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Measurement and analysis of excitation functions for alpha-induced reactions in copper

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Abstract. Excitation functions for the production of ^{68}Ga, ^{67}Ga, ^{66}Ga, ^{65}Ga + ^{65}Zn and ^{61}Cu from α-induced reactions in natural copper have been measured in the energy range $\approx$ 10–40 MeV using the stacked foil technique. A stack of nine copper foils was irradiated by a 40 MeV α-beam. The γ-rays emitted from the irradiated samples were recorded. Excitation functions have also been calculated theoretically using a statistical model with and without the inclusion of pre-equilibrium emission of particles. Pre-equilibrium component simulated by exciton model shows that the inclusion of pre-equilibrium emission gives better agreement between experimental and theoretical excitation functions. Pre-equilibrium fraction depends on the incident energy and the target mass number.

Keywords. Nuclear reactions; natural target; analysis of composite activity; statistical model; pre-equilibrium emission.

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1. Introduction

As part of our programme (Bhardwaj and Prasad 1986) to measure the excitation functions for α-induced reactions using stacked foil technique, excitation functions for the reactions (α, n), $(\alpha, 2n) + (\alpha, np)$, $(\alpha, 2n\alpha)$ in ^{63}Cu and (α, n), $(\alpha, 2n)$, $(\alpha, 3n)$ in ^{65}Cu have been measured in the energy range 10–40 MeV. Some of these measurements are available in literature but there are large discrepancies in the cross-section values for the same reaction reported by different groups (Porges 1956; Porile and Morrison 1959; Houck and Miller 1961; Bryant *et al* 1963; Stelson and McGowan 1964; Hille *et al* 1972; Graf and Muenzel 1974; Lin and Alexander 1977). Moreover, in most of these measurements errors associated with the cross-section values and energy are not mentioned. Porges (1956) who reported the excitation function for ^{65}Cu $(\alpha, 3n)$ ^{66}Ga reaction using natural copper foils did not discuss the procedure for separating the contribution from the reaction ^{63}Cu (α, n) which produces the same residual nucleus ^{66}Ga. In the present work, contributions of ^{63}Cu (α, n) and ^{65}Cu $(\alpha, 3n)$ reactions have been separated using the theoretical excitation function for ^{63}Cu (α, n) reaction. To the best of our knowledge excitation function for the ^{63}Cu $(\alpha, 2n\alpha)$ reaction is being reported for the first time.

These excitation functions in the past have been analysed mostly on the basis of the statistical equilibrium model. Both intuition and the results of some recent measurements indicate the presence of pre-equilibrium emission of particles at these moderate excitation energies (Blann 1975; Gadioli and Gadioli 1981; Hodgson 1982). In the

present analysis, excitation functions have been theoretically calculated using both the equilibrium and a mixture of equilibrium and pre-equilibrium mechanisms. Gamma competition and angular momentum effects have been explicitly considered. Theoretical calculations with an admixture of equilibrium and pre-equilibrium decay of compound system shows better agreement with the experimental excitation functions.

2. Experimental

Measurements have been done at the Variable Energy Cyclotron Centre, (VECC), Calcutta (India). Stacked foil technique has been used for these measurements. An α-beam of energy $40 \pm 0{\cdot}5$ MeV has been incident on a stack of nine copper foils (natural abundance, $^{63}Cu = 69{\cdot}2\%$ and $^{65}Cu = 30{\cdot}8\%$) fixed individually on aluminium sheets for immediate heat conduction. The exact beam energy was determined by the auxiliary experiment on α-scattering (Bhardwaj and Prasad 1986). Spectroscopically pure copper foils (purity better than 99·99%, thickness 23·27 mg/cm^2) have been used. The foil thickness corresponds to an energy degradation of about 2·5 MeV to 5·5 MeV from the first to the last foil (Northcliffe and Schilling 1970).

Various activities induced in the foils have been followed by a Ge(Li) detector coupled with a multichannel analyser. The detecting unit was calibrated using standard sources ^{57}Co, ^{60}Co, ^{137}Cs, ^{22}Na, ^{133}Ba, ^{54}Mn and ^{152}Eu for the efficiency and γ-ray energy. The dead time for counting has been kept $< 10\%$ by adjusting the sample-detector separation in these measurements and proper care of the dead time has been taken in the calculations. In general, several γ-rays arising from the same residual nucleus have been identified. A typical γ-ray spectrum from a natural copper target irradiated by $26{\cdot}3 \pm 1{\cdot}7$ MeV α-particles is shown in figure 1. Spectroscopic data of identified γ-rays are given in table 1. As a check in some cases, the relative intensities of identified γ-rays have also been measured. It can be seen that the presently measured relative intensities are in good agreement with the respective literature values (Lederer and Shirley 1978).

