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Authors
Loveland, W.
Casey, C.
Xu, Z.

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Lawrence Berkeley Laboratory
UNIVERSITY OF CALIFORNIA

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W. Loveland, C. Casey, Z. Xu, G.T. Seaborg,
K. Aleklett, and L. Sihver

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By

Walter Loveland, C. Casey, and Z. Xu
Oregon State University
Corvallis, OR 97331

Glenn T. Seaborg
Lawrence Berkeley Laboratory
Berkeley, CA 94720

K. Aleklett and L. Sihver
Studsvik Neutron Research Laboratory
S-611 82 Nykoping, Sweden

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W. LOVELAND, C. CASEY, Z.XU
Oregon State University
Corvallis, OR 97331, USA
(503)-754-2341

K. ALEKLETT, L. SIHVER
Studsvik Neutron Research Lab.
S-611 82 Nyköping, Sweden
+46-155-21000

G.T. SEABORG
Lawrence Berkeley Laboratory
Berkeley, CA 94720, USA
(415)-486-5661

ABSTRACT

We have measured the target fragment yields, angular and energy distributions for the interaction of 12-16 MeV/A ^{32}S with ^{165}Ho and ^{197}Au and for the interaction of 32 and 44 MeV/A ^{40}Ar with ^{197}Au. The Au fission fragments associated with the peripheral collision peak in the folding angle distribution originate in a normal, "slow" fission process in which statistical equilibrium has been established. At the two lowest projectile energies, the Au fission fragments associated with the central collision peak in the folding angle distribution originate in part from "fast" ($\tau \sim 10^{-23}$s), non-equilibrium processes. Most of the Ho fission fragments originate in non-equilibrium processes. The fast, non-equilibrium process giving rise to these fragments has many of the characteristics of "fast fission", but the cross sections associated with these fragments are larger than one would expect from current theories of "fast fission."

INTRODUCTION

Due to the large scale collective motions involved, fission is generally thought of as a "slow" process. For heavy nuclei excited by the resonance capture of neutrons, fission lifetimes can be measured to be $\sim 10^{-15}$ - 10^{-14}s. When one increases the temperature of the fissioning system, the fission lifetimes are expected to decrease, due to the overall decrease in the lifetime of the excited nucleus and the vanishing of the fission barrier. More specifically, for a heavy nucleus like ^{208}Pb, the neutron decay lifetime at a temperature of 5 MeV is estimated[1] to be $\sim 10^{-22}$s. The fission barrier of ^{208}Pb is expected[2] to vanish at T=5 MeV even with $l=0$. Thus there are reasons to expect that in highly excited nuclei, fission will become a much faster process especially if it is to compete with particle emission as a decay path for highly excited nuclei.

In this paper we report studies of fission induced by intermediate energy heavy ions that are fairly massive (S,Ar). Using the symmetry of the moving frame fragment angular distributions as a clock, we report the observation of a fast, non-equilibrium fission process whose lifetime is of the order of a nuclear relaxation time.

In the reactions induced by energetic S and Ar ions, the expected values of the nuclear temperatures are 4-6 MeV, possibly giving rise to an unusual setting for the fission process, i.e., a nonexistent fission barrier and competiting decay channels with short lifetimes.

EXPERIMENTAL

We have measured the target fragment yields, and angular distributions for the interaction of 12-16 MeV/nucleon ^{32}S with ^{165}Ho and ^{197}Au, 32 and 44 MeV/nucleon ^{40}Ar with ^{197}Au. Also the fragment energy spectra were measured for the interaction of 16 MeV/nucleon ^{32}S with ^{165}Ho. The experiments were performed at the LBL 88" cyclotron (^{32}S beam), the MSU National Superconducting Cyclotron (32 MeV/nucleon ^{40}Ar) and at GANIL (44 MeV/nucleon ^{40}Ar). The experimental apparatus, the methods used to acquire the data and to analyze it have been described previously[3,4]. The measurements were made using radioanalytical techniques. The corrections to the angular distributions for fragment scattering and the finite angular resolution of the detection apparatus are discussed in reference 4. The measurements of the target fragment production cross sections at LBL and MSU were made by a simple irradiation of a thick Ho or Au foil surrounded by ~15 mg/cm^2 carbon catcher foils. The radionuclide content of the irradiated foil stack was determined by off-line gamma ray spectroscopy. Production cross sections were calculated from end of bombardment activities[5]. (For the GANIL irradiation, the total nuclidic production cross sections were determined by integrating the measured fragment angular distributions.)

