Nuclear Dynamics in Heavy Ion induced Fusion-Fission Reactions

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Abstract

Heavy ion induced fission and fission-like reactions evolve through a complex nuclear dynamics encountered in the medium energy nucleus-nucleus collisions. In this paper, we shall discuss the studies of neutron-fragment angular correlations and fission fragment angular distributions in heavy ion induced fission with particular regard to the results from some recent studies carried out with the BARC-TIFR pelletron accelerator and to the information provided by these studies about the dynamics of nucleus-nucleus collision and the fission process. Deduction of times involved in nuclear dynamics through the use of "prescission neutron clock" will be presented for two typical target-projectile combinations.

1. INTRODUCTION

In the recent years, several studies of heavy-ion induced fusion-fission reaction have turned out to be extremely rich in providing information on the dynamical times during the various stages of nuclear shape changes involved in these reactions. In particular, measurements of the isotropic 'prescission neutron' component through the studies of fragment-neutron angular correlations have provided a very useful 'neutron clock' for the dynamical times, which have suggested that fission is a slow process implying a highly dissipative motion of the nuclear fluid.

It is also now well established[1-4] that in several cases of heavy ion induced fusion-fission reactions, the observed fragment angular distributions are highly anisotropic, and are not consistent with the predictions of the statistical transition state model. We have earlier presented[5] a model which explains these anomalous fragment angular distributions on the basis that in heavy-ion induced fusion-fission reactions, the observed fission events consist of an admixture of two types of events: (i) compound nucleus fission with the normal, statistical K-distribution at the saddle point given by $e^{-K^2/2K_0^2}$ ($K_0^2 = J_{eff}T/\hbar^2$, J_{eff} – effective moment of inertia of the transition state nucleus) and (ii) non-compound nucleus fission (NCNF) events in the sense that the dinuclear complex which is evolving towards the compound nucleus *minimum* energy configuration undergoes fission before equilibrating in the K-degree of freedom resulting in a much narrower K-distribution than that for events of type (i). Reaction mechanisms such as fast fission taking place for the case of composite systems with zero-fission barriers and quasi-fission taking place for composite systems with fission-barrier shapes more compact than the entrance-channel contact configuration, belong to fission-like events of type (ii). Another class of NCNF events proposed in our work[5] is the "pre-equilibrium fission" events occurring in a time scale comparable to the characteristic relaxation time in the K-degree of freedom when the fission-barrier heights become comparable to the temperature of the composite system. The K-distribution for these NCNF events was taken to be the $e^{-K^2/2\sigma_K^2}$ where $\sigma_K^2 = I^2\sigma_\theta^2$ and σ_θ^2 is a small angular variance representing misalignment of the symmetry axis of the fused system with respect to K=0 plane in NCNF events.

In the earlier work[5], the fragment angular distributions for a number of heavy ion induced fission reactions were fitted with a single set of parameters corresponding to a value of $\sigma_\theta = 0.06$ and a K-equilibration time $\tau = 8\times10^{-21}$s. This implied that fissions taking place prior to the time τ follow the narrow unequilibrated K-distribution and these events were identified as pre-equilibrium fission events. The fission taking place after this time are the normal compound nucleus fission events with the statistical K-width represented by the variance K_0^2. It was earlier also suggested[6] that a characteristic signature of fission before K-equilibration would be the observation of an entrance-channel dependence of the fragment anisotropies for target-projectile combinations across the Businaro-Gallone Ridge in mass/charge asymmetry degree of freedom. One also expects that the dynamical times particularly the times involved in the shape changes during the fusion process may also be dependent on the entrance channel mass asymmetry. Also the fraction fissioning without K equilibration is expected to have somewhat smaller dynamical times than the fraction of fissions going through fusion-compound nucleus-fission path. It is therefore of interest to also carefully investigate the entrance channel dependence of "prescission neutron components" and hence the dynamical times. To look for such entrance-channel dependence we have carried out measurements of both the fission fragment angular distributions and the prescission neutron multiplicities for several cases, which will be presented and discussed below:

2. Experimental Results

A. Fragment-neutron angular correlation

With the heavy-ion beams from BARC-TIFR pelletron accelerator, prescission neutron multiplicities for ^{11}B, ^{12}C and ^{16}O projectiles on ^{232}Th and ^{237}Np targets have been measured at several energies and details of these measurements are described elsewhere[7]. Pre and post scission neutron components were determined by fitting the observed neutron spectra at $0°$ and $90°$ with respect to the fragment direction. Fig. 1 shows a typical example of the observed neutron spectra and fits for the case of $^{12}C+^{232}Th$ at $E_{lab}=74$ MeV for the fragment-neutron angles θ_{nf} of $0°$ and $90°$ with respect to the two neutron detectors kept at the angles θ_n with respect of beam direction of $15°$ and $105°$ respectively. Fig. 2 shows the results on the deduced preneutron multiplicities ν_{pre} versus compound nucleus excitation energies for the two systems C+Th and O+Th. We also show in the same figure the values of ν_{pre} expected from the statistical model calculations of Γ_n/Γ_f for the values of the level density parameter shown. It is apparent that considerable number of excess neutrons ν_{excess} emitted during nuclear dynamics are present in both the cases. Fig. 3 gives the dynamical times needed to account for these excess neutrons as calculated from the statistical model programme PACE II. While there can be some uncertainty in the deduced dynamical times due to the dependence of the results on the assumed level density parameter, it is clear that the dynamical times involved are about few times 10^{-20}s. Specifically, the prescission time scales determined by the statistical model calculations for the $^{12}C+^{232}Th$ and $^{16}O+^{232}Th$ systems are 4.5×10^{-20}s and 5.5×10^{-20}s respectively for the level density parameter $a_n=a_f=A/10$ implying that $^{16}O+^{232}Th$ fissioning system is somewhat slower compared to $^{12}C+^{232}Th$ system. However, it is difficult to ascribe this difference to the differences in the dynamical times for fusion for the two cases, due to the presence of a small fraction of pre-equilibrium fission in the ^{16}O case.

