Chapter 4

Fission fragment mass distribution -
Analysis and results

4.1 Introduction

Phenomenon of nuclear fission [1] has been a topic of extensive theoretical and experimental study from the very beginning of its discovery. There are several reasons for this. It represents the most drastic re-arrangement of nuclear matter known today with both statistical and dynamical features coming into play. Fission is a rich laboratory within which one can explore the delicate interplay between macroscopic aspects of bulk nuclear matter and quantal effects of a finite number of Fermions.

Considerable effort has been made over the years in understanding various aspects of fission process. Fission fragment angular distribution measurement is a very effective probe to understand the time evolution of the composite system formed during the interaction as it relaxes in energy, mass, angular momentum and shape degrees of freedom. The standard statistical model [2–4] based on the assumptions of a well defined saddle point in the evolution of fissioning nucleus was able to explain most of the experimental data. However, heavy-ion fission experiments using actinide targets [5, 6] have shown anomalously large angular anisotropies over the predictions of the statistical model (saddle point model).

Nowadays many leading laboratories in the world are actively engaged in the production of super-heavy elements and super-heavy ERs. A major hurdle in the accomplishment of this effort is the presence of the above mentioned non-compound nucleus events, especially quasifission, which lead to the re-separation of the composite system after the capture process. In terms of the reaction time scale, quasifission
bridges the gap between deep inelastic collisions (DIC) and CN formation. Deep inelastic reactions [7] represent the energy relaxation mode, exhibiting a wide spectrum of kinetic energy losses. The width of the mass peaks increases with kinetic energy loss, while the centroid of these peaks shows a remarkable stability. On the other hand CN formation is characterized by full equilibration in all degrees of freedom. Quasifission is a fission like process that precedes the formation of a compact mono-nuclear system and is characterized by a total energy relaxation and a division of total mass between the two reaction partners ranging from initial entrance channel mass asymmetry and all the way to symmetry. This process dominates at lower excitation energies, just above the fusion threshold, where evaporation residue formation also maximizes, and, hence compete strongly with ER formation.

The dynamical models proposed in early eighties predicted the onset of quasifission process for heavier systems, when the product $Z_P Z_T > 1600$ (where $Z_P$ and $Z_T$ are the atomic charges of the projectile and target, respectively) [8–10]. The observation of anomalous angular anisotropies of fission fragments [11, 12] in reactions involving actinide targets over the predictions of transition state models (TSM) [2] necessitated the study of the dependence of quasifission on various entrance channel parameters. The first experimental signature of nuclear orientation and deformation on quasifission was reported by Hinde et al. [13, 14] in $^{16}$O + $^{238}$U reaction, where the measured fragment angular anisotropies were anomalous over the predictions of statistical models, though the charge product $Z_P Z_T$ (736 in this case) was much less than 1600. For explaining the data, orientation dependent quasifission process was hypothesized, according to which, if the reaction partners are deformed, at near barrier energies, tip to tip collisions lead to an enhanced probability for quasifission than collisions with the flattened sides. Similar experimental results were reported by various groups in reactions involving actinide targets [15, 16], supporting the effect of deformation and orientation. The experimental signatures of quasifission include a strong hindrance to ER formation [17, 18], anomalous fission fragment angular anisotropies, strong fragment mass angle correlation, and broadened mass distributions.

In this context, a systematic study of fission fragments is necessary to understand the reaction mechanism in general and quasifission in particular. The measurement of mass, energy and angular momentum of the fission fragments provides important information about the dynamic aspects of fusion-fission process. Large area, position sensitive gas detectors are the ideal detectors for measuring the mass, energy and angular distributions of the fragments produced in heavy ion reactions. However, the fragments
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from CN events must be very effectively separated from other non-compound nucleus channels (products of elastic, inelastic reactions and transfer fission). In the laboratory this can be very effectively achieved by keeping the detectors at proper folding angles. Folding angle between the fragments is the experimental signature of linear momentum transferred in the reaction process.

4.2 Folding angle

In heavy ion induced fission reactions, the emitted fragments will be at 180° separation with respect to each other in the centre-of-mass frame. However, this may not be the case in the laboratory frame. The emission angle between the two fragments in the laboratory frame is usually referred to as the folding angle, which depends on the velocity of the fragments and also on the recoil velocity of the fissioning nucleus. Hence, in reactions, fission following CN formation can be effectively separated from the fission following direct processes on the basis of different values of linear momentum transferred to the fissioning nucleus. Compound nucleus formation involves complete linear momentum transfer in the beam direction from the projectile to the fissioning nucleus. In contrast, the direct reactions produce the fissionable nuclei with only small linear momentum component along the beam axis. As a consequence, in CN formation the two fragments will be separated at a characteristic angle less than 180° with respect to one another in the laboratory system and will be co-planar with the beam axis. Fission fragments following direct processes will be separated along a nearly collinear axis ($\approx 180^\circ$). Hence the fragments from the two competing process can be very effectively separated by knowing the proper folding angle of the complimentary fragments.

The kinematics of symmetric fission from the CN is shown in Fig. 4.1.

