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Nucleon exchange mechanism of mass asymmetry relaxation in fission and other binary nuclear reactions

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Abstract. Mass asymmetry relaxation as manifested in fission and heavy ion-induced binary reactions is reviewed. In fission, the dynamics is characteristic of a fully damped case and is well described by a stochastic theory. In heavy ion deep inelastic collisions and quasi-fission, on the other hand, the relaxation is incomplete giving rise to the possibility of studying its time evolution.

Keywords. Fission; deep inelastic collisions; quasi-fission; mass asymmetry relaxation; stochastic theory; one-body dissipation.

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1. Introduction

It was exactly fifty years ago that Hahn and Strassman discovered the phenomenon of nuclear fission, a process in which a heavy nucleus, literally on the touch of a feather, splits into two nuclei of nearly equal masses. While the early interest in the process arose primarily from efforts to exploit the largest known energy release in any reaction to military and commercial purposes, it was soon realized that the physics of the process involving large scale rearrangement of nuclear matter is not only interesting but also unique. Fission, therefore, became in the following years, the subject of very intense experimental and theoretical studies. While the advent of particle accelerators in the last two decades capable of delivering a wide range of energetic heavy ions has considerably widened the scope of studies of nuclear collective motion in a more or less controlled way through nucleus-nucleus collisions, the interest in fission studies has not diminished, not only because fission is still considered to be an elementary collective process fit to serve as a test bed of many of the ideas being developed in heavy ion reaction studies, but also because it is the coldest collective process strongly influenced by nuclear shell and pairing effects. In fact, nuclear fission belongs to that rare class of discoveries of this century, on which scientific interest has remained active over several decades. It is therefore perhaps appropriate to take a historic perspective and review in somewhat broad terms what we have learnt about nuclear collective behaviour from these studies. For reasons of space, we restrict ourselves to a discussion of one specific aspect, namely, mass asymmetry relaxation in fission and other binary nuclear reactions.

One of the well-known and outstanding experimental features of low energy fission is the asymmetric mass division (Vandenbosch and Huizenga 1973). While it became fairly obvious very early in the history of fission research that nuclear shell effects of the nascent fragments play a part in deciding the fragment mass distributions, the mechanism leading to the observed distributions was far from understood. It was indeed puzzling to note that the mass division with the largest energy release did not have the maximum probability of occurrence or yield. The early success of the liquid drop model to explain qualitatively the gross behaviour of fission threshold energies on the basis of deformation potential energy surfaces leads naturally to an approach to include reflection asymmetric shapes in a description of the nuclear shape evolutions leading to fission. However, such an approach based on the structure of the static potential energy surface including asymmetric shapes was found to be inadequate, pointing to the role of nuclear dynamical effects on the fragment mass distributions. An early attempt to correlate the fragment mass yields to the available phase space at the scission point led to the well-known statistical theory of fission (Fong 1956). The qualitatively successful theory however failed again pointing out to the need to include dynamical effects. In particular, the quantities of interest apart from the deformation potential energy are the inertial tensor and the dissipation function for the shape parameters, which are the relevant collective coordinates. It was conjectured by Ramanna (1964) that a diffusion model with absorbing barriers may give a suitable description of the dynamics in the mass asymmetry degree of freedom. This corresponds to an overdamped case, with the motion governed entirely by the interplay of conservative driving forces and dissipative forces, and the inertial forces playing a negligible role. With the further assumption that the measured mass distributions correspond to the equilibrium distributions, one has only to know the relevant transport coefficients at the scission point. The resulting theory, the stochastic theory of fragment mass and charge distributions in fission, has been investigated in detail by the Trombay group (Ramanna *et al* 1965: Ramanna and Ramamurthy 1969; Ramamurthy 1971; Prakash *et al* 1980; Prakash 1980). With realistic inputs for the transport coefficients (the nucleon transfer probabilities), the theory was able to reproduce many of the known features of the experimental fragment mass and charge distributions in low energy fission. However, a few important questions were raised and remained unanswered. For example, the typical time for a nucleon transfer and whether there is enough time between the saddle and the scission points for many transfers to take place and establish equilibrium at scission were not known. In the last few years, with the advent of heavy ion reactions, convincing answers to some of these questions have become available and the mass asymmetry has come to be accepted as a relevant dynamical degree with well-defined transport coefficients in a description of both fission and heavy ion-induced binary reactions (Bromley 1984). It has further been shown that the transport coefficients are unlike any known for ordinary fluids but refer to an interesting new form of fluid dominated by the so-called "one-body" damping (Blocki *et al* 1978). We present here a brief review of the mass relaxation process as manifested in fission and other binary heavy ion reaction processes.

