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Significant Amount of Pre-Equilibrium Contribution in the $\alpha + {}^{93}$Nb System at Energies $\approx$ 18–40 MeV

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To study the role of a large pre-equilibrium emission of light nuclear particle(s) on the excitation functions of the reaction channels (α, n), (α, 2n), (α, 3n), (α, 4n), (α, 2p), (α, p3n), and (α, αn) in the $\alpha+{}^{93}$Nb system, an experiment has been performed using an α beam up to 40 MeV at the Variable Energy Cyclotron Center, Calcutta (India). In the present work, the experimental measurement is carried out using the stack foil activation technique followed by off-line high-purity Ge γ-ray spectroscopy methods. Results of the experimental measurements have been analyzed using the statistical model code of ALICE-91 and compared with the existing data for the same system [J. Phys. Soc. Jpn. **59**, 3916 (1990)]. The present analysis indicated that at higher energy points the pure compound nucleus calculations failed to reproduce the present experimental data at beam energies $\approx$ 18–40 MeV and revealed large contributions from pre-equilibrium emission. Furthermore, analysis of the data suggests that a summation of both equilibrium and pre-equilibrium emissions is needed to reproduce the present experimental data in the energy range considered.

We also calculate the pre-equilibrium fraction as a function of the normalized projectile energy, and a definite dependence has been observed on the projectile energy and on the number of emitted particle(s).

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I. INTRODUCTION

The process of pre-equilibrium emission in light-ion induced reactions is a subject extensively studied in the last few years. Although there have been many theoretical and experimental works on this subject, the process is still not fully understood, particularly where competition between the equilibrium (EQ) and pre-equilibrium (PEQ) emission of light particles is high [1–2].

The emission of light nuclear particles in the PEQ process followed by non-statistical γ-rays is assumed to arise from the interaction of the projectile with the target nucleons at the early stage of the reaction. To explain this subject many models have been proposed, including the internuclear cascade model [3–4] and the quasi free scattering model [5],

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and in the framework of the exciton model many other semi-classical models have been proposed [6–10]. Following the pioneering work of Griffin [11], the pre-equilibrium exciton model [7–8, 12] is considered to provide the most suitable description, especially for alpha-induced reactions. Apart from these semi-classical models of the nuclear reaction for the successful reproduction of the excitation function data, efforts are in progress to give a full quantum mechanical picture in the framework of the multistep theories proposed by Feshbach, Kerman, and Koonin [13] and others [14–15]. The models mentioned above have been used to describe various experimental data; however, the behavior of light ion induced reactions associated with the entrance channel, mass asymmetry, energy range, etc. is still not well understood. Furthermore, the light ion-induced nuclear reactions are also important in basic research for a better understanding of the reaction dynamics and to test the validity of various available and newly evolving statistical model codes. Moreover, the compiled experimental data on EQ and PEQ emission from various light ion-induced nuclear reactions may be applicable in applied research on nuclear energy generation, in the production of medically important radionuclides, and in advanced reactor technologies as well [16–17]. Inevitably this has led to a renewed interest in the study of nuclear reactions. The motive of the present work is to study the interplay between the EQ and PEQ emission processes in the production of ^{96}Tc, ^{95}Tc, ^{94}Tc, ^{93}Tc, ^{95}Nb, ^{93}Mo, and ^{92}Nb isotopes by precise measurements of excitation functions (EFs) using the stacked foil activation technique followed by off-line γ-ray spectroscopy. In Section II, the experimental procedures and results are described; in the sections to follow the measured EFs are compared with the theoretical model predictions. Results of the present analysis are presented, and finally conclusions are drawn.

II. EXPERIMENTAL PROCEDURE AND RESULTS

The experiment has been carried out at the Variable Energy Cyclotron Center (VECC), Calcutta (India). A stack containing pure ^{93}Nb samples was irradiated by a 40 MeV diffused beam of diameter 8 mm at the VECC. The irradiation of the stack was performed with a beam current of about 380 nA so as to ensure that only a few hours of well-controlled irradiation was sufficient to excite the required activities. The targets of ^{93}Nb of thickness 10.5 mg/cm^2 (measured by the α-transmission method) were fastened to an aluminum frame, having a circular hole of diameter 1.2 cm in the center backed by an Al-catcher of thickness 2 mg/cm^2.

