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MULTISTEP PROCESSES IN NUCLEAR REACTIONS

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Abstract

Recent applications of the theory of Feshbach, Kerman and Koonin to analyse multistep processes in nuclear reactions are described, and illustrated by detailed comparisons with a range of experimental data for neutron and proton reactions. The techniques used to distinguish between the multistep direct, multistep compound, compound and collective contributions to the cross-sections are described, and their effectiveness assessed. Particular attention is devoted to recent analyses that take into account the transitions from the multistep direct to the multistep compound chain and also to those that evaluate the collective contributions to the continuum region. The variation of the effective nucleon-nucleon interaction with incident energy and target nucleus is studied, and the possibility of a parameter-free calculation is discussed. The extensions of the theory to include multiparticle emission from the direct chain and also to studies of alpha-particle emission are described.

1. Introduction

For many years it was assumed that nuclear reactions take place either by a direct process or by a compound nucleus process or by both processes together. The direct process occurs rapidly, in the time taken by the projectile to traverse the target nucleus, whereas the compound nucleus process takes much longer, after full statistical equilibrium has been established.

In recent years this sharp division into direct and compound nucleus processes has proved inadequate, and there is much evidence for the presence of pre-equilibrium reactions that take place after the direct stage but long before the attainment of full statistical equilibrium (Gadioli and Hodgson, 1992).

Pre-equilibrium reactions take place in a number of stages or steps, corresponding to the excitation of successive particle-hole pairs by the interaction of the projectile with nucleons in the target. At each stage it is possible for emission to occur, and these are the pre-equilibrium reactions.

The first analyses of pre-equilibrium reactions were made using the exciton model, and this proved able to account for the total cross-sections in many reaction channels in a unified way (Blann, 1975; Gadioli *et al*, 1977). This model, and other semi-classical models were however less successful in accounting for the angular distributions of the emitted particles. The Boltzmann master equation theory of Harp, Miller and Berne (1968) has also been used to calculate the energy spectra of particles emitted in nucleon-induced reactions and also in heavy-ion reactions (Cervesato *et al*, 1992).

More recently, fully quantum mechanical theories of multistep pre-equilibrium reactions have been developed by Feshbach (1977, 1985, 1986, 1991) and by Feshbach, Kerman and Koonin (1980) (FKK), by Tamura, Udagawa and Lenske (1977, 1981, 1982) (TUL) and by Nishioka, Weidenmuller and Yoshida (1986, 1990) (NYY). The

relationship between these theories has been studied by Koning and Akkermans (1991), who derived them in a consistent way, showing how they follow from certain statistical assumptions. Thus the FKK theory uses leading particle statistics whereas the TUL and NXY theories use residual nucleus statistics. Furthermore, the additional assumptions in the FKK theory enable the cross-sections for the higher steps to be calculated easily whereas this becomes prohibitively complicated for the other theories beyond the second step. Moreover, a detailed intercomparison of the three theories by Koning and Akkermans (1991, 1992) showed that they fit the experimental data equally well. Since the FKK theory is easier to use it has therefore been compared with a wider range of data than the other two theories. The development and inter-relation of these theories has been discussed by Hodgson (1991).

These theories refer to multistep direct (MSD) reactions in which one particle always remains in the continuum, so that the memory of the incident direction is retained and the cross-sections are forward-peaked. The FKK theory also describes multistep compound (MSC) reactions in which all particles remain bound. A multistep compound theory has also been developed by Agassi *et al*, (1975) and Nishioka *et al* (1986) and a unified theory by Lenske *et al* (1994). These theories have also been used to analyse heavy-ion reactions, but this work is beyond the scope of this review (Lenske *et al*, 1983; Dean *et al*, 1984; Lenske and Wolter, 1992; Sudiak *et al*, 1994).

The theories of pre-equilibrium reactions range from very detailed formalisms that are difficult or impracticable to calculate numerically to phenomenological models or parametrisations that are easy to calculate and to fit to the data but contain little physics and so lack generality. Over the years, pre-equilibrium theories have been continually improved to incorporate as much physics as possible in a formally correct way while still allowing numerical calculations to be made.

This review is restricted to recent analyses of pre-equilibrium nuclear reactions made with the FKK theory. Earlier work has been reviewed by Gadioli and Hodgson (1992).