2. Mass relaxation in fission—The stochastic theory of fragment mass and charge distributions

Rearrangement collisions between quantal systems are known for a long time. In the Bohr model of the capture and loss of electrons in the penetration of atomic particles through matter, an electron which is initially bound to a nucleus is transferred to another, both the nuclei moving relative to each other all the time. It was proposed by Ramanna more than two decades ago (Ramanna 1964; Ramanna *et al* 1965) that a similar exchange of nucleons might take place between two nascent fragments formed during the fission process near the scission point. The application of the theory of random flights with absorbing barriers at shell closures then gave a natural explanation of the asymmetric mass distributions in low energy fission of actinide nuclei. Though the model appeared to be at complete variance with the then existing fission theories based on the liquid drop model, in retrospect, it is now clear that the nucleon exchange model is simply a discrete version of an overdamped dynamics in the mass asymmetry degree of freedom. If one assumes that the relative motion of the nascent fragments is slow compared to the characteristic time for nucleon transfers, the process can be treated as a stochastic process where not only the nascent fragments are in thermodynamic equilibration within themselves but also attain mutual equilibration among themselves with respect to nucleon and energy transfers. The observed distributions of the fragment mass and charge then correspond to the equilibrium distribution near the scission point. The problem is one of calculating the nucleon transfer probabilities. If the two nascent fragments are cold, the direction of spontaneous transfer of nucleons is from the fragment having the higher Fermi energy to the one having a lower Fermi energy. Even if the nascent fragments have some excitation energy, a tendency for a preferential transfer of nucleons in the direction of decreasing chemical potential persists. Figure 1 shows a schematic diagram illustrating the nucleon exchange mechanism between the light (L) and the heavy (H) fragment. $\rho_{L/H}(r)$ represents the diffuse matter densities of the fragments. $g_{L/H}$ are the respective single particle level densities and $f_{L/H}$ are the Fermi-Dirac occupation probabilities. $\mu_{L/H}$ are the chemical potentials of the two fragments. A quantitative estimate of the relative probabilities of nucleon transfers in both directions can be made as (Ramanna and Ramamurthy 1969; Ramamurthy 1971; Prakash *et al* 1980,

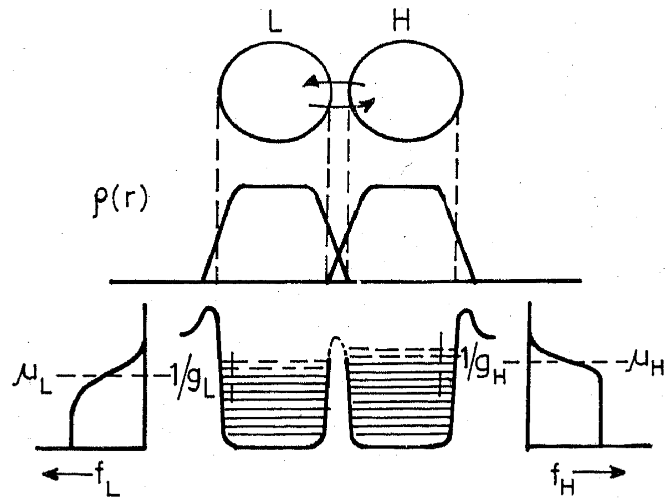

Figure 1. Schematic diagram illustrating the nucleon exchange mechanism between two nascent fission fragments.