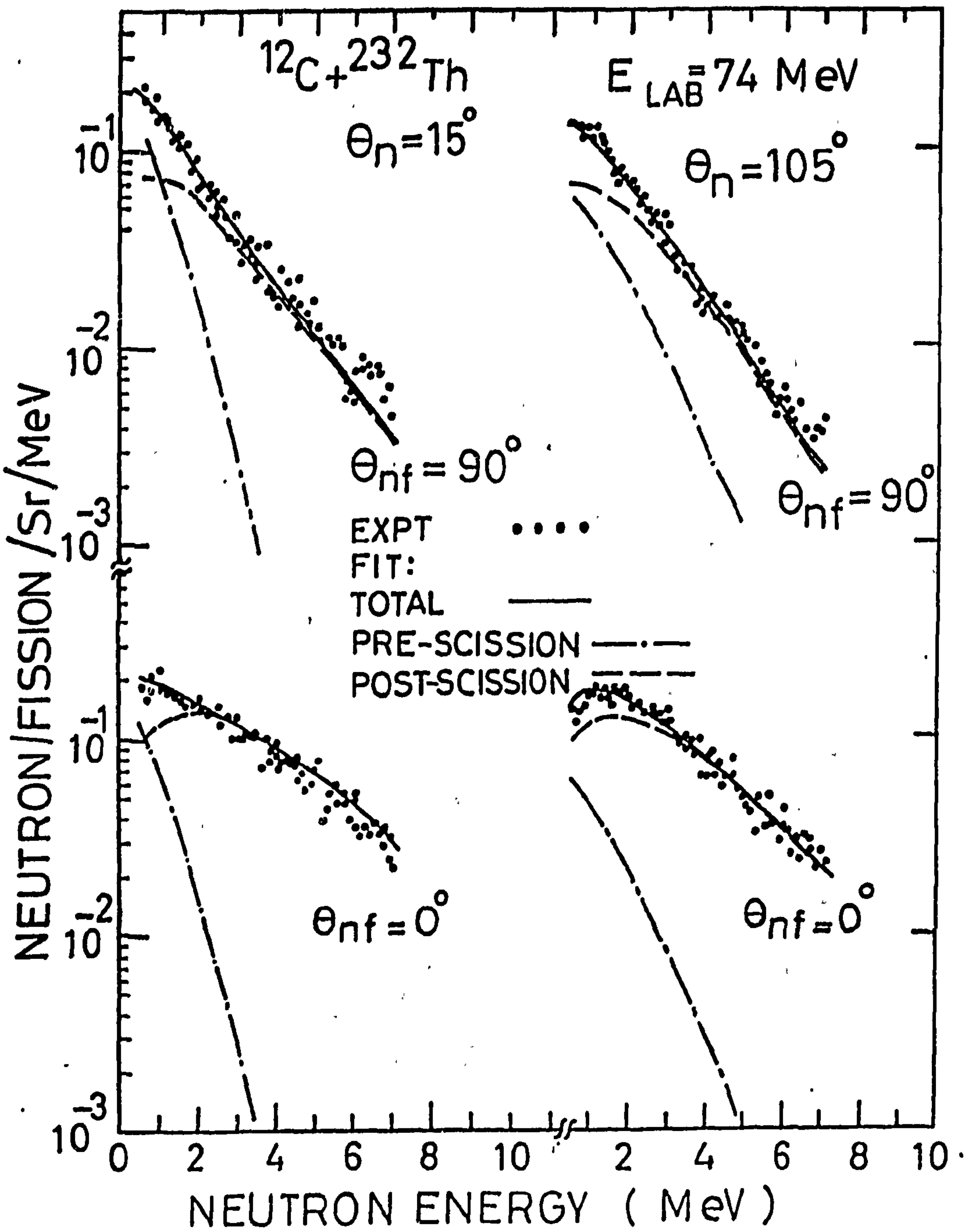

Fig. 1: Experimantally measured neutron lab energy spectrum for the two neutron detectors kept at 15° and 105° respectively with respect to the beam direction and the fits obtained from a least square fits to the $\theta_{nf}=0°$ and $\theta_{nf}=90°$ with respect to each of the two detectors under the assumption of isotropic emission in the rest frame of the moving compound nucleus and the two moving fragments.

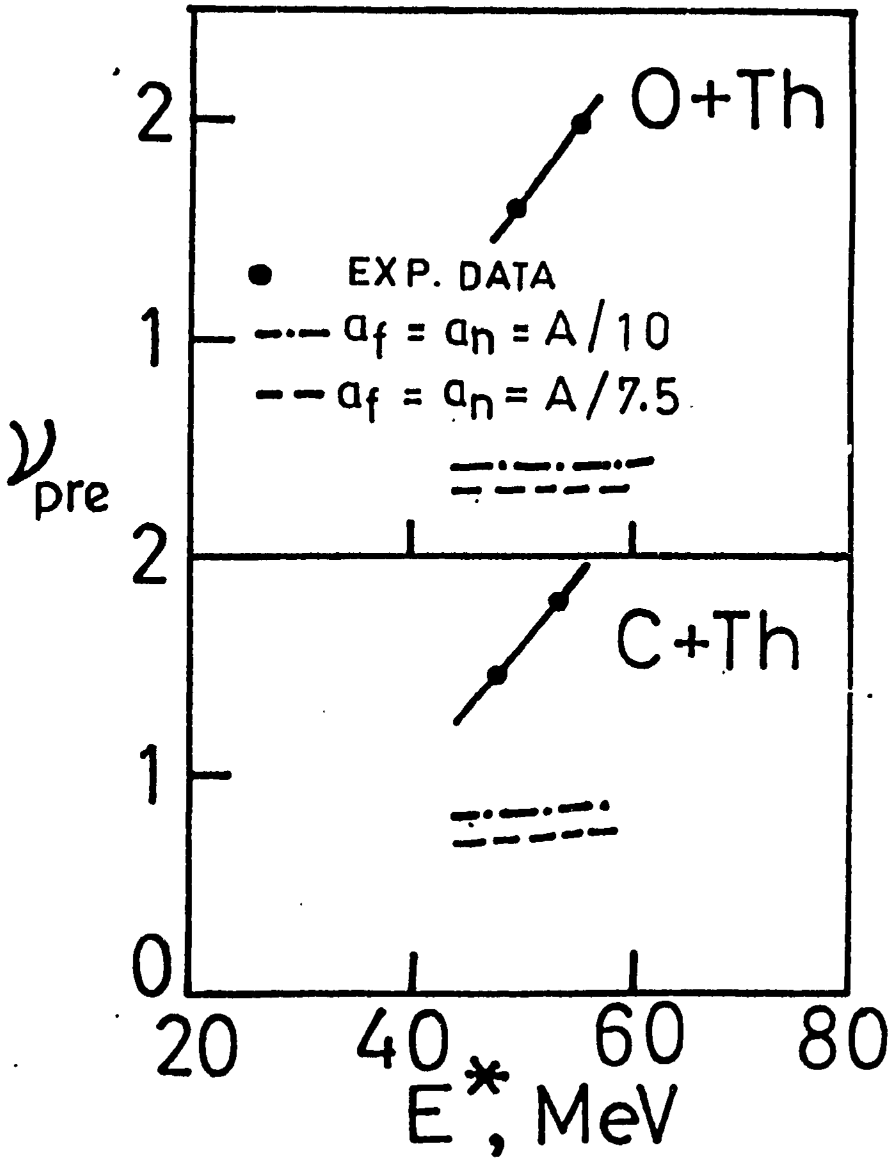

Fig. 2: Prefission neutron multipiicity deduced from fitting experimental neutron spectrum are plotted. The dashed line and dash-dotted line represent the statistical model calculations for two different level density parameters.

