same order as for the f_v on card 3. These are fixed point numbers. (I10 Format, with 16 in Columns 79-80.)

Card Type 17

- a. There is a set of type 17 cards for each atom and the sets are listed in the order on card 16. However, if an atom is not treated by E.A. its set is omitted.

Any set lists the $T_{v_{tx}}$, $T_{v_{ty}}$, $T_{v_{tz}}$ for each mode treated by E.A. The mode index corresponds to that on Card 2, and is punched in Columns 1-10 as an integer (I Format). Columns 11-20 give $T_{v_{tx}}$, Columns 21-30 give $T_{v_{ty}}$, and Columns 31-40 give $T_{v_{tz}}$. The T values are punched with decimal points (F Format). A 17 is inserted in 79, 80.

IV. Illustrative Applications

Table I shows the input needed to calculate $\sigma(E_0, E, \mu)$ vs E for CH_4 . Input parameters are based on Pope's normal coordinate analysis [6]. All modes are treated by the E.A. procedure. The E.A. and K.N. results are compared in Fig. 1.

Table II gives the input data for an E.A. calculation of $S(\kappa^2, \beta)$ of C_2H_6 . Only the rotations and the torsional vibration of .038eV are given an E.A. treatment. Table III compares the E.A. and K.N. results with unpublished data of K. A. Strong. The E.A. method improves the agreement with the experiments in most cases.

Table I

Sigma CH₄ Exact Average

MACS 22 PROGRAM TO COMPUTE TOTAL AND PARTIAL DIFFERENTIAL CROSS SECTIONS AND SCATTERING LAW

H L MCMURRY, L J GANNON, D H SPEAS

LATEST COMPILATION 12/11/64

OPTION 1 S KAPPA SQUARED
 2 SIGMA (EO,E,MU)
 3 SIGMA (EO,E)
 4 SAME AS OPT. 3 AND P(L),L=0,3
 INPUT DATA, CARD TYPE IN COLS 79-80, TITLE CARD IS ZERO

SIGMA CH ₄ EXACT AVERAGE				TABLE 1		MARCH 5, 1965								
OPTION	ENERGIES	ATOMS	KMAX	TEMP	G1	G3	C1	C2	C3	C4	NOU	NCHI	IPCH	CD
2	13	2	5	100.0000	0.0500	0.0100	0.	0.3000	0.3000	0.	15	15	-0	1
CHARACTERISTIC ENERGIES, CARD TYPE 2														
0.	0.	0.	0.	0.	0.16200	0.16200	0.16200				2			
0.18800	0.18800	0.36100	0.	0.37400	0.37400	0.37400	-0.				2			
FRACTIONS OF ATOM NU, CARD TYPE 3														
4.00000	1.00000	-0.	-0.	-0.	-0.	-0.	-0.				3			
SCATTERING AMPLITUDES FOR ATOM NU (A2+C2), CARD TYPE 4														
6.53000	0.41000	-0.	-0.	-0.	-0.	-0.	-0.				4			
MASS OF ATOM, CARD TYPE 5														
1.00000	12.00000	-0.	-0.	-0.	-0.	-0.	-0.				5			
AMPLITUDE FACTORS DELTA(NU,TAU), CARD TYPE 6														
0.18750	0.37500	0.37500	0.	0.20970	0.20970	0.20970					6			
0.25000	0.25000	0.25000	0.22760	0.22760	0.22760	-0.					6			
0.18750	0.	0.	0.	0.01342	0.01342	0.01342	0.01342				6			
0.	0.	0.	0.00742	0.00742	0.00742	-0.					6			
FN	F MIN	DEF	F	F MAX	ARRMAS	MMIT	CARD 9							
0.30600	0.02000	0.02000	0.24000	3.20000		3	9							
MU (COSINE OF SCATTERING ANGLE) TABLE, CARD 10														
-0.86603	0.	0.50000	-0.	-0.	-0.	-0.					10			
NO. OF MODES TO BE AVERAGED EXACTLY FOR ATOM NU, CARD TYPE 16														
11	0	-0	-0	-0	-0	-0	-0				16			
MODE INDX	TX	TY	TZ											
2	0.	-0.61237	0.								17			
3	0.61237	0.	0.								17			
5	0.05574	-0.09654	0.44420								17			
6	0.05574	0.09654	0.44420								17			
7	-0.11148	0.	0.44420								17			
8	0.	0.50000	0.								17			
9	-0.50000	0.	0.								17			
10	0.	0.	0.50000								17			
11	0.22147	-0.38360	0.17718								17			
12	0.22147	0.38360	0.17718								17			
13	-0.44294	0.	0.17718								17			

7040 TIME FOR THIS CASE 2.8610E-02 MINUTES

Table II C₂H₆ Gas Exact Average

MACS 22 PROGRAM TO COMPUTE TOTAL AND PARTIAL DIFFERENTIAL CROSS SECTIONS AND SCATTERING LAW

H L MCMURRY, L J GANNON, D H SPEAS

LATEST COMPILATION 12/11/64

OPTION 1 S KAPPA SQUARED
2 SIGMA (EO,E,MU)
3 SIGMA (EO,E)
4 SAME AS OPT. 3 AND P(L),L=0,3
INPUT DATA, CARD TYPE IN COLS 79-80, TITLE CARD IS ZERO