Prakash 1980)

$$P_{L-H} = \frac{2\pi}{\hbar}\int g_L(E) f_L(E) g_H(E)[1 - f_H(E)]|M_{LH}|^2\,dE,$$

$$P_{H-L} = \frac{2\pi}{\hbar}\int g_H(E) f_H(E) g_L(E)[1 - f_L(E)]|M_{HL}|^2\,dE.$$

The matrix elements of transfer M_{H-L} and M_{L-H} can be taken to be equal because of microscopic reversibility. Since the main contribution to the integral comes from a narrow band of energy around the chemical potentials of the two nascent fragments, the quantities g_L, g_H and the matrix element of transfer can be calculated at the mean chemical potential and taken out of the integral. One then has

$$P_{L-H} = \frac{2\pi}{\hbar}|M|^2 g_L(\bar{\mu}) g_H(\bar{\mu}) I_{L-H},$$

where

$$I_{L-H} = \int f_L(E)[1 - f_H(E)]\,dE,$$

with an analogous expression for P_{H-L}. $\bar{\mu}$ is the mean chemical potential. For a nearly degenerate system like a nucleus having a temperature much less than the mean chemical potential, one gets

$$I_{L-H} = (\mu_H - \mu_L)/\{\exp[(\mu_H - \mu_L)/T] - 1\}.$$

Thus the main driving force for the net transfer of nucleons from one of the fragments to the other is the difference in their chemical potentials though a finite temperature induces nucleon transfers in both directions. Nuclear shell structure effects influence the nucleon transfer probabilities through their influence on the single particle densities $g_L(E)$ and $g_H(E)$. In fact, it is now well known that it is the deviations of $g(E)$ from an average behaviour which are responsible for the shell effects on many of the observables. This will also influence the chemical potentials which are in general temperature-dependent. $g_L(\bar{\mu})$ and $g_H(\bar{\mu})$ are to be taken as an average of $g_L(E)$ and $g_H(E)$ around μ over an energy range of the order of the temperature. It is obvious that with increasing temperatures, shell effects on all the quantities including the nucleon transfer probabilities will disappear. The stochastic theory has been investigated in all its details by the Trombay group and it was shown that the theory can reproduce many of the known features of fragment mass and charge distributions in fission. Figure 2 shows a few typical mass distributions calculated on the basis of the stochastic theory.

One of the early criticisms of the stochastic theory was whether there is enough time available before scission for sufficient number of nucleon transfers to take place to establish equilibrium in the mass asymmetry degree. Related to this criticism were the questions: what is the typical time for nucleon exchange and what is the time between the saddle and the scission points? Direct experimental evidence for substantial nucleon exchanges between two nuclei in interaction times of the order of 10^{-21} s has now come from the studies of heavy ion collisions (Schroder and Huizenga 1984). It has also been established now that the motion between the saddle

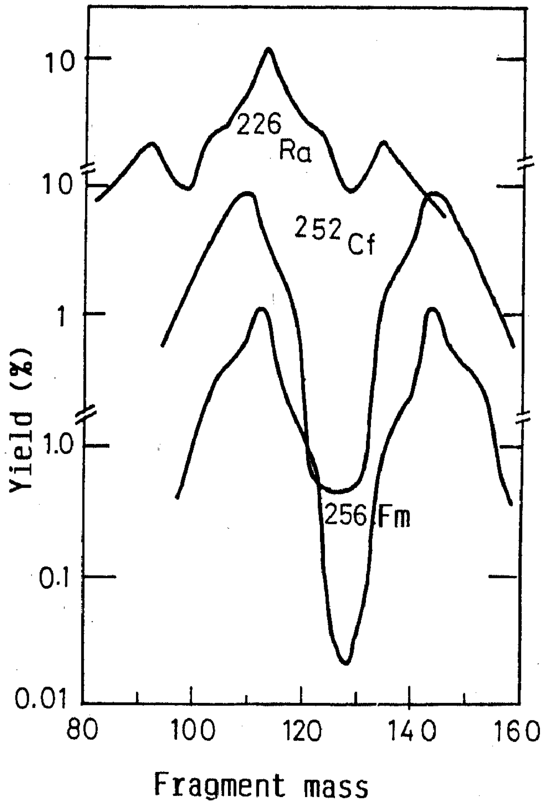

Figure 2. Typical fragment mass distributions calculated on the basis of the stochastic theory.

and the scission point is viscous allowing enough time for a number of nucleon exchanges to take place. In addition, a few other questions were also raised and remained unanswered. For example, the relation between the stochastic theory and the conventional statistical theory was not very clear. In particular, in spite of both the theories being based on the equilibrium assumption, the predicted mass distributions were different appreciably.

