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# Comparison of 14 MeV Isomer Production of $^{178m2}$Hf and $^{179m2}$Hf Using Feshbach-Kerman-Koonin and Exciton Preequilibrium Models

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**The $^{178m2}$Hf($16^+$) isomeric state has a 31-yr half life and could pose serious radioactive problems in nuclear fusion reactors if its production in 14 MeV neutron-induced reactions is significant. We present statistical/preequilibrium model calculations for the production of this isomer in the $^{179}$Hf$(n,2n)^{178m2}$Hf reaction, as well as the 25-days $12.5^-$ isomer in the $^{179}$Hf$(n,n')^{179m2}$Hf reaction, using two different preequilibrium models: the exciton model and the Feshbach-Kerman-Koonin (FKK) theory. Our calculations which use the exciton model agree well with measurements, but those with the FKK theory underestimate measurements. Our calculations are the first to probe angular momentum transfer effects in the FKK theory and suggest that, as it is presently applied, high spin-transfer reactions are underestimated. We suggest modifications to the FKK statistical averaging procedure which may result in an improved agreement with experiment.**

## I. INTRODUCTION

The International Atomic Energy Agency (IAEA) Nuclear Data Section has established a Coordinated Research Programme (CRP) on activation cross sections for the generation of long-lived radionuclides of importance in radioactive waste problems in fusion reactor technology [1]. A number of reactions of particular importance were selected, and the CRP organized both experimental and theoretical efforts to determine production cross sections for these reactions. One reaction being considered is the $^{179}$Hf$(n,2n)^{178m2}$Hf ($J^\pi = 16^+$), the isomeric state having a 31 year half life. There is a possibility that small amounts of tungsten and tantalum be included in the first-wall material of the fusion reactor, and since the neutron fluxes are expected to be so high these nuclides might first be transmuted to hafnium and then undergo $(n, 2n)$ reactions to produce the long-lived $16^+$ isomer. This state, if formed in sufficient quantity, could lead to the first wall being active for a long time after it is removed from the reactor.

To assess the production of this isomer both experimental measurements and theoretical calculations are being performed. Indeed, in 1990 we performed calculations of this isomer [2] with the GNASH nuclear model code before experimental measurements were made, and obtained results which agreed to within a factor of two with the later measurement by Patrick *et al.* [3]. Taking into account the very small cross sections for isomer production (due to the high spin), and the uncertainties in the theoretical modeling (particularly in the nuclear structure and optical potentials) our results were very encouraging. Since then, more measurements [4] have been made which have verified Patrick *et al.*'s initial result. Furthermore, a number of developments to the GNASH code have been made which led us to recalculate isomer production cross section. Even though experimental data exists at 14 MeV, they do not at lower energies, and thus theoretical calculations are of importance since they can be normalized to the measurements at 14 MeV and used to provide cross sections for lower neutron energies.

In this paper we apply the improved GNASH code system to calculate 14 MeV neutron reactions on $^{179}$Hf to both the $16^+$ isomer in $^{178}$Hf and the $12.5^-$ isomer in $^{179}$Hf. Two calculations were compared which utilize different modifications to the code: firstly, the exciton model preequilibrium calculation was modified so that the spin distribution of residual nuclei after preequilibrium decay is obtained from the exciton spin-dependent level density [5], and not from the compound-nucleus spin distribution; and secondly, a version of the code, FKK-GNASH [6], which uses the quantum mechanical Feshbach-Kerman-Koonin (FKK) [7] preequilibrium model.

The FKK theory has been used with success in calculating a range of different reactions, with incident nucleon energies from 10 to 200 MeV [8]. The present work is the first application of the FKK theory to probe angular momentum transfer effects in the multistep direct preequilibrium emission, and enables a comparison to be made between angular momentum effects in the exciton and FKK models, since the amount of high-spin isomer production depends sensitively on these effects. Most exciton models predict the spin-distribution of residual nuclei after preequilibrium emission from simple phase-space arguments and thus it is interesting to investigate the predictions of the FKK theory, which explicitly determines the spin-transfer contributions to the preequilibrium cross section. Soon after FKK presented their theory, Herman, Marcinkowski and Stankiewicz [9] applied the multistep compound theory to calculate $(n, 2n)$ reactions and some isomer production cross sections. However, recent work [6] on this theory has shown that the multistep compound mechanism is much less important than multistep direct mechanism even for incident energies below 20 MeV.