For the reaction of 16 MeV/nucleon ^{32}S with ^{197}Au, the angular distributions of 49 different target fragments were measured along with the production cross sections for 102 different nuclides. For the reaction of 32 MeV/nucleon ^{40}Ar with ^{197}Au, angular distributions were measured for 40 fragments while the yields of 83 fragments were measured. In the reaction of 44 MeV/nucleon ^{40}Ar with ^{197}Au, the angular distributions and yields of 78 different target fragments were measured. In the reaction of 12-16 MeV/nucleon ^{32}S with ^{165}Ho, the yields of 75 different radionuclides were measured along with the angular distributions of 82 different target fragments. From the measured target fragment production cross sections, fragment mass yield distributions were deduced using techniques described previously[6].

EVIDENCE FOR UNUSUAL ASPECTS OF FISSION

In Figures 1 and 2, we show the deduced mass yield curves for the two reactions of 12-16 MeV/nucleon ^{32}S with ^{165}Ho and ^{197}Au. Also shown in these figures are the isobaric yield distributions from reactions induced by similar velocity ^{16}O ions (as well

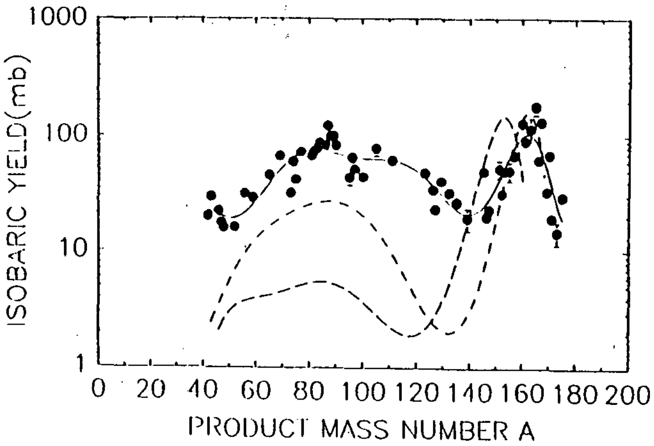

Figure 1. Isobaric yield distributions for the fragmentation of ^{165}Ho by (a) 12 MeV/nucleon ^{32}S, solid points, solid line (b) 17 MeV/nucleon ^{16}O, short dashed line (c) 442 MeV ^{12}C, long dashed line.

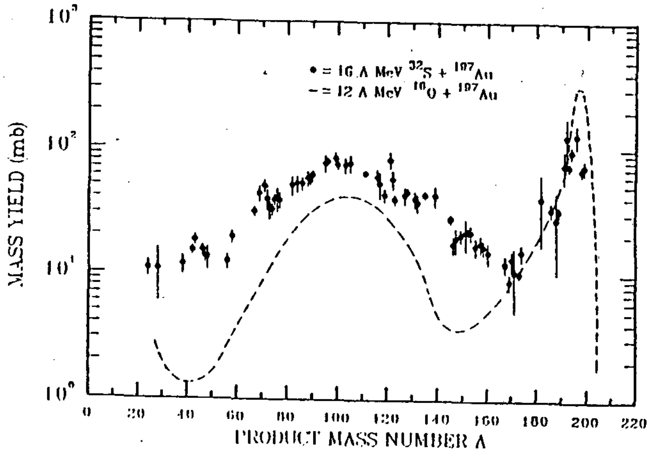

Figure 2. Comparison of isobaric yield distributions for the fragmentation of ^{197}Au by (a) 12 MeV/nucleon ^{16}O, dashed line and (b) 16 MeV/nucleon ^{32}S.

