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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 $^{68}\text{Ga}$, $^{67}\text{Ga}$, $^{66}\text{Ga}$, $^{65}\text{Ga} + ^{65}\text{Zn}$ and $^{61}\text{Cu}$ from $\alpha$-induced reactions in natural copper have been measured in the energy range $\alpha10-40$ MeV using stacked foil technique. A stack of nine copper foils, each having a thickness of $23.27\ \text{mg/cm}^2$ was irradiated by a $\alpha40$ MeV $\alpha$-beam of the Variable Energy Cyclotron Centre (VECC), Calcutta (India). The $\gamma$-rays emitted from the irradiated samples were recorded by a Ge(Li) detector system. Excitation functions have also been calculated theoretically using Statistical model with and without the inclusion of pre-equilibrium emission of particles. Pre-equilibrium component has been simulated by exciton model. It has been found that the inclusion of pre-equilibrium emissions gives better agreement between experimental and theoretical excitation functions. Pre-equilibrium fraction has been found to depend on the incident energy and the target mass number.

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1. Introduction

In a programme of measuring the excitation functions for $\alpha$-induced reactions using stacked foil technique, excitation functions for the reactions $(\alpha,n)$, $(\alpha,2n)+(\alpha,np)$, $(\alpha,2n\alpha)$ in $^{63}\text{Cu}$ and $(\alpha,n)$, $(\alpha,2n)$, $(\alpha,3n)$ in $^{65}\text{Cu}$ have been measured in the energy range $\approx 10-40$ MeV. Some of these excitation functions have already been measured in past, but there are large discrepancies in the cross-section values for the same reaction reported by different groups. Moreover, in most of these measurements errors associated with the cross-section values and energy are not mentioned. Porges has reported the excitation function for $^{65}\text{Cu}(\alpha,3n)^{66}\text{Ga}$ reaction using natural copper foils, but he has not discussed the procedure for separating the contribution from the reaction $^{63}\text{Cu}(\alpha,n)$ which produces the same residual nucleus $^{66}\text{Ga}$. In the present work, contributions of $^{63}\text{Cu}(\alpha,n)$ and $^{65}\text{Cu}(\alpha,3n)$ reactions have been separated using theoretical excitation function for $^{63}\text{Cu}(\alpha,n)$ reaction. To the best of our knowledge excitation function for the $^{63}\text{Cu}(\alpha,2n\alpha)$ reaction is being reported for the first time.

The analysis of these excitation functions, in past, has been done mostly on the basis of the statistical equilibrium model. Both intuition and the results of some recent measurements indicate the presence of post-equilibrium emission of particles at these moderate excitation energies. In the present analysis, excitation functions have been calculated theoretically.
using both the equilibrium and a mixture of equilibrium and pre-equilibrium mechanisms. Gamma competition and angular momentum effects have been taken into account explicitly. Theoretical calculations with an admixture of equilibrium and pre-equilibrium decay of compound system is found to agree better 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 a-beam of energy ~ 40 MeV has been made to incident on a stack of nine copper foils (natural abundance, $^{63}$Cu = 69.2% and $^{65}$Cu = 30.8%) fixed individually on aluminium sheets for immediate heat conduction. Spectroscopically pure copper foils of purity better than 99.99% and of thickness 23.27 mg/cm$^2$ have been used. The foil thickness correspond to an energy degradation of about 2.5 MeV to 5.5 MeV from first to the last foil$^{(13)}$.

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, $^{137}$Co, $^{22}$Na, $^{133}$Ba, $^{54}$Mn and $^{152}$Eu for the efficiency and $\gamma$-ray energy. The dead time for counting has been kept less than 10% by adjusting the sample-detector separation in these measurements and proper care of the dead time has been taken in calculations. In general, several $\gamma$-rays are
from the same residual nucleus have been identified. Spectroscopic
data of identified \( \gamma \)-rays are given in table 1. As a check in
some cases, the relative intensities of identified \( \gamma \)-ray have
also been measured. It can be seen from this table that presently
measured relative intensities are in good agreement with respective
literature values\(^{14}\).

