UNIVERSIDADE DE SÃO PAULO

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BRASIL

PUBLICAÇÕES

IFUSP/P-708

A MATRICIAL METHOD TO OBTAIN SUB-BARRIER FUSION CROSS SECTION FROM OFF-LINE X-RAY MEASUREMENT

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Maio/1988

A matricial method to obtain sub-barrier fusion cross section from off-line X-ray measurement

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Abstract

A matricial method to obtain heavy ion fusion cross sections from off-line detection of X-rays emitted by the evaporation residues is presented. The method uses analytical functions for the decay expressions. It can treat all decay branchings and any number of decay chains or any number of nuclides.

1) Introduction

Heavy ion fusion cross sections can be measured through the off-line detection of the characteristic X-rays emitted by the evaporation residues in their decay[1]. These X-rays identify the elements present in the decay and the analysis of the time dependence of the decay using the known half-lives and the absolute X-ray yields furnishes the isotopic identification. Until now, this analysis has been carried out by fitting simultaneously all experimental data with only the activity of each residue at the end of the bombardment as unknowns[1,2,3,4]. However, it is possible to make the analysis in a more precise and elegant way through matrix algebra, as we show in this paper. Moreover, our method gives the possibility to simulate the physical process to choose irradiation bombarding exposure time and counting time intervals to obtain the lowest statistical errors.

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1 Supported in part by CNPq of Brasil.
2 Supported in part by FAPESP of Brasil.

The mathematical development of the method is presented in section 2. Section 3 deals with its application to an experimental example.

2) Mathematical development

In order to introduce the mathematical problem we begin with a short description of the experiment. It consists of bombarding a target with a beam of particles during a certain time. As a result of this bombardment compound nuclei are formed and after evaporation of particles the residues are trapped in a catcher foil. This foil is then removed from the vacuum and placed in front of a detector. The X-rays produced by the evaporation residues in their decay path to stability are recorded at different time intervals.

Now let us take a hypothetical example to establish the differential equations of the problem. A target with two isotopes ${}_aX_c$ and ${}_aX_{c-1}$ is bombarded by a beam of particles ${}_bX_d$ with a flux F(t), during a time t_b. M and M' are the number of isotopes ${}_aX_c$ and ${}_aX_{c-1}$ hit by the beam, respectively. The fusion process of these nuclei leads to the compound nuclei ${}_ZX_N$ and ${}_ZX_{N-1}$, where Z = a + b and N = c + d. These compound nuclei evaporate one or two neutrons producing the residues ${}_ZX_{N-1}$, ${}_ZX_{N-2}$ and ${}_ZX_{N-3}$, as shown in figure 1. Let $\sigma^1_{I(II)}$ and $\sigma^2_{I(II)}$ be the cross sections to produce the residues after the evaporation of 1 neutron or 2 neutrons from the compound nucleus ${}_ZX_N$ (${}_ZX_{N-1}$). Now, calling $N_i(t)$ the quantity of the i-nuclide (evaporation residues and its daugthers) present at time t in the process and λ_i its decay constant we can write that[5]

$$\begin{aligned}
\frac{d}{dt} N_1(t) &= -\lambda_1 N_1(t) + F(t)\, M\, \sigma^1_I \\
\frac{d}{dt} N_2(t) &= -\lambda_2 N_2(t) + F(t)\, (\, M\, \sigma^2_I + M'\, \sigma^1_{II}\,) \\
\frac{d}{dt} N_3(t) &= -\lambda_3 N_3(t) + F(t)\, M'\, \sigma^2_{II} \\
\frac{d}{dt} N_4(t) &= -\lambda_4 N_4(t) + \lambda_1 N_1(t) \\
\frac{d}{dt} N_5(t) &= -\lambda_5 N_5(t) + \lambda_2 N_2(t) \\
\frac{d}{dt} N_6(t) &= -\lambda_6 N_6(t) + \lambda_3 N_3(t)
\end{aligned} \qquad (1)$$

where $F(t) \neq 0$ for $0 \leq t \leq t_b$ and $F(t) = 0$ otherwise.