The incident flux of the α-beam was determined from the charge collected in the Faraday Cup, as well as from the counts of the two charge integrators. A copper foil of thickness 10.68 mg/cm^2 was also used as a flux monitor, and the values were found to agree with each other within the range of 4–8%. The activities induced in the catcher-target assembly were followed off-line, using the pre-calibrated CANBERA's HPGe detector coupled to the Ortec's PC-based multi-channel analyzer. The nuclear spectroscopic data used in the evaluations and measurements of cross-sections have been adopted from the radioactive isotopes data table [18]. The activities induced in the assembly were recorded,

leading to the production probability measurement of individual evaporation residues. The sample-detector separation was carefully adjusted so as to keep the dead time of the counting geometry at less than 10%. The background spectra were also recorded for different time intervals in order to check the presence of any background γ-line due to contamination of the detector with the surroundings. The spectrometer was calibrated for energy and efficiency using various standard sources, i.e., ^{152}Eu, ^{60}C, ^{57}Co, and ^{133}Ba. The details of the geometry dependent efficiency measurements used in this work are similar to those of Ref. [19].

TABLE I: Decay characteristics of the nuclides studied.

Reaction	Nuclide	Half-life	$\gamma-$ray energy (KeV)	Iγ γ/decay	Q-value (MeV)
^{93}Nb(α, n)	^{96}Tc^{g}	4.35 d	778.0 850.0	0.991 0.970	-7.04
^{93}Nb(α, 2n)	^{95}Tc^{m} ^{95}Tc^{g}	60.0 d 20.0 h	204.0 765.8	0.662 0.930	-14.91
^{93}Nb(α, 2p)	^{95}Nb^{m} ^{95}Nb^{g}	90.0 h 34.9 h	235.7 765.8	0.975 0.998	-12.58
^{93}Nb(α, 3n)	^{94}Tc^{m} ^{94}Tc^{g}	52.5 min 293.0 min	871.0 702.6 871.0 850.0	1.000 1.000 1.000 0.980	-24.84
^{93}Nb(α, p3n)	^{93}Mo^{m}	6.9 h	263.0 1477	0.580 0.983	-29.48
^{93}Nb(α, αn)	^{92}Nb^{m}	10.1 h	934.0	0.973	-8.83

The evaporation residues produced through different reaction channels were identified by their characteristic γ-rays, Q-values, and decay-curve analysis. The details of the experimental arrangements, formulations, and data reduction procedures used in the present work are similar to those in Ref. [20].

III. RESULTS AND DATA ANALYSIS

In the present work we have made an attempt to measure the excitation functions for the (α, n), (α, 2n), (α, 3n), (α, 4n), (α, 2p), (α, p3n), and (α, αn) reaction channels set by the interaction of α with Nb, using the activation technique followed by off-line γ spectroscopy. A list of reaction channels, Q-values, half lives, energy of identified γ-rays, and their branching ratios are given in Table I. The cross-section for a given reaction channel was determined using the observed intensities of characteristic γ-rays, arising from the same residue nucleus. The reported cross-sections in Table II are the weighted average of the

TABLE II: Cross-section in mb for ^{93}Nb(α, x) measured experimentally.

Beam energy, (MeV)									
Reaction	Isomer	Nuclide	23.3	26.1	28.2	30.9	33.2	35.3	37.4
^{93}Nb(α, n)		^{96}Tc	64.7	30	24.5	19.2	16.7	17.9	14.3
^{93}Nb(α, 2n)	^{95}Tc^{m} ^{95}Tc^{g}	^{95}Tc	74 932 1006	61.5 750.8 812.3	35.7 466 501.7	64 333.6 397.6	30 263.3 293.3	22.2 224.9 247.1	10.7 211.9 222.6
^{93}Nb(α, 2p)	^{95}Nb^{m} ^{95}Nb^{g}	^{95}Nb	- - -	- - -	- - -	- - -	0.46 0.09 0.55	0.73 0.10 0.83	1.06 0.24 1.30
^{93}Nb(α, 3n)	^{94}Tc^{m} ^{94}Tc^{g}	^{94}Tc	3.4 20.8 24.2	2.9 15.6 18.5	2.8 54.9 57.7	41.7 197.7 239.4	29 620 649	21 814 835.2	17.3 895 912.3
^{93}Nb(α, p3n)	^{93}Mo^{m}		-	-	-	-	-	1.36	16.8
^{93}Nb(α, αn)	^{92}Nb^{m}		4.3	6.5	9.1	11.7	13.1	17.3	13.6