In an effort to understand this apparent discrepancy, Kataria *et al* (1985) studied the stochastic model with the inclusion of transfers of 2-, 3- and 4-nucleon clusters. The cluster transfer probabilities were calculated using expressions analogous to the single nucleon transfer probability expressions with appropriately chosen chemical potentials for different clusters. Figure 3 shows the calculated equilibrium mass distributions in the fission of two typical nuclei ^{160}Dy and ^{120}Te under different model assumptions. These calculations had assumed the same overlap form factors for all the clusters and no shell effects. It is seen that with only one-nucleon transfer probabilities, the stochastic model predictions are much narrower compared to the statistical model results. However, with the inclusion of multi-nucleon cluster transfers, the stochastic model predictions converge to the statistical model results. Similar conclusions were drawn with the inclusion of shell effects when not only the width but also the peak of the equilibrium mass distribution depends on whether or not one includes cluster transfers. In the final stages of the fission process near the scission point, cluster transfers are highly inhibited as compared to single-nucleon transfers due to the large differences in the corresponding overlap form factors. Therefore, the stochastic model calculations dominated by single-nucleon transfers are more likely

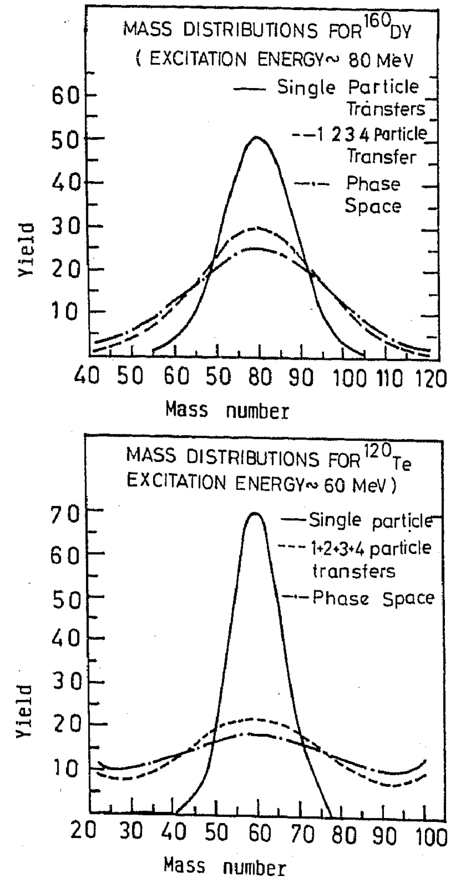

Figure 3. Calculated equilibrium mass distributions under different model assumptions.

to result in equilibrium mass distributions in agreement with the experimental distributions, as shown by the calculations of Prakash *et al* (1980).

3. Mass asymmetry relaxation in binary heavy ion reactions

Nucleus-nucleus collisions at moderate bombarding energies exhibit a rather broad spectrum of reaction mechanisms ranging from direct processes to fully equilibrated compound nucleus formation. Another striking feature of nuclear collisions is the clear hierarchical ordering of the different reaction channels with respect to relaxation in different collective coordinates. The new reaction channels which have been identified in medium energy heavy ion collisions are deep inelastic collisions, fast and quasi-fission reactions and pre-equilibrium fission reactions. In deep inelastic collisions, there is dissipation of relative kinetic energy and angular momentum but not of mass asymmetry. On the other hand, the fast and the quasi-fission reactions have full kinetic energy relaxation accompanied by a gradual relaxation of the mass asymmetry coordinate. Pre-equilibrium fission events have fully relaxed kinetic energy and mass asymmetry but have incomplete relaxation of the K quantum number associated with the tilting mode of the reaction plane. Finally, complete fusion leading to compound nucleus formation corresponds to complete relaxation of all relevant collective degrees of freedom. Figure 4 shows schematic illustrations of the three reaction types, namely, deep inelastic collisions, quasi-fission and compound nucleus