We define now the vector $\mathbf{N}(t)$ as

$$\mathbf{N}(t) = \begin{bmatrix} N_1(t) \\ N_2(t) \\ N_3(t) \\ N_4(t) \\ N_5(t) \\ N_6(t) \end{bmatrix} , \qquad (2)$$

$\mathbf{R}$ as

$$\mathbf{R} = \begin{bmatrix} M\,\sigma_I^1 \\ M\,\sigma_I^2 + M'\,\sigma_{II}^1 \\ M'\,\sigma_{II}^2 \\ 0 \\ 0 \\ 0 \end{bmatrix} \qquad (3)$$

and the triangular matrix Λ as

$$\Lambda = \begin{bmatrix} -\lambda_1 & & & & & \\ 0 & -\lambda_2 & & & & \\ 0 & 0 & -\lambda_3 & & & \\ \lambda_1 & 0 & 0 & -\lambda_4 & & \\ 0 & \lambda_2 & 0 & 0 & -\lambda_5 & \\ 0 & 0 & \lambda_3 & 0 & 0 & -\lambda_6 \end{bmatrix} . \qquad (4)$$

Then the system of equations (1) can be written as

$$\frac{d}{dt}\mathbf{N}(t) = \Lambda\,\mathbf{N}(t) + F(t)\,\mathbf{R} \qquad \text{for} \quad 0 \leq t \leq t_b \qquad (5)$$

$$\frac{d}{dt}\mathbf{N}(t) = \Lambda\,\mathbf{N}(t) \qquad \text{for} \quad t > t_b . \qquad (6)$$

More general processes can be described by the equations (5) and (6) with a suitable construction of the vectors $\mathbf{N}(t)$ and $\mathbf{R}$ and the

matrix Λ. Suppose, for example, a process in which the i-nuclide decays to j-, k- and m-nuclides with partial decay constant λ_i^j, λ_i^k and λ_i^m, respectively. To take this into account, the i-column of the matrix Λ has the following elements $\Lambda_{ii} = -\lambda_i$, $\Lambda_{ji} = \lambda_i^j$, $\Lambda_{ki} = \lambda_i^k$ and $\Lambda_{im} = \lambda_i^m$. As a consequence of

$$\lambda_i = \sum_n \lambda_i^n \qquad \text{for all i} \qquad (7)$$

the matrix Λ has the property

$$\sum_j \Lambda_{ij} = 0 \qquad (8)$$

for all columns, except those whose respective nuclide is the last one considered in the chain.

The formal solution of equation (5) through the integrand factor method with the initial condition $\mathbf{N}(0) = 0$ can be expressed as:

$$\mathbf{N}(t) = e^{\Lambda t} \int_0^t e^{-\Lambda t'}\,F(t')\,\mathbf{R}\,dt' \qquad (9)$$

The exponential of the matrix Λ can be written as

$$e^{\Lambda t} = C\,[e^{\lambda t}]\,C^{-1} \qquad (10)$$

where C is the matrix that diagonalizes Λ and $[e^{\lambda t}]$ is the diagonal matrix

$$[e^{\lambda t}] = \begin{bmatrix} e^{\lambda_1 t} & & & & & \\ & e^{\lambda_2 t} & & & & \\ & & e^{\lambda_3 t} & & & \\ & & & e^{\lambda_4 t} & & \\ & & & & e^{\lambda_5 t} & \\ & & & & & e^{\lambda_6 t} \end{bmatrix} , \qquad (11)$$

so, inserting equation (10) in equation (9) and remembering that C and R are independent of t', we have

$$\mathbf{N}(t) = C \int_0^t [e^{\lambda(t-t')}]\,F(t')\,dt'\,C^{-1}\,\mathbf{R} \qquad (12)$$

where $[e^{\lambda(t-t')}]$ is expressed as equation (11).

The equation (12) can be written in a compact form as

$$N(t) = C\ \Phi(t)\ C^{-1}\ R \qquad (13)$$

with

$$\Phi(t) = \int_0^t [e^{\lambda(t-t')}]\ F(t')\ dt' \ . \qquad (14)$$

At this point we have the solution of equation (5) once the function F(t) is known.

