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Some aspects of fission and quasifission processes

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Abstract. The discovery of nuclear fission in 1938–1939 had a profound influence on the field of nuclear physics and it brought this branch of physics into the forefront as it was recognized for having the potential for its seminal influence on modern society. Although many of the basic features of actinide fission were described in a ground-breaking paper by Bohr and Wheeler only six months after the discovery, the fission process is very complex and it has been a challenge for both experimentalists and theorists to achieve a complete and satisfactory understanding of this phenomenon. Many aspects of nuclear physics are involved in fission and it continues to be a subject of intense study even three quarters of a century after its discovery. In this talk, I will review an incomplete subset of the major milestones in fission research, and briefly discuss some of the topics that I have been involved in during my career. These include studies of vibrational resonances and fission isomers that are caused by the second minimum in the fission barrier in actinide nuclei, studies of heavy-ion-induced fission in terms of the angular distributions and the mass–angle correlations of fission fragments. Some of these studies provided evidence for the importance of the quasifission process and the attendant suppression of the complete fusion process. Finally, some of the circumstances around the establishment of large-scale nuclear research in India will be discussed.

Keywords. Fission history; fission discovery; quasifission; angular distribution.

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1. Some early fission history

In this section, an abbreviated sequence of the events that led up to the discovery of nuclear fission and rapid theoretical understanding of the process that took place in the late 1938 and 1939 will be reviewed. This history is reported in further detail by one of the participants, Frisch in his book [1], which is the main source for this information.

The events leading up to the discovery of the nuclear fission process took place at the end of 1938. The chemistry studies of the products generated in thermal neutron radiation of uranium were carried out at the Kaiser Wilhelm Institute in Berlin by the group of Otto Hahn and Lise Meitner. The puzzling result was that the chemical separation seemed to isolate the induced radioactivity in the barium-like fraction and not in the fractions with properties expected for elements near uranium. Unfortunately, the political situation in Germany forced Lise Meitner to leave Germany with the help of her Dutch colleagues. She took up a position in Stockholm sometime in the fall of 1938 and could therefore not participate in the final work on the neutron activation of uranium in Berlin.

Otto Hahn and Lise Meitner did, however, stay in contact by mail about the results of the continuing uranium experiments and just before Christmas, Lise Meitner received a letter from Otto Hahn confirming that the activity created by neutron bombardment of uranium appeared to have the chemical properties of Ba. Lise Meitner had been invited to spend the Christmas holidays with Swedish friends in Kungälv, just a little north of Göteborg in Sweden where her nephew, Otto Frisch, also joined her from Niels Bohr's institute in Copenhagen.

During a long walk in the woods, Meitner and Frisch discussed these new findings and started to entertain the thought that barium could indeed have been produced in the process by a cleavage of the uranium into two large fragments. They determined that the energetics of division into two fragments is indeed possible and that it would release a large kinetic energy of ~200 MeV based on the nuclear mass formula and the idea of the nucleus as a liquid drop.

After Christmas, Frisch returned to Copenhagen. Niels Bohr was preparing to leave for New York for a long stay in the United States, but he had just enough time to hear about the new results from Berlin and Meitner and Frisch's conjecture. Bohr exclaimed "Oh! what idiots we all have been! Oh! but this is wonderful! This is just as it must be! Have you and Lise Meitner written a paper about it?" ? and he sailed off for America (figure 1).

Meitner and Frisch wrote the paper while staying in contact via telephone calls between Copenhagen and Stockholm and Frisch performed the physical measurements of the energetic fission fragments emitted in the $\mathrm{U}(n, f)$ reaction using an ionization chamber.

On January 6, Hahn and Strassmann published the chemical observation of a Ba fraction resulting from neutron bombardment of uranium [2] leaving Meitner off the author list although she had been heavily involved in the work leading up to the final result. In short order, Meitner and Frisch published a letter in *Nature* [3] providing the correct explanation for the puzzling result, and on February 18, Otto Frisch published his observation of the physical measurement of the energetic, heavy fission fragments [4]. At this point, the long-standing puzzle concerning the appearance of products with barium-like chemical properties in neutron bombardments of uranium samples had effectively been solved and the 'atomic age' was launched with all its consequences for mankind over the following decades. Even today, newspaper articles appear almost daily, the subject of which have their root in this monumental discovery.