$\mathbf{v}_1$ and $\mathbf{v}_2$ are the velocities of the fragments (FF1 and FF2 respectively) in the laboratory frame and $\mathbf{v}_{cm}$ is the centre-of-mass velocity. $\mathbf{V}_{rec}$ is the recoil velocity of the CN. $\theta_1$ and $\theta_2$ are the fragment angles in laboratory frame with respect to the beam axis. By definition, folding angle is given by

$$\theta_{fold} = \theta_1 + \theta_2$$  \hspace{1cm} (4.1)

If we know $\theta_1$, $\theta_2$ can be calculated and hence the folding angle. From geometry,

$$\tan \theta_2 = -\tan(\pi - \theta_2) = -\frac{SR}{SO} = -\frac{SR}{QS - QO}$$  \hspace{1cm} (4.2)
Substituting for SR, QS and QO from Fig. 4.1,

$$\tan \theta_2 = -\frac{v_1 \sin \theta_1}{v_1 \cos \theta_1 - 2V_{rec}}$$

(4.3)

The folding angle can be expressed as,

$$\theta_{fold} = \theta_1 + \tan^{-1} \left[ \frac{v_1 \sin \theta_1}{2V_{rec} - v_1 \cos \theta_1} \right]$$

(4.4)

Hence From Eq. 4.1, it is clear that total folding angle can be calculated if $\vec{v}_1$ and $V_{rec}$ are known. The recoil velocity of the CN can be calculated from the recoil energy.

$$V_{rec} = \sqrt{\frac{2E_{rec}}{A_{CN}}}$$

(4.5)

where $E_{rec}$ is the CN recoil energy and $A_{CN}$ is the CN mass. The CN recoil energy in turn can be obtained from the incident momentum $P_{inc}$.

$$E_{rec} = \frac{P_{inc}^2}{2A_{CN}}$$

(4.6)
From the above figure it is obvious that $\vec{v}_{cm} = \vec{v}_1 - \vec{V}_{rec}$. Hence,

$$v_{cm}^2 = v_1^2 + V_{rec}^2 - 2v_1V_{rec}\cos\theta_1$$  \hspace{1cm} (4.7)

Re-arranging the equation in quadratic form and solving we get two solutions

$$v_1 = \frac{1}{2} \left[ 2V_{rec}\cos\theta_1 \pm \sqrt{4V_{rec}^2\cos^2\theta_1 + 4(v_{cm}^2 - V_{rec}^2)} \right]$$  \hspace{1cm} (4.8)

We consider the positive solution only because with the increase in $\theta_1$, $v_1$ becomes negative for the negative root, which is physically impossible.

$$v_1 = V_{rec}\cos\theta_1 + \sqrt{V_{rec}^2\cos^2\theta_1 + v_{cm}^2 - V_{rec}^2}$$  \hspace{1cm} (4.9)

The centre-of-mass velocity $v_{cm}$ of the fragment is obtained using Viola systematics [19]. The fragment kinetic energy is the result of Coulomb repulsion of two spheres in contact. Viola systematics assume symmetric fission for calculating the average kinetic energy and is given by,

$$<E_K> = \left[ 0.01189 \frac{Z^2}{A_{CN}^{1/3}} + 7.3(\pm1.5) \right] \text{MeV}$$  \hspace{1cm} (4.10)

Experimental folding angle distribution is observed to be Gaussian in shape for CN fission. If there is any contribution from incomplete fusion followed by fission such as transfer-fission, these events can be seen as a hump on both sides of the full linear momentum transfer events. At higher energies, this incomplete fusion component appears at larger folding angles whereas at sub-barrier energies they correspond to smaller folding angles as compared to the complete fusion events. Fig. 4.2 shows $\theta_1$ versus folding angle plot for the two reactions $^{16}\text{O} + ^{194}\text{Pt}$ and $^{24}\text{Mg} + ^{186}\text{W}$ calculated using the above mentioned method.

## 4.3 Fission fragment mass distribution analysis

Fission fragments produced in the two reactions were collected using two large area MWPCs mounted on the two arms of the scattering chamber. These detectors were mounted at the folding angles. Details of the experimental setup used are discussed in chapter 3. These detectors provide fast position (XL, XR, YU and YD) and timing (anode) signals. The position signals (X and Y), energy loss of the fragments and the time-of-flight of the fragments were recorded event by event.
4.3.1 Offline detector calibration

The gas detectors were calibrated before the experiment using a fission source ($^{252}$Cf) of known strength. A mask with holes of 1 mm diameter separated by 5 mm in XY plane, was placed in between the source and the detector and fragments were collected. The data was collected in singles mode. The detectors were operated at a gas pressure of about 4 Torr during this calibration run. The anode voltage was +550V and cathode voltage was -200V. The position signals were recorded and digitized using Philips 16 channel TDC, using anode signal as the start signal. The position resolution of the detectors were better than 1.5 mm. Fig. 4.3 shows X-position spectra of one of the detectors with the mask in between the detector and the source. Angular calibration was also obtained in a similar fashion. For this, one row of holes (both in X and Y) was masked using some tape and data were collected.

The position calibrations of these detectors were further performed using the known positions of the edges of the illuminated areas of the detectors during the experiment.
4.3.2 Fragment mass ratio distributions

The position signals XL and XR were used to obtain the X-position of the particle incident on the detector. This position signal was gated using the X-sum spectra (XL + XR) for cleaning the spectra from the background. Similarly YU and YD signals were used for constructing the Y-position of the particle. Y-sum spectra was used for cleaning the spectrum. The calibrated positions (X and Y) were converted to polar angles $\theta$ and $\phi$. Fig. 4.4 shows the correlation between $\theta_1$ and $\theta_2$ of the fragments collected at the two MPWCs. The fission fragments from CN reaction were exclusively determined from the distributions of polar ($\theta$) and azimuthal ($\phi$) angles. This $\theta_1$ and $\theta_2$ correlation was generally used to separate the non-compound nucleus fission (such as transfer fission) from CN fission as the correlation carries the signature of linear momentum transferred.
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during the reaction process. This distribution of fission fragments in \((\theta, \phi)\) plane is shown in Fig. 4.5. It is obvious that in \(^{16}\text{O} + ^{194}\text{Pt}\) reaction, transfer-fission events are absent as the Fig. 4.5 has only a single contour which correspond to CN fission. The projection of this distribution on \(\theta\)-axis will be peaked at total folding angle \((\theta_{fold})\) and that on the \(\phi\)-axis will be centered at 180°.

![Image](image.png)

*Figure 4.4:* Correlation between \(\theta_1\) and \(\theta_2\) (in degrees) of the two complimentary fragments in \(^{16}\text{O} + ^{194}\text{Pt}\) reaction at 101.8 MeV beam energy.