Now we solve the equation (6). Its initial condition is the solution of equation (5) at time t_b, so

$$N(t_b) = C\ \Phi(t_b)\ C^{-1}\ R \qquad (15)$$

To make the notation easier we take t = 0 for equation (6) at the end of the bombardment. Then the initial condition, equation (15), can be rewritten as

$$N(0) = C\ \Phi(t_b)\ C^{-1}\ R \qquad (16)$$

The formal solution of the equation (6) is then

$$N(t) = e^{\Lambda t}\ N(0) \ . \qquad (17)$$

Substituting the equation (16) in (17), leads to

$$N(t) = e^{\Lambda t}\ C\ \Phi(t_b)\ C^{-1}\ R \ , \qquad (18)$$

or using equation (10)

$$N(t) = C\ [e^{\lambda t}]\ \Phi(t_b)\ C^{-1}\ R \ . \qquad (19)$$

The equation (19) gives the quantity of all nuclides present in the process at time t. Now we can obtain the X-ray recorded by a detector with efficiency ϵ at the energy of this X-ray in the time interval between t_p and $t_p + \Delta t$.

The X-ray activity of the element of atomic number Z at time t

can be written as:

$$A^Z(t) = \sum_i I_i^Z\ \lambda_i^Z\ N_i(t) \qquad (20)$$

with the sum restricted only to the i-nuclides that produce X-ray of element Z in its decay, λ_i^Z is its partial decay constant and I_i^Z is the corresponding absolute X-ray yield.

Using equation (19) in equation (20) we have

$$A^Z(t) = \sum_i I_i^Z\ \lambda_i^Z\ \{C\ [e^{\lambda t}]\ \Phi(t_b)\ C^{-1}\ R\ \}_i \qquad (21)$$

where $\{\ \}_i$ stands for the i-vector component.

Calling f^{Zp} the number of X-ray counts of element Z expected in the time interval between t_p and $t_p + \Delta t$ by a detector of efficiency ϵ to this X-ray, we have

$$f^{Zp} = \epsilon \int_{t_p}^{t_p+\Delta t} A^Z(t)\ dt \ . \qquad (22)$$

Inserting equation (21) into equation (22) leads to

$$f^{Zp} = \epsilon \sum_i I_i^Z\ \lambda_i^Z\ \{C \int_{t_p}^{t_p+\Delta t} [e^{\lambda t}]\ dt'\ \Phi(t_b)\ C^{-1}\ R\ \}_i \ . \qquad (23)$$

The vector R includes known and unknown cross sections, so we split it as

$$R = \sum_{n>1} M_n\ \vec{\sigma}^{kn} + M_1\ \vec{\sigma}^{u} \qquad (24)$$

where $\vec{\sigma}^{kn}$ for n>1 stands for the known cross sections and $\vec{\sigma}^{u}$ for unknown one.

Substituting equation (24) into equation (23), one has

$$f^{Zp} = \epsilon \sum_i I_i^Z\ \lambda_i^Z\ \{\sum_{n>1} Q^{pn}\ \sigma^{kn} + Q^{p1}\ \sigma^{u}\ \}_i \ , \qquad (25)$$

where

$$Q^{pn} = C \int_{t_p}^{t_p+\Delta t} [e^{\lambda t}]\ dt'\ M_n\ \Phi(t_b)\ C^{-1} \ . \qquad (26)$$

The vector $\vec{\sigma}^{u}$ can be optimally obtained from the measured counts, y^{Zp}, through a least squares procedure because f^{Zp} is a linear

function of $\vec{\sigma}^u$ and the probability density function of the experimental values are approximately normal. The estimates of $\vec{\sigma}^u$ are unique and the calculated variances, due to statistical errors in y^{Zp}, are almost exact. Then, let us construct the function

$$S = \sum_{Zp} (f^{Zp} - y^{Zp})^2 w^{Zp} \qquad (27)$$

where $(w^{Zp})^{-1/2}$ is the error in y^{Zp}. So, $\vec{\sigma}^u$ is then obtained from the minimum condition in S

$$\frac{\partial}{\partial \sigma_j^u} S = 0 \qquad (28)$$

Substituting equation (25) in equation (27) and writing

$$Y^{Zp} = y^{Zp} - \epsilon \sum_i I_i^Z \lambda_i^Z \sum_m \sum_{n>1} Q_{im}^{pn} \sigma_m^{kn} \qquad (29)$$

and

$$F_m^{Zp} = \epsilon \sum_i I_i^Z \lambda_i^Z Q_{im}^{p1} \qquad (30)$$

and carrying out the partial derivative of S with respect to σ_j^u, we have

$$\frac{\partial}{\partial \sigma_j^u} S = 2 \sum_{Zp} (\sum_m F_m^{Zp} \sigma_m^u - Y^{Zp}) F_j^{Zp} w^{Zp} . \qquad (31)$$

