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Analysis of fission excitation functions and the determination of shell effects at the saddle point

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Abstract. A method is proposed to deduce the shell correction energy corresponding to the fission transition state shape of nuclei in the mass region around 200, from an analysis of the first chance fission values of the ratio of fission to neutron widths, $(\Gamma_f/\Gamma_n)_1$. The method is applied to the typical case of the fissioning nucleus ^{212}Po, formed by alpha bombardment of ^{208}Pb. For the calculation of the neutron width, the level densities of the daughter nucleus after neutron emission were obtained from a numerical calculation starting from shell model single particle energy level scheme. It is shown that with the use of standard Fermi gas expression for the level densities of the fission transition state nucleus in the calculation of the fission width, an apparent energy dependence of the fission barrier height is required to fit the experimental data. This energy dependence, which arises from the excitation energy dependence of shell effects on level densities, can be used to deduce the shell correction energy at the fission transition state point. It is found that in the case of ^{212}Po, the energy of the actual transition state point is higher than the energy of the liquid drop model (LDM) saddle point by (3 ± 1) MeV, implying significant positive shell correction energy at the fission transition state. Further, the liquid drop model value of level density parameter a is found to be a few per cent smaller for the saddle point shape as compared to its spherical shape.

Keywords. Fission excitation function, ^{212}Po; shell effects at saddle point.

1. Introduction

It is now well known that shell corrections to the liquid drop model (LDM) potential energy of a nucleus are, in general, present at all deformations. It is in fact this feature which leads to a double-humped fission barrier for nuclei in the actinide region. In the region of nuclei with mass numbers around 200, although single particle effects do not lead to any significant secondary minimum in the nuclear deformation potential energy due to a much steeper variation of the LDM energy with deformation, a significant shell correction to the LDM energy may be present at the saddle point deformations of these nuclei, as indicated by some calculations (Bolsterli *et al* 1972, Mosel and Schmitt 1971, Pauli *et al* 1971). In this work it is shown that experimental information regarding the shell correction energy at the saddle point deformation of nuclei with mass numbers around 200 can be deduced from an analysis of their measured fission excitation functions.

Fission excitation functions for a number of nuclei with mass numbers around 200 have been measured by Thompson and his collaborators (Burtnett *et al* 1964, Khodai Joopari 1966, Thompson 1966) over a wide excitation energy range of the compound nuclei. In earlier studies (Huizenga *et al* 1962, Burnett *et al* 1964, Thompson 1966) analysis of the fission excitation functions was made with the standard Fermi gas model expressions for the level densities of the fission transition state nucleus and the residual nucleus after neutron emission, on which the magnitudes of the fission width, Γ_f and the neutron width, Γ_n are sensitively dependent. In these studies, theoretical expressions based on Fermi gas model level densities were fitted to the experimental data by treating the level density parameters a_f and a_n corresponding to the transition state nucleus and the nucleus after neutron emission as free parameters. Good fits to the data then required a value significantly greater than unity for the ratio a_f/a_n and it was also found necessary (Thompson 1966) to treat this ratio as energy dependent in order to find a good fit over a wide energy range. These results on a_n and a_f were qualitatively understood earlier in terms of shell effects on level densities.

In recent years, numerical calculations of nuclear level densities starting from shell model single particle energy level schemes have become possible (Huizenga and Moretto 1972, Ramamurthy *et al* 1970). Although calculations of single particle levels for both spherical and deformed shapes of nuclei have been carried out, uncertainties associated with these calculations for the highly deformed saddle point shapes can be much larger particularly due to the problems of shape parametrization. The numerical calculations of Γ_f, based on single particle levels of highly deformed saddle shapes may, therefore, have much larger uncertainties than the numerical calculations of Γ_n for spherical shapes. In fact some of the recently reported analysis, while using the numerically calculated level densities for the calculations of Γ_n, retain the standard Fermi gas expression for Γ_f (Moretto *et al* 1972). This simple procedure is justified only if there are no shell effects at the saddle point shape, which may not be true. In this work, we suggest a method of analysis of the experimental Γ_f/Γ_n data, which is valid even if there is significant shell correction at the saddle point shape. In fact the present method deduces not only the fission barrier height, but also the contribution of shell correction energy to it. In the following sections, after a brief description of the method, the available data on Γ_f/Γ_n for a typical compound nucleus ^{212}Po formed by alpha bombardment on ^{208}Pb are analysed with the present method and the results of analysis are discussed.