Fragment velocities in the laboratory frame \((\vec{v}_1\) and \(\vec{v}_2)\) were re-constructed using TOF, \(\theta\) and \(\phi\) informations. The energy loss correction of the fragments in the target was performed assuming that the interaction takes place at the midpoint of the target. Since the targets used were very thin, the energy loss correction would not make any appreciable change (the changes were less than 2%) in the mass ratio distributions of the fragments. Similarly the effects produced by the energy loss of the fragments at the entrance foil was also negligible. The centre-of-mass velocities \(v_{1cm}\) and \(v_{2cm}\) of the fragments were then obtained from laboratory velocities using kinematic transformations. The mass distributions and mass ratio distributions of the fragments were obtained event by event by using the expressions given below ref. [20].

Fig. 4.6 shows the kinematic representation of fusion-fission process. The projectile (P) fuses with the target (T), leading to the formation of the CN, which undergoes fission. \(\theta_1\) and \(\theta_2\) are the emission angles of the fragments of masses \(m_1\) and \(m_2\) with
Figure 4.5: Distribution of complimentary fission fragments in (θ, φ) plane at 101.8 MeV beam energy.

respect to the beam axis. Using the conservation of linear momentum,

\[ p_1 \cos \theta_1 + p_2 \cos \theta_2 = M_{CN} V_{CN} \]  \hspace{1cm} (4.11)

\[ p_1 \sin \theta_1 = p_2 \sin \theta_2 \]  \hspace{1cm} (4.12)

which leads to,

\[ p_1 = \frac{M_{CN} V_{CN}}{\cos \theta_1 + \sin \theta_1 \cot \theta_2} \]  \hspace{1cm} (4.13)

and

\[ p_2 = \frac{p_1 \sin \theta_1}{\sin \theta_2} \]  \hspace{1cm} (4.14)

where \( p_1 \) and \( p_2 \) are the linear momenta of the two fragments of masses \( m_1 \) and \( m_2 \) respectively. \( V_{CN} \) is the velocity of the CN of mass \( M_{CN} \). The time of flight difference between the two fragments entering the two detectors can be expressed as,
Figure 4.6: Diagram representing the fusion-fission kinematics.

\[ t_1 - t_2 = \frac{d_1}{v_1} - \frac{d_2}{v_2} = \frac{d_1 m_1}{p_1} - \frac{d_2 M_{CN}}{p_2} \]  \hspace{1cm} (4.15)

Re-arranging,

\[ m_1 = \frac{(t_1 - t_2) + \delta t_0 + M_{CN} \frac{d_2}{p_2}}{\frac{d_1}{p_1} + \frac{d_2}{p_2}} \]  \hspace{1cm} (4.16)

where \( d_1 \) and \( d_2 \) are the flight paths of the fragments. \( \delta t_0 \) is the electronic time delay between the two timing signals. Also from the linear momentum conservation,

\[ m_1 v_{1cm} = m_2 v_{2cm} \]  \hspace{1cm} (4.17)

The mass ratio is given by,

\[ M_R = \frac{m_2}{m_1 + m_2} = \frac{v_{1cm}}{v_{1cm} + v_{2cm}} \]  \hspace{1cm} (4.18)

During the analysis, the time difference method was used for obtaining the mass ratio distributions of the complimentary fragments. In the case of \( {}^{16}\text{O} + {}^{194}\text{Pt} \) reaction, we have used TOF method for getting the mass ratio distributions for
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some energies. It was verified that both TOF and time difference methods were giving the same results. Hence, the latter method was adopted for the analysis, for consistency. The basic assumption in time difference method is the presence of only full momentum-transfer fission events, and cannot be adopted for reactions where incomplete momentum-transfer events such as transfer induced fission are present. But in the present measurements the targets used were not fissile. Thus the probability for transfer induced fission was absent.

Fig. 4.7 shows the two-dimensional plot of anode signal versus cathode signal of the MWPC mounted in the forward direction in $^{16}$O + $^{194}$Pt reaction at 101.8 MeV beam energy. Fission events are shown inside the cut. Fig. 4.8 shows the time correlation between the complimentary fission fragments collected in the two MWPCs. Fig. 4.9 shows the 2D plot of X-position versus Y-position of one of the detectors at 101.8 MeV beam energy.

![Figure 4.7](image)

**Figure 4.7:** Two dimensional plot between the energy loss (cathode) and TOF (anode) signals.

The delay $\delta t_0$ in ref. [20], was corrected for each energy during the analysis for $^{16}$O + $^{194}$Pt run assuming symmetric mass division and Viola systematics [19]. However in the case of $^{24}$Mg + $^{186}$W reaction, the $^{16}$O + $^{197}$Au reaction, which is expected to undergo fission through pure CN formation, was used as the calibration system for measuring the electronic time delay involved in the measurements. In this calibration run, both gas detectors were kept at 90° in centre-of-mass frame and the fragments were collected at 90 MeV beam energy. The electronic time delay was then obtained
through an iterative method, by imposing the condition that mass ratio distribution is reflection symmetric about 0.5 at $\theta_{cm} = 90^\circ$, a condition true for all reactions. Fig. 4.10 shows the mass ratio distribution for $^{16}$O + $^{197}$Au reaction with different $\delta t_0$ values used in the iteration. It can be noticed that at $\delta t_0 = 51.5$ ns, the distribution is symmetric with the mass ratio centered at 0.5.