The condition of minimum, equation (28), becomes

$$\sum_{Zp} Y^{Zp} F_j^{Zp} w^{Zp} = \sum_m \sum_{Zp} F_m^{Zp} F_j^{Zp} \sigma_m^u w^{Zp} . \qquad (32)$$

or in a compact form

$$D_j = \sum_m [V^{-1}]_{jm} \sigma_m^u \qquad (33)$$

or in vector form

$$D = V^{-1} \vec{\sigma}^u \qquad (34)$$

where

$$D_j = \sum_{Zp} Y^{Zp} F_j^{Zp} w^{Zp} \qquad (35)$$

and

$$[V^{-1}]_{jm} = [V^{-1}]_{mj} = \sum_m \sum_{Zp} F_m^{Zp} F_j^{Zp} w^{Zp} . \qquad (36)$$

V is the covariance matrix due only to the counting statistics.

Finally we have the unknown cross section as

$$\vec{\sigma}^u = V D \qquad (37)$$

This procedure can be extended with little modification to obtain more than one unknown fusion cross section since the evaporation process produces different decay chains to each target isotope.

The total covarince matrix V^0 is obtained as

$$V^0 = V + V' \qquad (38)$$

where V' is the covariance matrix due to the deviation associated with the parameters I_i^Z and λ_i^Z. The element V'_{ij}, covariance between the cross sections σ_i^u and σ_j^u originated by these parameters, can be estimated in a linear approximation by [6]

$$V'_{ij} = \sum_k \frac{\partial \sigma_i^u}{\partial p_k} s_{p_k}^2 \frac{\partial \sigma_j^u}{\partial p_k} \qquad (39)$$

where p_k stand for the X-ray yields I_i^Z and for all independent partial decay constants λ_i^Z, see equation (7), and s_{p_k} stand for the corresponding standard deviation of p_k. The derivatives in equation (39) are evaluated numerically. The total standard deviation s_i of σ_i^u is giving as

$$s_i = (V_{ii}^0)^{1/2} \qquad (40)$$

and the standard deviation of the total cross section as

$$s = \left(\sum_{ij} V^0_{ij} \right)^{1/2} \qquad (41)$$

The standard deviation s_i and s can be also estimated by Monte Carlo method. To this, it is only necessary to calculated $\bar{\sigma}^u$ with several sets of the parameters I_i^z, λ_i^z and y^{zp} randomly drawed around their experimental values assuming gaussian distributions with the variances equal to the experimental measured ones. The standard deviation s_i are estimated from the obtained cross section histograms.

3) Application to an experimental example

Experimentally the beam flux F(t) and the number of isotopes hit by the beam are not measured. Instead of these quantities the Rutherford scattered beam particles and the beam current as a function of time are measured to carry out the calculations.

The beam current, I(t), is recorded as the beam charge collected in a Faraday cup in a multiscaler form (see figure 2), so it is represented by the step function

$$I(t) = \frac{\tau_i}{\Delta t} \qquad \text{for} \quad 1 \le i \le m \qquad (42)$$

where τ_i is the charge collected between times t_n and $t_n+\Delta t$, $\Delta t = t_b/m$ and m is the total number of steps. The Rutherford scattered beam particles, N_{Ruth}, are detected through two detectors placed in the horizontal plane symmetrically to the beam axis at 45 degree.

Suppose now a chemically pure target that contains the isotopes T_1, T_2, T_3, . . . , T_n with mass percentage P_1, P_2, P_3, . . . , P_n, respectively. It is easy to deduce the equality to each isotope

$$M_n\, F(t) = \frac{N_{Ruth}\, P_n}{\sigma_{Ruth} \sum_{i=1}^{m} \tau_i}\, I(t) \qquad (43)$$

where σ_{Ruth} is the differential Rutherford cross section integrated over the solid angle embeded by the two detectors.