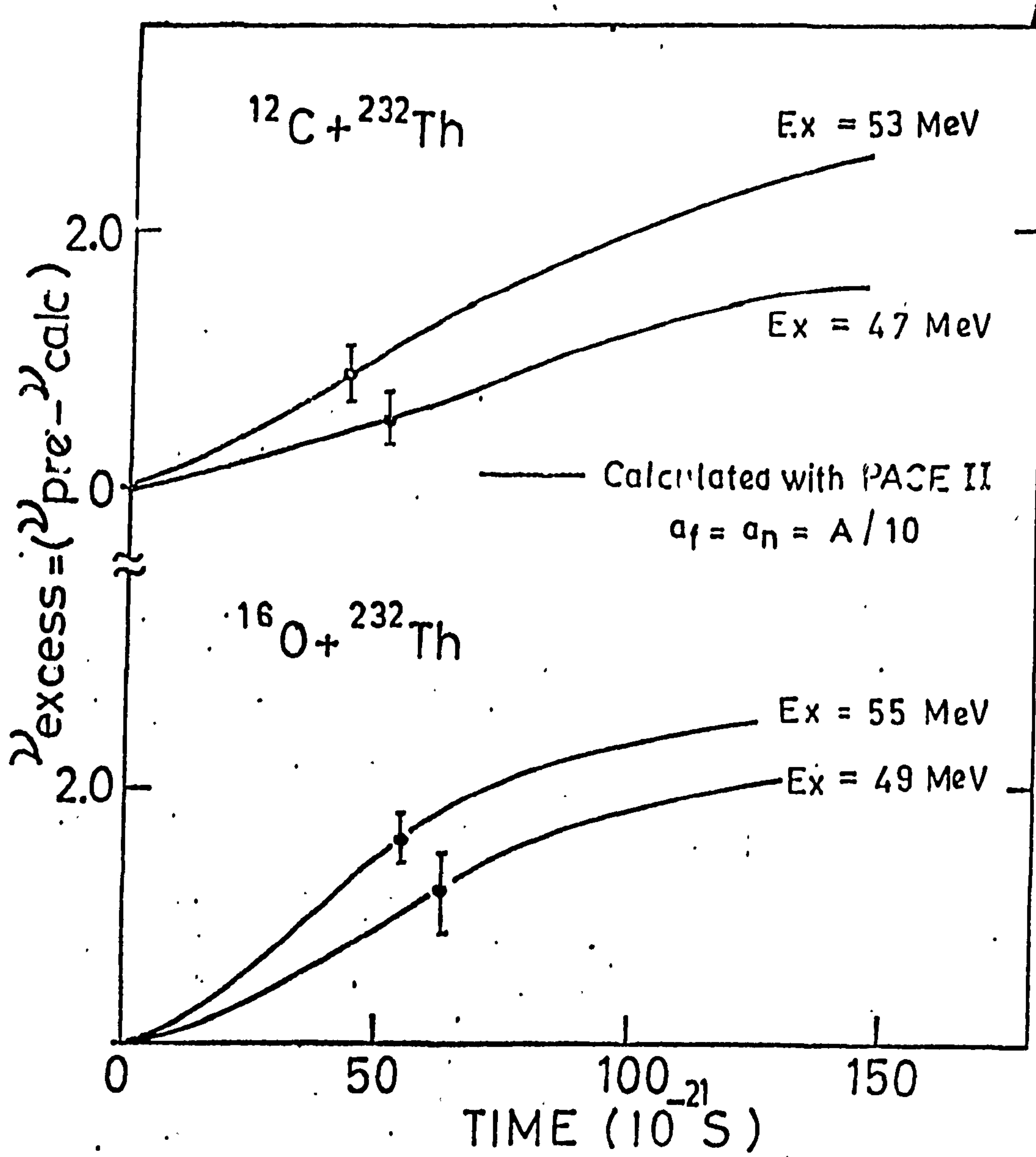

Fig. 3: The excess number of neutrons emitted are compared with the PACE II calculations after suppressing the fission for the time shown in the figure.

Further work is required to deduce heavy ion fusion time scales using "neutron clock" by reaching the same compound nucleus through different entrance channels for the cases in which the contribution from pre-equilibrium fission is absent or small.

B. Fission Fragment Angular Distributions

To look for such entrance-channel dependence we recently carried out[3] measurements of the fission fragment angular distributions in reactions of ^{10}B, ^{12}C, ^{16}O + ^{232}Th and ^{237}Np and ^{19}F + ^{237}Np. The results of these measurements are shown in Fig. 4, where it can be clearly seen that while the measured anisotropies in B- and C- induced fissions are in agreement with the prediction of the statistical model, they are anomalously large in the case of O- and F- induced fission. Observation of this entrance-channel dependence of fragment anisotropies provides experimental support for the occurance of "pre-equilibrium fission" in those cases where the dinuclear complex has nuclear temperatures comparable to fission barriers and is expected to equilibrate via mass-symmetric configurations.

In this paper, we further discuss the interpretation of the fission fragment angular distributions also taking into consideration the Kramer effect of nuclear dissipation on the fission process, namely the effect that the diffusion towards fission from the equilibrium configuration to saddle point occurs over a finite transient time, and the quasistationary value of the fission width is reduced over that predicted by the Bohr-Wheeler theory which is based on the considerations of phase space alone.

2 DYNAMICAL EFFECTS ON THE FRAGMENT ANGULAR DISTRIBUTIONS

In the transition state model based on phase-space consideration, the fragment anisotropy is determined by the mean square angular momentum $\langle I^2 \rangle$ imparted to the nucleus and a statistical quantity K_o^2 determined by the expected K-distribution of the transition state. The anisotropy is given by