2. Outline of the present method of analysis

The deformation energy curve for a nucleus with mass number around 200 is shown schematically in figure 1 for the general case where shell effects are present at the saddle point. The standard theoretical expressions for the calculation of Γ_f and Γ_n are given in the Appendix. In the present method, the values of Γ_n are computed from a numerical calculation of the level densities of the residual nucleus. From these values of Γ_n, and the experimental Γ_f/Γ_n data, we then determine the excitation energy variation of Γ_f. As is described below, an analysis of this deduced Γ_f vs excitation energy can give not only the fission barrier height but also the shell correction energy and the value of LDM a parameter at the saddle point shape.

It has been shown earlier (Ramamurthy *et al* 1970) that for a calculation of the entropy at the deformed saddle shape with the inclusion of shell and pairing effects, one can use a modified Fermi gas expression $S^2=4a_f\, E_x^{s\prime}=4a_f(E_x^s+\delta)$, where a_f is the LDM value of the level density parameter at the saddle point, E_x^s is the excitation energy at the saddle point and δ is an excitation energy dependent correction term. It

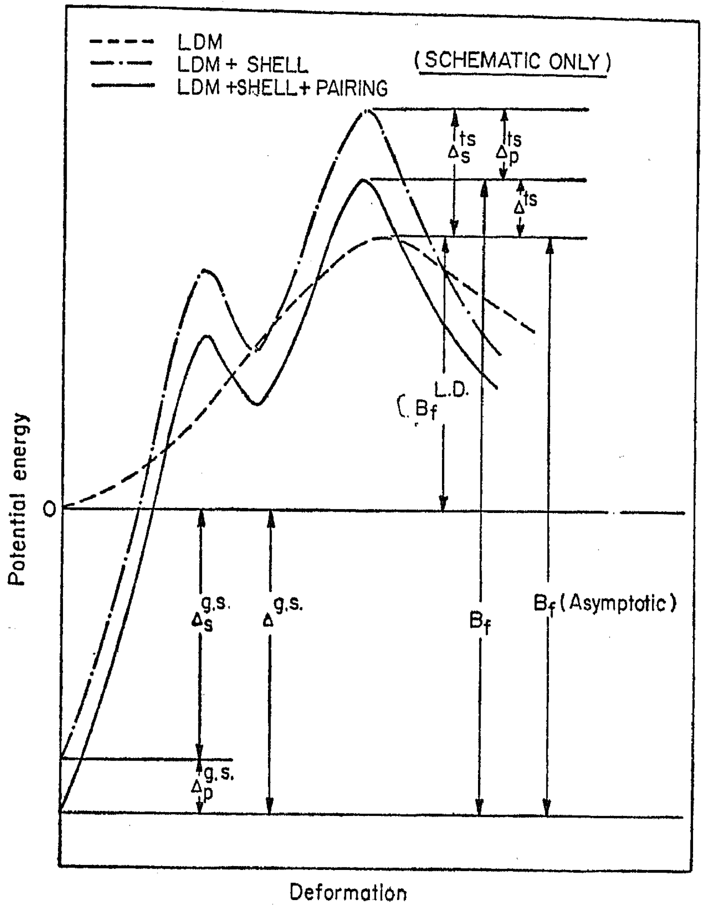

Figure 1. A schematic representation of potential energy of deformation of a nucleus with residual single particle effects at the fission barrier. $\triangle_s^{gs}$, $\triangle_p^{gs}$ and $\triangle^{gs}$ represent the shell, pairing and shell plus pairing energy corrections respectively in the ground state, while $\triangle_s^{ts}$, $\triangle_p^{ts}$ and $\triangle^{ts}$ are the corresponding quantities for the transition state nucleus.