Now to complete the analysis to obtain $\bar{\sigma}^u$ it is necessary to evaluate $M_n\, \Phi(t_b)$ to insert into equation (26). So, taken $\Phi(t_b)$ from equation (14) and using the equality (43), we have

$$M_n\, [\Phi(t_b)]_{nn} = \frac{N_{Ruth}\, P_n}{\sigma_{Ruth} \sum_{i=1}^{m} \tau_i} \int_0^{t_b} [e^{\lambda(t_b - t')}]_{nn}\, I(t')\, dt' \qquad (44)$$

Substituting equation (42) into equation (44) we have

$$M_n\, [\Phi(t_b)]_{nn} = \frac{N_{Ruth}\, P_n}{\sigma_{Ruth} \sum_{i=1}^{m} \tau_i} \sum_{j=1}^{m} \frac{\tau_j}{\Delta t} \int_{(j-1)\Delta t}^{j\Delta t} [e^{\lambda(t_b - t')}]_{nn} dt' \qquad (45)$$

or

$$M_n\, [\Phi(t_b)]_{nn} = \frac{N_{Ruth}\, P_n}{\sigma_{Ruth} \sum_{i=1}^{m} \tau_i} \sum_{j=1}^{m} \frac{\tau_j}{\Delta t}\, \frac{e^{\lambda_n (t_b - j\Delta t)}}{\lambda_n} \left(e^{\lambda_n \Delta t} - 1 \right) \qquad (46)$$

Now we apply the method to the fusion experiment $^{16}O + ^{149}Sm$ carried out at the Buenos Aires Tandem Accelerator, TANDAR, with the participation of one of us (V.R.V.). The complete description of the experiment will be published elsewhere.

The fusion process of this system leads to the compound nucleus ^{165}Yb, that after evaporation of 2 to 6 neutrons decays producing in its decay chain the elements Tm, Er, Ho and Dy. The decays are β decays or EC, except to some Ho isotopes that have isomeric transition also. Table 1 presents the nuclides produced in the process with its half-live and corresponding X-rays yield. The $K_{\alpha_1} + K_{\alpha_2}$ X-rays counts and counting times from the experiment for the energy E_{cm} = 59.64 Mev are presented in table 2. The peaks of the spectrum are fitted with a gaussian function plus a first degree polynomial using the computer code IDEFIX[10]. The beam time profile recorded as the collected charge by the Faraday cup in bins of 1 minute is show in figure 2. The calculated Rutherford cross section of the monitor detector is 0.0724 mb and the corresponding recorded counts is 5.16×10^4.

In the present work the Dy isotopes are not included due to the large error in the counts or the lack of them. The total fusion cross

section and the partial evaporation residue cross section after evaporation of x neutrons are shown graphically in figure 3.

The effect of uncertainties in the half-lives and in the X-ray yields evaluated by the Monte Carlo method show a standard deviation around 11% on the final cross sections and evaluated by the linear approximation around 11.5%. In both cases the uncertainties of 4% of the X-ray detector efficiency and 2% of the recorded Rutherford scattered beam particles are included.

This method was also applied with success to obtain the cross section for the reaction $^{141}Pd(\gamma,xn)$[11].

Conclusions

We show in this paper a matricial method to obtain sub-barrier fusion cross sections from off-line X-rays measurement. Among the features of the method are: 1) all the data can be used simultaneously, 2) the decay expressions are analytic, 3) all decay branchings (for example: β decay, EC, IT and α decay) can be treated, 4) it can be used with any particle evaporation (for example: neutrons, protons, α), 5) the deviation in the half-lives and in the X-rays yields are taken in account to obtain the deviations in the final cross sections, and 6) any number of decay chains or any number of nuclides can be treated. The method can be applied also to obtain the cross section of (γ,xn) reactions.

Acknowledgements

The authors thank Prof. S.J. Skorka for the critical reading of the manuscript.

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TABLE 1

Half-live and X-ray yields of the fragments and its daugthers produced by the reaction ^{16}O on ^{149}Sm. Half-lives are giving in minutes and those greater than 2 days are not printed. Intensities, printed in *italic*, are expressed as the number of X-rays per 100 decays. The nuclides decays through β-decay and or EC. The data are from reference [7], exceptions are indicated.