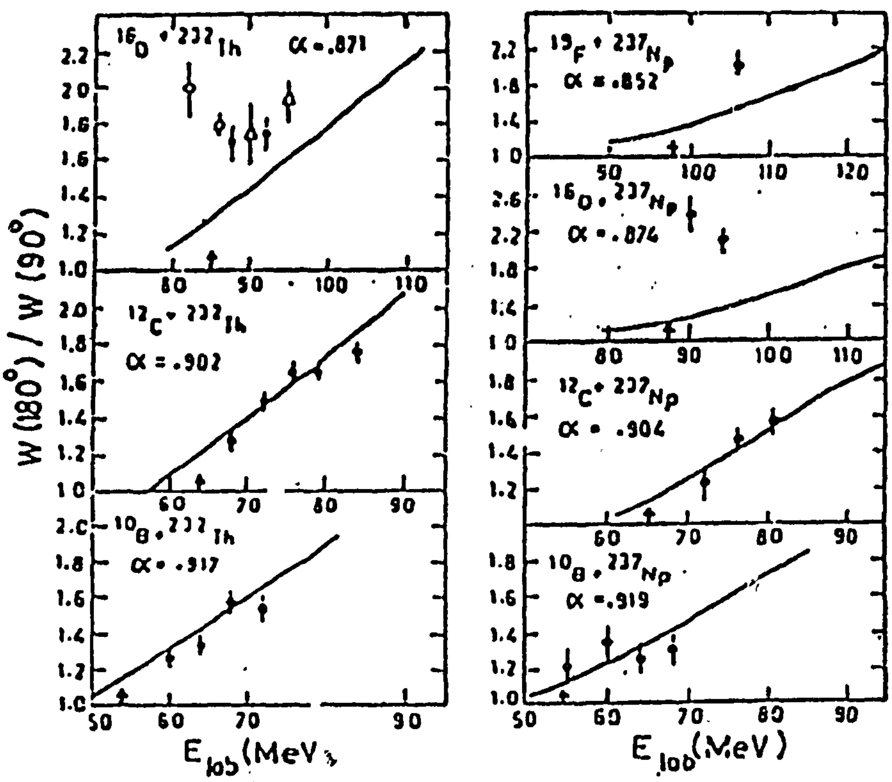

Fig. 4: Experimental and calculated fission fragment anisotropies as a function of bombarding energy for various target-projectile systems. The solid lines represent the calculations based on the statistical theory. The calculated fusion barriers for various systems are marked by arrows on the energy axis. (Figure taken from Ref. 3).

$$W(0^\circ)/W(90^\circ) \approx 1+\langle I^2\rangle/4K_o^2 \quad (1)$$

where $K_o^2 = J_{eff}T/\hbar^2$ (2)

It is now well established from pre-fission neutron emission data[8,9] that the fission width based on pure phase space considerations should be modified to include dynamical times from compound nucleus to the saddle point. The above statistical transition state model naturally does not include dynamical effects.

The theoretical framework for dynamical effects on fission probabilities has been developed by Grange, Weidenmuller and Coworkers[10-12] based on the pioneering work of Kramers[13] Treating nuclear fission as a diffusion process over the fission barrier, Kramers showed that the fission width is reduced with respect to the Bohr-Wheeler value by a factor (Kramer factor) given by

$$f_K = \left[(1+\gamma^2)^{1/2} - \gamma\right] \quad (3)$$

where $\gamma = \beta/2\omega_o$; β- coefficient of nuclear dissipation and ω_o is the harmonic oscillator frequency corresponding to the saddle point. It has been earlier shown[10-12] that the diffusion from the equilibrium configuration to the saddle point also takes over a finite transient time t_1, which can be defined as the time during which the fission width rises from 10% to 90% of its final quasi-stationary limit. Thus the disintegration constant λ_f for fission becomes a time dependent quantity eventually reaching the asymptotic value λ_f^∞. For mathematical simplicity, we assume λ_f of the same form as that for charging of a condenser:

$$\lambda_f(t) = \lambda_f^\infty \left[1 - e^{-t/t_1}\right] \quad (4)$$

where

$$\lambda_f^\infty = \frac{\Gamma_f^{B.W}}{\hbar}\left[(1+\gamma^2)^{1/2} - \gamma\right] \quad (5)$$

where $\Gamma_f^{B.W}$ is the Bohr-Wheeler value.

For over damped motion $\gamma>1$, the transient delay time t_1, has been approximately expressed[11,14] as

$$t_1 \approx \frac{\beta}{2\omega_1^2} \ln\left(\frac{10E_f}{T}\right) \quad (6)$$

where E_f = fission Barrier height

T = Nuclear temperature at saddle

ω_1 = is an oscillator frequency relating to the equilibrium configuration of the compound nucleus. For a given fission barrier height, the value of ω_1 will be inversely proportional to the distance from the minimum in the pocket describing compound nucleus configuration to the saddle point.

We discuss below the consequences of including this effect on the fission fragment angular distribution in heavy ion induced fission.

2.1 Change of Temperature due to Extra Preneutron Emission

The first important consequence of the dynamical effects as also pointed out recently by Rossner[15] is to delay the fission process, resulting in a much larger number of prefission neutrons than given by the statistical model based on phase space consideration alone. Thus the saddle point temperature T needs to be calculated taking into account additional neutrons expected to be emitted due to the fission delay t_1 and the reduction in the value of Γ_f. This will lead to a lower value of K_0^2 and hence a somewhat larger calculated anisotropy on the basis of Eq. (1). We have reanalysed[17] the experimental data of Ref. 3, by recalculating saddle point temperatures taking into account the number of prefission neutrons as measured by us[7] or as taken from other published data by suitable interpolation/ extrapolation. In order to find the maximum effect, we have additionally assumed that all additional prefission neutrons are emitted prior to saddle point. Figs. 5 and 6 shows the results of this reanalysis. Since our analysis is based on J_{eff} deduced from alpha induced fragment angular distribution data, appropriate correction for presaddle neutron emission must also be made for the case of alpha induced fission, as there is experimental evidence[18] that 2-3 neutrons are emitted in this case also prior to fission. The different theoretical curves in Figs. 5 and 6 refer to different assumed values of prefission neutrons in alpha induced fission case. Further details of this analysis can be seen in Ref. 17. While the prefis-