In Section II we describe the FKK theory, the exciton model, and the compound nucleus model used, and in Section III we present our results, comparing both calculations with each other and with data. Our conclusions and suggestions for future work are given in Sec. IV.

## II. THEORY

### A. The FKK Theory

The quantum mechanical multistep theory of FKK describes preequilibrium reactions as passing through a series of particle-hole states of increasing complexity towards equilibrium. Emission from the early particle-hole states yields the preequilibrium emission. Two different mechanisms are considered: multistep compound (MSC) and multistep direct (MSD) emission, which we describe below.

MSD reactions occur when the unbound chain of states is populated so that at least one particle is in the continuum. This particle maintains a 'memory' of the initial direction of the projectile as it creates $p$-$h$ states through scatterings with bound nucleons, and results in forward-peaked emission.

MSD theory represents an extension of distorted-wave-Born-approximation theory (DWBA) into the continuum, and can be derived from a Lippmann-Shwinger expansion of the transition amplitude [10]. For the incident energies that we consider, only one-step scattering needs to be included [6], and the one-step cross section is given by

$$\frac{d^2\sigma(E,\Omega \leftarrow E_0,\Omega_0)}{d\Omega dE}_{1-step} = \sum_l (2l+1)\rho(1p,1h,E_0-E,l)\left\langle\left[\frac{d\sigma(E,\Omega \leftarrow E_0,\Omega_0)}{d\Omega}\right]_l^{DWBA}\right\rangle, \tag{1}$$

where:-

- $l$ is the orbital angular momentum transfer, which equals the spin of the $1p$-$1h$ excitation spin since we follow FKK in assuming zero spin-transfer. As we discuss later, we remove the usual FKK assumption of a target spin-zero, and instead we couple in the target nucleus ground-state spin to that of the $p$-$h$ excitations when determining the residual nucleus spin distribution after preequilibrium emission.
- $\rho(1p,1h,E_0-E,l)$ is the density of $1p1h$ states with energy $E_0-E$ and spin $l$. In general the density of states for a $p$-particle $h$-hole system can be partitioned into the energy-dependent density multiplied by a spin distribution, $\rho(p,h,E,l) = \omega(p,h,E)\ R_n(l)$. We use the Williams [11] equidistant expression with finite well-depth restrictions [12]

$$\omega(p,h,E) = \frac{g^n}{p!h!(n-1)!}\sum_{j=0}^{h}\binom{h}{j}(E-\Delta-A_{ph}-j\epsilon_F)^{n-1}\Theta(E-\Delta-A_{ph}-j\epsilon_F), \tag{2}$$

where $n = p + h$, and we take the single particle spacing as $g = A/13$. The Pauli-blocking factor is $A_{ph} = [p^2 + h^2 + p - 3h]/4g$, $\epsilon_F$ is the Fermi energy which we take as 35 MeV, and the pairing energy corrections ($\Delta$) of Dilg *et al.* [13] are used. The $\Theta$-function is unity if its argument is greater than zero, and zero otherwise. A Gaussian angular momentum distribution is assumed,

$$R_n(l) = \frac{2l+1}{2\sqrt{2\pi}\sigma_n^3}\exp\left[-\frac{(l+1/2)^2}{2\sigma_n^2}\right], \tag{3}$$

with spin cut-off, $\sigma_n^2 = 0.24nA^{2/3}$ [14].

- $\left\langle \left[\frac{d\sigma(E,\Omega\leftarrow E_0,\Omega_0)}{d\Omega}\right]_l^{DWBA} \right\rangle$ is the average of DWBA cross sections exciting $1p1h$ states of energy $E_0 - E$ consistent with angular momentum and parity conservation. The $1p1h$ states are obtained from a spherical Nilsson model [15].