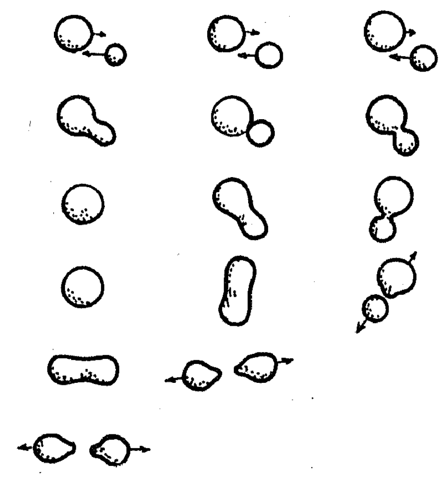

Figure 4. Schematic illustration of the three reaction channels that occur in strongly interacting heavy ion collisions at medium energies.

formation. Since both DIC and quasi-fission reactions proceed at a time scale comparable to the time for one revolution of the dinucleus complex, it is possible to determine the reaction time from an analysis of the angle versus kinetic energy loss and angle versus mass distributions of the reaction products. Information on mass relaxation can therefore come from studies of the time evolution of the mass asymmetry in deep inelastic collisions and fast and quasi-fission reactions. The mass relaxation in deep inelastic collisions and quasi-fission reactions exhibit different characteristics both because of the different reaction times involved and because of the different strength of interaction between the two partners resulting in mass exchange.

3.1 *Mass exchange in deep inelastic collisions*

It was first observed in a number of heavy ion-induced deep inelastic collisions that the double differential cross-sections $d^2\sigma/(dE \times dZ)$ cut at a fixed final energy or energy loss have nearly a Gaussian shape. Figure 5 shows the Z distributions of projectile-like fragments in $^{209}Bi + {}^{136}Xe$ reactions at $E_{lab} = 940$ MeV for a succession of 26 MeV wide bins in total kinetic energy with centroids as indicated at each distribution (Schroder and Huizenga 1984). The experimental spectra are described rather strikingly well by Gaussian fit curves drawn through the data points. The development of the fragment Z distributions with increasing energy loss illustrated in figure 5 is quite suggestive of a classical nucleon diffusion mechanism where individual nucleons are exchanged between the reaction partners in random directions. As the interaction time proceeds and as the kinetic energy is dissipated, both the mass and the charge distributions of the final fragments broaden significantly. One has also observed in these studies a net change in the element distribution without an associated drift in the average fragment mass consistent with a fast equilibration of the isospin mode during a short reaction time of about 1×10^{-21} s. These features offer a unique opportunity to study nucleon transport processes occurring between two interacting nuclei.