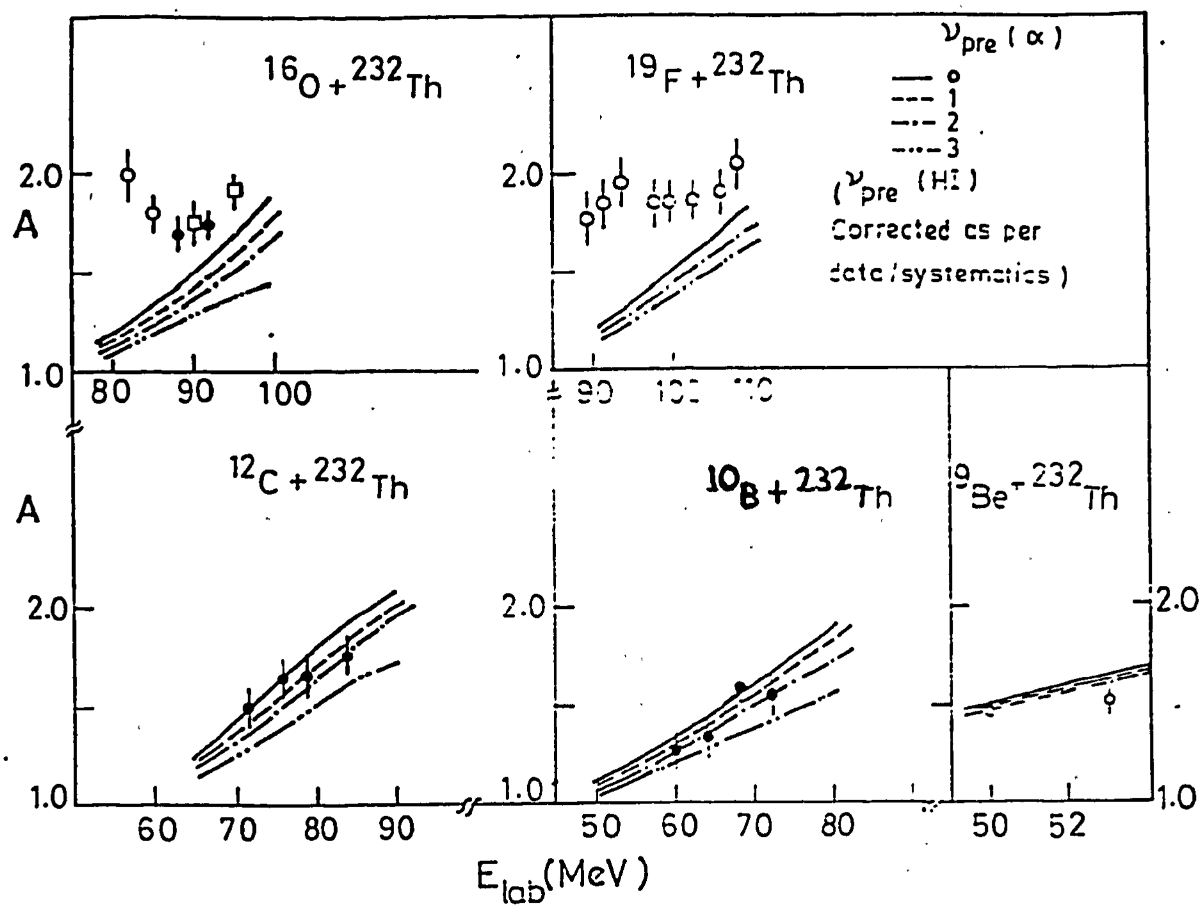

Fig. 5: The experimental and calculated anisotropies A for various target-projectile systems. The curves represent calculations which take into account additional pre-fission neutrons emitted in calculating temperature at the transition point. It is assumed that all additional prescision neutrons are emitted prior to saddle point. As the calculated anisotropies are based on J_{eff} deduced from alpha induced fission, prefission neutron correction is also applied in this case. Various curves correspond to different assumptions on prefission neutrons in alpha induced fission case.

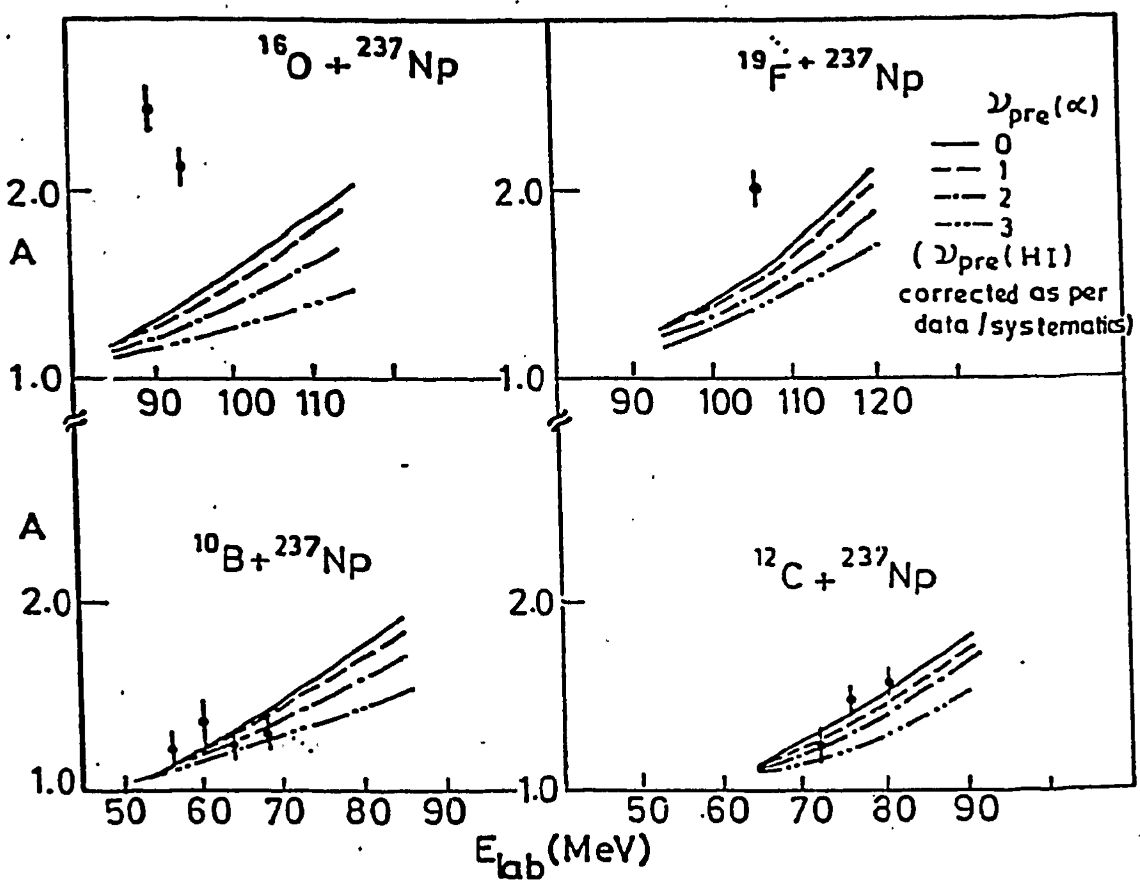

Fig. 6: Same as Fig. 5., but for different target-projectile combinations.

sion neutron correction slightly reduces the descrepancy with the predictions of transition state model it is clear from Figs. 5 and 6 that the observed anisotropies for O- and F- induced fission continue to be anomalously large and the angular anisotropy still shows a discontinuous behaviour with respect to entrance channel mass/charge asymmetry as can be seen from the Fig. 7. Recently Rossner et al[14] have reported analysis of fission fragment angular distributions for $^{208}Pb(^{16}O,f)$ with the transition state model including the nuclear temperature reduction caused by prefission neutron emission, and they find that this system shows anisotropies consistent with the model at low and near barrier energies. In their analysis Rossner et al[14] have used calculated values of J_{eff}, while in the present analysis we have used the values of J_{eff} deduced from alpha induced fission case. It is to be ascertained whether a different conclusion arrived at by Rossner et al is due to the difference in the systems investigated or due to the use of calculated J_{eff}.