We calculate the DWBA form factors for the various transitions with DWUCK4 [16] using a Yukawa potential of range 1 fm, and strength $V_0$, for $1p1h$ excitations. We have used $V_0$=30 MeV in accordance with systematics developed in FKK analyses [17]. In this calculation, for each 1 MeV energy bin, we average about ten microscopic cross sections for each $l$-transfer), and we have found it important to include twelve values of $l$-transfer in Eq. (1) to ensure that all possible $1p1h$ excitation strength is accounted for (and we include microscopic form factors leading to both neutron $p$-$h$ and proton $p$-$h$ excitations). When calculating the form factors, unbound-state wavefunctions were obtained from optical-potential scattering states using the Wilmore-Hodgson potential [18], and bound-states from a real Wood-Saxon potential well with radius parameter 1.2 fm and diffuseness 0.6 fm. We apply a Gaussian smoothing to our calculated MSD cross sections of width 2 MeV, to remove artificial fluctuations which would not arise if we used deformed Nilsson single-particle states.

MSC processes occur when a chain of $p$-$h$ states is populated in which all the particles are bound. The MSC contribution is greatest for low incident energies, becoming insignificant at high incident energies (above about 50 MeV), due to the decreasing probability of forming bound preequilibrium states with increasing energy. Matrix elements for MSC emission involving different total angular momenta, parity, and other quantum numbers, are assumed random so that no interference terms remain on averaging, yielding angular distributions that are symmetric about 90°.

Recent analyses of 14 MeV $(n, n')$ reactions has pointed to the minor importance of MSC in comparison with MSD, and our calculations for $^{179}$Hf$(n, xn)$ confirm this: our calculated MSC spectrum is less than 20% of the MSD spectrum. Due to the relative unimportance of MSC we do not present the MSC formalism here, but refer to Ref. [6] for full details.

Our calculations also include crossover processes in which preequilibrium states in the MSD chain enter the MSC chain before nucleon emission, a mechanism which a number of recent works have provided evidence for [6, 19].

## B. The Exciton Model

The GNASH code system uses the exciton model of Kalbach [20] to calculate preequilibrium emission. This exciton model is spin-independent, and until recently the code assumed that the spin distribution of residual nuclei left after preequilibrium emission can be taken from the Hauser-Feshbach calculated spin distribution. We have removed this assumption, and obtain the spin distribution of residuals after preequilibrium emission by coupling ($(2J+1)$ times) the exciton model $p$-$h$ spin distribution (Eq. 3) to the target, projectile, and ejectile spins. This procedure was found to be important [5] when analyzing specific gamma-ray transitions in residual nuclei in $^{208}$Pb$(n, xn\gamma)$ ($x$=1 to 9) reactions for a white neutron source at LAMPF/WNR, since in this case angular momentum effects are also very important.

We used default parameters for the exciton model calculation: namely, the damping matrix element was taken as 150 MeV$^3$ and single-particle level densities as $A/13$ MeV$^{-1}$.

## C. Compound Nucleus Emission

Compound nucleus decay occurs in cases where the the chain of particle-hole states reaches equilibrium without undergoing preequilibrium emission. It also accounts for the decay of residual nuclei left in excited states after preequilibrium (or equilibrium) emission. We use the Hauser-Feshbach theory to describe compound nucleus decay, which conserves angular momentum and parity in the reaction.

Our Hauser-Feshbach calculations follow closely those in Ref. [2]. In brief, the Gilbert-Cameron [21] level density model with pairing energies from the Cook systematics [22] was used to describe the continuum states, and matched on to low-lying discrete levels. Rotational bands of experimentally undetected states were built upon the $16^+$, $14^-$ levels in $^{178}$Hf and the $12.5^-$ level in $^{179}$Hf, and embedded within the continuum states. Gamma-ray transmission coefficients were calculated with the generalized Lorenzian model of Kopecky and Uhl [23]. The coupled-channel optical potential developed and checked against elastic and total scattering cross sections in Ref. [2] was used. For full details, see our earlier paper [2].

## III. RESULTS

In Fig. 1 we show the neutron emission spectrum for $^{179}$Hf$(n, xn)$ calculated with both the FKK and the exciton models. It is evident that the two models yield rather similar preequilibrium spectras. This suggests that the value of 30 MeV that we used for the residual interaction strength in the MSD calculation, and that we took from systematics, is reasonable.

When we compare the spin distributions of residual nuclei after preequilibrium emission (Fig. 2), significant differences are seen. In both cases the spin distribution peaks at $J = 4.5\hbar$, which is the target (and residual) nucleus ground-state spin. However, the exciton model spin distribution extends to much higher values than the FKK distribution. As discussed earlier, the exciton model residual nucleus spin distribution was obtained solely from phase space. However, the spin distribution from the FKK theory rests upon two factors: the strength of microscopic DWBA transition exciting $p$-$h$ states for various spin transfers; and the $1p1h$ phase space (see Eq. 1). Since the averaged microscopic DWBA cross sections are dependent upon the spin-transfer, the FKK and exciton models yield differing residual nucleus spin distributions. This is likely to result in differences in the production cross sections of high-spin isomers, with higher cross sections being expected for the exciton model calculations.