2. The Feshbach-Kerman-Koonin Theory

This theory distinguishes between two reaction chains, the multistep direct and the multistep compound. In multistep direct reactions (MSD) at least one particle, usually the projectile, remains in the continuum, whereas in the multistep compound reactions (MSC) all nucleons remain bound. These are called the *P* and *Q* chains respectively.

The FKK formalism was programmed for electronic computation by Avaldi *et al* (1980) and by Bonetti *et al* (1981) and used to calculate the MSD cross-sections of several (*p, n*) reactions from 25 to 45 MeV. Subsequently the formalism was extended to include the analysing powers (Bonetti *et al*, 1982a) and used to analyse experimental data for ^{58}Ni (*p, p'*) at 65 MeV.

At lower energies it is necessary to include the contribution of the MSC process, and this was done by Bonetti *et al* (1980, 1981, 1982b, 1983). These early analyses showed that an incoherent superposition of the MSD and MSC cross-sections is able to give a good overall account of the cross sections of pre-equilibrium reactions from 10 to 50 MeV with consistent values of the parameters (Bonetti and Colombo, 1983).

In the following years many analyses were made of MSC reactions (Herman *et al*, 1984; Field *et al*, 1986; Chadwick *et al* 1988, 1989; Koumdjieva and Hodgson 1989 and Demetriou *et al* 1993, 1994) and MSD reactions (Holler *et al*, 1985; Mordhorst *et al*, 1986; Trabandt *et al*, 1988, 1989; Marcinkowski *et al* 1989; Scobel *et al* 1990, Cowley *et al* 1991, Richter *et al* 1992, 1994) and these established the validity of the FKK theory over a wide range of energies and target nuclei. Extensive reviews of the FKK formalism

have been published for both the MSC (Bonetti *et al.* 1991) and the MSD (Bonetti *et al.* 1994) reactions.

With increasing precision of the analysis it has become clear that the FKK theory must be modified and developed in several ways. These include the incorporation of transitions from the P to the Q chain after the initial interaction, more accurate ways of separating the contributing reaction mechanisms, the inclusion of collective excitations in the continuum region, multiple emission of nucleons and the emission of alpha-particles. These developments will now be summarised.

Fig. 1. P to Q chain transitions according to (a) the model of Chadwick and Young (1993) and (b) the gradual absorption model of Marcinkowski *et al* (1991, 1993) for three $P \rightarrow Q$ transitions. More transitions can be added at higher energies.

In the original FKK formalism the P and Q chains remain distinct after the initial interaction, but many subsequent analyses showed that this gives MSC cross-sections that are too large to fit the experimental data. This is the result of allowing all the compound nucleus cross-section to pass through the 2p1h state, and may be avoided by allowing transitions from the P to the Q chain to take place after the initial interaction (Chadwick, 1989; Marcinkowski *et al.* 1991, 1993; Chadwick and Young, 1993; Marcinkowski and Kielan 1994, Marcinkowski, 1994; Lenske *et al.*, 1994). This has been done in two ways, firstly as a single transition and secondly by summing the transitions at each stage. Chadwick and Young (1993) evaluated the fraction R of the incident flux that goes immediately to the Q chain as the ratio ρ_1^B / ρ_1 of the bound to the total phase space of 1p-1h excitations. The fraction going to the P chain is then $(1 - R)$. Not all this flux, however, is emitted by the MSD process, but only a fraction $(1 - R')$ of the incident flux. The remaining flux $(R' - R)$ must be added to the Q -chain and is the additional P to Q transition, as shown in Fig.1(a).

A gradual absorption model, considering the P to Q transitions at each stage of the P -chain, has been formulated by Marcinkowski *et al* (1991, 1993). The MSD flux is $(1 - R')$ as before, but now the remaining flux is divided using the same phase space factor so that $R'\rho_1^B/\rho_1$ goes to the Q chain and $R'(1 - \rho_1^B/\rho_1)$ to the P chain. This latter flux all goes eventually to the Q -chain, an amount $R'(1 - \rho_1^B/\rho_1)\rho_2^B/\rho_2$ at the second stage and so on. Summing all these gives a total flux $R'(1 - \rho_1^B/\rho_1)$ to the Q -chain after the first stage, as shown in Fig.1(b).