2.2 Dynamical effects on fragment angular distribution theory

It was earlier suggested[17] that flexible rotor model which includes the dependence of the transition-state shape on orientation of the symmetry axis is more appropriate model for the description of the fragment angular distributions. In this model, the number of open channels at the saddle point being proportional to the density of intrinsic states at the transition state is given by

$$\rho(I,K) \propto \exp\left[E - {}_tE_{def} - {}_tE_{rot}\right]/T \quad (7)$$

where E is the total excitation energy for the compound nucleus, and ${}_tE_{def}$ and ${}_tE_{rot}$ are deformation and rotational energies at the transition state. ${}_tE_{rot}$ is given by

$${}_tE_{rot} = \frac{\hbar^2}{2J_\perp}(I^2 - K^2) + \frac{\hbar^2}{2J_\parallel}K^2 \quad (8)$$

The above expressions are based purely on the con-

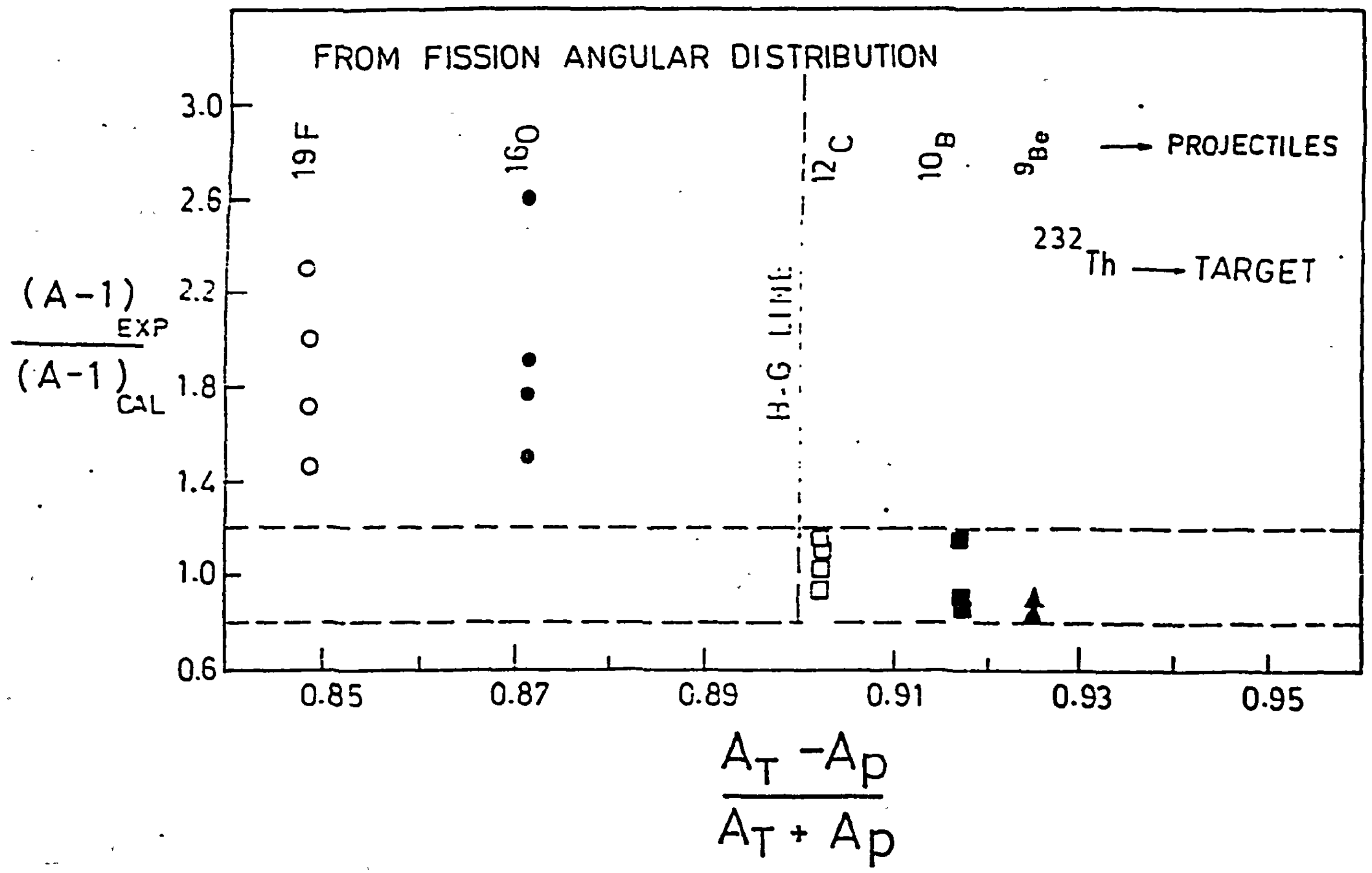

Fig.7: The ratio of experimental and calculated anisotropy $(A-1)_{exp}/(A-1)_{cal}$ has been plotted as a function entrance channel mass asymmetry of various projectile beams on ^{232}Th target

sideration of phase space at the transition state and does not take into account the dynamical effects, which in general, are expected to be dependent on I and K as well. It was earlier shown that the fission transition state shape for the fissioning nucleus of a given fissility and angular momentum varies somewhat with the value of K; the transition state shapes become more elongated as K increases. For a typical case of ^{225}Pa and I=40, for the extreme cases of K = 0 and K = 40 the nuclear elongation of the saddle configuration was found to vary from c ≈ 1.5 to 1.8, where c is a parameter describing nuclear alongation. For larger values of K this resulted in a slight decrease of $\ln P_f(I,K)$ versus K^2 from the straight line behaviour corresponding to the value of fission probability $P_f(K)$ given by $e^{-K^2/2K_0^2}$ expected purely on the consideration of available phase space at the saddle point. However, this effect was found to increase the calculated anisotropy by only 10-20% in most cases. Thus on the phase-space considerations alone the statistical flexible rotor model can not explain the anomalously large observed anisotropies.