In Table I we compare FKK and exciton model calculations with experimental data. The different spin distributions obtained using FKK and exciton models leads to significantly different isomer cross sections. The theoretical calculation of high-spin isomer cross sections is notoriously difficult [2, 24], and uncertainties in the optical model and in the nuclear structure result in theoretical uncertainties of about a factor of 2 to 3 [2]. But even after this cautionary comment, it is clear from Table I that the calculations using an exciton model agree well with the measurements, and those with the FKK theory underpredict the measurements.

By considering the microscopic DWBA cross sections in the FKK calculation we can determine why the spin distribution after FKK preequilibrium emission is so great at low spins. For the case of a residual energy of 5 MeV, as shown in Fig. 2, we find particularly strong DWBA cross sections for $l$-transfers of 1 and 3. In the case of $l$-transfer=1, this is due to particularly strong transitions exciting the neutron particle-hole state $2d3/2 - 2f5/2$ and the proton particle-hole state $2p1/2 - 2d3/2$. These strong low $l$-transfer transitions, when coupled in with the ground-state spin of 4.5, yield residual nucleus spin distributions which are large near 4.5, and smaller for high spins, as seen in Fig. 2.

Why, then, does the FKK theory fail in this case? We think that this may happen because of the simplifying assumptions made in evaluating the theoretical predictions computationally. Our approach to determining the averaged MSD cross section for a given $l$-transfer follows that of other works, and is as follows: One averages a sample of basic DWBA cross sections and multiplies the average by a spin-dependent level density. A more accurate averaging procedure would be to omit the usual level density term and instead determine all basic DWBA transitions, multiplying by the number of possible states for each transition, obtained from a deformed Nilsson scheme. Such an improved averaging procedure, whilst computionally more involved and computer-intensive, may enhance the contributions from high-spin transfer processes.

## IV. CONCLUSIONS

With the recent modifications to the exciton model calculations in GNASH we obtain high-spin isomer production cross sections that are in good agreement with measurements, and agree more closely with measurements than those we published in 1991 [2]. However, our calculations which utilize the FKK theory yield smaller high-spin-transfer transitions than found with the exciton model, and underpredict high-spin isomer production. We doubt that this underprediction represents a failure of the basic physics of the FKK theory, but rather the inadequacy of its present implementation. Our formalism for implementing the FKK theory has been successful in describing nucleon emission cross sections and angular distributions, and the present investigation is the first to be sensitive to the transfer of large angular momenta. More work is needed to investigate spin transfer effects in the FKK theory, and we suggested a new method for averaging microscopic cross section which may remove the discrepancies. The calculation of high-spin isomer production presents a particularly difficult challenge to the theory, and it would be useful in future to consider processes in which lower angular momenta are involved. Two different phenomena which could be investigated are the production of lower spin isomers, and the production of various discrete gamma rays in $(n, xn\gamma)$ reactions [5], both of which are sensitive to angular momenta effects.