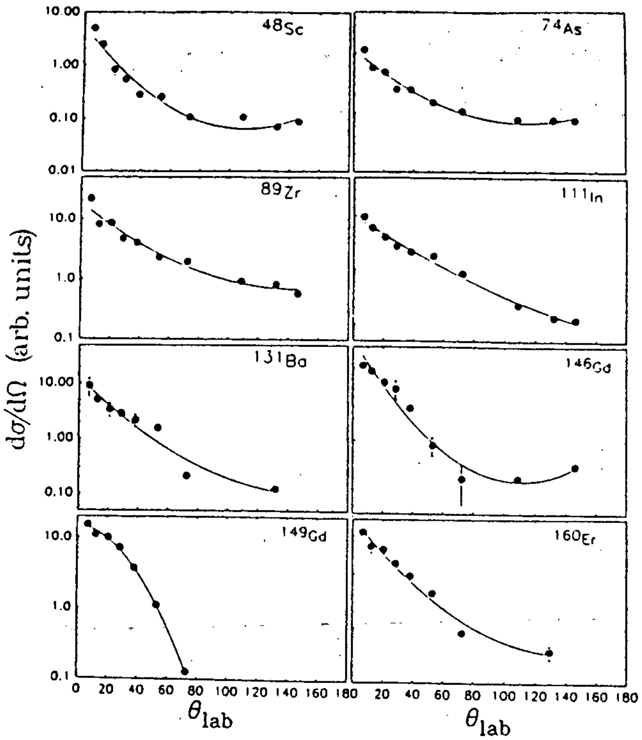

Figure 3. Laboratory frame angular distributions for representative fragments from the reaction of 16 MeV/nucleon ^{32}S with ^{165}Ho.

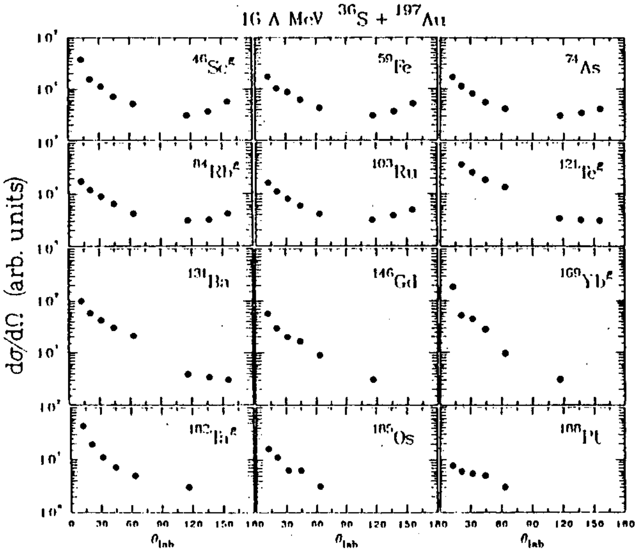

Figure 4. Laboratory frame angular distributions for representative fragments from the reaction of 16 MeV/nucleon ^{32}S with ^{197}Au.

as ^{12}C ions of similar total projectile kinetic energy interacting with ^{165}Ho). One notes two prominent peaks in the mass distribution, a fission peak (A~50-150) and a heavy residue peak(A>150). The fission cross section (σ_f(Ho)=2060 mb, σ_f(Au)=2600 mb). is enhanced for the ^{32}S induced reactions relative to the other (C,O) reactions . The yields of the heavier fission products are especially enhanced. The overall width of the fission mass distribution is unusually broad.

In Figures 3 and 4, we show the laboratory frame angular distributions for a series of typical fragments from the interaction of 16 MeV/nucleon ^{32}S with ^{165}Ho and ^{197}Au. (For lack of space, we omit a detailed discussion of the angular distributions for the two higher energy reactions although similar conclusions can be reached for these reactions'.) The laboratory frame angular distributions are all strongly forward-peaked. For the reactions involving the Au target, the light mass fission fragments (A=40-106) have a similar "dipper" shape while the heavier members of this distribution (A=111-169) have a very different shape. This latter group of fragments exhibits more forward-peaked distributions similar to those observed for the heavy residues.

Each fragment angular distribution was integrated from 0 to $\pi/2$ and $\pi/2$ to π to obtain the ratio of fragments recoiling forward (F) to those recoiling backward (B). To extract further information from the data, the laboratory frame angular distributions were transformed into the moving frame of the target residue following the initial target-projectile encounter. To do this we have assumed that the final velocity of the fragment in the laboratory system can be written as $V_{lab} = V + v$, where the velocity v is the velocity of the moving frame and V is the velocity kick given the target fragment by particle emission or fission at an angle θ_{MF} with respect to the beam direction in the moving frame. The vector v has components of $v_{\parallel}$ and v_{t}, parallel and perpendicular to the beam direction.

In lieu of detailed information about v_t, the forward-peaked nature of the distributions and the difficulty of getting information about v_t, we have assumed $v_t=0$. We have used standard formulas[8] to make transformations for $d\sigma/d\Omega$ and θ.