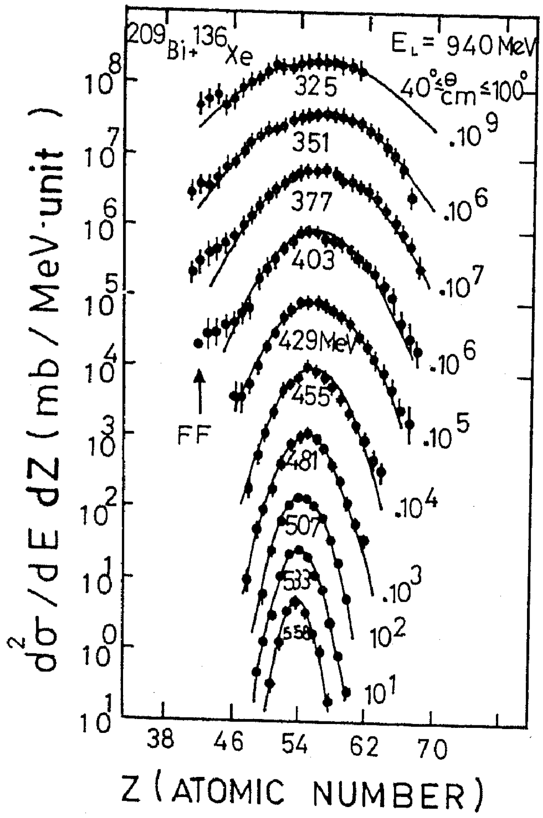

Figure 5. Charge distributions of projectile-like fragments in deep inelastic collisions for specific energy losses.

In a simplified model of nuclear shape evolutions (Randrup 1978, 1979), the heavy ion reaction dynamics is described by the time evolution of a set of macroscopic coordinates q_i specifying the relative separation, neck opening, mass, charge and mass-to-charge asymmetry, angular momentum etc. One can then assume that the mean values of these coordinates follow the classical Lagrange–Rayleigh equations of motion determined by conservative, dissipative and inertial forces

$$\left[\frac{\mathrm{d}}{\mathrm{d}t}\frac{\partial}{\partial \dot{\bar{q}}_i}\frac{\partial}{\partial \bar{q}_i}\right]L = -\frac{\partial F}{\partial \dot{\bar{q}}_i},$$

where L is the Lagrangian. The dissipation function F describes the irreversible conversion of kinetic energy of relative motion into internal excitation owing to changes in the binding energies induced by nucleon exchanges, recoil momentum as well as damping of other collective motions. The equations of motion for the mean values of the collective coordinates can also be coupled to a Fokker–Planck equation for the time evolution of the distributions around the mean values. For the case of the mass and charge asymmetry, one has

$$\frac{\mathrm{d}}{\mathrm{d}t}P(N,Z,t) = \left[-\frac{\partial V_N}{\partial N} - \frac{\partial V_Z}{\partial Z} + \frac{\partial^2 D_{NN}}{\partial N^2} + \frac{\partial^2 D_{ZZ}}{\partial Z^2}\right] P(N,Z,t),$$

where $P(N,Z,t)$ is the joint probability of finding N neutrons and Z protons in one of the fragments. As mentioned earlier, the physical information on the microscopic nucleon transfer probabilities at each instant of time t is contained in the drift and the diffusion coefficients V and D respectively. The average neutron and proton numbers are expected to change with rates determined only by the drift coefficients as

$$\partial \bar{N}/\partial t = V_N \quad \text{and} \quad \partial \bar{Z}/\partial t = V_Z,$$

whereas the growth of the corresponding covariances of the probability distributions depends on the drift and the diffusion coefficients and the covariance already accumulated. Evaluated along the mean trajectory $[\bar{N}(t), \bar{Z}(t)]$ these growth rates can be written as

$$\frac{\partial}{\partial t}\sigma_N^2 = 2\left(D_{NN} + \sigma_N^2 \frac{\partial V_N}{\partial N} + \sigma_{NZ}\frac{\partial V_N}{\partial Z}\right),$$

$$\frac{\partial}{\partial t}\sigma_Z^2 = 2\left(D_{ZZ} + \sigma_Z^2 \frac{\partial V_Z}{\partial Z} + \sigma_{NZ}\frac{\partial V_Z}{\partial N}\right),$$

$$\frac{\partial}{\partial t}\sigma_{NZ} = 2D_{NZ} + \sigma_N^2 \frac{\partial V_Z}{\partial N} + \sigma_Z^2\frac{\partial V_N}{\partial Z} + \sigma_{NZ}\left(\frac{\partial V_N}{\partial N} + \frac{\partial V_Z}{\partial Z}\right),$$