Element \ A	164	163	162	161	160	159
Yb	75.8±.7[a] *83.±13.*	11.05±.25 *57.7±1.3*	18.87±.19 *66.8±1.6*	4.2±.2 *76.8±8.4*	4.8±.2 *75.*[b]	1.5±.2 *75.*[b]
Tm	2.0±.1 *44.2±3.5*	108.6±3.6 *115.7±16.8*	21.7±.2[c] *72.8±1.2*	38.±4. *148.9±4.7*	9.2±.4 *70.*[b]	9.0±.4 *114.±8.*
Er		75.00±.42 *62.6±9.5*		194.40±2.40 *71.4±11.6*	1715.±6. *58.5±1.3*	36.±1. *63.7±3.6*
Ho				149.±3. *69.6±18.*	[d]	33.±1. *117.±11.*

a) data taken from reference [8]

b) data taken from reference [2], standard deviation of 20% was assumed

c) data taken from reference [9]

d) no data available

TABLE 2

$K_{\alpha_1} + K_{\alpha_2}$ X-rays counts of the reaction $^{16}O + ^{149}Sm$ at $E_{CM} = 59.64$ MeV. T_i is the starting time measured after bombardment end and T_c is the counting time. Results with bad fit are no printed.

T_i min	T_c min	Dy	Ho	Er	Tm
5.58	5.0	34±17	79±27	1380±49	1035±44
11.01	5.0		84±21	1366±49	953±44
16.35	5.0		46±14	1271±43	777±36
21.68	5.0		63±15	1176±41	629±33
27.01	5.0		40±13	1094±40	516±30
32.35	5.0		25±12	1067±38	389±27
37.93	20.0	99±30	218±33	3184±68	988±45
58.25	20.0	28±17	205±29	2012±55	460±33
78.59	20.0	103±22	146±23	1369±43	194±23
98.93	20.0	91±18	213±22	862±37	113±21
119.26	20.0	59±16	183±20	542±28	63±14
139.59	20.0			405±24	

FIGURE CAPTIONS

FIGURE 1

Diagram of a hypothetical fusion followed by neutron evaporation and decay. See the text for details.

FIGURE 2

Beam charge collected in a Faraday cup for the energy E_{CM} = 59.64 MeV.

FIGURE 3

Fusion cross sections for the fusion of ^{16}O + ^{149}Sm. Squares are the total fusion cross section, triangles are the partial evaporation residue cross section with evaporation of 2 neutrons, circles of 3 neutrons, lozenges of 4 neutrons, crosses of 5 neutrons and × of 6 neutrons. The error bars are only due to the counting statistics.