For the value of $\eta_\parallel$ ($=v_\parallel/V$) needed to make such transformations, we have used values of $\eta_\parallel$ derived from integrating the angular distributions. To get the value of $\eta_\parallel$ from F and B, we assume the angular distributions of the fission fragments in the moving frame can be represented as $1 + \alpha \cos^2 \theta_{MF}$. In this case, it can be shown[8] that

$$F=\tfrac{1}{2}[1+(1+\eta^2\alpha/3)\eta/(1+\alpha/3)]$$

$$B=\tfrac{1}{2}[1-(1+\eta^2\alpha/3)\eta/(1+\alpha/3)]$$

These equations were solved numerically using a Levenberg - Marquardt method to give values of η and α. (The values of α range from 0 to 0.2 but were mostly ~0).

The values of $\eta_\parallel$ obtained from this procedure for the reactions involving Au targets are shown in Figure 5. The values of $\eta_\parallel$ change as a function of fragment mass number with high $\eta_\parallel$ values being associated with the heavy fission fragments and low $\eta_\parallel$ values being associated with the lighter fission fragments. The average values of $\eta_\parallel$ for each fragment group agree well with previous measurements of $\eta_\parallel$ for the same or similar reactions using the fission fragment folding angle technique[9-11]. Thus the heavy fission fragments for the two lower projectile energies appear to have $\eta_\parallel$ values characteristic of central collisions (high momentum transfer) while the light mass fission fragments appear to have $\eta_\parallel$ values characteristic of peripheral collisions(low momentum transfer). For the highest projectile energy (44 A MeV ^{40}Ar), where the fission mass distribution extends from A=70 to A=110) fission only occurs with $\eta_\parallel$ values characteristic of peripheral collisions. The $\eta_\parallel$ values associated with the Ho fission fragments are all large and characteristic of central collisions. That the Ho fission events are associated with high linear momentum transfer events is not surprising given the small fissionability of Ho and the need to impart substantial amounts of

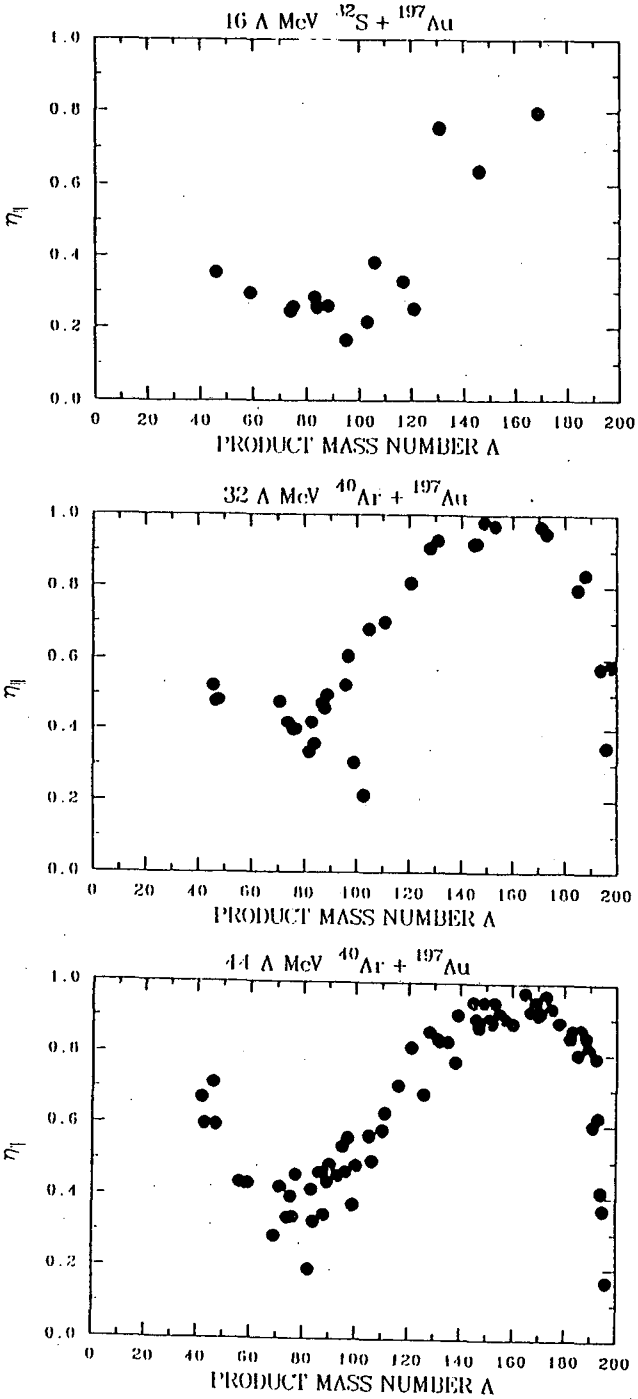

Figure 5. Values of $\eta_\parallel$ as a function of product mass number for all the reactions involving Au targets.