Reaction cross-section $\sigma_r(E)$ has been calculated using the following expression,

$$\sigma_r(E) = \frac{A\lambda \exp(\lambda t_2)}{N_0 \phi \cdot (G\varepsilon)\theta K[1-\exp(-\lambda t_1)][1-\exp(-\lambda t_3)]},$$

where $K = [1-\exp(-\mu d)]/\mu d$ is the correction for self-absorption of γ-ray in the sample of thickness d(g/cm^2) and of the absorption coefficient μ(cm^2/g); A is the area under photopeak, λ the decay constant of the product nucleus, t_1 the irradiation time, t_2 the time between the end of irradiation and beginning of counting, t_3 the counting period, N_0 the number of nuclei in the sample, ϕ the incident flux, $(G\varepsilon)$ the geometry-dependent efficiency of the detecting unit and θ the branching ratio for the specific γ-ray. The factor $(A\lambda \exp(\lambda t_2)/[1-\exp(\lambda t_3)]$, the count rate of the induced activity just at the stop of irradiation, is denoted by $C_{t=0}$. The factor $[1-\exp(-\lambda t_1)]$, called saturation correction, accounts for the decay of the activity during irradiation.

Activation cross-section for a given reaction has been determined from the intensities of the various identified γ-rays arising from the same residual nucleus. The reported value is the weighted average (Mughabghab *et al* 1981) of the various cross-section values so obtained. The statistical error given in the results is the larger one of the

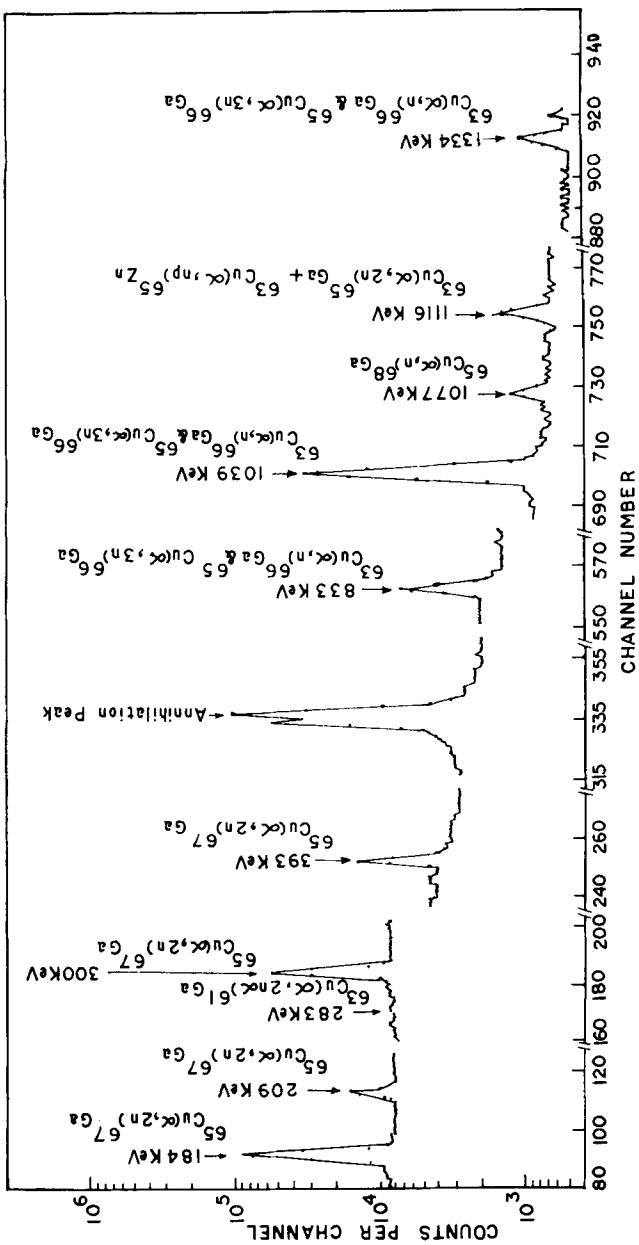

Figure 1. Observed γ-ray spectrum from ^{63,65}Cu irradiated by 26·3 MeV α-particles.

Table 1. Spectroscopic data and the measured relative intensities of γ-rays.

γ-ray energy E_γ(keV)	Absolute** abundance I_γ(%)	Normalized relative intensity: Present measurement	Normalized relative intensity: Literature value**
Residual nucleus ^{68}Ga of $T_{1/2}$ = 68·33 min.			
1077	3·0	—	100
Residual nucleus ^{67}Ga of $T_{1/2}$ = 78·26 hr.			
184·6	23·56	62·7 ± 6·4	62 ± 3
209	2·7	7·3 ± 0·2	7·1 ± 0·4
300·2	19	50·0 ± 0·3	50* ± 3
393·6	5·32	13·4 ± 0·9	14 ± 1
Residual nucleus ^{66}Ga of $T_{1/2}$ = 9·45 hr.			
833·6	6·1	14908 ± 1465	15950 ± 160
1039·3	38	1 × 10^5 ± 320	1 × 10^5*
1333·4	1·2	3265 ± 432	3260 ± 30
1419	0·64	1791 ± 358	1680 ± 20
Residual nucleus ^{65}Zn of $T_{1/2}$ = 244·1 days			
1116	50·75	—	—
Residual nucleus ^{61}Cu of $T_{1/2}$ = 3·4 hr.			
283	13·2	100 ± 0·6	100* ± 6
656	11·7	89·5 ± 7·2	88·7 ± 3·6
1185	4·88	32·7 ± 0·7	37 ± 1·5

*Normalized with respect to this value of literature; **Lederer and Shirley (1978).