$$\sigma_A^2 = \sigma_N^2 + \sigma_Z^2 + 2\rho\sigma_N\sigma_Z,$$

where $\rho = \sigma_{NZ}/\sigma_N\sigma_Z$ is the correlation coefficient. The above growth rate expressions take into account the constraints imposed on the transport process by the variation of the drift coefficients with N and Z arising mainly due to the curvature and the alignment of the underlying time-dependent potential energy surface and therefore lead to the eventual attainment of the equilibrium variances in the N and Z distributions. Although the mixed diffusion coefficient D_{NZ} is zero owing to the statistical independence of individual nucleon exchanges, a covariance $\rho\sigma_N\sigma_Z$ develops with time because of the misalignment of the underlying potential energy surface with respect to the $N-Z$ coordinate system. Randrup (1978) has given a parameter-free calculation of the transport coefficients including their form factors for peripheral collisions based on the mean field approximation. While the underlying mechanism of nucleon exchange between two nuclei is very similar to that employed by Ramanna *et al*, the Randrup model explicitly takes into account the relative motion of the two nuclei and evaluates all transport coefficients. Analysis of the energy loss versus mass/charge widths in deep inelastic collisions has brought out not only the dominant role played by the nucleon exchange mechanism both in the damping of relative motion and in the relaxation of the mass asymmetry mode but also the intricate interplay between the static driving forces and the dynamic characteristics of the nucleon exchange process (Kapoor and De 1982; De and Kapoor 1983; Schroder and Huizenga 1984).

3.2 *Mass equilibration in quasi fission reactions*

In the hierarchy of strongly interacting heavy ion collisions, quasi-fission is the mass asymmetry relaxation mode (Toke *et al* 1985). Figure 6 illustrates the topology of the potential surface which makes it possible to sample various stages of motion even though the observation method is restricted to a recording of the final states only.

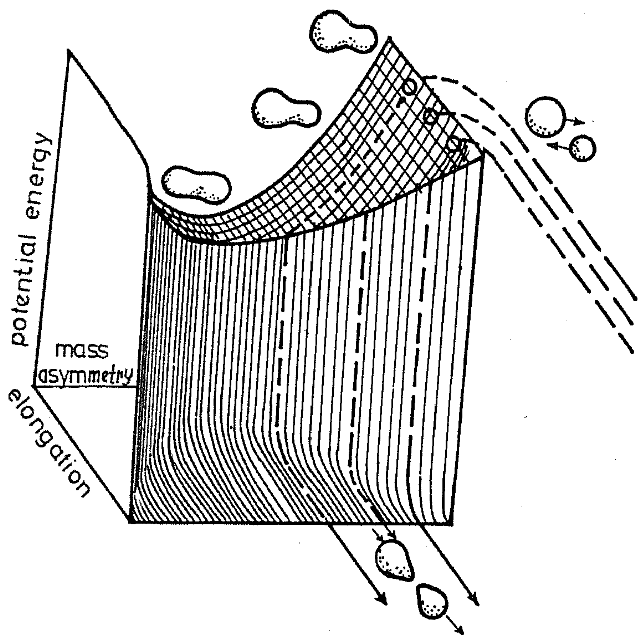

Figure 6. Topology of the potential energy surface appropriate to mass asymmetry relaxation in quasi fission.

The target and the projectile after making contact go through various dinuclear states when the relative motion is brought essentially to a stop resulting in an excited asymmetric mononuclear configuration. The combined Coulomb and surface forces then drive the system towards symmetry but the system will come apart at various stages of this process giving rise to the observed broad mass-angle distributions. Since the reaction proceeds also in this case at a time scale comparable to the time for one revolution, it is possible to determine the reaction time from an analysis of the angle versus mass distributions of the products. Extensive measurements of the mass drift in ^{238}U-induced quasi-fission reactions on targets of ^{32}S, ^{40}Ca, ^{48}Ca and ^{nat}Zn at several bombarding energies ranging from 4·6 MeV/u to 7·5 MeV/u have recently been reported by Toke *et al* (1985). From the analysis of the triple differential cross-sections $d^3\sigma/dA\,d\theta\,dE_k$ for binary events and the resulting plot of the mass asymmetry versus time, shown in figure 7, three distinct features were deduced:

(i) The mass asymmetry relaxation can be described by an exponential relaxation law.
(ii) Within experimental error, the time constant for the relaxation is the same for all systems varying in total mass between $A = 270$ and $A = 302$ and in total charge between $Z = 108$ and $Z = 122$.
(iii) Within error, the relaxation time is independent of the excitation energy during the reaction.