angular momentum to cause fission[11]. It would be an oversimplification, however, to believe that for Au, the high linear momentum transfer events lead strictly to heavy mass fragments and low momentum transfer events lead to light mass fragments. The situation is probably more complicated as we shall discuss presently.

To gain further insight into what might be happening, we show (in Figures 6 and 7) the moving frame angular distributions corresponding to the data shown in Figures 3 and 4. All the events with high values of η_l have angular distributions that are asymmetric with respect to 90° in the moving frame while the events with low values of η_l have symmetric distributions. Symmetry in the moving frame implies a "slow" process in which statistical equilibrium has been achieved while the lack of symmetry implies a "fast" process in which statistical equilibrium has not been established. (The terms "slow" and "fast" are to be taken relative to the time required for the establishment of statistical equilibrium which has been estimated[13] to be 2-3 x 10^{-23}s.) Furthermore it can be shown that for many of the fragments no choice of a value (or a set of values) of η_l will lead to a symmetric distribution in the moving frame. We believe that this unique observation suggests the occurrence of a fast, non-equilibrium mode of fission for these fragments.

As to the puzzling observation that the heavy mass Au fission fragments preferentially show this "fast" production mechanism, it can be argued that "normal, slow" fission will always occur to produce these fragments. But that these fragments are also produced by a fast, non-equilibrium process. The fast non-equilibrium process is assumed to have an unusually broad mass distribution, thus making its relative importance greater for the more asymmetric fission events. Using the 32 MeV/nucleon ^{40}Ar + ^{197}Au reaction as an example, this argument may be carried further. If we assume the distribution for the light mass fragment ^{87}Y is representative of normal fission, we can normalize it to the distribution of a typical heavy mass fragment

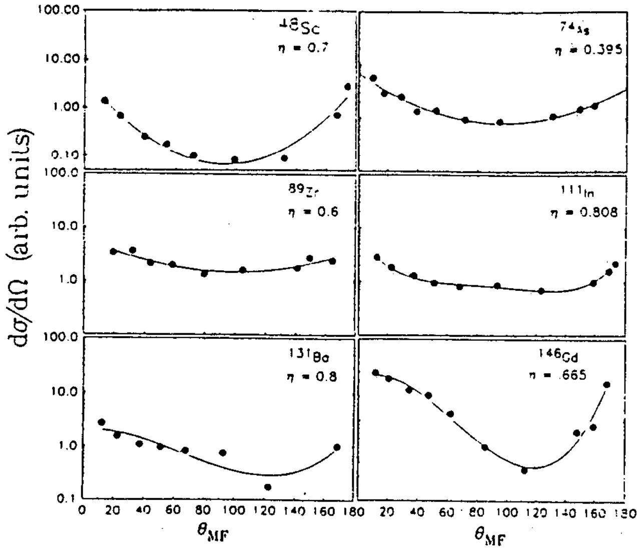

Figure 6. Moving frame angular distributions for representative fragments from the interaction of 16 MeV/nucleon ^{32}S with ^{165}Ho.