various cross-section values [21]. The errors quoted in the production cross-sections arise mainly from uncertainties in the target thickness, detector efficiency, the beam current integration, counting statistics, and evaluating the γ-ray intensity and the background subtraction. Proper care was taken in order to keep the beam current constant; however, all the fluctuations that were identified during the irradiation time and the beam flux were separately calculated for the duration of fluctuations. The error from the fluctuations in the beam current was found to be around 3%. The overall error in the measured cross-section is found to be less than 16%. However, the uncertainties in the branching ratio decay constant, half-lives, etc., which are taken from the table of isotopes [18], have not been taken into account in the measured cross-sections. The analysis of experimentally measured EFs was done using the statistical model code ALICE-91 [22], which employs the Weisskopf-Ewing model [23] to calculate the EQ cross-sections and a geometry dependent hybrid model to calculate the PEQ cross-sections [10], as shown in Fig. 1. For a better analysis of the experimental data the code ALICE-91 is frequently used [20, 24]. This may be attributed to the fact that the input parameters of this code are few and well defined. Moreover, the theoretical analyses with ALICE-91, in general, are found to give reasonably good agreement with the experimental data. The Myers-Swiateck/Lysekil mass formula [25] is used. Like other classical models, in ALICE-91 for PEQ emissions the equipartition of energy among the initial excited particles and holes is assumed. The mean free path for the internuclear transition rates may be calculated either from the optical potential parameters of Bechetti and Greenless [26] or from Pauli-corrected n-n cross-sections [27–28].

The input parameters, initial exciton number n_o, and mean free path multiplier *MFP* largely govern the PEQ emission contribution, while the level density parameter a (defined as $a = A/K$, where A is the composite system nucleon number and K is an adjustable constant) affects the EQ compent. These are the important input parameters that may be