Table 2. Typical values of various factors used for calculating the cross-section for ^{63}Cu(α, *n*) ^{66}Ga reaction.

Identified gamma (E_γ) (keV)	Area under photopeak (A)	Counting time in seconds	Count rate at zero time ($C_{t=0}$)	Branching ratio (θ)	Detection efficiency ($G\varepsilon$)	Cross-section (σ_r) mb	Error in cross-section ($\Delta\sigma_r$) mb
833	2489 ± 1011	276	14·3	0·0610	0·00820	18·09	7·35
1039	12691 ± 5153	276	73·0	0·3800	0·00695	17·46	7·09
1334	353 ± 143	276	2·0	0·0120	0·00520	20·56	8·33
833	1435 ± 583	300	9·9	0·0610	0·00820	12·48	5·07
1039	9859 ± 4003	300	67·8	0·3800	0·00695	16·24	6·59
1334	247 ± 101	300	1·7	0·0120	0·00520	17·22	7·04
1419	149 ± 60	300	1·0	0·0064	0·00470	21·61	8·70

Incident energy = 29·6 ± 1·6 MeV; Number of target nuclei = 0·77350 × 10^{20}; Incident flux = 0·56811 × 10^{12}; Half-life of product nucleus = 34020 s.; Irradiation time = 1800 s.; Saturation correction = 0·0360026.

Weighted average of cross section = 16·679 mb; internal error = 2·598 mb; external error = 0·454 mb.

Table 3. Measured cross-sections for α-induced reactions. (a) Target nucleus ^{63}Cu.

E_α (MeV)	$\sigma(\alpha, n)$ (mb)	$\sigma(\alpha, 2n) + (\alpha, np)$ (mb)	$\sigma(\alpha, 2n\alpha)$ (mb)
38·2 ± 1·3	3·5 ± 0·2	598·5 ± 72·2	35·7 ± 0·4
35·5 ± 1·4	5·4 ± 0·4	854·7 ± 56·1	17·0 ± 0·5
32·6 ± 1·5	6·1 ± 0·6	929·9 ± 45·8	5·5 ± 0·3
29·6 ± 1·6	16·7 ± 2·6	1158·6 ± 36·4	1·6 ± 0·2
26·3 ± 1·7	107·8 ± 0·8	1160·9 ± 47·0	0·4 ± 0·2
22·8 ± 1·8	250·6 ± 0·8	829·8 ± 81·7	
18·9 ± 2·1	500·0 ± 1·4	403·6 ± 102·1	
14·4 ± 2·4	497·1 ± 1·4		
9·0 ± 3·0	55·1 ± 1·8		

(b) Target nucleus ^{65}Cu.

E_α (MeV)	$\sigma(\alpha, n)$ (mb)	$\sigma(\alpha, 2n)$ (mb)	$\sigma(\alpha, 3n)$ (mb)
38·2 ± 1·3	15·1 ± 5·5	284·9 ± 3·6	338·4 ± 2·5
35·5 ± 1·4	—	481·7 ± 3·1	248·9 ± 3·1
32·6 ± 1·5	38·0 ± 27·6	641·1 ± 3·0	107·6 ± 4·5
29·6 ± 1·6	75·2 ± 12·2	974·4 ± 19·6	72·1 ± 6·1
26·3 ± 1·7	112·5 ± 8·6	1174·8 ± 14·5	
22·8 ± 1·8	355·0 ± 60·3	724·3 ± 4·1	
18·9 ± 2·1	687·7 ± 127·4	301·0 ± 5·3	
14·4 ± 2·4	662·1 ± 80·4		
9·0 ± 3·0	87·6 ± 8·6		

internal and external errors (Mughabghab *et al* 1981). In general, these errors are $< 10\%$ except for few points. Typical values of different factors for (α, n) reaction in ^{63}Cu target at incident energy 29·6 ± 1·6 MeV are tabulated in table 2. while the measured cross-section values are given in table 3.

3. Results and discussion

The measured excitation functions for the reactions ^{63}Cu(α, n), ^{63}Cu$(\alpha, 2n) + {}^{63}$Cu(α, np), ^{63}Cu$(\alpha, 2n\alpha)$, ^{65}Cu(α, n), ^{65}Cu$(\alpha, 2n)$ and ^{65}Cu$(\alpha, 3n)$ have been shown in figures 2–7 with closed circles. The size of the circle includes the magnitude of statistical error, if no error bar is plotted. The bars depicting the energy spread in these figures refer to energy loss in the actual thickness of the foils. Straggling effects are expected to be negligibly small (Ernst *et al* 1982). The present measurements have also been compared with the respective literature data in these figures. In general, the literature values of these excitation functions have been found to be quite different from the present measurements and also different from each other. However, earlier results (Bryant *et al* 1963; Stelson and McGowan 1964; Hille *et al* 1972) have been found to be close to our results in cases of ^{63}Cu(α, n), ^{65}Cu(α, n) and ^{65}Cu$(\alpha, 2n)$ reactions, but their measurements cover only a small energy range.