is also known from earlier studies (Ramamurthy *et al* 1970, Kapoor and Ramamurthy 1973) that the transition state point is to be identified as the point of minimum entropy along the fission path, and the effective excitation energy $E_x^{s'}$ is to be measured from a reference energy surface which coincides with the actual potential energy surface at low excitation energies and from the LDM surface at higher excitation energies where shell effects have disappeared. It then follows that δ asymptotically approaches the value $\triangle^{ts}$ at these higher excitation energies, where $\triangle^{ts}$ is the energy difference between the maximum of the actual potential energy surface and the LDM energy surface (figure 1). Since the quantity $\triangle^{ts}$ can be identified with the sum of shell and pairing correction energies at the LDM saddle shape if the maximum of the actual potential energy surface coincides with the LDM saddle shape, we shall, refer to the quantity $\triangle^{ts}$ as the shell and pairing correction energy at the fission transition point shape. One can, therefore, write

$$E_x^{s'} = E_x^s + \delta = (E_x - B_f + \delta) = E_x - B_f'$$

where, at sufficiently higher excitation energies.

$$B_f' = (B_f - \triangle^{ts}) = (B_f^{\mathrm{LDM}} + \triangle^{gs}) = \text{constant.}$$

This implies that if an analysis of Γ_f is carried out on the basis of the standard Fermi gas expression for $\rho^*(X)$ it would necessitate the use of an apparent fission barrier B_f' which changes with energy in such a way that at higher excitation energies, it asymptotically approaches a constant value equal to $(B_f - \Delta^{ts})$. It may be stressed here that B_f' will reach this constant value only if the chosen value a_f corresponds to the correct LDM value for the transition state shape of the nucleus. For nuclei with no single particle effects at the saddle point, this constant value of B_f' will be realised starting from zero excitation energy of the transition state nucleus, and in this case, one can treat a_f as a free parameter to search for this single constant value of B_f' by a least square fit to the data. However, as shown earlier, for a nucleus with single particle effects at the saddle point one expects a constant value of B_f' only at excitation energies, sufficiently large to wipe out shell effects. The expected constancy of B_f' at higher excitation energies can also be used as a criterion to obtain the correct value of a_f. Figure 2 shows schematically the expected energy dependance of the apparent fission barrier B_f' for different values of a_f for a nucleus having a positive value for the shell plus pairing energy correction to its LDM deformation energy near the saddle point deformation. It can be seen that B_f' reaches a constant value only for a single value of a_f which is to be identified as the correct LDM value for the transition state shape of the nucleus. It is, therefore, clear that such an analysis