The second part of the story took place in the US. Niels Bohr and Leon Rosenfeld, a colleague at his institute, arrived in New York harbour where they were met by John A Wheeler from Princeton University, and where Bohr was planning to spend an extended period of time discussing the fundamental tenets of quantum physics with Albert Einstein, who had serious conceptual problems with this new description of Nature. Wheeler escorted Rosenfeld on the train ride to Princeton, while Bohr stayed in New York visiting colleagues at Columbia University, and presumably had discussions with Enrico Fermi, who had just arrived there instead of returning to Italy after accepting the Nobel Prize in Stockholm in December 1938 [5].

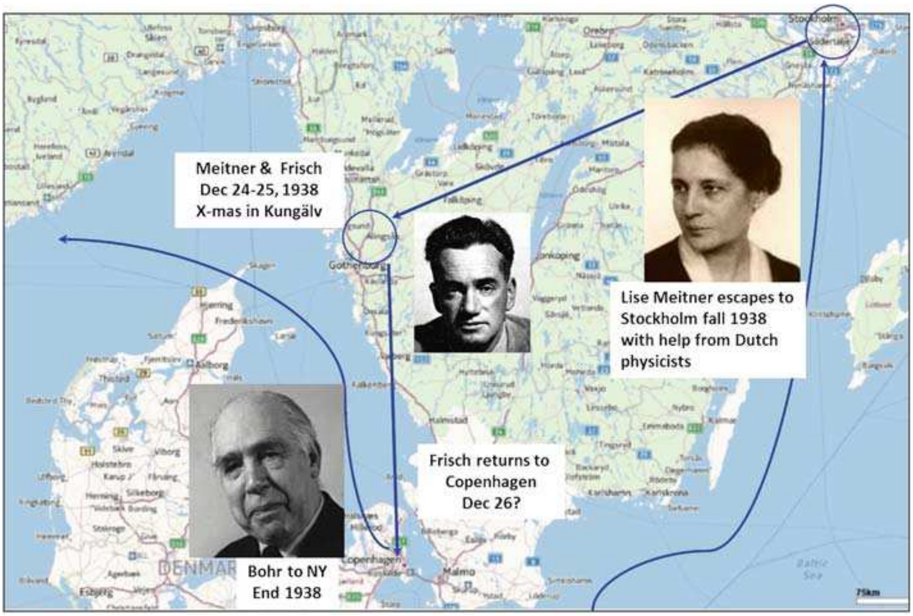

Figure 1. Illustration of some of the interactions between Lise Meitner, her nephew Otto Frisch, and Niels Bohr that took place in southern Sweden and Denmark during the Christmas break in 1938. See text for details.

Before leaving Copenhagen, Bohr had promised Otto Frisch to keep quiet about the discovery of nuclear fission until he had completed the physical measurement of energetic fission fragments. However, while Bohr had long and detailed discussions about the subject during their voyage across the Atlantic, he apparently forgot to tell Rosenfeld about this promise, and so on the train ride to Princeton together with Wheeler, he told him about the exciting new development and soon after they arrived at Princeton, Rosenfeld gave a 'journal club' about it at the Palmer Laboratory, may be even on the evening of his arrival.

Bohr stayed in New York a couple of days before going to Princeton, where he immediately started theoretical work in order to understand the results. He soon identified the minor ^{235}U isotope as responsible for $\mathrm{U}(n_{\mathrm{th}}, f)$ cross-section [6]. Bohr and Wheeler started to work on the theory of fission and submitted their ground-breaking paper to *Phys. Rev.* on June 28 [7], almost six months after Bohr first learned about the discovery. In reading this paper today, one cannot help but being struck by the depth of understanding of this new phenomenon and the detailed description of many of the central aspects of fission that the authors were able to achieve based on the still very meager but rapidly expanding body of experimental data. At this point, the study of fission was widespread and it is well known that it soon led to the demonstration of the nuclear chain reaction by Fermi at the University of Chicago, and, of course, the start of the Manhattan project that culminated in the assembly of the first two atomic bombs in Los Alamos. In the following sections, some of the developments in fission research that took place over the following decades are highlighted.

2. Transition state model and angular distributions

At the first conference on the ‘Peaceful uses of atomic energy’ that was sponsored by the United Nations and held in Geneva, Switzerland in 1955, the many advances in fundamental fission research as well as the development of nuclear power reactors were reviewed. One important contribution to this conference was the Aage Bohr’s analysis of the angular distribution of fission fragments from photofission as well as fission induced by fast neutrons [8] based on the concepts of the transition state model of fission (figure 2). Further theoretical developments of the theory was given at the second conference in this series by Halpern and Strutinski [9] who showed that the angular distribution of fragments carries information on the shape (via moments of angular momenta) of the fission saddle point when the energy above the barrier is sufficiently high to allow for a statistical treatment of the problem.