Fig. 2. Multistep calculations of the $^{93}\text{Nb}(n, n')$ cross-section at 14.1 MeV (a) without and (b) including P to Q chain transitions using the gradual absorption model. The inclusion of these transitions enhances the MSD cross-section and greatly reduces the MSC cross-section. The cross-section due to the first neutron emitted from the compound nucleus is CN1, to the second CN2 and to the neutron following proton emission CP2. (Marcinkowski and Kielan, 1994).

As shown in Fig.2 the inclusion of P to Q transitions substantially reduces the MSC cross-section and improves the fit to the $^{93}\text{Nb}(n, n')$ cross-section. A similar comparison (Fig.3) for the corresponding $^{93}\text{Nb}(n, p)$ reaction shows a somewhat improved fit and again a lowered MSC cross-section; in this case it is the angular distribution (Fig.4) for outgoing energies around 12 MeV that shows more clearly the dominance of the MSD reaction in this energy region. A similar result was obtained by Lenske *et al* (1994).

The cross-sections corresponding to these two models of P to Q transitions are compared in Fig.5. It is notable that there are substantial differences both in shape and magnitude between the MSC cross-sections given by the two models. Since the gradual absorption model is more general, and is easy to calculate, it is preferred over the simpler model.

Fig. 3. Energy spectra for the $^{93}\text{Nb}(n, p)$ reaction (Traxler *et al*, 1985; Grimes *et al* 1978) compared with FKK calculations (a) without and (b) with gradual absorption. The curves labelled CN1 and CN2 correspond to the first and second compound nucleus protons (Kielan and Marcinkowski, 1994).

Fig. 4. Angular distribution of the outgoing protons with energies between 10 and 12 MeV from the $^{93}\text{Nb}(n, p)$ reaction (Traxler *et al*, 1985) compared with FKK calculations with (MFKK) and without P to Q chain transitions (Kielan and Marcinkowski, 1994).

Fig. 5. Angle-integrated cross-sections for the inelastic scattering of 25.6 MeV protons by ^{98}Mo compared with FKK calculations with two modes of P to Q chain transitions (Watanabe *et al*, 1995).

Fig. 6. Analyses of the ^{93}Nb (n, n') angle-integrated spectra at 14 MeV including the four possible P to Q chain transitions and normal and non-normal DWBA matrix elements (Arbanas *et al.* 1995).

A more detailed formalism for P to Q chain transitions that uses the FKK transition matrix instead of a phase space factor has been developed by Arbanas *et al* (1995). By removing an approximation made by FKK, they express the MSC cross-section as a sum of four terms taking into account the possible P to Q transitions. Calculations were made with both normal and non-normal matrix elements, and the results are compared with the angle-integrated cross-section of the ^{93}Nb (n, n') reaction at 14 MeV in Fig.6.

This comparison shows that a better fit is obtained with the non-normal DWBA matrix elements and this was also found for the ^{107}Ag (n, n') reaction at 14 MeV by Chadwick *et al* (1994) contrary to the work of Kumabe *et al* (1984). However some calculations of the $^{90}\text{Zr}(p, n)$ reaction at 80 MeV, by the same authors showed a large calculated excess cross-section for emission energies below 40 MeV, indicating that multistep reactions are greatly over-estimated. Until the reason for this is established and corrected, calculations with normal DWBA matrix elements are preferred.

The relative contributions of the different reaction mechanisms are shown by studies of reactions on a series of isotopes. The total cross-sections of the (n, p) reaction on five ruthenium isotopes have been measured using the activation technique and analysed using the FKK theory without and with gradual absorption (Marcinkowski *et al*, 1990; Kielan, Marcinkowski and Garuska, 1993; Kielan and Marcinkowski, 1994), and the results of the latter analysis are shown in Fig.7. It is notable that the compound nucleus cross-section falls rapidly as the Q -value increases, relative to the MSD and MSC cross-sections and this affects the shape of the excitation function around 14 MeV. A similar analysis has been made for the Zn isotopes (Kielan and Marcinkowski 1995).