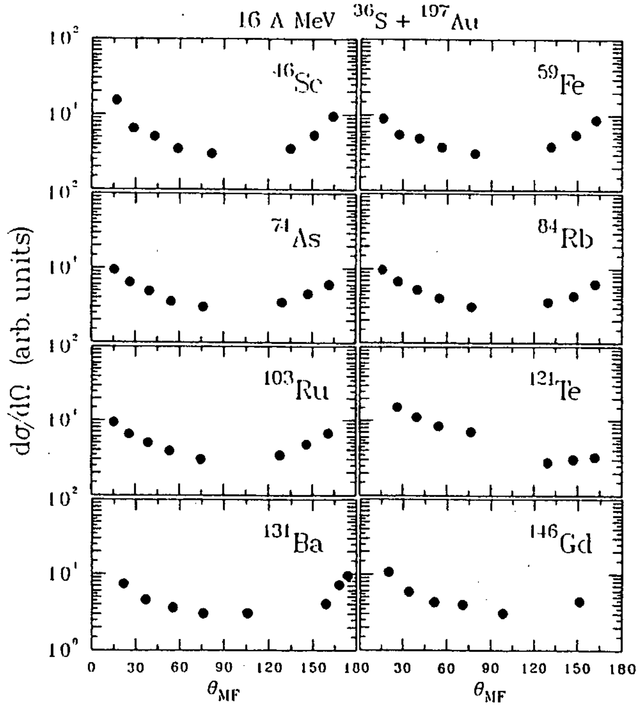

Figure 7. Moving frame angular distributions for representative fragments from the interaction of 16 MeV/nucleon ^{32}S with ^{197}Au.

(^{131}Ba) at backward angles (Figure 8a). The difference between the two distributions (Figure 8b) can be taken as the contribution of a "fast" direct fission mechanism to the production of ^{131}Ba and other heavy mass fragments.

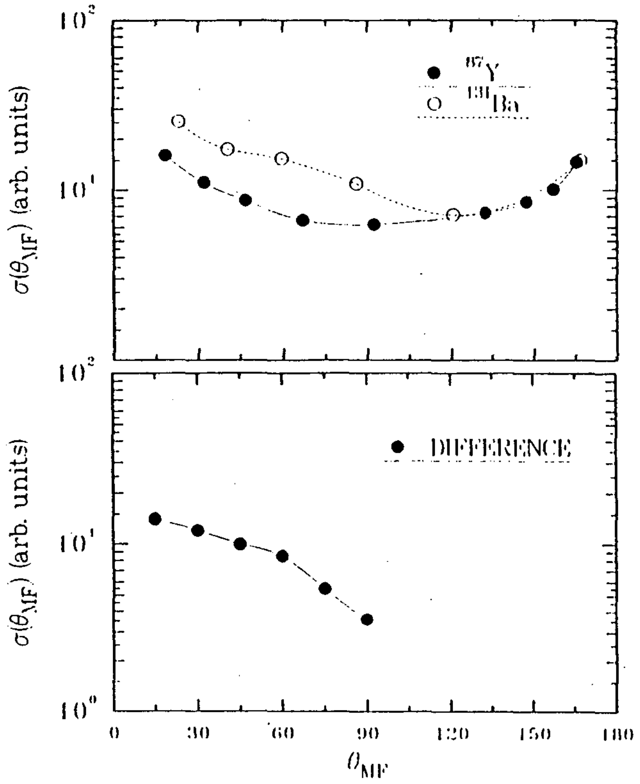

Figure 8. (a) The moving frame distributions of ^{87}Y and ^{131}Ba from the 32 MeV/nucleon ^{40}Ar + ^{197}Au reaction normalized at back angles. (b) The difference between the distributions in (a).

SPECULATIONS ABOUT THE REACTION MECHANISMS INVOLVED.

A known nuclear reaction mechanism[14] for low energy nuclear collisions, "fast fission" or "quasifission" is a possible candidate for the suggested non-equilibrium mechanism. In this mechanism, all partial waves between the l-wave at which the fission barrier vanishes, $l_{Bf=0}$, and the critical angular momentum, l_{crit} go via fast fission. In these events, the fusing system never reaches a configuration inside the fission saddle point and the resulting event is fast. Experimental signatures for such events are the lack of symmetry of the moving frame angular distributions and a broader than normal fission mass distribution. The principal difficulty with this explanation is the magnitude of the measured cross sections for the "fast" events. By using the difference technique described above, one estimates "fast" cross sections for the Au target reactions that are ~2.9x the expected[14] fast fission cross section. For the Ho target reaction, the measured "fast" cross section is about the twice the expected fast fission cross section.

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REFERENCES

1. H. DELAGRANGE, C. GREGOIRE, F. SCHEUTER, and Y. ABE, "Dynamical Decay of Nuclei at High Temperature: Competition between Particle Emission and Fission Decay," Z. Phys. A323, 437 (1986).

2. X. CAMPI and S. STRINGARI, "Temperature Dependence of Nuclear Surface Properties," Z. Phys. A309, 239 (1983).

3. K. ALEKLETT, M. JOHANSSON, L. SIHVER, W. LOVELAND, H. GROENING, P.L. MCGAUGHEY, and G.T. SEABORG, "Heavy Residue Spectra in the Interaction of 85 A MeV ^{12}C with ^{197}Au," Nucl. Phys. A (in press).