### 4.3.3 Mass-angle correlations

It is known that quasifission is intermediate between deep in-elastic processes and fission following CN formation. Deep in-elastic processes preserve the mass asymmetry of the ingoing channel on the average and has focused angular distributions. On the contrary, fission following CN formation has nearly symmetric mass division and forward-backward symmetric angular distributions. When quasifission is the dominant reaction mode it can be easily recognized experimentally. However, when it occurs as a less prominent component, side by side with CN fission process and deep in-elastic process, it is rather a serious problem to handle. It is experimentally observed that as the projectile gets heavier and heavier, less cross section appears in the capture channel and more in the deep in-elastic channel. Capture processes are dominated by quasifission, which emerges as the mass-drift mode in heavy ion reactions, exploring the entire mass range between contact and full symmetry. Depending on Z-value and centre-of-mass energy, the evolution of the systems towards symmetry finds itself
in more or less severe competition with a tendency of re-separation. As a result, mass distributions vary from a nice peak centered at symmetry through essentially flat distributions to tails extending inwards from the target-projectile-like peaks and leaving a broad minimum at symmetry [21–23]. A strong correlation of fragment mass with emission angle and broadened mass distributions are hence treated as the experimental signatures of quasifission.

The mass ratio plotted against centre-of-mass angle of the fragments from the reaction $^{16}\text{O} + ^{194}\text{Pt}$ at $E_{\text{c.m.}}$ values 94.0 MeV (well above the Coulomb barrier, $V_B = 76.3 \text{MeV}$) and 74.6 MeV (below $V_B$) are shown in Fig. 4.11(a) and Fig. 4.11(b) respectively. Fig. 4.11(c) and Fig. 4.11(d) are the similar plots for $^{24}\text{Mg} + ^{186}\text{W}$ reaction, for $E_{\text{c.m.}} = 110.6 \text{MeV}$ and 100.0 MeV, respectively. No evidence of mass angle correlation has been observed for either of the systems, within the angular and energy ranges studied. Since the finite geometry of our detection system limits the most forward and most backward events, a software cut ($120^\circ$ to $130^\circ$ for $^{16}\text{O} + ^{194}\text{Pt}$ reaction in Fig. 4.11(a) and (b) and $125^\circ$ to $135^\circ$ for $^{24}\text{Mg} + ^{186}\text{W}$ reaction, in (c) and (d) ) was made in the mass density plots as shown in figures and only those events which were falling inside the window were taken for obtaining the mass ratio distributions. This was very important to avoid any biasing of the data coming from the geometrical limitations of the experimental setup.
Figure 4.10: Calibration procedure for determining $\delta t_0$. Iteration method was followed to fix $\delta t_0$ by imposing the condition $M_R = 0.5$ when both detectors were placed at 90° in centre-of-mass frame. (A) $\delta t_0 = 41.5$ ns and $M_R = 0.44$, (B) $\delta t_0 = 51.5$ ns and $M_R = 0.50$, (C) $\delta t_0 = 67.5$ ns and $M_R = 0.61$ and (D) $\delta t_0 = 61.5$ ns and $M_R = 0.56$

4.3.4 Mass ratio distributions

From the mass density plots, experimental mass ratio distributions can be obtained by projecting the distribution on to the X-axis. The experimental mass ratio distributions for $^{16}$O + $^{194}$Pt and $^{24}$Mg + $^{186}$W reactions at different beam energies are shown in Fig. 4.12 and Fig. 4.13, respectively. Since shell effects are expected to be washed out at higher excitation energies, fragment mass ratio distributions should be symmetric, with the fission like fragments centered at $M_R = 0.5$. The width of this distribution increases smoothly with temperature for an equilibrated CN, and a sudden change could be a signature of departure from equilibration. The experimental mass ratio distributions can easily be represented by a Gaussian function with the standard deviation ($\sigma_m$) representing the width of the mass ratio distribution. Table 4.1 and Table 4.2 show the width of the mass ratio distribution ($\sigma_m$) and error involved, at different laboratory beam energies for $^{16}$O + $^{194}$Pt and $^{24}$Mg + $^{186}$W, respectively.
Figure 4.11: (a) and (b) are the mass ratio versus centre-of-mass angle plots for the reaction $^{16}\text{O} + ^{194}\text{Pt}$ at $E_{cm} = 96.0$ MeV and 74.6 MeV, respectively. (c) and (d) are similar plots for the reaction $^{24}\text{Mg} + ^{186}\text{W}$ at $E_{cm} = 110.6$ MeV and 100.0 MeV, respectively.

Table 4.1: Width of the mass ratio distribution ($\sigma_m$) and error involved at different laboratory beam energies for $^{16}\text{O} + ^{194}\text{Pt}$ reaction.

<table>
<thead>
<tr>
<th>$E_{lab}$ (MeV)</th>
<th>$\sigma_M$</th>
<th>$\sigma_M$ (error)</th>
</tr>
</thead>
<tbody>
<tr>
<td>101.8</td>
<td>$6.13 \times 10^{-2}$</td>
<td>$2.84 \times 10^{-4}$</td>
</tr>
<tr>
<td>97.8</td>
<td>$5.55 \times 10^{-2}$</td>
<td>$1.87 \times 10^{-4}$</td>
</tr>
<tr>
<td>93.8</td>
<td>$5.48 \times 10^{-2}$</td>
<td>$1.91 \times 10^{-4}$</td>
</tr>
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<td>89.8</td>
<td>$5.21 \times 10^{-2}$</td>
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<td>85.8</td>
<td>$5.29 \times 10^{-2}$</td>
<td>$3.39 \times 10^{-4}$</td>
</tr>
<tr>
<td>82.8</td>
<td>$5.21 \times 10^{-2}$</td>
<td>$1.95 \times 10^{-4}$</td>
</tr>
<tr>
<td>80.8</td>
<td>$5.04 \times 10^{-2}$</td>
<td>$2.98 \times 10^{-4}$</td>
</tr>
<tr>
<td>76.7</td>
<td>$4.96 \times 10^{-2}$</td>
<td>$6.93 \times 10^{-4}$</td>
</tr>
<tr>
<td>74.7</td>
<td>$4.65 \times 10^{-2}$</td>
<td>$1.16 \times 10^{-3}$</td>
</tr>
</tbody>
</table>

Mass ratio widths, ($\sigma_m$), plotted against the CN excitation energy ($E^*$) are shown.
Figure 4.12: Fission fragment mass ratio distributions at different beam energies for $^{16}\text{O} + ^{194}\text{Pt}$ reaction. The distribution is symmetric and centered around $M_R = 0.5$. Solid line is the Gaussian fit to experimental mass ratio distribution at 101.8 MeV beam energy.