In fact, the liquid-drop model (LDM) prediction that the fission saddle point becomes more compact for heavy nuclei was beautifully demonstrated by a series of α-induced fission angular distribution measurements by Reising *et al* [10]. This result is shown in figure 3, where the saddle-point deformation is given in terms of the ratio of the moments of inertia of the sphere to the effective moment of inertia defined by

$$\frac{1}{\mathcal{J}_{\text{eff}}} = \frac{1}{\mathcal{J}_{\parallel}} - \frac{1}{\mathcal{J}_{\perp}}, \tag{1}$$

where $\mathcal{J}_{\parallel}$ and $\mathcal{J}_{\perp}$ refer to the moments of inertia parallel and perpendicular to the nuclear symmetry axis, respectively.

The discrepancy between the data and the theory arises from the choice of parameters for the LDM at the time, which have since been readjusted to correctly predict the value of Z^2/A for which the fission barrier vanishes. This observation firmly established the fact that the saddle-point shape controls the fission anisotropy as proposed by Halpern and Strutinski [9].

3. Shell effects, fission isomers and the double-humped barrier

A dramatic expansion of the understanding of fission took place with the discovery of fission isomers [11] and fission shape resonances [12], both of which found their explanation

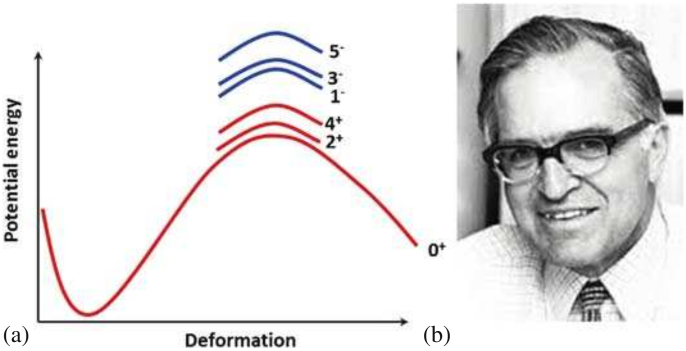

Figure 2. (**a**) Schematic of the transition state model. (**b**) Aage Bohr.

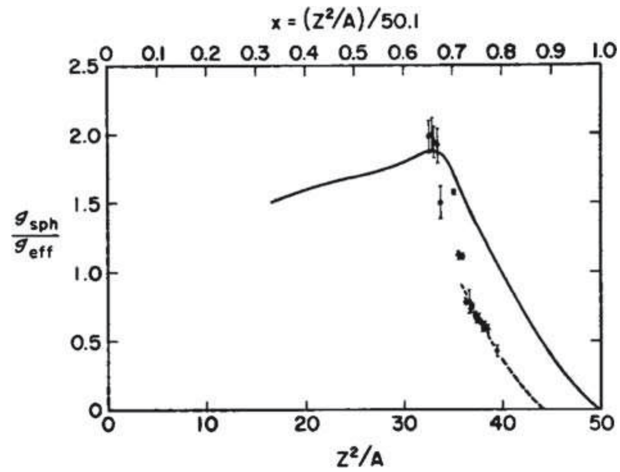

Figure 3. The deformation of the saddle point is given as a function of Z^2/A for the compound nucleus. Solid points are the data compared with the LDM prediction represented by the solid curve.

as manifestations of the nuclear shell effects on the shape of the fission barrier [13]. The shape resonances were, however, observed about a decade earlier by Northrup *et al* [14] and they were even seen in ^{232}Th(n, f) data (figure 3b) that were presented in a declassified report which appeared in 1952 [15]. The correct interpretation of these resonances was first given in [12]. This happened just as I started my Ph.D. studies at the Niels Bohr Institute in Copenhagen, where many of the key players in the newly invigorated field of fission studies were resident at that time, both theorists and experimentalists. I got involved in fission probability measurements using charged particle reactions to derive fission barrier parameters both in Copenhagen [16,17] and in Los Alamos [18–21]. During my stay at Los Alamos it was realized that several other processes, besides the standard (d, p) single neutron transfer reaction, could be used to measure fission barriers for many other species that had not been studied earlier (figure 4).