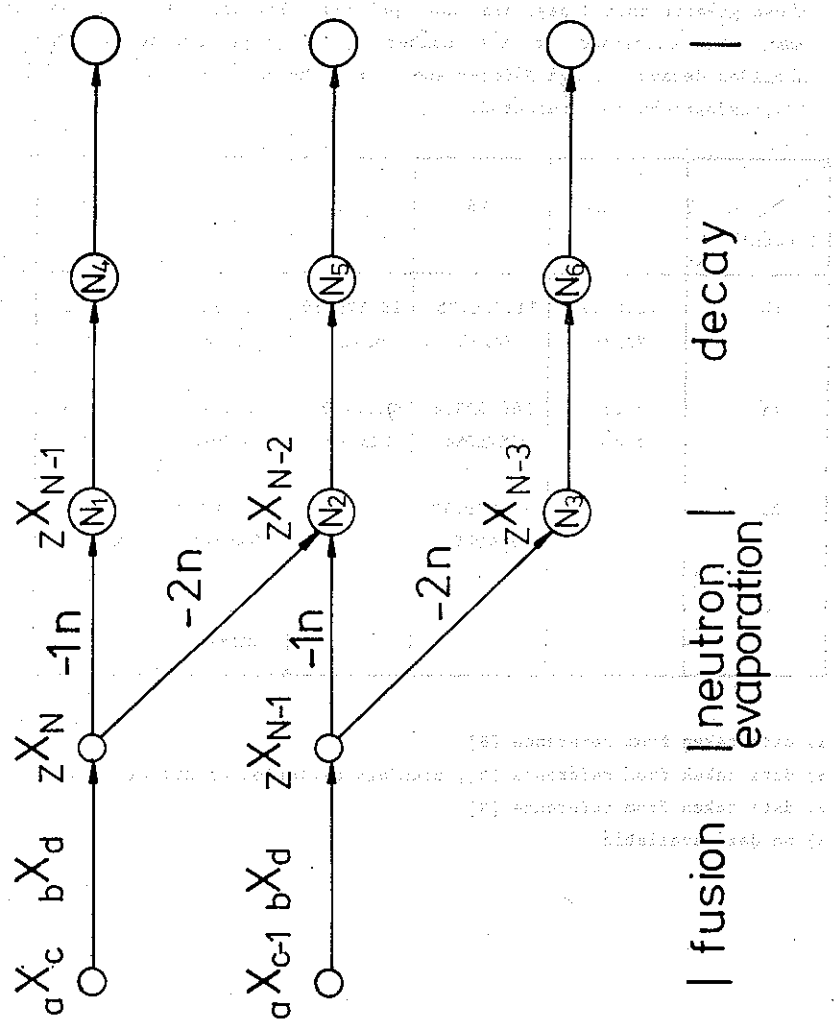

FIG. 1

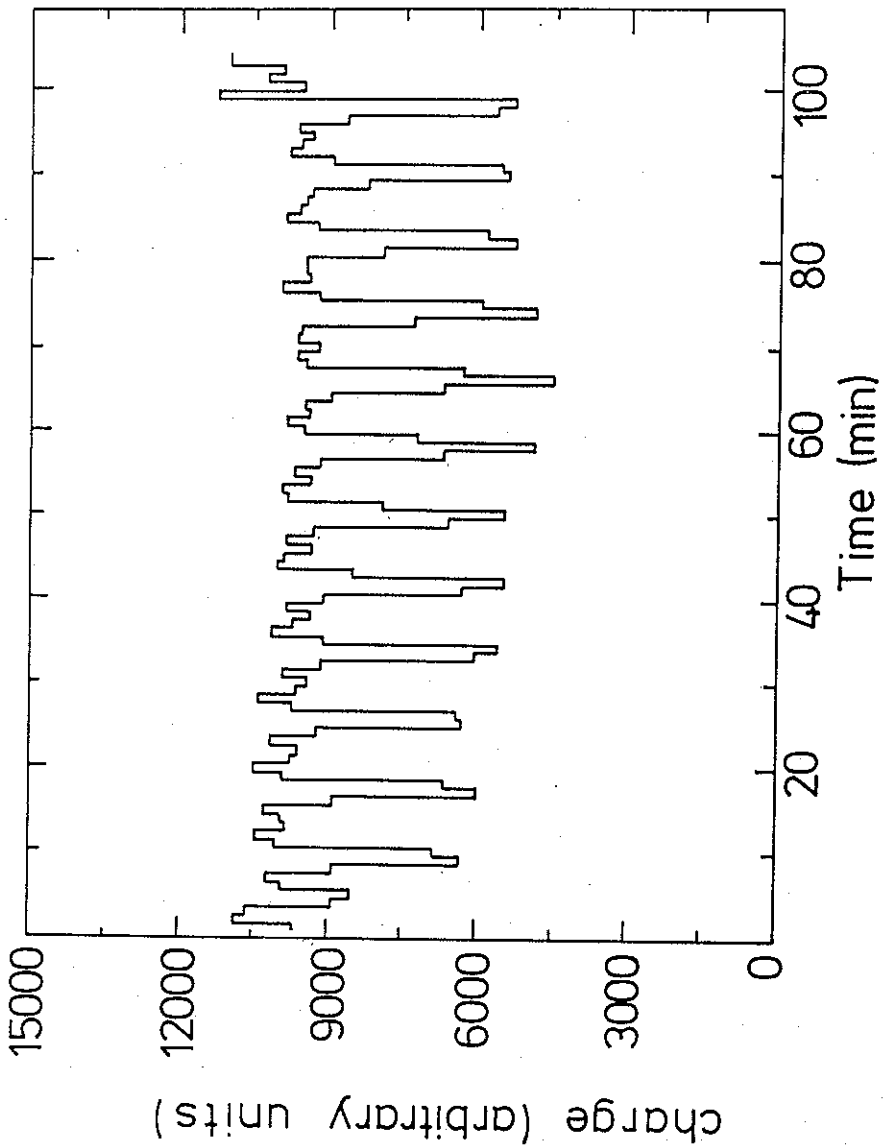
charge (arbitrary units)
15000
12000
9000
6000
3000
0
20
40
60
80
100
Time (min)