## V. ACKNOWLEDGMENTS

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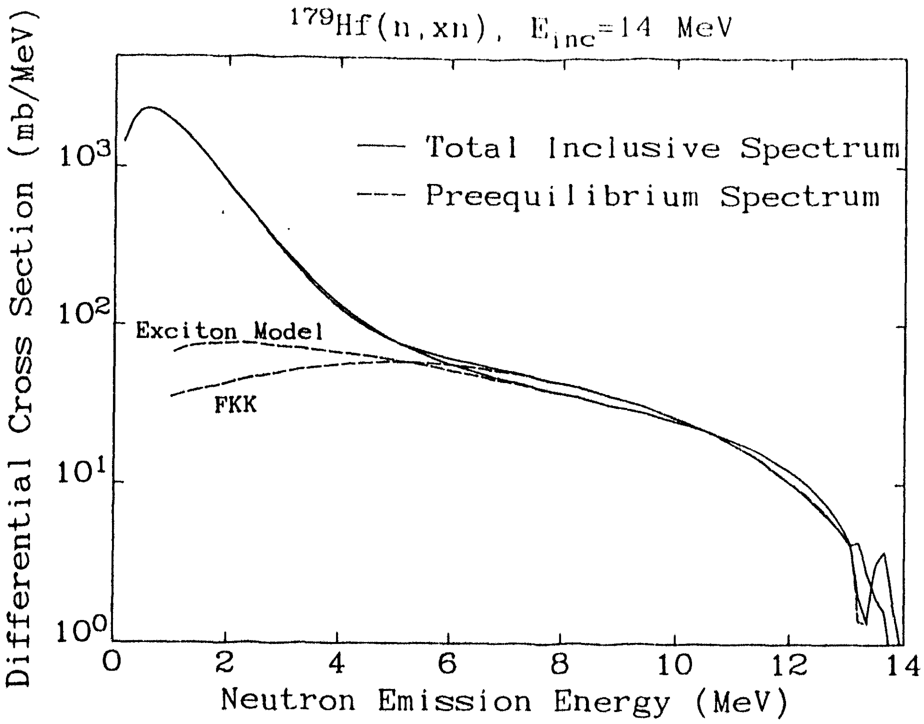


Fig. 1. Comparison of total and preequilibrium neutron emission spectra in $n+^{179}$Hf using FKK and exciton preequilibrium models.

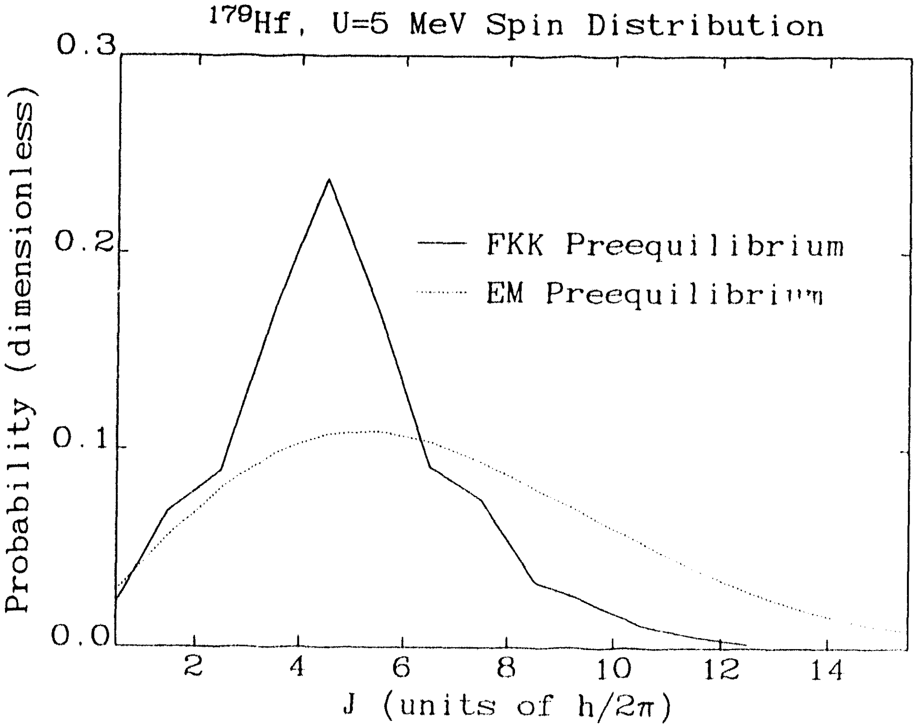


Fig. 2. Comparison of residual nucleus spin distribution at an excitation energy of 5 MeV in $^{179}$Hf using FKK and exciton preequilibrium models.

## TABLES

TABLE I. Theoretical 14 MeV Neutron-Induced Cross Sections for the Production of Isomeric States in Hafnium Compared with Experimental Data

| Reaction | FKK Preeq. | Exciton Preeq. | Experiment |
|---|---|---|---|
| $^{179}$Hf$(n,2n)^{178m2}$Hf$(16^+)$ | 2.11 mb | 9.72 mb | 6.29 ± .35 mb [4] |
| $^{179}$Hf$(n,n')^{179m2}$Hf$(12.5^-)$ | 1.29 mb | 11.92 mb | 12.8 ± 1.5 mb [3] |

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