4. Heavy-ion fission angular distributions

A central theme in nuclear physics was the study of heavy-ion-induced reactions, which was enabled by several new facilities set up in the 1970s. Heavy-ion beams allow one to impart large angular momenta to the compound nucleus and to reach much heavier systems for which the fission barrier was predicted to vanish according to the LDM. This gave the incentive to study fission angular distribution in ^{32}S bombardment of some heavy targets to test these naive predictions [22]. The results showed a strong deviation from theory and further experiments were carried out to determine whether this deviation was caused by the much larger angular momenta brought into the system compared to the previous measurements. Subsequent measurements which compared the ^{16}O+^{238}U and ^{32}S+^{208}Pb systems [23], both of which lead to compound systems with a fissility of $x = 0.84$ and therefore should have identical fission barrier shapes, clearly established that the

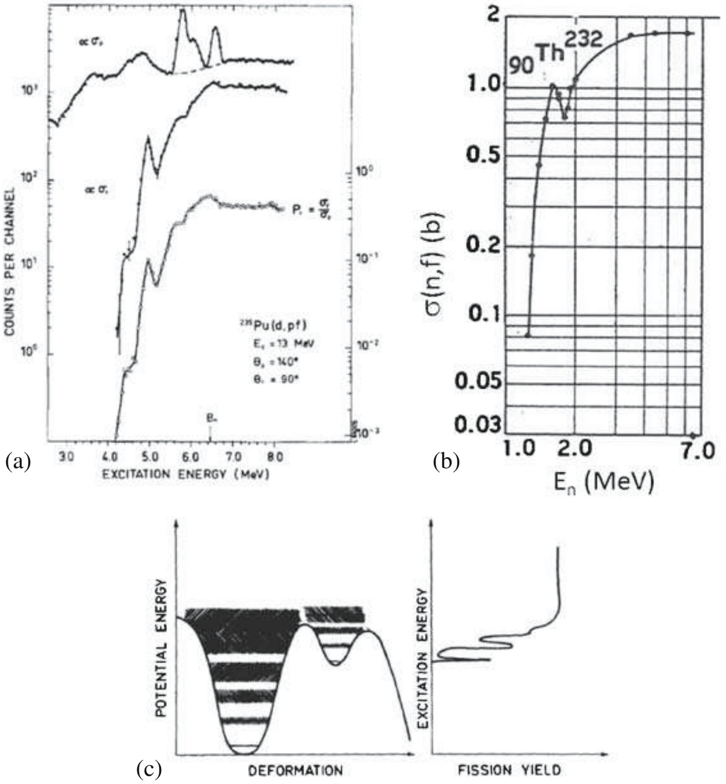

Figure 4. (**a**) ^{239}Pu(d, pf) showing a strong shape resonance at $E_x \sim 5$ MeV [16]. (**b**) Resonance in ^{232}Th(n, f) at $E_n \sim 1.5$ MeV [15]. (**c**) Illustration of the enhancement of the fission cross-section at the location of beta-vibration resonances in the second well of the fission barrier.

excess anisotropy was not associated with the angular momentum but rather was an effect of the mass asymmetry in the entrance channel. The higher anisotropy observed in the ^{32}S+^{208}Pb system was thus a clear evidence that the K-distribution is not fully relaxed to the value that reflect a statistical equilibrium at the fission saddle point but that this relaxation has been interrupted by a shorter, dynamical time evolution of the system [24] which is consistent with the expectations for the quasifission process (figure 5).

5. Mass–angle distributions and quasifission

The conclusion about quasifission appeared to be quite well justified. However, it relied on the expectations of the LDM and it was clearly desirable to obtain data that were devoid of this model dependency. However, almost concurrently, incontrovertible evidence for the dynamical nature of the quasifission process was obtained by Bock *et al* [27] at GSI in Germany. As shown in figure 6, a strong correlation between the fragment mass and the scattering angle is seen in the ^{208}Pb+^{58}Fe reaction, which clearly