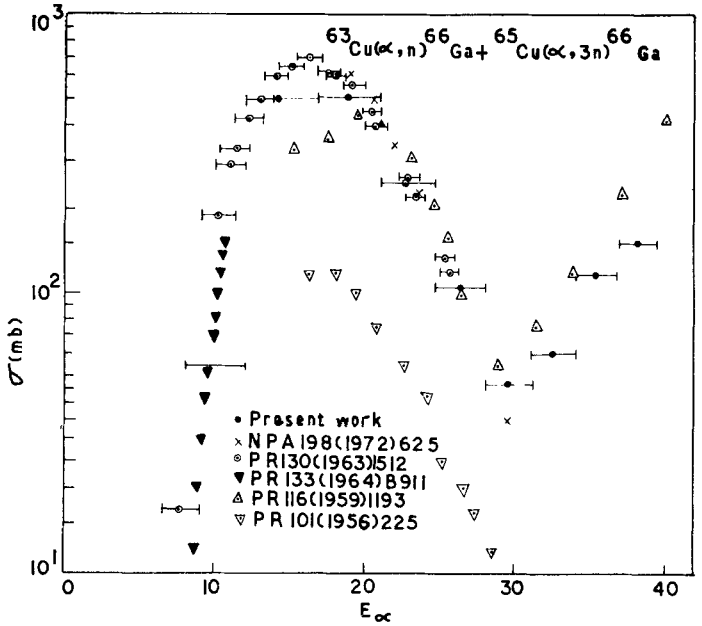

Figure 2. Excitation functions (presently measured) and other literature values.

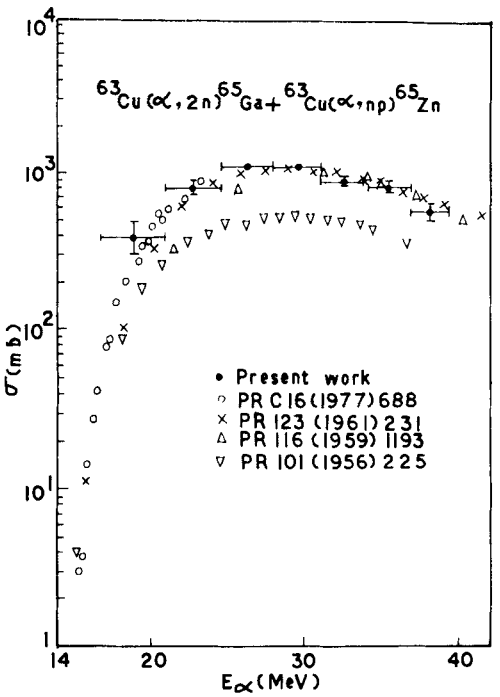

Figure 3. Excitation functions (presently measured) and other literature values.

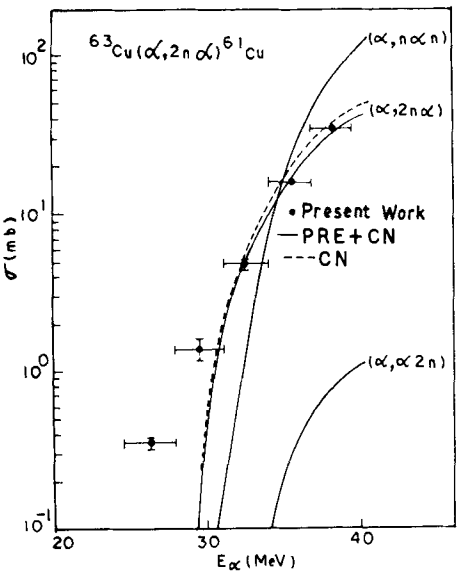

Figure 4. Experimentally measured and theoretically calculated excitation functions.

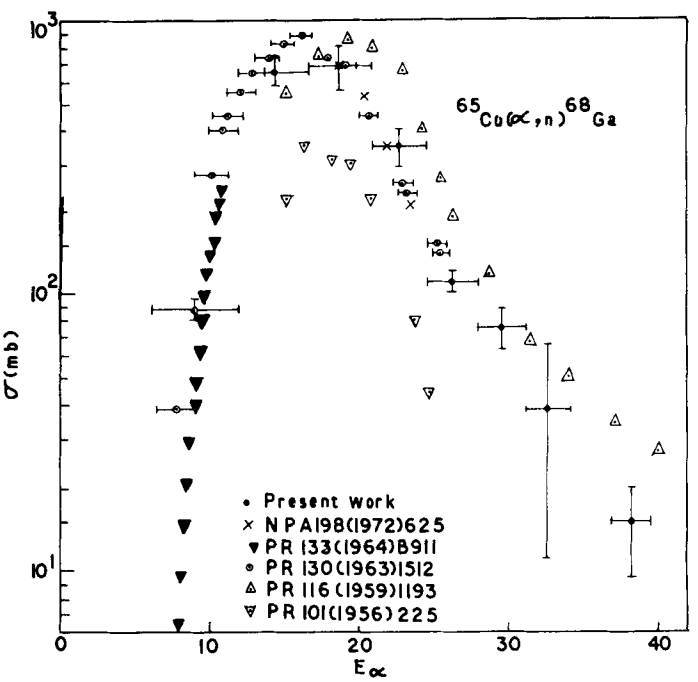

Figure 5. Excitation functions (presently measured) and other literature values.