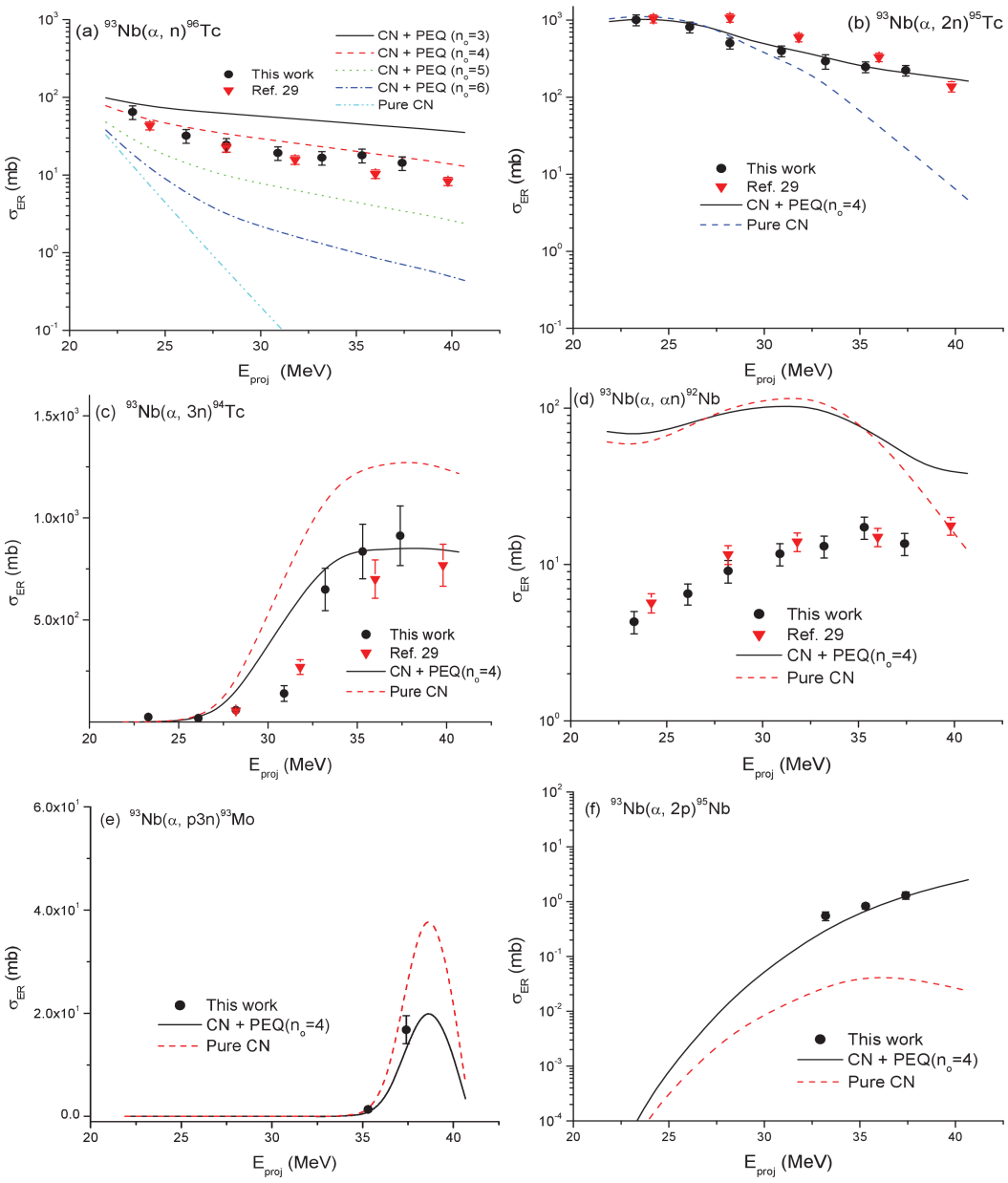

FIG. 1: (Colour online) Experimentally measured and theoretically calculated excitation functions for evaporation residues produced in the α + ^{93}Nb reaction using the code ALICE-91.

varied to reproduce the experimental data. In the Hybrid model, the intermediate states of the system are characterized by the excitation energy, the number of excited particles n_p, and the number of excited holes n_h. Particles and holes are defined relative to the ground state of the nucleus and are called excitons. In PEQ reactions, the initial exciton configuration is a crucial quantity. The influence of this initial exciton configuration on the

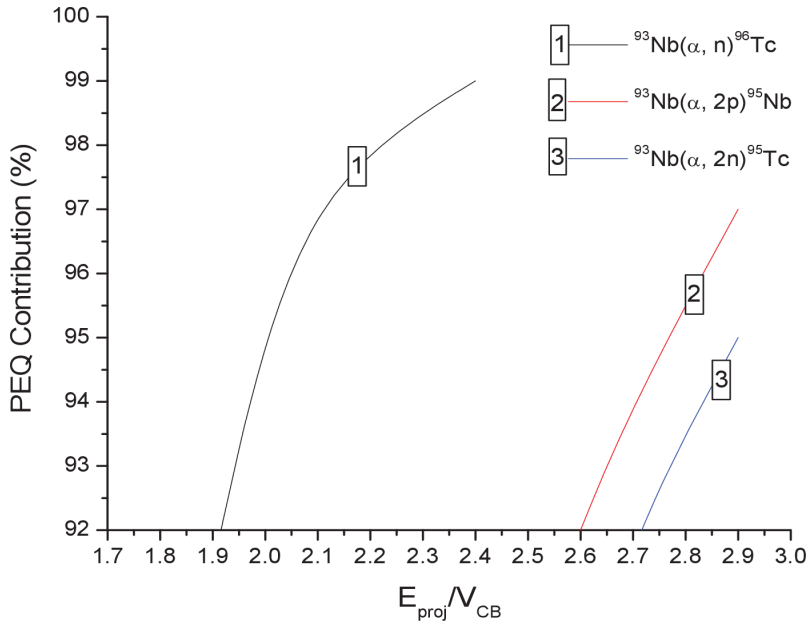

FIG. 2: (Colour online) The percentage of estimated PEQ contribution as a function of normalized projectile energy.

calculated EFs was investigated by varying the initial exciton configuration n_o, which is described by the number of neutrons (n) and protons (p) in excited states and the number of holes (h) after the first collision. The total exciton number n_o is equal to the sum of n, p, and h. To see the effects of n_o on the EFs and to find a nicely fitted value from it, calculations at different n_o values were performed.

As a representative channel, calculated EFs for different values of n_o ranging from 3 to 6 with the configurations $n_o = 3(2p+n+0h)$, $n_o = 4(2p+2n+0h)$, $n_o = 5(2p+2n+h)$, and $n_o = 6(3p+2p+h)$ for the reaction channel (α, n) are shown in Figure 1(a). It may be pointed out that, in PEQ emission, the initial exciton number is an important quantity and determines the shape of the PEQ component. For α-induced reactions Blann [28] has given justification for the values of the initial exciton number n_o =4 or 5, since a lower value of n_o means a large number of two-body interactions prior to the equilibration of the composite system, leading to a significant PEQ contribution. In the present work, it may be deduced that a set of $K = 8$, $n_o = 4$ with MFP=1 gives the best combination in reproducing satisfactorily the experimental data, in general. Throughout this work the same data set has been consistently used to calculate the cross-section values from the code. It is very clear from Fig. 1 that the CN calculations do not satisfactorily reproduce the experimental data at higher energies, where PEQ emission may be important. However, in the tail portion of the EFs the aggregate of the EQ and PEQ emissions reproduce the experimental data to a satisfactory level.