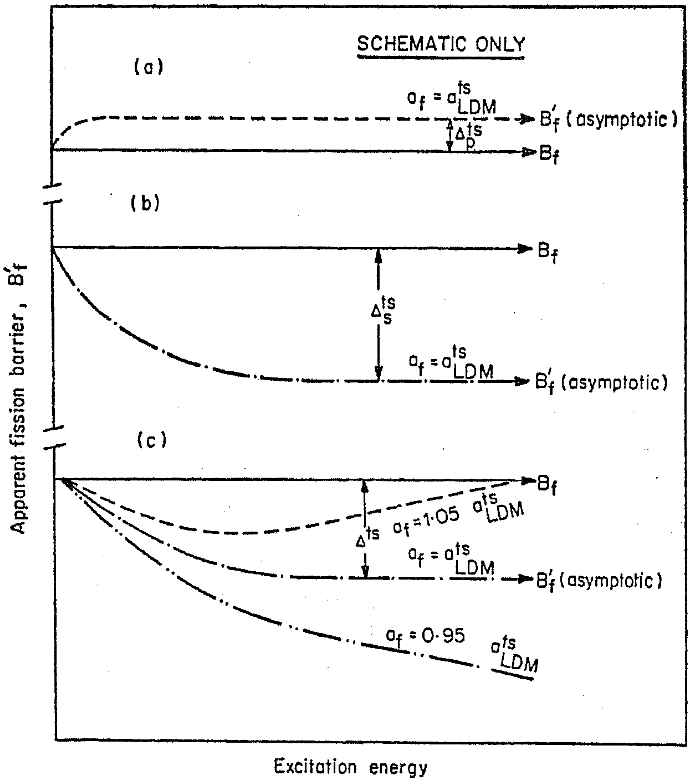

Figure 2. Expected variation of the apparent fission barrier height B_f' for a nucleus considering (a) pairing energy correction Δ_p^{ts} alone (b) a positive shell energy correction Δ_s^{ts} alone and (c) pairing plus shell energy correction Δ^{ts} respectively. In 2(c), the apparent fission barrier B_f' versus excitation energy is shown for three values of a_f, and it can be seen that a constant value of B_f' is reached asymptotically only if $a_f = a_{\mathrm{LDM}}^{ts}$.

provides the saddle point shell plus pairing energy correction which is simply equal to the actual fission barrier B_f minus the asymptotic value of the apparent fission barrier B'_f at high excitation energies.

3. Results for ^{212}Po

The present method was used to analyse the data of Thompson (1966) on Γ_f/Γ_n for the nucleus ^{212}Po. The values of Γ_f/Γ_n as deduced by Thompson (1966) from their measurements of alpha induced fission cross-sections of ^{208}Pb and the reaction cross sections calculated from optical model formed the input data. In order to compare with the calculated first chance fission values of (Γ_f/Γ_n), the above measured values need corrections for multiple chance fissions. The first chance fission values $(\Gamma_f/\Gamma_T)_1$ for ^{212}Po were obtained from the measured (Γ_f/Γ_T) $(\Gamma_T=\Gamma_f+\Gamma_n)$ for ^{212}Po and ^{211}Po on the assumption that $(\Gamma_f/\Gamma_T)_{2+3+\cdots}$ for ^{212}Po at excitation energy E_x is equal to the measured (Γ_f/Γ_T) for ^{211}Po at excitation energy $E_x - B_n - 2T$, where B_n is the neutron binding energy for ^{212}Po and T is the average nuclear temperature for the daughter nucleus after neutron emission. It is seen that the magnitude of this correction is negligibly small below 40 MeV and increases rapidly with increase in the excitation energy. For example, at an excitation energy of 60 MeV, the second chance correction is found to be 70% of the measured value pointing out the importance of correction of the data for multiple chance fission, if the analysis is extended to the data points up to 60 MeV or more. In the present procedure the data might be undercorrection by a few percent due to the angular momentum differences for ^{212}Po and ^{211}Po at the same excitation energies, but this residual error is expected to cancel the effect of any small direct interaction effects, if present. In order to keep to a minimum the uncertainties associated with the multiple chance corrections and possible direct interaction effects at higher bombarding energies, the data points for excitation energies only up to 60 MeV were used in the present analysis.

The values of Γ_n vs excitation energies were computed with eq. (A2) with the values of neutron binding energies obtained from the known ground state masses, and taking $r_0=1{\cdot}2\times10^{-13}$ cm. The nuclear level densities $\rho^{**}(E_x)$ for the residual nucleus were computed by a numerical calculation of both the entropy and the pre-exponential factor starting from a shell model single particle level scheme as described earlier (Ramamurthy 1971). Calculations were carried out for two sets of single particle levels, one given by Seeger and Perisho (1967) for a modified harmonic oscillator potential and the other given by Bolsterli *et al* (1972) for a folded-Yukawa potential. The nuclear pairing effects on level densities were taken into account by replacing the excitation energy E_x by $(E_x-\Delta_p^{gs})$, where Δ_p^{gs} is the ground state pairing energy† of the residual nucleus. Since the level densities which enter into the calculation of Γ_n are mainly for the residual nucleus excitation energies exceeding about 10 MeV, this procedure for including pairing interactions is justified, (Ramamurthy 1971) since the pairing effects quickly disappear with excitation energy and the level density of a nucleus corresponds to the excitation energy measured from the ground state stripped off its pairing energy Δ_p^{gs}. The calculations for Γ_f were carried out with a Fermi gas expression for the level densities and involved two adjustable parameters, a_f and