FIG. 2

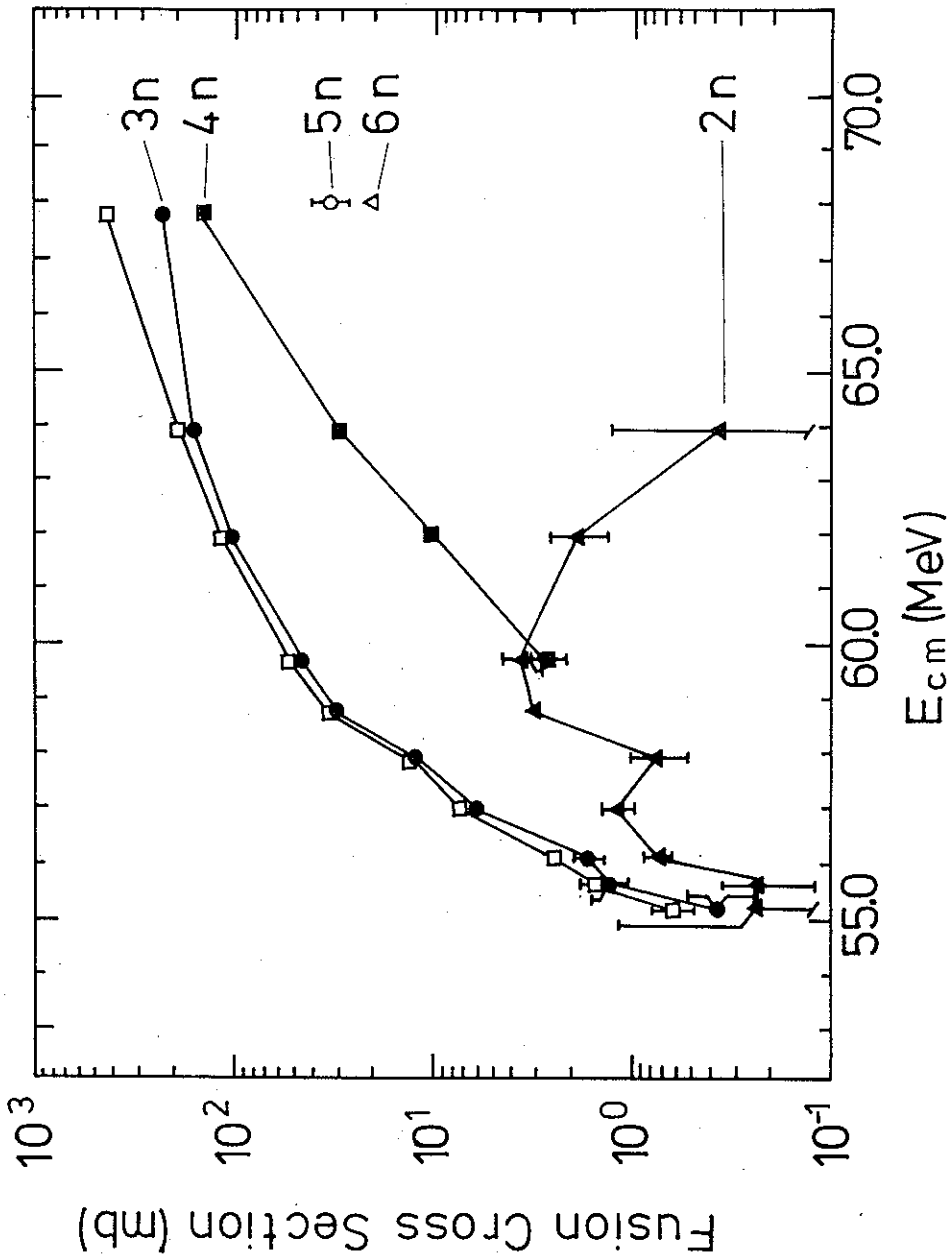
Fusion Cross Section (mb)
10^3
10^2
10^1
10^0
10^{-1}
55.0
60.0
65.0
70.0
E_{cm} (MeV)
3n
4n
5n
6n
2n

FIG. 3