4. R.H. KRAUS,JR.,W. LOVELAND, K. ALEKLETT, P.L. MCGAUGHEY, T.T. SUGIHARA, G.T. SEABORG, T. LUND, Y. MORITA, E. HAGEBO, AND I.R. HALDORSEN, "Target Fragment Angular Distributions for the Interaction of 86 MeV/A ^{12}C with ^{197}Au," Nucl. Phys. A432, 525 (1985).

5. D.J. MORRISSEY, D. LEE, R.J. OTTO, and G.T. SEABORG, "Measurement of the Product Mass Distributions from Heavy-Ion Induced Nuclear Reactions," Nucl. Inst. Meth. 158, 499 (1978)

6. D.J. MORRISSEY, W. LOVELAND, M. de SAINT-SIMON, and G.T. SEABORG, "Target residues from the reaction of 8 GeV ^{20}Ne with ^{181}Ta and ^{197}Au," Phys. Rev. C21, 1783 (1980).

7. K. ALEKLETT, W. LOVELAND, L. SIHVER, Z.XU, C. CASEY, D.J. MORRISSEY, J.O. LILJENZIN, M. de SAINT-SIMON, and G.T. SEABORG, "Changes in Target Fragmentation Mechanisms with Increasing Projectile Energies in Intermediate Energy Nuclear Collisions," Phys. Rev. C (submitted for publication).

8. J.M. ALEXANDER, in Nuclear Chemistry, Vol I, L. Yaffe, ed., (Academic, New York, 1968) pp273-357.

9. E.C. POLLACCO, M. CONJEAUD, S. HARAR, C. VOLANT, Y. CASSAGNOU, R. DAYRAS, R. LEGRAIN, M.S. NGUYEN, H. OESCHLER, and F. SAINT-LAURENT, "High Momentum and Energy Transfer induced by 1760 MeV ^{40}Ar on ^{197}Au and ^{232}Th Targets," Phys. Lett. 146B, 29 (1984).

10. Y. PATIN, S. LERAY, E. TOMASI, O. GRANIER, C. CERRUTI, J.L. CHARVET, S. CHIODELLI, A. DEMEYER, D. GUINET, C. HUMEAU, P. LHENORET, J.P. LOCHARD, R. LUCAS, C. MAZUR, M. MORJEAN, C. NGO, A. PEGHAIRE, M. RIBRAG, L. SINOPOLI, T. SUOMIJARVI, J. UZUREAU and L. VAGNERON, "Fission Fragment-Light Particle Coincidences and Linear Momentum Transfer," Nucl. Phys. A457, 146 (1986).

11. G. BIZARD, R. BROU, H. DOUBRE, A. DROUET, F. GUILBAULT, F. HANAPPE, J.M. HARASSE, J.L. LAVILLE, C. LEBRUN, A. OUBAHADOU, J.P. PATRY, J. PETER, G. PLOYART, J.C. STECKMEYER, and B. TAMAIN, "Reaction Mechanisms in the ^{40}Ar + ^{197}Au Collisions at 35 MeV/nucleon," Nucl. Phys. A456, 183 (1986)

12. C.CASEY, W. LOVELAND, Z.XU, L. SIHVER, K. ALEKLETT, and G.T. SEABORG, "Non-Equilibrium Fission and Heavy Residue Production in the Interaction of 12-16 MeV/nucleon ^{32}S with ^{165}Ho," Phys. Rev. C (submitted for publication)

13. K. ALEKLETT, W. LOVELAND, T. LUND, P.L. MCGAUGHEY, Y. MORITA, G.T. SEABORG, E. HAGEBO and I.R. HALDORSEN, "Fast and slow processes in the fragmentation of ^{238}U by 85 MeV/nucleon ^{12}C," Phys. Rev. C33, 885 (1986).

14. C. GREGOIRE, C. NGO, AND B. REMAUD, "Fast Fission Phenomenon, Deep Inelastic Reactions and Compound Nucleus Formation Described Within a Dynamical Macroscopic Model," Nucl. Phys.A383, 392 (1982).

LAWRENCE BERKELEY LABORATORY
TECHNICAL INFORMATION DEPARTMENT
1 CYCLOTRON ROAD
BERKELEY, CALIFORNIA 94720