These observations are compatible with a picture of the mass asymmetry relaxation in terms of motion in a harmonic driving potential subject to strong dissipative forces of the one-body type. If k is the force constant describing the parabolic potential and m the mobility, the time constant τ is given by

$$\tau^{-1} = km.$$

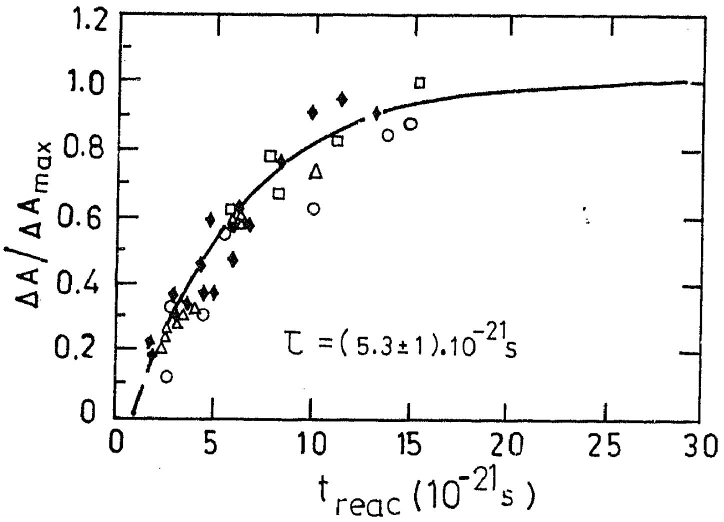

Figure 7. Mass asymmetry relaxation versus time in several ^{238}U-induced reactions. The mass relaxation is expressed as $[\langle A\rangle-(A_P+A_T)/2]/[A_P-(A_P+A_T)/2]$ with $A_P=238$ and $\langle A\rangle$ is the mean heavy fragment mass.

The mobility is itself a product of a geometrical form factor and a dissipation coefficient describing the medium. Thus

$$\tau^{-1} = (\text{force const.}) \times (\text{geom. fac.}) \times (\text{dissipation coeff.}).$$

The observed "universal" exponential relaxation law implies that the product of these three factors is an approximate constant of motion. While the fact that the different target-projectile combinations fit the same relaxation curve is an indication that the product (force const.) × (geom. factor) remains constant within the relatively narrow range of total mass and charge sampled by the experiment, the near-constancy of the relaxation time for different values of the bombarding energy and hence excitation energy indicates that the dissipation coefficient must also be independent of excitation energy. This is indeed what one expects from the one-body dissipation theory where the dominant dissipation mechanism is due to collisions of the long mean-free path particles with the nuclear surface as it tries to move towards its equilibrium shape. It is quite different from the picture of two-body collisions in the interior of the nucleus giving rise to the dissipation that varies as $1/T^2$ (equivalently as $1/E^*$). One also obtains a satisfactory fit by assuming a common delay of about 1×10^{-21} s before mass drift sets in. This delay one can associate with the initial and the final dinuclear stages of the collision, where there is essentially no mass asymmetry motion.

4. Summary and conclusions

Mass asymmetry is one of the important collective degrees of freedom in a macroscopic model of nuclear shape evolutions describing fission and medium energy heavy ion reactions. The dynamics in this coordinate seems to be dominated by a strong viscosity of the one-body type, a unique feature of the quantum nuclear fluid. While the heavy ion reactions have provided very detailed information on the time evolution of the mass asymmetry, the deduced viscosity is structureless without shell and pairing effects because of the high excitation energies of the systems involved. Mass distributions in low energy fission, on the other hand, are strongly influenced by shell and pairing effects but there is no direct way of studying their time evolutions as in the case of heavy ion reactions. While a complete understanding of the relaxation of the mass asymmetry degree of freedom over the full range of excitation energies is yet to emerge, the nucleon exchange mechanism seems to provide an appropriate description of the relaxation process.

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