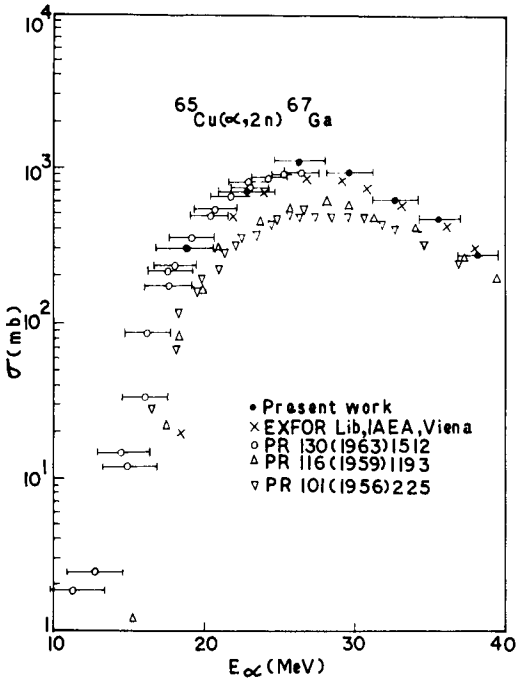

Figure 6. Excitation functions (presently measured) and other literature values.

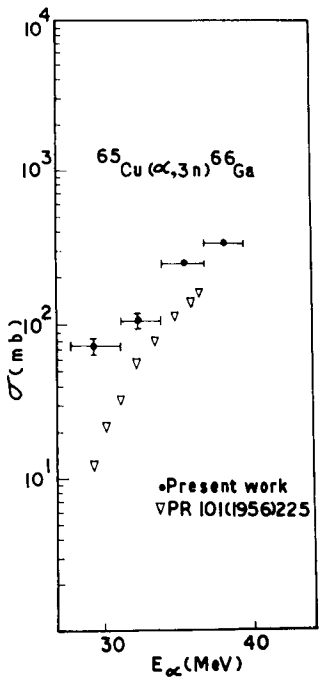

Figure 7. Excitation function (presently measured) and other literature values.

These excitation functions have been calculated theoretically using the statistical model with and without the inclusion of pre-equilibrium emission of particles. For the equilibrium part of analysis, the statistical model of Hauser-Feshbach (1952) has been adopted, while the exciton model of Griffin (1966) has been used for simulating pre-equilibrium decay of the compound system. A computer code ACT (Bhardwaj and Prasad 1986) developed on the lines of code STAPRE has been used for these calculations. The parent code has been described by Uhl and Strohmaier (1981). The computer code takes sequential evaporation of particles and considers pre-equilibrium emissions only at the first step of deexcitation of the compound system.

Level densities of residual nuclei play an important role in deciding the shapes and absolute values of the excitation functions. Level densities are calculated using the spin-dependent Lang expression (Lang 1966). Level density parameter a, fictive ground state energy Δ and the effective moment of inertia Θ for the various nuclei are taken from the backshifted Fermi gas model tables of Dilg *et al* (1973). In the cases where parameters were not available in the tables, their values are interpolated. In all calculations, the effective moment of inertia has been taken consistently equal to the rigid body value. Various separation energies, needed for calculations, are taken from the tables of Wapstra and Bos (1977). The decay schemes of various nuclei used in these calculations are from the table of isotopes (Lederer and Shirley 1978). Transmission coefficients for incident and outgoing particles, needed for these calculations, are generated using global optical potentials (Blann and Vonach 1983).

In the pre-equilibrium formulations, the configuration of the compound system is defined by its exciton number $n(p, h)$. In these calculations, no distinction has been made between neutrons and protons. Theoretical calculations have been done taking different values of the initial exciton number. It has been found that the initial configuration of 6-exciton state (5-particles, 1-hole) gives the best fit to the experimental data for α-induced reactions. Pre-equilibrium contributions are sensitive to the choice of the square of absolute value of the average effective matrix element for two-body residual interactions ($|M|^2$). The expression $|M|^2 = FM \cdot A^{-3}E^{-1}$ (Kalbach-Cline 1973) has been used to estimate its value. Here A and E are the mass number and excitation energy of the compound system respectively. In general $FM(\text{MeV}^3)$ has been treated as an adjustable parameter and the values between 95 and 7000 MeV^3 have been proposed for it in the literature (Gudima *et al* 1983). In our earlier studies on (n, p) (Gupta *et al* 1985) and (α, xn) (Bhardwaj and Prasad 1986) reactions, the best value of FM was 430 MeV^3. In the present analysis also, we have adopted the same value of FM ($= 430\ \text{MeV}^3$).

In order to match the excitation functions for various reactions induced by α-particles in a given target simultaneously, the level density parameters (a and Δ) in some cases were required to be varied within 10% from the values given by Dilg *et al* (1973). These variations are justified keeping in view the fact that the parameters of Dilg *et al* (1973) are good only in the energy range of 10 to 20 MeV and since these are of an empirical nature.