<table>
<thead>
<tr>
<th>$E_{lab}$ (MeV)</th>
<th>$\sigma_M$</th>
<th>$\sigma_M$ (error)</th>
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<td>124.9</td>
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</tr>
<tr>
<td>122.9</td>
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</tr>
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<td>121.2</td>
<td>5.94$\times10^{-2}$</td>
<td>3.37 $\times10^{-4}$</td>
</tr>
<tr>
<td>118.9</td>
<td>5.95$\times10^{-2}$</td>
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</tr>
<tr>
<td>116.9</td>
<td>5.91$\times10^{-2}$</td>
<td>3.86 $\times10^{-4}$</td>
</tr>
<tr>
<td>114.9</td>
<td>5.82$\times10^{-2}$</td>
<td>3.73 $\times10^{-4}$</td>
</tr>
<tr>
<td>112.9</td>
<td>5.77$\times10^{-2}$</td>
<td>4.31 $\times10^{-4}$</td>
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<td>110.9</td>
<td>5.57$\times10^{-2}$</td>
<td>4.94 $\times10^{-4}$</td>
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</table>

Table 4.2: Width of the mass ratio distribution ($\sigma_m$) and error involved at different laboratory beam energies for $^{24}\text{Mg} + ^{186}\text{W}$ reaction.

in Fig. 4.14. It can be seen that $\sigma_m$ increases with increase in excitation energy for both reactions. However, at the same excitation energies, the magnitude of $\sigma_m$ for $^{24}\text{Mg} + ^{186}\text{W}$ reaction is higher than that of $^{16}\text{O} + ^{194}\text{Pt}$ reaction. This difference
is more pronounced at lower excitation energies. In the energy range studied in the present measurement, the contribution from fast fission reaction is negligible, as the angular momentum ($l$) values populated in both reactions are much lower than the critical angular momentum, at which the liquid drop fission barrier $B_f$ [24] vanishes. Pre-equilibrium fission, which is characterized by a non-equilibration of $K$ degrees of freedom and hence an enhanced fragment angular anisotropy [25], does not change the mass distribution of the fragments, as mass equilibration is expected to take place before $K$ equilibration and the system would pass over the unconditional fission barrier.

4.3.5 Dependence of $\sigma_m$ on excitation energy and angular momentum

In the case of an equilibrated CN, the variance ($\sigma_m^2$) of the fragment mass distribution is linearly related to the nuclear temperature ($T$) and the mean square angular
momentum $< l^2 >$ [26-29]. Hence, it was very important to confirm whether this angular momentum dependence is responsible for the difference in $\sigma_m$ for the two systems studied. To verify this, variation of $< l^2 >$ of the composite system is plotted as a function of excitation energy $E^*$ in Fig. 4.15. $< l^2 >$ values were calculated using the coupled-channels code CCFULL [30] including the rotational couplings ($\beta_2$) of the target nucleus. CCFULL is a coupled channels code in which the coupled-channels equations are solved to compute the fusion cross sections and mean angular momenta of the CN, taking into account couplings to all orders. In this program incoming wave boundary condition is applied and the barrier penetrability is calculated for each partial wave. The code relies on the assumption that fusion process is predominantly governed by the quantum tunneling process and treats the vibrational coupling in the harmonic limit and rotational coupling with pure rotor.

The barrier parameters of the coupled channels code were fixed by reproducing the capture excitation function for $^{16}\text{O} + ^{197}\text{Au}$ reaction [31, 32] and these values were scaled to get the parameters for the reactions $^{16}\text{O} + ^{194}\text{Pt}$ and $^{24}\text{Mg} + ^{186}\text{W}$. Table 4.3
lists various CCFULL parameters used in the calculations for the two reactions $^{16}\text{O} + ^{194}\text{Pt}$ and $^{24}\text{Mg} + ^{186}\text{W}$. From Fig. 4.15, it is clear that, at similar $E^*$-values, the $<l^2>$ value for the reaction $^{24}\text{Mg} + ^{186}\text{W}$ is lower than that of $^{16}\text{O} + ^{194}\text{Pt}$ reaction, which would not explain the increased mass ratio widths for the former reaction. As mentioned earlier, the mass variance ($\sigma_m^2$) of the fragments from an equilibrated composite system is linearly proportional to $T$ and $<l^2>$ \[33\]. That is,

$$\sigma_m^2 = \lambda T + \kappa <l^2>$$ \tag{4.19}$$

where, $\lambda$ and $\kappa$ are proportionality constants. Since the CN undergoes decay via fission as well as particle evaporation, fission $<l^2>$ values used in the calculations were calculated at energies above the Coulomb barrier, using CCFULL and statistical model code PACE3 \[34\]. Fusion $l$-distribution obtained from CCFULL was used as the input to PACE3 in trace back mode and fission $<l^2>$ values were calculated. For further avoiding any discrepancy in $<l^2>$ values, the calculations were restricted to higher excitation energies (above 50 MeV excitation, where fission cross section is dominant over ER cross section) as lower $l$ values may lead to the formation of ERs.

The estimation of nuclear temperature essentially depends upon the assumption, whether the properties of the fragments are determined at the saddle point or scission point. In light compound systems, saddle point and scission point are close to each other. However, for heavy nuclei the descent from saddle point to scission point has an essential extension. The estimates by Knyazheva et al, \[33\] in the mass $\sim$ 200 region have shown that saddle point and scission point temperatures are very similar, and, in our calculations we have used the saddle point as our reference point.