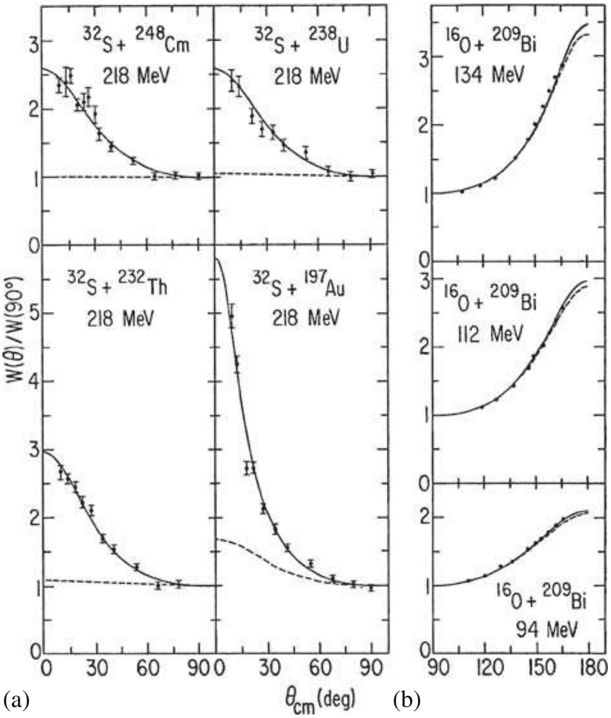

Figure 5. (**a**) The fission angular distributions in ^{32}S-induced reactions (solid points and curves) [22] are compared with the predictions of the LDM [25] (dashed curves). (**b**) Same as panel (**a**) but for ^{16}O-induced fission [26].

demonstrates that the quasifission fragments are emitted on a time-scale that is significantly shorter than the rotational period of the system. For a completely fused system, one would expect that the memory of the initial orientation of the ^{208}Pb+^{58}Fe complex would be lost before the subsequent fission process would occur.

Subsequent studies of many heavy systems [28–33] have shown that the quasifission process is a ubiquitous feature of heavy-ion reactions that depletes the cross-section for complete fusion. For the heaviest systems, including both the heavy targets and the projectiles, that are needed to synthesize new, superheavy elements, this depletion contributes to the strong suppression of the formation cross-section, in addition to the loss to normal compound fission competition during the neutron-emission cascade to reach the ground state of such nuclei. One way to estimate the maximum cross-section that can come from true compound-nucleus fission was suggested in [32] and is illustrated in figure 7, where the cross-sections are shown for different fragment mass bins as $\mathrm{d}\sigma/\mathrm{d}\theta$ for the ^{60}Ni+^{154}Sm system at three different energies. As required for a two-body final state, one observes that the cross-section for symmetric mass division obeys forward–backward symmetry, while for asymmetric mass splits, the violation of this symmetry increases with mass asymmetry. As the compound nucleus fission fraction must obey the forward symmetry, only a decreasing fraction of the fission-like cross-section can be attributed to this process for the more asymmetric mass splits. Although this type of

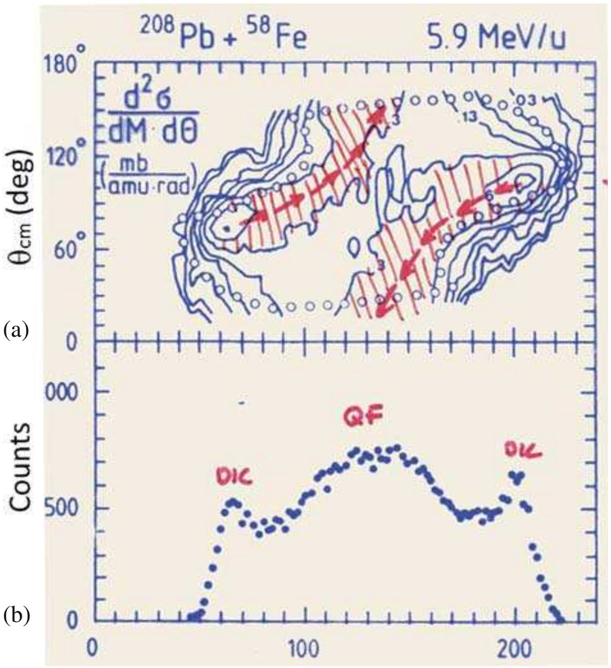

Figure 6. (**a**) Mass–angle contour map showing the quasifission region (highligted) for the ^{208}Pb+^{58}Fe reaction at 5.9MeV/u. The open circles represent the region of acceptance of the detector system. (**b**) Mass distribution for the same reaction. The contributions from deep inelastic scattering and quasifission are indicated [27].