S C2HG GAS EXACT AVG.				TABLE 2				DEC. 14 64							
OPTION	ENERGIES	ATOMS	KMAX	TEMP	G1	G3	C1	C2	C3	C4	NOU	NCHI	IPCH	CD	
1	22	2	5	293.0000	0.1000	0.0100	0.	0.2000	0.2000	0.	20	20	-0	1	
CHARACTERISTIC ENERGIES, CARD TYPE 2															
0.	0.	0.	0.	0.	0.12600	0.18000	0.37800							2	
0.03760	0.17900	0.38000	0.10200	0.10200	0.18900	0.18900								2	
0.38900	0.38900	0.15400	0.15400	0.19200	0.19200	0.39400								2	
0.39400	-0.	-0.	-0.	-0.	-0.	-0.	-0.							2	
FRACTIONS OF ATOM NU, CARD TYPE 3															
6.00000	2.00000	-0.	-0.	-0.	-0.	-0.	-0.							3	
SCATTERING AMPLITUDES FOR ATOM NU (A2+C2), CARD TYPE 4															
6.53000	0.41000	-0.	-0.	-0.	-0.	-0.	-0.							4	
MASS OF ATOM, CARD TYPE 5															
1.00810	12.00390	-0.	-0.	-0.	-0.	-0.	-0.							5	
AMPLITUDE FACTORS DELTA(NU,TAU), CARD TYPE 6															
0.09981	0.09308	0.05100	0.16533	0.03514	0.13204	0.16351	H							6	
0.16533	0.13261	0.16480	0.30269	0.01451	0.02548	0.28290	H							6	
0.30000	0.	0.15124	0.06501	0.11524	0.21459	0.30188	H							6	
0.	-0.	-0.	-0.	-0.	-0.	-0.	H							6	
0.09981	0.02350	0.02350	0.	0.03280	0.00839	0.00046	C							6	
0.	0.00825	0.00014	0.00171	0.00171	0.00281	0.00281	C							6	
0.00387	0.00387	0.01443	0.01443	0.00010	0.00010	0.00362	C							6	
0.00362	-0.	-0.	-0.	-0.	-0.	-0.	C							6	
KAP MIN KAP MAX G2 ARB. MASS NBETA CARD TYPE 7															
20.00000	80.00000	4.00000	1.00000		3									7	
BETA VALUES FOR KAPPA SQUARED CALCULATION, CARD TYPE 8															
0.40000	0.20000	0.	-0.	-0.	-0.	-0.								8	
NO. OF MODES TO BE AVERAGED EXACTLY FOR ATOM NU, CARD TYPE 16															
4	-0	-0	-0	-0	-0	-0	-0							16	
MODE INDX TX TY TZ															
2	0.22583	0.	-0.20547					YROTH	17						
3	0.	-0.22583	0.					XROTH	17						
4	0.	0.40662	0.					ZROTH	17						
8	0.	0.40662	0.					VIB H	17						

7040 TIME FOR THIS CASE 3.1944E-02 MINUTES

Table III

Comparison of E.A. and K.N. values of $S(\kappa^2, \beta)$ for C_2H_6

$\kappa^2(A^{-2})$	β	$S(\kappa^2, \beta)$ (barns/evx ster. x mol.)		
		Obs'd ^a	E.A.	K.N.
130	0.0	94	90.8	75.2
130	0.2	100	89.8	74.6
130	0.4	89	86.7	73.1
130	0.7	89	79.1	68.9
130	1.0	69	69.0	63.2
130	1.5	56	51.6	51.8
58	1.0	175	173	178
58	1.5	107	102	118

a. Unpublished data from K. A. Strong.

Figure 1. $\sigma(E_0, E, \mu)$ Vs. E For CH_4

REFERENCES

1. H. L. McMurry, Nuc. Sci. Eng. **15**, 429 (1963).
2. H. L. McMurry and L. J. Gannon, A Machine Program for Calculating Slow Neutron Scattering Cross Sections with Application to Water, IDO 16984, July, 1964.
3. T. J. Krieger and M. S. Nelkin, Phys. Rev. **106**, 290 (1957).
4. E. B. Wilson, Jr., J. C. Decius, and P. C. Cross, Molecular Vibrations, McGraw-Hill, New York, 1955, p. 285.
5. H. L. McMurry. To be published in Spectrochem. Acta. See also, Ref. 2, p. 6. However the notation there is ambiguous. It would have been better to use T_{VT_x} , T_{VT_y} , T_{VT_z} , and T_{VR_x} , T_{VR_y} , and T_{VR_z} to denote the three element column vectors associated with translations along the molecular axes, and rotation about these axes, respectively.
6. N. K. Pope, Can. Jour. Phys. **30**, 597 (1952). These are included in work by H. L. McMurry, G. W. Griffing, W. A. Hestir, and L. J. Gannon in IDO 16692, August, 1961.
7. An IBM 7040 program suited to this purpose is available from Phillips. For description of input see PPCO 40.0206.

**PHILLIPS
PETROLEUM
COMPANY**

ATOMIC ENERGY DIVISION