†Throughout this text, the pairing energy correction refers to the difference in the total potential energy with and without pairing interaction.

$\hbar\omega$. It was found that the experimental data on $(\Gamma_f/\Gamma_n)_1$, cannot be satisfactorily fitted to the calculations with a single value of B_f over the entire energy range for any value of a_f implying the existence of shell effects at the saddle point. The values of the apparent fission barrier B'_f at any excitation energy was then deduced from a fit to the experimental first chance fission $(\Gamma_f/\Gamma_n)_1$ data. The values of B'_f vs excitation energy were thus deduced for a range of values of a_f, and that value of a_f which led to the expected constany of B'_f at higher excitation energies was obtained from these calculations. Figure 3 shows the deduced values of B'_f for various compound nucleus excitation energies and for values of barrier penetration parameter $\hbar\omega$ equal to zero and 1·5 MeV and for that value of a_f which led to a constant value of B'_f at higher excitation energies. These values of fission barriers have been corrected for a small effective decrease in the fission barrier heights due to the angular momentum brought in by the incident alpha particles using the tabulated (Plasil 1963) Pick-Pichak energies. The results of the analysis are summarized in table 1.

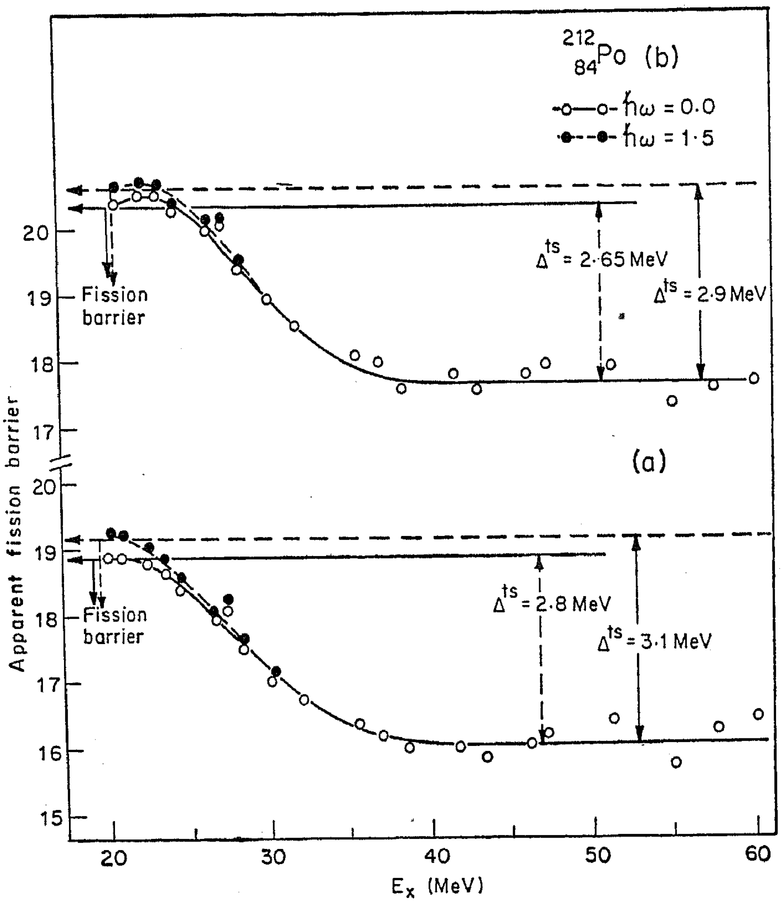

Figure 3. The apparent fission barrier height B'_f as a function of compound nucleus excitation energy E_x for the fissioning nucleus ^{212}Po obtained from the analysis of first chance $(\Gamma_f/\Gamma_n)_1$ data. The calculation of Γ_n with the modified harmonic oscillator levels of Seeger and Perisho (1967), and the folded Yukawa potential levels of Bolsterli *et al* (1972), yields the results shown in figures 3 (a) and (b) respectively. The results of analysis based on the assumptions of barrier penetration factor $\hbar\omega$=0·0 and 1·5 MeV are shown for comparison with each other. It can be seen that with the correct value of a_f chosen, the apparent fission barrier reaches a constant value asymptotically with increasing excitation energies and in all the cases the pairing plus shell energy correction at the transition state is found to have a significant positive value of about 3 MeV.

Table 1. Summary of the results of analysis of first chance $(\Gamma_f/\Gamma_n)_1$ data of ^{212}Po fissioning nuclei.