Let us now discuss the consequences due to the inclusion of dynamical effects. We shall consider the correction factors due to dynamical effects in the fission probabilities for fissions through barriers of different K, over those expected from phase-space considerations alone. The time dependent fission width $\Gamma_f(t)$ which depends on the shape of the fission barrier will therefore, depend on the quantum numbers I and K characterizing the fission barrier. We can write

$$\Gamma_f(t,I,K) = \Gamma_f^{B.W}(I,K)\, f_{dyn}(t,I,K) \qquad (9)$$

$$= \Gamma_f^{B.W}(I,K)\, f_k(I,K)\left[1 - e^{-t/t_1(I,K)}\right] \qquad (10)$$

For the purpose of determining the behaviour of $f_k(I,K)$, we shall use the results of calculations of Ref. 19. These are shown in Fig. 8, where deformation energy curves and saddle point deformations are shown for I=K=0, and K=0, 20 and 40 for I=40 for the compound nucleus ^{225}Pa.

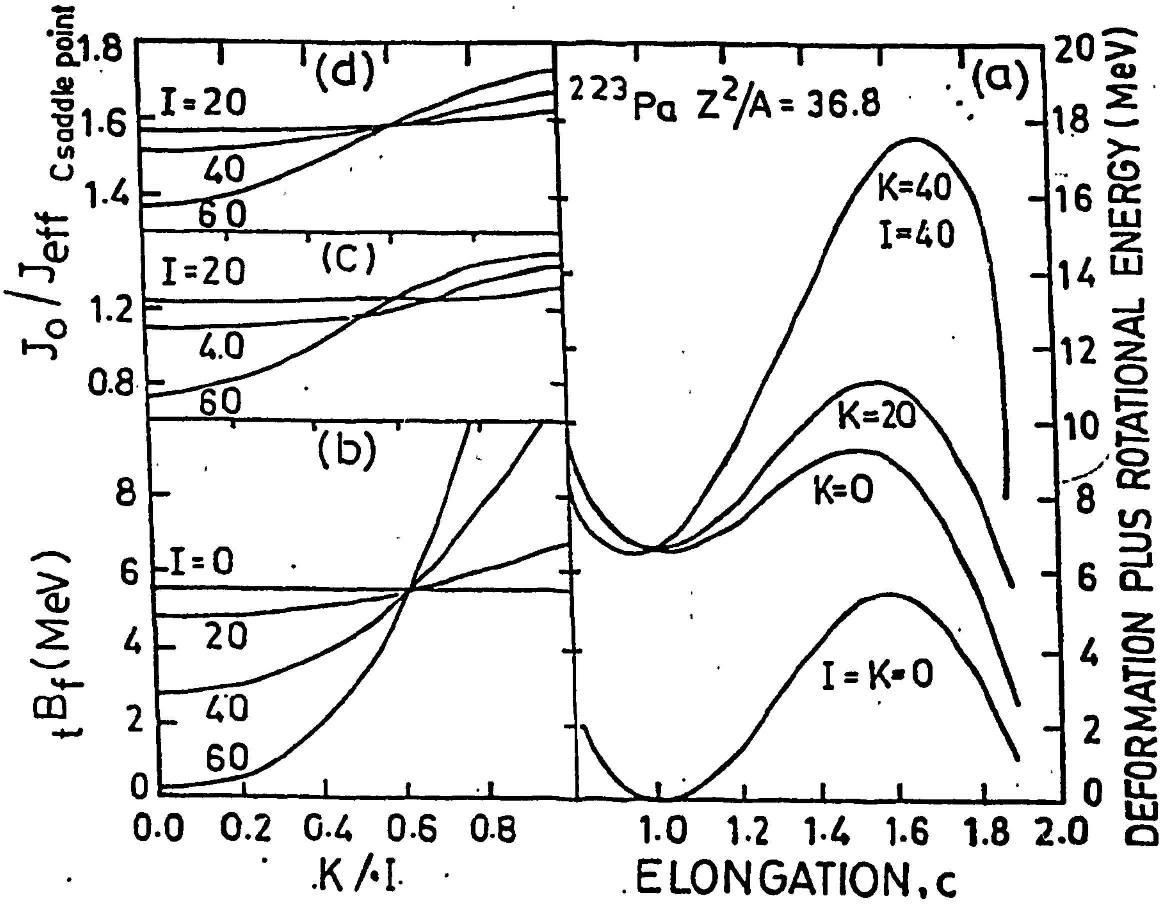

Fig. 8: Calculated deformation plus rotational energy as a function of the elongation parameter c, for fixed spin (I) but different orientation (or K). (b) – (d) Fission barrier B_f , effective moment of intertia J_{eff} (J_o refers to a rigid sphere); and elongation c of the saddle-point configuration as a function of K/I, for different spins I. (Figure taken from Ref. 19)

Effect on $\langle I^2 \rangle$

Considering K=0 case, we can quantitatively compare ω_o for the extreme cases of I=0 and 40 on the basis of curvatures at the fission barrier, taking inertial parameter to be independent of I. It is seen that $\omega_o(I=40) < \omega_o(I=0)$
Hence, Kramer factor will be smaller for I = 40, and this will reduce the fission probabilities due to dynamical effects by a larger fraction for I=40 than for I=0. Thus, for cases of $B_f > T$, where $\langle I^2 \rangle$ should be calculated including fission probabilities for each I, it is important to also include dynamical effects while calculating $\langle I^2 \rangle$ for fissioning nuclei. If the value of $\langle I^2 \rangle$ for fissioning nuclei is caluclated including dynamical effects on $\Gamma_f(I)$,

$$\langle I^2 \rangle = \frac{\int f_{dyn}(I)\, \Gamma_f^{B.W}(I)\, P_f(I)\, dI}{\int P_f(I)\, dI} \qquad (11)$$

While the Bohr-Wheeler expression favours fission for large I values, the dynamical effect favours fission for smaller I-values. While this effect is not expected to significantly change the value of $\langle I^2 \rangle$, the dynamical effects can only somewhat reduce the value of $\langle I^2 \rangle$, and hence this cannot be the basis of explanation of observed anomalous fragment anisotropies.