The saddle point temperature $T$ is given by

$$T = \sqrt{\frac{E_{cm} + Q - B_f - E_{rot} - E_{pre}}{a}}$$ \tag{4.20}$$

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where, \( Q \) is the 'Q'-value of the reaction, \( B_f \) is the fission barrier at average angular momentum and \( E_{\text{rot}} \) is the average rotational energy at equilibrium deformation. \( B_f \) and \( E_{\text{rot}} \) were calculated using Sierk Model [24]. \( E_{\text{pre}} \) is the average energy taken away by the neutrons, calculated using the compilation of Saxena et al. [35]. \( a \) is the level density parameter, given by \( A_{\text{CN}}/10 \text{ MeV}^{-1} \).

In Table 4.4 and Table 4.5 various parameters of the two reactions \( {}^{16}\text{O} + {}^{194}\text{Pt} \) and \( {}^{24}\text{Mg} + {}^{186}\text{W} \) are listed.

As we could not observe any mass angle correlation in \( {}^{16}\text{O} + {}^{194}\text{Pt} \) reaction and the mass ratio width increases smoothly with \( E^* \), this reaction was assumed to proceed through the formation of a true CN. Next step was to determine the proportionality constants \( \lambda \) and \( \kappa \) to reproduce the experimental mass ratio variance. These constants were obtained using the least square fitting technique. The fitting was performed for data above 50 MeV excitation energy, where fission cross section is more than the ER cross section. This was to minimize the deviations between the capture \( < I^2 > \)
Table 4.4: Various parameters used in the analysis and calculations of $^{16}\text{O} + ^{194}\text{Pt}$ reaction.

<table>
<thead>
<tr>
<th>$E_{lab}\text{(MeV)}$</th>
<th>$E^*(\text{MeV})$</th>
<th>$&lt;l&gt;$</th>
<th>$B_f(l)\text{(MeV)}$</th>
<th>$E_{rot}\text{ (MeV)}$</th>
<th>$N_{pre}$</th>
<th>$&lt;l^2 \geq ccfull$</th>
</tr>
</thead>
<tbody>
<tr>
<td>101.8</td>
<td>64.3</td>
<td>28</td>
<td>6.468</td>
<td>3.628</td>
<td>2.7</td>
<td>903</td>
</tr>
<tr>
<td>97.8</td>
<td>60.6</td>
<td>26</td>
<td>6.707</td>
<td>3.129</td>
<td>2.7</td>
<td>746</td>
</tr>
<tr>
<td>93.8</td>
<td>56.9</td>
<td>23</td>
<td>7.040</td>
<td>2.452</td>
<td>2.67</td>
<td>589</td>
</tr>
<tr>
<td>89.8</td>
<td>53.2</td>
<td>19</td>
<td>7.433</td>
<td>1.677</td>
<td>2.65</td>
<td>431</td>
</tr>
<tr>
<td>85.8</td>
<td>49.5</td>
<td>16</td>
<td>7.686</td>
<td>1.191</td>
<td>2.65</td>
<td>279</td>
</tr>
<tr>
<td>82.8</td>
<td>46.8</td>
<td>12</td>
<td>7.964</td>
<td>0.672</td>
<td>2.62</td>
<td>176</td>
</tr>
<tr>
<td>80.8</td>
<td>44.9</td>
<td>10</td>
<td>8.075</td>
<td>0.467</td>
<td>2.62</td>
<td>118</td>
</tr>
<tr>
<td>78.7</td>
<td>43.1</td>
<td>8</td>
<td>8.169</td>
<td>0.300</td>
<td>2.6</td>
<td>74</td>
</tr>
</tbody>
</table>

Table 4.5: Various parameters used in the analysis and calculations of $^{24}\text{Mg} + ^{186}\text{W}$ reaction.

<table>
<thead>
<tr>
<th>$E_{lab}\text{(MeV)}$</th>
<th>$E^*(\text{MeV})$</th>
<th>$&lt;l&gt;$</th>
<th>$B_f(l)\text{(MeV)}$</th>
<th>$E_{rot}\text{ (MeV)}$</th>
<th>$N_{pre}$</th>
<th>$&lt;l^2 \geq ccfull$</th>
</tr>
</thead>
<tbody>
<tr>
<td>124.9</td>
<td>63.9</td>
<td>25</td>
<td>6.920</td>
<td>2.669</td>
<td>2.68</td>
<td>700</td>
</tr>
<tr>
<td>122.9</td>
<td>61.9</td>
<td>22</td>
<td>7.144</td>
<td>2.245</td>
<td>2.65</td>
<td>591</td>
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<tr>
<td>121.2</td>
<td>60.6</td>
<td>21</td>
<td>7.240</td>
<td>2.046</td>
<td>2.65</td>
<td>505</td>
</tr>
<tr>
<td>118.9</td>
<td>58.6</td>
<td>18</td>
<td>7.521</td>
<td>1.505</td>
<td>2.65</td>
<td>407</td>
</tr>
<tr>
<td>116.9</td>
<td>56.8</td>
<td>17</td>
<td>7.605</td>
<td>1.343</td>
<td>2.65</td>
<td>351</td>
</tr>
<tr>
<td>114.9</td>
<td>55.0</td>
<td>15</td>
<td>7.762</td>
<td>1.041</td>
<td>2.62</td>
<td>278</td>
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<tr>
<td>112.9</td>
<td>53.2</td>
<td>12</td>
<td>7.964</td>
<td>0.672</td>
<td>2.60</td>
<td>177</td>
</tr>
<tr>
<td>110.9</td>
<td>51.5</td>
<td>9</td>
<td>8.124</td>
<td>0.379</td>
<td>2.6</td>
<td>108</td>
</tr>
</tbody>
</table>

and fission $<l^2>$ values. The best fitting parameters were found to be $\lambda = 2.6 \times 10^{-3} \pm 0.3 \times 10^{-3}$ and $\kappa = 4.9 \times 10^{-7} \pm 0.8 \times 10^{-7}$. Table 4.6 and Table 4.7 show excitation energy, saddle point temperature and fission $<l^2>$ values for the two reactions $^{16}\text{O} + ^{194}\text{Pt}$ and $^{24}\text{Mg} + ^{186}\text{W}$, respectively. If the reaction $^{24}\text{Mg} + ^{186}\text{W}$ proceeded via CN formation, the obtained constants should reproduce the experimental mass ratio variance for this reaction as well. Fig. 4.16 shows the experimental and calculated $\sigma_m$ versus CN excitation energy for the compound system $^{210}\text{Rn}$ at excitation energy above 50 MeV. It can be seen that the calculated values using $\lambda$ and $\kappa$ do not reproduce the experimentally observed mass width for $^{24}\text{Mg} + ^{186}\text{W}$ reaction.
Fission fragment mass distribution - Analysis and results

**Table 4.6:** Excitation energy, saddle point temperature and fission $<l^2>$ values for $^{16}$O + $^{194}$Pt reaction.