Fission-ing nucleus	Fission barrier, (MeV) B_f		B_f' MeV (asym-ptotic)	Δ^{ts} (MeV)		a_f, MeV^{-1}	Sphe-rical a_{LDM}, MeV^{-1}	a_f/a_{LDM}	Re-marks
	$\hbar\omega=$ 0·0 MeV	$\hbar\omega$ 1·5 MeV		$\hbar\omega$ 0·0 MeV	$\hbar\omega$ 1·5 MeV				
^{212}Po	18·9	19·2	16·1	2·8	3·1	26·1	27·8	0·94	a
	0·4	20·6	17·7	2·6	2·9	19·21	20·9	0·92	b

(a) Modified harmonic oscillator levels (Seeger and Perisho 1967) for calculation of Γ_n
(b) Folded-Yukawa potential single particle levels (Bolsterli *et al* 1972) for calculation of Γ_n.

4. Discussion

It can be seen from figure 3 and table 1 that the asymptotic value of B_f' is less than the actual fission barrier by a few MeV, irrespective of the details of the single particle level scheme used to calculate Γ_n and the input values of the parameter $\hbar\omega$. This implies that ^{212}Po nucleus has a significant positive value for the sum of the shell and pairing correction energy at the saddle point similar to the cases of actinide nuclei. It is further seen from figure 3 that in the present case where shell correction energy has a positive sign, most of the shell effects have disappeared at a moderate excitation energy E_x^s of about 20 MeV. It is estimated that the deduced values of a_f have an uncertainty of less than 3% due to the scatter in the experimental data points and uncertainty in the choice of r_0 used to calculate Γ_n. The corresponding uncertainty in the derived values of Δ^{ts} is estimated to be within 1 MeV. It is therefore inferred that for the nucleus ^{212}Po, the sum of the shell and pairing corrections at the transition state shape is $+(3\pm1)$ MeV. In table 1, the deduced values of a_f are compared with the LDM value of the parameter a for the *spherical shape* of the nucleus ^{212}Po. These LDM a values corresponding to each single particle level scheme were obtained by generating its corresponding uniform level scheme with a value of 1·0 for the Strutinsky parameter γ, and correction terms up to sixth order, and then by determining the single particle level density at the Fermi level for the uniform level schemes. It is seen that the derived values of a_f are less by 6-8% than the corresponding LDM values for the spherical shapes. A reduction of this order in the LDM value of a_f for the deformed saddle point shape as compared to spherical shape is in fact expected on the basis of the recently found evidence (Kataria *et al* 1977) for the dependence of a_f on nuclear surface areas.