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

\[
\sigma(E) = \frac{A \lambda \exp(\lambda t_2)}{N_0 \lambda (G \xi) \Theta K [1-\exp(-\lambda t_1)] [1-\exp(-\lambda t_2)]}
\]

where \( K = \frac{[1-\exp(-\mu d)]}{\mu d} \) is the correction for self absorption
of \( \gamma \)-ray in the sample of thickness \( d(\text{gm/cm}^2) \) and of absorption
coefficient \( \mu(\text{cm}^2/\text{gm}) \). \( A \) is the area under photopeak, \( \lambda \) is the
decay constant of the product nucleus, \( t_1 \) is the irradiation time,
\( t_2 \) is the time lapsed between stop of irradiation and start of
counting, \( t_3 \) is the counting period, \( N_0 \) is the number of nuclei
in the sample, \( \Theta \) is the incident flux, \( (G \xi) \) is the geometry
dependent efficiency of the detecting unit and \( \Theta \) being the
branching ratio for the specific \( \gamma \)-ray.

Activation cross-section for a given reaction has been
determined from the intensities of the various identified \( \gamma \)-rays
arising from the same residual nucleus. The reported value is
the weighted average\(^{15}\) of the various cross-section values
so obtained. The statistical error given in the results is the
larger one of the internal and external errors\(^{15}\). In general,
these errors are less than 10% except for few points. The measured values are given in table 2.

3. Results and Discussion

Measured excitation functions for the reactions $^{63}$Cu($\alpha$,n), $^{63}$Cu($\alpha$,2n) + $^{63}$Cu($\alpha$,np), $^{63}$Cu($\alpha$,2n$\alpha$), $^{65}$Cu($\alpha$,n), $^{65}$Cu($\alpha$,2n) and $^{65}$Cu($\alpha$,3n) have been shown in figures 1-6 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\(^{(6)}\). Present measurements have also been compared with respective literature data\(^{(2-9)}\) 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 with each other. However, results of Ville et al.\(^{(3)}\), Bryant et al.\(^{(6)}\) and Stelson et al.\(^{(5)}\) have been found to be close to our results in cases of $^{63}$Cu($\alpha$,n), $^{65}$Cu($\alpha$,n) and $^{65}$Cu($\alpha$,2n) reactions, but their measurements cover only a small energy range.

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\(^{(17)}\) has been adopted, while exciton model\(^{(18)}\) formalism has been used for simulating pre-equilibrium decay of the compound system. A computer code ACT\(^{(1)}\) developed on the lines of code STAPPRE\(^{(19)}\) has
been used for these calculations. A detailed description of the parent code may be found elsewhere. 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 important role in deciding the shapes as well as the absolute values of the excitation functions. Level densities are calculated using spin dependent Lang expression. Level density parameter 'a' fictive ground state energy 'Δ' and effective moment of inertia 'Θ' for the various nuclei are taken from the back-shifted Fermi gas model tables of Dilg et al. For those cases in which these parameters were not available in these tables, their values are interpolated. In all calculations, 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 Mapstra et al. The decay schemes of various nuclei used in these calculations are from table of isotopes. Transmission coefficients for incident and outgoing particles, needed for these calculations, are generated by an optical model code TLK developed on lines of code MB2 which uses global optical potentials.

In the pre-equilibrium formulations, the configuration of the compound system is defined by its exciton number n(n, 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 \( \alpha \)-induced reactions. Pre-equilibrium contributions are sensitive to the choice of square of absolute value of average effective matrix element for two-body residual interactions \( |M|^2 \). Following expression \( |M|^2 = \frac{FM}{A^2 E^{-1}} \)

Here \( A \) and \( E \) are the mass number and excitation energy of the compound system respectively. In general \( FM(MeV^3) \) has been treated as an adjustable parameter and values between 95 to 7000 MeV\(^3\) have been proposed for it in literature \( (26) \). In our earlier studies on \( (n,p) \) \( (27) \) and \( (\alpha, xn) \) \( (1) \) reactions, the best value of \( FM \) was found to be 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 \( \alpha \)-particles in a given target simultaneously, it was required to vary the level density parameters \((a, A)\) in some cases within 10\% from the values given by Dilg et al. \( (21) \). These variations are justified keeping in view the fact that Dilg et al. parameters are good only in the energy range of 10 to 20 MeV\( (21) \) and also these are of empirical nature.

Excitation functions calculated with and without the inclusion of pre-equilibrium emission of particles are shown in Figs. 3, 7, 11. As can be seen in these figures, in general,
excitation functions calculated with the consideration of pre-equilibrium emission of particles are in better agreement with the ones measured presently over full energy range. However, some composite excitation functions, measured presently, require special mention.