<table>
<thead>
<tr>
<th>$E^*$ (MeV)</th>
<th>$T$ (MeV)</th>
<th>$&lt;l^2&gt;_{fission}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>64.3</td>
<td>1.1387</td>
<td>937</td>
</tr>
<tr>
<td>60.6</td>
<td>1.0644</td>
<td>797</td>
</tr>
<tr>
<td>56.9</td>
<td>0.9938</td>
<td>633</td>
</tr>
<tr>
<td>53.2</td>
<td>0.9163</td>
<td>473</td>
</tr>
<tr>
<td>49.5</td>
<td>0.8214</td>
<td>322</td>
</tr>
<tr>
<td>46.8</td>
<td>0.7539</td>
<td>218</td>
</tr>
</tbody>
</table>

**Table 4.7:** Excitation energy, saddle point temperature and fission $<l^2>$ values for $^{24}$Mg + $^{186}$W reaction

<table>
<thead>
<tr>
<th>$E^*$ (MeV)</th>
<th>$T$ (MeV)</th>
<th>$&lt;l^2&gt;_{fission}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>63.9</td>
<td>1.1439</td>
<td>743</td>
</tr>
<tr>
<td>61.9</td>
<td>1.1171</td>
<td>666</td>
</tr>
<tr>
<td>60.6</td>
<td>1.0868</td>
<td>561</td>
</tr>
<tr>
<td>58.6</td>
<td>1.0536</td>
<td>473</td>
</tr>
<tr>
<td>56.8</td>
<td>1.0103</td>
<td>405</td>
</tr>
<tr>
<td>55.0</td>
<td>0.9761</td>
<td>324</td>
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<tr>
<td>53.2</td>
<td>0.9412</td>
<td>217</td>
</tr>
<tr>
<td>51.5</td>
<td>0.8988</td>
<td>144</td>
</tr>
</tbody>
</table>

### 4.4 Discussion

The dramatically different behaviour of mass ratio width of $^{24}$Mg + $^{186}$W reaction at similar excitation energies could be a strong signature of the deviation of reaction mechanism from equilibrated CN process. In the case of an equilibrated CN, the ratio between the experimental ($\sigma_{mexp}^2$) and calculated ($\sigma_{mcal}^2$) mass ratio variance is expected to be equal to unity. This ratio plotted against the CN excitation energy for both reactions is displayed in Fig. 4.17. The line at $\frac{\sigma_{mexp}^2}{\sigma_{mcal}^2} = 1$ represents the expected results from CN assumption. It can be clearly seen that for the reaction $^{16}$O + $^{194}$Pt the values of $\frac{\sigma_{mexp}^2}{\sigma_{mcal}^2}$ are scattered around this line, while the reaction $^{24}$Mg + $^{186}$W shows a remarkable deviation from the line. This deviation, more significant at lower excitation energies which correspond to near and below Coulomb barrier energies, is an obvious signature of the onset of quasifission process in this reaction.

It is now well known that if the two colliding nuclei are heavy, merely overcoming the Coulomb barrier is not the sufficient condition to ensure the formation of a CN, as
the electric repulsion dominates over the attractive nuclear force during the dynamical evolution after contact. Hence the effective barrier to overcome is not the Coulomb barrier, but the saddle point in a multi-dimensional potential energy surface, the same which determines the potential energy barrier that protects the CN against fission decay. Hence some extra energy (extra-extra-push [8, 9]) is required to induce fusion into CN, which depends on the mean fissility $\chi_m$ of the composite system. The mean fissility represents the degree of re-separability of the binary system, which is macroscopically given by the Coulomb repulsion in the initial stage of the reaction process ($\chi_e$) and the balance between the Coulomb and surface energies of the CN $\chi$). That is, the mean fissility $\chi_m$, is a mixture of CN fissility $\chi$ and the effective fissility $\chi_e$, given by $\chi_m = \frac{2}{3} \chi + \frac{1}{3} \chi_e$ [8, 9]. It is known that quasi-fission is strongly dependent on $\chi_m$ and the extra-extra-push energy calculated [9] shows a dramatic increase around $\chi_m = 0.723$, which was attributed to the onset of quasi-fission. Though the properties
of the entrance channel is partly incorporated in mean fissility $\chi_m$ through effective fissility, $\chi_e$, the effect of deformation is not included. Here, we compared our results with two other reactions, $^{30}\text{Si}+^{186}\text{W}$ [17] and $^{34}\text{S}+^{186}\text{W}$ [23]: both are reported to show deviation from CN behaviour. In table 4.8, we have listed the calculated values of CN fissility $\chi$, effective fissility $\chi_e$, mean fissility $\chi_m$ and static quadrupole deformation ($\beta_2$) of projectiles and targets [36]. Though it is quite clear from the calculations that $\chi_m$ values for all the three reactions $^{24}\text{Mg}+^{186}\text{W}$, $^{30}\text{Si}+^{186}\text{W}$ and $^{34}\text{S}+^{186}\text{W}$ are less than 0.723, they show clear evidence of onset of quasifission process. This suggests that the entrance channel parameters such as Coulomb repulsion ($Z_PZ_T$), mass asymmetry, static deformation of the reactions partners etc, play important role in deciding the onset of quasifission process. However more experimental evidences are required to understand the relative importance of these parameters.
Table 4.8: Comparison of different parameters of three reactions using the same target $^{186}$W.