Excitation functions calculated with and without the inclusion of pre-equilibrium emission of particles are shown in figure 4 and figures 8–12. As can be seen excitation functions calculated by considering the pre-equilibrium emission of particles are in general in better agreement with the ones measured presently over the full energy range. However, some composite excitation functions now measured require special mention.

Same residual nucleus ^{66}Ga is produced by reactions ^{63}Cu(α, n) and ^{65}Cu$(\alpha, 3n)$. As a

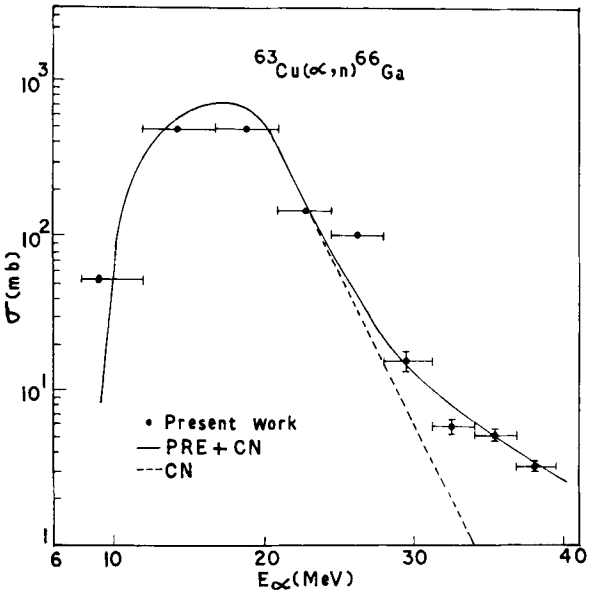

Figure 8. Experimentally measured and theoretically calculated excitation functions.

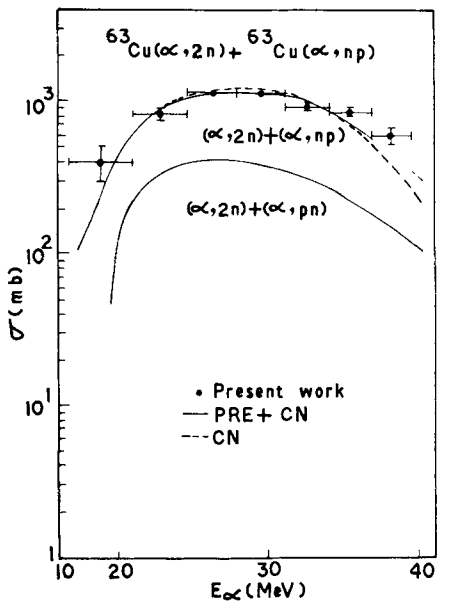

Figure 9. Experimentally measured and theoretically calculated excitation functions.

result, the observed activity of ^{66}Ga in the irradiated samples of natural copper is the composite activity due to the two reactions. The contributions from these two reactions have been separated by the theoretical analysis of the data. Below the threshold of $^{65}Cu(\alpha, 3n)$ reaction (26·9 MeV), the measured excitation function is due to $^{63}Cu(\alpha, n)$ reaction only. Therefore, the measured excitation function for $^{63}Cu(\alpha, n)$ reaction below the threshold of $^{65}Cu(\alpha, 3n)$ reaction has been first reproduced from theoretical

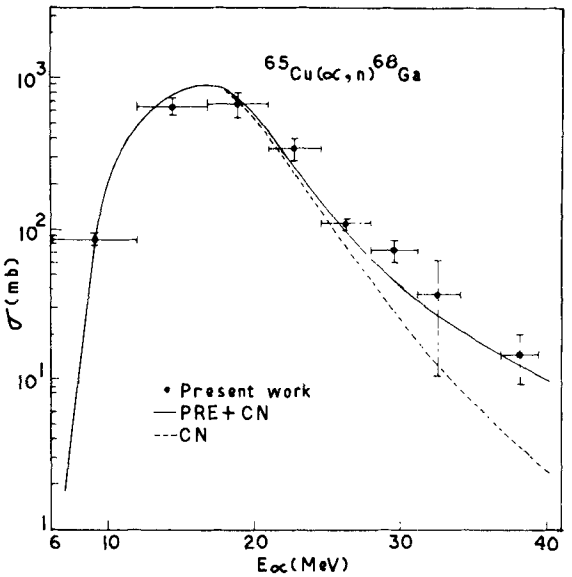

Figure 10. Experimentally measured and theoretically calculated excitation functions.

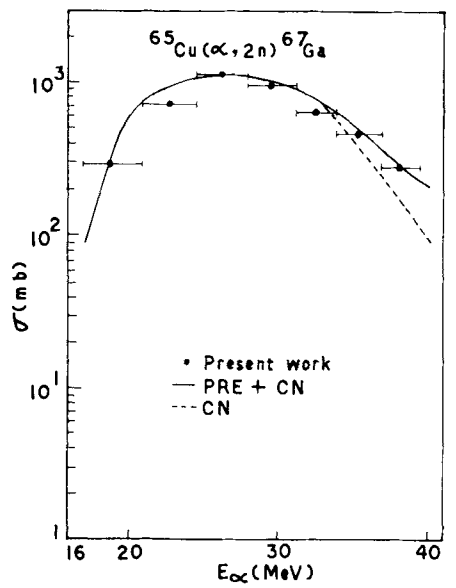

Figure 11. Experimentally measured and theoretically calculated excitation functions.