The existence of shell and pairing energy corrections at the saddle point of the nucleus ^{212}Po, as shown by the present analysis, implies that the LDM fission barrier for this nucleus is appreciably lower than the values deduced earlier (Thompson 1966) with the neglect of shell effects at the saddle point. Consequently the Myer-Swiatecki semiempirical mass formula (Myers and Swiatecki 1966) whose coefficients are determined by a fit to the fission barriers deduced earlier with the neglect of shell effects at the saddle point is expected to overestimate the LDM fission barrier heights. Indications to the effect that LDM fission barriers are smaller than those obtained from the calculations have recently come from other investigations (Shelino *et al* 1972, Methasiri and Johansson 1971, Beckerman and Blann 1977).

In conclusion, the present work has shown that the magnitude of the single particle effects at the fission transition state deformation of nuclei in the region of masses around 200 can be deduced from an analysis of the first chance fission values, $(\Gamma_f/\Gamma_t)_1$. The analysis carried out for the case of a typical nucleus ^{212}Po has shown that appreciable single particle effects exist at its transition state shape. The maximum in the actual potential energy surface is found to be (3±1) MeV higher than the LDM saddle point energy. It is also found that the LDM value of the level density parameter *a*, is lesser by a few per cent at the deformed saddle shape as compared to spherical shape indicating the dependence of the parameter *a* on nuclear surface.

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Appendix

Theoretical expressions for the fission and neutron widths

Analysis of fission excitation functions is based on the Bohr-Wheeler transition state theory for the evaluation of fission width Γ_f and the statistical theory of neutron evaporation for the evaluation of neutron width Γ_n. The theoretical expressions for the fission and neutron widths used in the present investigations are the same as those earlier (Khodai Joopari 1966). The neutron width Γ_n is given by

$$\Gamma_n=\hbar \int_0^{E_x-B_n} W_n(t)\, dt \tag{A1}$$

where

$$W_n(t)=\frac{\sigma(E_x, t)\, g\, mt\, \rho^{**}(X)}{\pi^2\hbar^3\, \rho(E_x)}$$

Here $\sigma(E_x, t)$ is the cross section for the inverse process, g is the statistical weight which applies to the spin states of the neutrons, namely 2, and m is the neutron mass. The level density of the residual nucleus following neutron emission at excitation X is $\rho^{**}(X)$ and $\rho(E_x)$ is the level density of the compound nucleus at excitation energy E_x. The neutron kinetic energy t is related to the total excitation energy E_x, the neutron binding energy B_n and the excitation energy of the residual nucleus X, by

$$t=E_x-B_n-X.$$

By taking the geometric cross section for $\sigma(E_x, t)$ one gets

$$\Gamma_n=\frac{1}{\pi\rho(E_x)}\int_0^{E_x-B_n} \rho^{**}(X)\,\frac{t}{t_0}\, dX \tag{A2}$$

where

$$t_0 = \frac{\hbar^2}{2mr_0 A^{2/3}}$$

For the fission width, the Bohr-Wheeler expression obtained on the basis of the standard theory of reaction rates applied to fission, with the fissioning nucleus at the saddle point configuration, is given by

$$\Gamma_f = \frac{1}{2\pi\rho(E_x)} \int_0^{E_x - B_f} \rho^*(X) dX$$

where $\rho^*(X)$ is the level density of the saddle point configuration at the excitation energy X which is the energy in the non-fission degree of freedom. The relation of X to the total excitation energy E_x and the potential and kinetic energies in the fission degree of freedom is given by

$$E_x = B_f + T + X$$

where B_f is the fission barrier and T is the kinetic energy in the fission degree of freedom. At excitation energies close to the fission barrier, one has to take into account the quantum mechanical penetrability of the barrier. Including this effect, one gets

$$\Gamma_f = \frac{1}{2\pi\rho(E_x)} \int_0^{E_x} \frac{\rho^*(X) dX}{[1 + \exp\{-2\pi(E_x - B_f - X)/\hbar\omega\}]} \qquad \text{(A3)}$$

where the quantity $\hbar\omega$ is a measure of the thickness of the barrier, which is assumed to be parabolic.

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