Further, in some cases the common γ-ray energy is reproduced by various residues,

and hence the observed count rate may be the sum of contributions from different paths. For example the 850 KeV γ-ray is common to the ^{93}Nb (α, n)^{96}Tc and ^{93}Nb(α,3n)^{94}Tc channels, where the Q-values of these reactions are -7.04 MeV and -24.84 MeV, respectively. Thus, in the analysis of (α, n) the 850 KeV photopeak was not used at energies above the threshold for the (α, 3n) channel, as this γ-ray is also associated with the (α, 3n) channel. The 778 KeV γ-ray is common to the ^{96}Tc isomers from the (α, n) channel. However, this peak can be used in the analysis after the decay of metastable state activity. The metastable state of ^{96}Tc decays to the ground state with 95% isomeric transition and 2% electron capture and β^+ decay. The half-life of the metastable state is much shorter than that of the ground state. Therefore, the cross-sections for the ground state producing reaction as obtained above is almost the total cross section for the production of ^{96}Tc isomers, as the activities were measured after the decay of the metastable state.

Furthermore, the comparison of the measured cross-sections for the residues ^{96}Tc, ^{95}Tc, and ^{94}Tc via the (α, n), (α, 2n), and (α, 3n) channels, respectively, has also been made with the results of other workers in Ref. [29] and are given in Figs. 1(a)–1(c). It can be seen from these figures that our results are in good agreement with those of Singh *et al.* [29], both in magnitude and pattern, in general.

The excitation function for the (α, αn) channel were measured only for the metastable state of ^{92}Nb. As can be seen from Fig. 1(d), the present data points are in good agreement with the literature values of [29]. In fact, a comparison of the experimental results with the theoretical one is inappropriate, since we were only able to measure the metastable state cross-sections. However, it is possible from the measured and calculated EFs to have some idea on the reaction mechanisms involved in this channel. Fig. 1(d) shows that the calculated EFs exhibit a peak while the measured peak shows saturation. The slow variation in cross-sections may be attributed to the fact that the reaction mechanisms involved in the (α, αn) channel come from a direct knockout process, where the projectile knocks out a neutron from the target nucleus, leaving the residual in an excited state.

It may further be pointed out that the measured data set is well fitted with the prediction of the code ALICE-91. In general, it is clear from Fig. 1 that the PEQ emission is significantly observed in most channels of the present study. Thus, a systematic attempt has been made to estimate the PEQ contribution at a given energy for a particular reaction channel. It may be defined as the ratio of the PEQ cross-section (defined as the cross-section difference between the measured evaporation residue and the pure CN prediction of ALICE-91 (σ_{ER}- σ_{CN})) to the evaporation residue cross section (σ_{ER}). The percentage PEQ contribution deduced from the systematic used in the analysis of the data for different reaction channels are plotted in Fig. 2 as a function of the normalized projectile energy (E_{poj}/V_{CB}). It can be seen clearly from Fig. 2 that the percentage of the PEQ contribution is found to increase with normalized projectile energy. Furthermore the threshold of PEQ emission for the various reaction channels is found to be different, depending on the corresponding Q-value. However, it may be pointed out that the PEQ contribution is found to be greater for the channels that consist of fewer PEQ particle(s), even at smaller projectile energy.

IV. CONCLUSION

It may be summarized that the measured data and the predictions of the statistical model code ALICE-91 disclose a significant contribution from the PEQ emission process at the studied energies of the present system. It may be further pointed out that a set of $K = 8, n_o = 4(2p + 2n + 0h)$ with *MFP=1* is found to give a satisfactory reproduction of the experimental data. The percentage PEQ contribution for individual reaction products is found to be sensitive to the Q-value of the reaction channels and/or the PEQ particle multiplicity. As such, we conclude that the PEQ emission is an important mode of reaction in light ion induced reactions at the studied energies. Further information about the PEQ emission may be obtained by the PEQ particle(s) multiplicity and energy spectra measurement in the EQ and PEQ emission process, leading to an estimate of the entry point from PEQ emission to EQ processes.

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