calculations. The calculated excitation function was then extended in the region of overlap, i.e. above 26·9 MeV, using the same set of level density parameters. The contribution of extrapolated part of calculated excitation function beyond 26·9 MeV for $^{63}Cu(\alpha, n)$ reaction was subtracted from the composite decay curve to get the counting rate for $^{65}Cu(\alpha, 3n)$ reaction. This method for the separation of two activities is justified since the excitation functions for reactions $^{63}Cu(\alpha, n)$ (figure 8) and for $^{65}Cu(\alpha, 3n)$

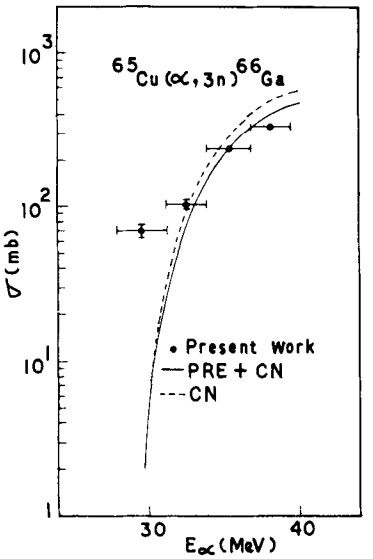

Figure 12. Experimentally measured and theoretically calculated excitation functions.

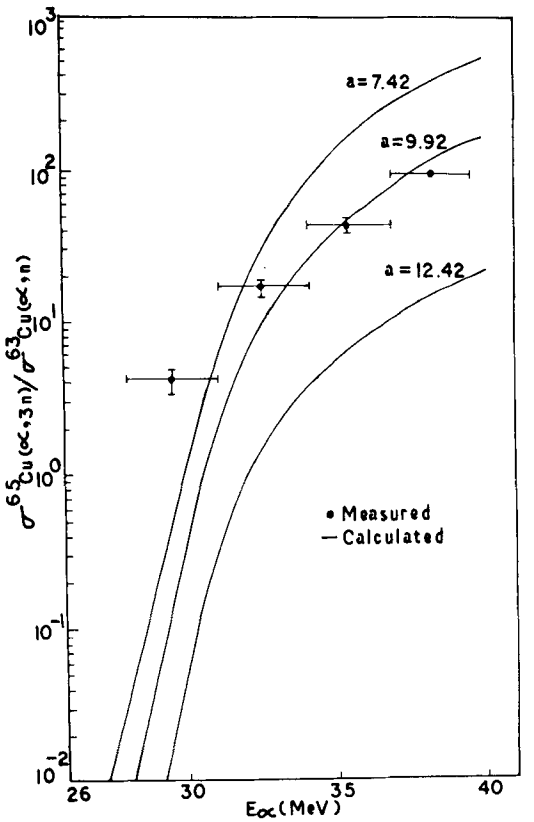

Figure 13. Dependence of the ratio $[\sigma\,^{65}\mathrm{Cu}(\alpha, 3n)/\sigma\,^{63}\mathrm{Cu}(\alpha, n)]$ on the choice of level density parameter a.

(figure 12) so resolved are reproduced individually by theoretical calculations using the same set of level density parameters for all residual nuclei produced in the evaporation chain. The analysis is thus self-consistent. To check the sensitiveness of this method to resolve the excitation functions for these two reactions to the value of level density parameter a for ^{66}Ga, the ratio of cross-section for ^{65}Cu(α, 3n) and ^{63}Cu(α, n) reactions has been calculated and plotted in figure 13 for the three different values of a for ^{66}Ga (a = 7·42, 9·92 and 12·42 MeV^{-1}) as a function of incident α-particle energy in the energy range ≈ 27–40 MeV. As can be seen, the value of a = 9·92 MeV^{-1}, used in these calculations, gives best fit with the experimental data. This further justifies the validity of the method of resolving the composite activity.

Similarly reactions ^{63}Cu(α, 2n) ^{65}Ga and ^{63}Cu(α, np) ^{65}Zn also gave a composite decay curve. This is because ^{65}Ga decays with a relatively small half-life (15 min) into ^{65}Zn which in turn decays with a half-life of 244 days. In the present measurements a comparatively longer time elapsed between the end of irradiation and the beginning of the counting and therefore the shortlived activity of ^{65}Ga could not be observed separately. Besides, the residual nucleus ^{65}Zn can be produced in two ways, i.e. evaporating first a neutron followed by a proton or a proton followed by a neutron. Theoretical calculations have been done to fit the measured composite excitation function by ^{63}Cu(α, np) + ^{63}Cu(α, 2n) or ^{63}Cu(α, pn) + ^{63}Cu(α, 2n) reactions. Excitation functions for reactions (α, 2n), (α, np) and (α, pn) in ^{63}Cu have been calculated individually using the same set of level density parameters for the intermediate nuclei. The sum of excitation functions for (α, 2n) reaction with (α, np) and with (α, pn) reactions are compared with the excitation function now measured in figure 9. This figure shows that the measured composite excitation function is in better agreement with the