<table>
<thead>
<tr>
<th>Reaction</th>
<th>$\chi$</th>
<th>$\chi_e$</th>
<th>$\chi_m$</th>
<th>$\alpha$</th>
<th>$\alpha_{BG}$</th>
<th>$\beta_2$ (projectile)</th>
<th>$\beta_2$ (target)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{24}$Mg + $^{186}$W</td>
<td>0.7350</td>
<td>0.5239</td>
<td>0.6646</td>
<td>0.771</td>
<td>0.851</td>
<td>0.605</td>
<td>0.2257</td>
</tr>
<tr>
<td>$^{30}$Si + $^{186}$W</td>
<td>0.7504</td>
<td>0.5547</td>
<td>0.6851</td>
<td>0.722</td>
<td>0.865</td>
<td>-0.315</td>
<td>0.2257</td>
</tr>
<tr>
<td>$^{34}$S+ $^{186}$W</td>
<td>0.7688</td>
<td>0.5977</td>
<td>0.7117</td>
<td>0.691</td>
<td>0.874</td>
<td>0.252</td>
<td>0.2257</td>
</tr>
</tbody>
</table>

According to Swiatecki [8], there are three milestone configurations which play very important role in fusion process, which are, contact configuration, conditional saddle point configuration and unconditional saddle point configuration. The contact configuration is the point at which the nuclei are in contact with each other and the neck degree of freedom becomes unfrozen. Conditional saddle point configuration corresponds to the maximum of the potential energy under the constraint that mass asymmetry remains frozen to its initial value. Unconditional saddle point configuration is the configuration with no constraint on mass asymmetry. If the contact configuration is less elongated than conditional saddle point, then the system will fuse to form a mono-nucleus, and if the contact configuration is less elongated than unconditional saddle point configuration, an equilibrated CN will be formed. On the other hand, if the contact configuration is more elongated than the unconditional saddle point configuration, which can happen at near barrier energies for deformed reaction partners, the mono-nucleus will re-separate before complete equilibration, which is the quasifission process. In the present measurements, the different behaviour of the mass ratio distributions of two systems populating the same composite system is indicative of the difference in the dynamical evolution of the two systems over the multi-dimensional potential energy surface, showing the dependence of entrance channel parameters. In the case of $^{16}$O + $^{194}$Pt reaction, where $Z_PZ_T$ is 624, the trajectories were reaching the fully equilibrated CN, after the contact process. The absence of any non-compound nucleus behaviour in this reaction suggests that the effect of deformation and orientation are not very significant in this case. However, the anomalous behaviour of $^{24}$Mg+$^{186}$W system with $Z_PZ_T$ equal to 888, shows experimental signatures of deviation from the CN behaviour. It may be conjuctured that in this case the trajectories may not be completely reaching the compact configuration and thus a substantial portion may be escaping, as a combined effect of elongated contact configuration and subsequent Coulomb repulsion during the dynamical evolution after contact.
4.5 Summary and conclusions

We have measured fission fragment mass ratio distributions for the two reactions
\(^{16}\text{O}+^{194}\text{Pt}\) and \(^{24}\text{Mg}+^{186}\text{W}\), both forming the same CN \(^{210}\text{Rn}\) at near barrier energies.

The CN excitation energy was matched for the two reactions at many energy points during the measurements. The entrance channel mass asymmetry of \(^{24}\text{Mg}+^{186}\text{W}\) reaction is much lower than the critical Businaro-Gallone mass asymmetry value, while that of \(^{16}\text{O}+^{194}\text{Pt}\) is pretty close to \(\alpha_{BG}\). As the targets used in the experiments were not fissile, time difference method was successfully used in the analysis. Both the reactions studied did not show any mass angle correlation. However, the measured mass ratio variance for \(^{24}\text{Mg}+^{186}\text{W}\) reaction is much higher than that of \(^{16}\text{O}+^{194}\text{Pt}\) reaction at similar excitation energies. Larger mass width is a signature of re-separation before complete equilibration. For the system to equilibrate in mass, it should remain together longer than \(5 \times 10^{-21}\) seconds and complete several rotations before separation. As we could not observe any mass-angle correlation in any of the systems studied, it is clear that system completes several rotations after capture, but re-separate before \(5 \times 10^{-21}\) seconds in the case of \(^{24}\text{Mg}+^{186}\text{W}\) reaction.

This different behaviour of mass widths in the two reactions \(^{16}\text{O}+^{194}\text{Pt}\) and \(^{24}\text{Mg}+^{186}\text{W}\) shows the difference in the evolution of the trajectories in the composite system over the multi-dimensional potential energy surface, implying a strong dependence on the entrance channel properties. In the case of \(^{24}\text{Mg}+^{186}\text{W}\), trajectories are more likely to be deflected away from the compact CN stage, may be due to the elongated contact configuration followed by the Coulomb repulsion during the dynamical evolution. It is well known that the onset of quasifission is strongly dependent on the mean fissility, \(\chi_m\). However, calculations show that the mean fissility for \(^{24}\text{Mg}+^{186}\text{W}\), \(^{30}\text{Si}+^{186}\text{W}\) and \(^{34}\text{S}+^{186}\text{W}\), all reactions which show conclusive evidence of quasifission, are less than 0.723. These experimental results suggest that in any dynamical model which attempts to explain the fission fragment mass and angular distributions of fissile and less fissile systems should include \(Z_PZ_T\) and the deformations of the collision partners in a systematic way. The apparent observation of larger mass variances for the very asymmetric reaction, \(^{24}\text{Mg}+^{186}\text{W}\), necessitates further investigations with different projectile-target combinations at near barrier energies.
Bibliography