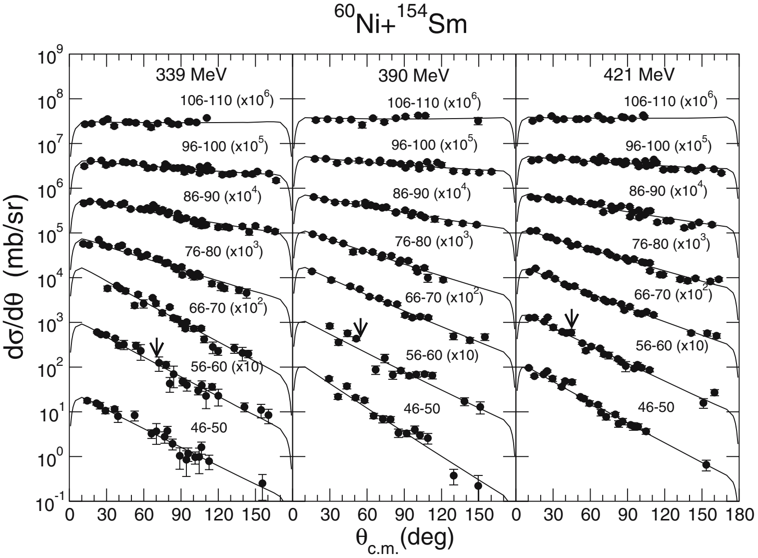

Figure 7. Differential cross-sections for fission-like fragments for the ^{60}Ni+^{154}Sm reaction for different fragment mass bins. The solid points correspond to the mass bin as indicated. The solid lines represent best fits to the data [32].

analysis gives some information about the complete fusion cross-section, it is important to keep in mind that it provides only an upper bound on this quantity; the fact that the angular distribution at mass symmetry exhibits forward–backward symmetry is only a simple kinematic constraint for a two-body exit channel and it does not provide any indication of whether a compound nucleus was formed as an intermediate step in the process. The true compound nucleus cross-section could have orders of magnitudes lower than observed.

6. Early development of fission research in India

After this discussion of the fission process, I would like to return briefly to the historical perspective, but this time in the context of Indian science. We are celebrating the 75th anniversary of the discovery of the fission process at the research centre that is named after Homi Bhabha, who played a central role for the development of nuclear science in India. It is interesting to reflect upon the connections and circumstances that resulted in this development. In 1939, Dr Bhabha was returning to India for a vacation from his research in Cambridge, but with the start of World War II that year, he decided to stay in India and take up a position as reader at the Indian Institute of Science

Figure 8. The men behind the early research on fission in India. Homi Bhabha (**a**) took advantage of his close friendship with Jawaharlal Nehru (**b**) and his family connections to J R D Tata, Trustee of the Tata Foundation (**c**) that was established by his uncle, Sir Dorabji Tata (**d**).

in Bangalore. Meanwhile, he realized the potential of exploiting the practical uses of the recently discovered fission process and worked to establish a nuclear research programme in India. Fortunately, Homi Bhabha was very well connected to people in power through his friendship with Jawaharlal Nehru, senior member of the Congress Party and later the first Prime Minister of India and J R D Tata, a distant relative, who was a trustee of the Tata Trust that had been established in 1932 by Bhabha's uncle, industrialist Sir Dorabji Tata (see figure 8).

Bhabha's efforts led to the establishment of the Tata Institute of Fundamental Research, which since 1962 occupies a beautiful building (designed by a Chicago-based architect, Helmuth Bartsch) on military land on the Colaba peninsula in Mumbai that Bhabha was able to acquire through his political connections. When the need for more space to build a larger nuclear installation became apparent, Homi Bhabha was again instrumental in establishing the Atomic Energy Establishment in Trombay, which was later re-named the Bhabha Atomic Research Centre in his honour, the venue of this conference.

With this celebration of the discovery of fission 75 years ago, one cannot help but reflect on the enormous effects this discovery has had on human society and science. As scientists, we often take it for granted that governments give strong support and substantial funding to fundamental, curiosity-driven science, but I think that it is fair to say that this level of government support was really started with the discovery of nuclear fission. This discovery demonstrated that obscure research, carried out by university professors and their assistants in small laboratories that seem to have no other purpose than to satisfy the curiosity of the people engaged in the work, can have dramatic, society-altering consequences. Homi Bhabha realized this connection and prepared the ground for a large and very successful effort in fundamental research into the fission process as well as many other disciplines in India.

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