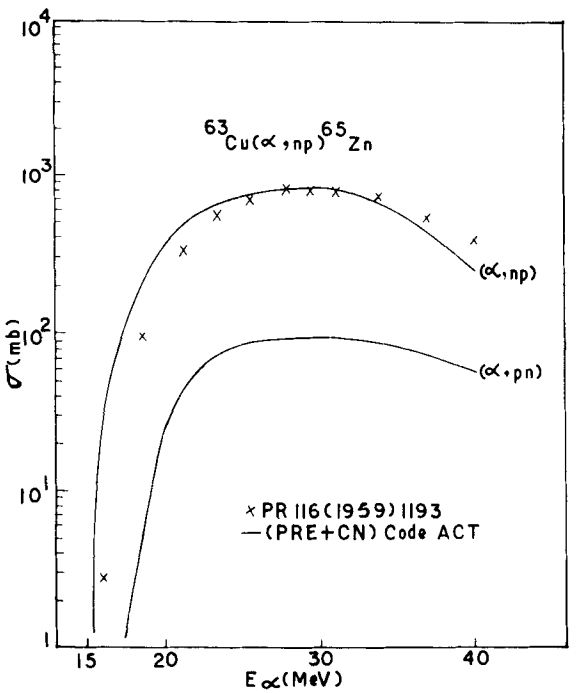

Figure 14. Theoretically calculated excitation functions and other literature data.

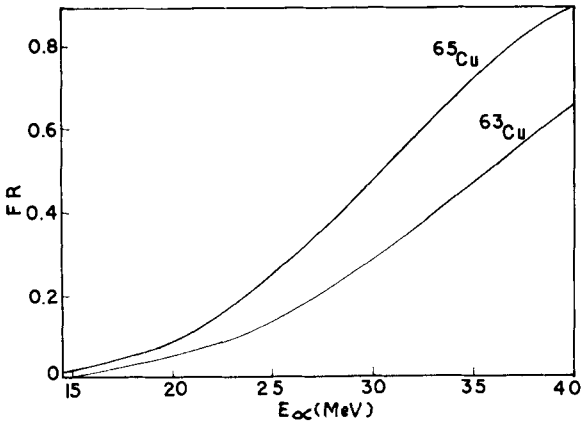

Figure 15. Variation of pre-equilibrium fraction FR, as a function of incident energy of α-particle.

combination ^{63}Cu(α, *np*) + ^{63}Cu(α, 2*n*). The calculated excitation functions for reactions (α, *np*) and (α, *pn*) in ^{63}Cu have also been compared in figure 14 with the experimental data available in literature (Porile and Morrison 1959). As can be seen the literature data are close to the calculated excitation function for the reaction ^{63}Cu(α, *np*) rather than for ^{63}Cu(α, *pn*). The analysis indicates that the first chance emission of neutron is favoured as compared to the proton even when the separation energy of neutron (11·2 MeV) in the compound system ^{67}Ga is larger than that for the proton (5·3 MeV). In this reference the measured excitation function for the production of ^{61}Cu from ^{63}Cu is of considerable interest. The residual nucleus ^{61}Cu may be produced either by (α, 2*n*α), (α, *n*α*n*) or (α, α2*n*) reactions in ^{63}Cu. Calculations have been done for these three possible ways of producing ^{61}Cu. It may be seen from figure 4 that the measured excitation function for the production of the residual nucleus ^{61}Cu from target ^{63}Cu is better reproduced if one considers a major contribution from the reaction path as ^{63}Cu(α, 2*n*α) ^{61}Cu. Thus, the first chance emission of neutron in comparison to the first chance emission of charged particles is again observed.

The present studies clearly show considerable pre-equilibrium contributions in α-induced reactions. Pre-equilibrium fraction (FR) is a measure of the relative weight of the pre-equilibrium component needed to reproduce experimental excitation functions and reflects the relative importance of pre-equilibrium and equilibrium processes. In these calculations, FR is inherently energy-dependent. This dependence is derived from the consideration of internal transition rates and of continuum decay rates. The FR is taken to be proportional to the commutative sum of the probability of finding any particle in the continuum for every possible configuration during the process of equilibration. The calculated FRs for the system of targets ^{63}Cu and ^{65}Cu are shown in figure 15 as a function of bombarding energy (E_α) in the energy range ≈ 15–40 MeV. It can be seen that FR increases with incident α-ion energy in both cases. Further, the threshold for pre-equilibrium emission is higher for the system of lower mass number.

4. Conclusion

The present results lead us to conclude that α-induced excitation functions have high energy tails, which in general, cannot be accounted for by pure equilibrium reaction mechanism. Proper admixture of equilibrium and pre-equilibrium processes is needed to reproduce the experimental data. The exciton model is quite adequate to describe the pre-equilibrium process. For α-induced reactions, the choice of 6-exciton state ($5p$, $1h$) for the initial configuration of the compound system gives satisfactory results. The $FM = 430\,\text{MeV}^3$ favours projectile-independent prescription for the average matrix element. The relative magnitude of the pre-equilibrium component has been found to depend on the compound system mass number and its excitation energy. Moreover, the present analysis indicates a preference for the first chance neutron emission.

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