Effect on the width of K-distribution

On the basis of potential energy curves shown in Fig. 8 and taking inertial parameter to be independent of K, it is estimated that the values of ω_o increases by a factor of 2 in going from K = 0 to K = 40. From the studies of preneutron emission in fission, the nuclear dissipation coefficient has been estimated to be quite large, within the range of $\beta = 8 \times 10^{21} s^{-1}$ to $22 \times 10^{21} s^{-1}$. Since the typical values of ω_o are $1 \times 10^{21} s^{-1}$, the values of γ can range from 4 to 11. For value of γ = 4 to 11 (for K = 0) it is seen that $f_k(K=0)/f_k(K=40) \approx 0.5$. Similarly, we also estimate that $\omega_1(K=0)/\omega_1(K=40) \approx 0.55$. This implies that inclusion of dynamical effects have the consequence that the values of $\Gamma_f(K)$ will be larger by about a factor of 2 for the extreme case of $K = 40$ in relation to value for $K = 0$, as compared to that given by phase space considerations alone. Also quasi-stationary values for $K = 40$ are reached faster in time by about a factor of 2 than for $K = 0$. Considering that the K-distribution has a Gaussian shape, a change by a factor of two for extreme cases of $K = 40$ can only lead to a marginal change in the shape of the K-distribution for the quasistationary case, as compared to that given by phase space consideration. Moreover, this small change will be only in the directionof increasing the width of the K-distribution. In short, it is clear from the preceding discussion that in compound nucleus fission due to viscosity controlled dynamical effects, the K-distribution at the saddle point accessable to the fissioning nucleus can become only marginally different and wider with respect to Gaussian K-distribution expected from phase space consideration. The above dynamical effects thus do not provide a mechanism for a narrow K-distribution needed to explain anomalous fragment angular distributions. The explanation put forward earlier to explain the anomalous fragment angular distributions on the basis of 'pre-equilibrium fission' therefore continues to hold even if we take into consideration the dynamical effects. As shown below, the only consequence of dynamical effects can be to change our earlier estimates of the relaxation time.

3. DEDUCTION OF K-EQUILIBRATION TIME WITH INCLUSION OF DYNAMICAL EFFECTS

In the earlier work[8], we had deduced the non-compound nucleus fission probabilities P_{NCNF} versus $B_F(I,K)/T$ that were needed to fit the experimental data on the fragment anisotropies on the assumpiton that the fraction P_{NCNF} has a very narrow un-equilibrated K-distribution corresponding to $\sigma_K^2 = I^2\sigma_0^2$, while the equilibrium fission fraction $(1-P_{NCNF})$ has the standard Gaussian K-distribution at the saddle point with the variance given by K_0^2. While the evolution of K distribution is continuous, for simplicity it was assumed in the above analysis that fissions taking place upto a time less than τ are K-unequilibrated NCNF, while those taking place after time τ are K-equilibrated compound nucleus fissions. The fraction of NCNF taking place in time is given by

$$P_{NCNF} \approx \left[1 - \exp(-\lambda_f \tau)\right] \qquad (12)$$

It is therefore expected that the product of λ_f and τ will determine the observed values of P_{NCNF}. In the previous calculations, when λ_f was calculated on Bohr-Wheeler phase space considerations a value of $\tau \sim 8\times10^{-21}$s was found to fit the data, as shown in Fig. 9. Now if the value of Γ_f is calculated taking into consideration dynamical effects, the value of λ_f will be reduced depending on the value of dissipation parameter and moreover the value of may not reach its full quasi-equilibrium value in this time and hence λ_f will be smaller than that calculated by Bohr-Wheeler formula. Consequently withthe dynamical effects included, the value of τ required to fit the observed values of P_{NCNF} is expected to be larger than the value of 8×10^{-21}s estimated earlier.

For the value of γ in the range 4 to 11 the values of λ_f can get reduced by a factor of 0.12 to .05 over the Bohr-Wheeler estimate. Thus if the dynamical effects are also taken into consideration, the value of τ can be an order of magnitude larger than that estimated in Ref. 5, and a reasonable estimate of τ would be about few times 10^{-20}s.

SUMMARY AND CONCLUSIONS

Measurements of prescission neutron multiplicities have suggested that fission is a slow process and dynamical times of few times 10^{-20}s may be involved in the dynamics of nucleus-nucleus collision from contact configuration to compound nucleus formation to fission. The prescission time scales determined by the statistical model calculations for the $^{12}C+^{232}Th$ and $^{16}O+^{232}Th$ systems are found to be 4.5×10^{-20}s and 5.5×10^{-20}s for assumed level density parameters $a_n=a_f=A/10$. Further

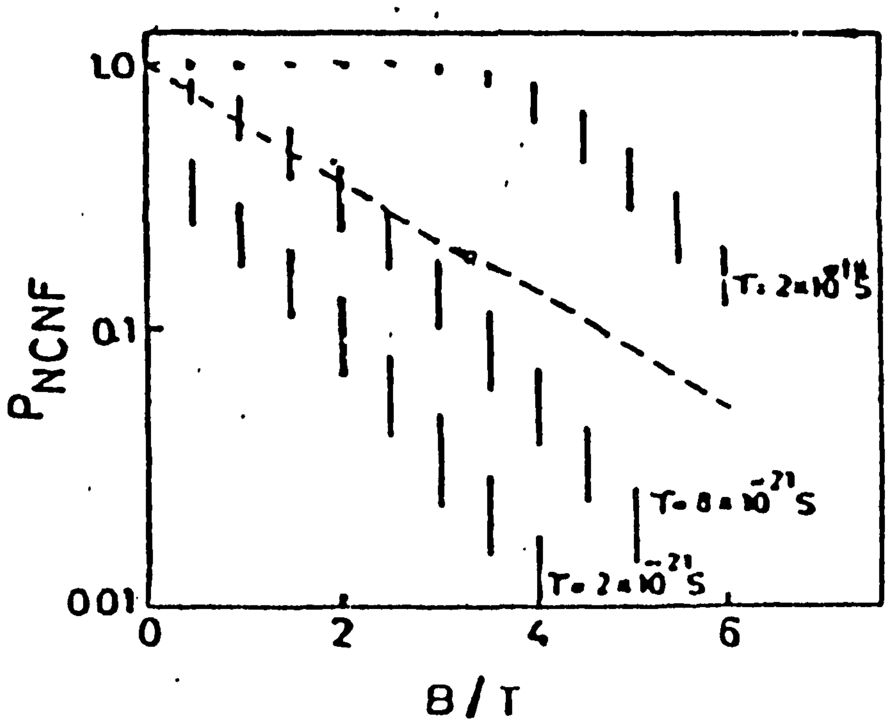

Fig.9: Plots of P_{NCNF} versus $B_f(I,K=0)/T$ for different values. The dashed line represent $P_{NCNF}(I)=exp(-0.5B_f(I,K=0)]$. (Figure taken from Ref. 5)

studies of this type where pre-equilibrium fissions are absent or small in every case, will be more useful to isolate differences in the dynamical times from contact configuration to fusion for various entrance channel mass-assymetries.

The larger anisotropies observed in our measurements for the cases of O- and F- induced fissions continue to remain anomalous with respect to transition state model predictions, even after incorporating cooling at the saddle point due to the emission of prescission neutrons. It is also shown that the nuclear dissipation governed diffusion over the fission barrier does not lead to a reduction in the width of the K-distribution and the explanation put forward earlier to explain the anomalous fragment angular distributions on the basis of "pre-equilibrium fission" continues to hold. The only consequence of dynamical effects is to increase our earlier estimate of the relaxation time in the K-degree of freedom to few times 10^{-20}s.

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