Same residual nucleus $^{66}$Ga is produced by reactions $^{63}$Cu($\alpha$,n) and $^{65}$Cu($\alpha$,3n). As a 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 calculations. The calculated excitation function was then extended in the region of overlap, i.e. above 26.9 MeV, using same set of level density parameters. The contribution of extrapolated part of calculated excitation function beyond 29.9 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) (fig. 7) and for $^{65}$Cu($\alpha$,3n) (fig. 11) 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. In this way the analysis is self consistent. To check the sensitiveness of this method for the resolution of the excitation functions for these two reactions to the value of level density parameter 'a' for $^{66}\text{Ga}$, the ratio of cross-section for $^{65}\text{Cu}(\alpha,3n)$ and $^{63}\text{Cu}(\alpha,n)$ reactions has been calculated and plotted in fig. 12 for the three different values of a for $^{66}\text{Ga}$ ($a = 7.42$, 9.92 and 14.42 MeV$^{-1}$) as a function of incident $\alpha$- particle energy in the energy range 27-40 MeV. As can be seen, the value of $a = 9.92$ MeV$^{-1}$, which has been used in these calculations, gives best fit with the experimental data. This further justifies the validity of the method of resolving the composite activity.

In a similar way reactions $^{63}\text{Cu}(\alpha,2n)$ $^{65}\text{Ga}$ and $^{63}\text{Cu}(\alpha,np)$ $^{65}\text{Zn}$ also gave a composite decay curve. It is because $^{65}\text{Ga}$ decays with a relatively small half life (15 min.) into $^{65}\text{Zn}$ which in turn decays with a half life of 244 days. In the present measurements comparatively long time was lapsed between the stop of irradiation and the start of counting and so, the short lived activity of $^{65}\text{Ga}$ could not be observed separately. Besides, the residual nucleus $^{65}\text{Zn}$ 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 $^{63}\text{Cu}(\alpha,np) + ^{63}\text{Cu}(\alpha,2n)$ or $^{63}\text{Cu}(\alpha,\text{pn}) + ^{63}\text{Cu}(\alpha,2n)$ reactions. Excitation
functions for reactions (α,2n), (α,np) and (α,pn) in ⁶³Cu have been calculated individually using same set of level density parameters for the intermediate nuclei. Sum of excitation functions for (α,2n) reaction with (α,np) and with (α,pn) reactions are compared with the excitation function measured presently in fig. 8. As can be seen in this figure, the measured composite excitation function is in better agreement with the combination ⁶³Cu (α,np) + ⁶³Cu(α,2n). The calculated excitation functions for reactions (α,np) and (α,pn) in ⁶³Cu have also been compared with experimental data available in literature in fig. 13. As can be seen the literature data is close to the calculated excitation function for the reaction ⁶³Cu(α,np) rather than for ⁶³Cu (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 ⁶⁷Ga is larger than that for the proton (5.3 MeV). In this reference measured excitation function for the production of ⁶¹Cu from ⁶³Cu is of considerable interest. Residual nucleus ⁶¹Cu may be produced either by (α,2nα), (α,nαn) or (α,α2n) reactions in ⁶³Cu.Calculations have been done for these three possible ways of producing ⁶¹Cu. It may be seen in fig. 3 that the measured excitation function for the production of the residual nucleus ⁶¹Cu from target ⁶³Cu is better reproduced if one considers a major contribution from the reaction path as ⁶³Cu (α,2nα)⁶¹Cu. Thus, first chance emission of neutron in comparison to the first chance emission of charged particles is observed again.
Present studies indicate clearly the presence of considerable amount of pre-equilibrium contributions in $\alpha$-induced reactions. Pre-equilibrium fraction (FR) is a measure of relative weight of the pre-equilibrium component needed for the reproduction of experimental excitation functions and it 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 system of targets $^{63}\text{Cu}$ and $^{65}\text{Cu}$ are shown in fig. 14 as a function of bombarding energy ($E_{\alpha}$) in the energy range $\approx 15$-40 MeV. It can be seen in this figure that FR increase with incident $\alpha$-ion energy in both cases. Further, the threshold for pre-equilibrium emission is higher for system of lower mass number.

It may, therefore, be concluded that alpha induced excitation functions have high energy tails, which, in general, can not be accounted for by pure equilibrium reaction mechanism. Proper admixture of equilibrium and pre-equilibrium processes is needed to reproduce the experimental data. Exciton model is quite adequate for the description of the pre-equilibrium process. For $\alpha$-induced reactions, 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 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 first chance neutron emission.

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