Most of the early analyses were designed to test either the MSC or the MSD formalism, and so the data were chosen so that either one or the other process dominates. Thus MSC analyses were made of the energy spectra in the backward direction from reactions at low energies, and MSD analyses were made of double differential cross-sections at high energies. In most cases, however both processes contribute and in addition there are contributions from compound nucleus emission and collective excitations. The earlier analyses used the optical model transmission coefficients to calculate the MSC cross-section and chose a value of the effective nucleon-nucleon interaction strength V_0 to fit the MSD cross-section. The collective excitations are assumed to be responsible only for the resolved peaks at the higher outgoing energies, and the

Fig. 7. Total cross-sections for the (n, p) reaction on ruthenium isotopes compared with FKK calculations using the gradual absorption model. The relative contributions of the multistep direct (MSD), multistep compound (MSC) and compound nucleus (CN) reactions varies markedly along the isotopic sequence (Kielan and Marcinkowski, 1994).

compound nucleus cross-section are calculated from statistical theory.

It is however desirable to develop more precise methods of separating these contributions. Thus Chadwick and Young (1993) evaluated the MSC and MSD cross-sections and adjusted their normalisations to optimise the fit in the intermediate energy region. Demetriou *et al* (1993) removed the compound nucleus and MSC contributions, which are symmetric about 90° in the CM system, by subtracting double differential cross-sections for pairs of complementary angles, thus leaving only the MSD and the collective cross-sections. Assuming that the collective contributions are confined to the peaks at higher emission energies, the remaining cross-section is MSD only, and may be compared with similarly-subtracted FKK calculations. This is shown in Fig.8 for the ^{93}Nb (n, n') reaction at 14 MeV. The fit is good except in the region of the collective peaks, and the values of the effective nucleon-nucleon interaction strength V_0 obtained by normalising to the data are consistent for each angle pair. A more severe test of the subtraction method is provided by the (p, n) reaction which does not have appreciable collective contributions. This is shown by the analysis of the ^{98}Mo (p, n) reaction at 25.6 MeV in Fig.9. The subtraction method is sensitive to the accuracy of the fit to the angular distributions, and also assumes that there are no collective excitations in the continuum region. Providing the MSC cross-section is isotropic, the method of Chadwick and Young is identical to the subtraction method over the same energy region, and gives a result integrated over all angles.

The possibility of such collective excitations has been studied by Marcinkowski *et*

Fig. 8 Subtracted double differential cross-sections in the CM system for 14 MeV neutrons inelastically scattered by ^{93}Nb for four pairs of complementary angles. The curves are similarly subtracted cross-sections calculated with the multistep direct theory of Feshbach, Kerman and Koonin (1980). The peaks at higher outgoing energies are due to the excitation of collective states, a process not included in the calculations. The curves are normalised to the data, giving the values of the effective interaction strength V_0 given in each box. The experimental points are obtained from the data of Takahashi *et al* (1992) (Demetriou *et al*, 1993).

Fig. 9 Subtracted double differential cross-sections for the $^{98}\text{Mo}(p,n)$ reaction at 25.6 MeV for four pairs of complementary angles compared with similarly-subtracted FKK MSD calculations. The curves are normalised to the data and the values of the effective interaction V_0 are given for each angle pair: these are in satisfactory agreement with each other (Watanabe *et al.*, 1995).

al (1995ab). The cross-sections of (p,n) reactions on several nuclei from 9 to 27 MeV were analysed by the FKK theory. Since collective contributions to this reaction are small, this establishes the value of V_0 . Using the same parameters, the corresponding (n,n') cross-sections were calculated, and there was a shortfall compared with the data, indicating the presence of collective contributions to the continuum. This was confirmed by calculating the collective contributions using the experimental values of the energies and strengths of the low-lying collective states, and including the contributions of the giant multipole resonance, if any, using the energy-weighted sum rule. The results of this calculation, when added to the other contributions, is in good accord with the data, as shown in Fig.10 (Marcinkowski *et al* 1995b). In this way a consistent analysis of (p,n) and (n,n') reactions can be made that includes all the contributing processes. Further studies of the contributions of collective excitations (Demetriou *et al.*, 1995) showed that they are able to account for the resonance structure of the (n,n') energy spectra (see Fig.11). This structure was also reproduced by Lenske *et al* (1994), using a microscopic model.

Fig. 10 Multistep calculations of the $^{93}\text{Nb}(n, n')$ cross-section at 14.1 and 20 MeV showing the contribution of the collective excitations in the continuum (Marcinkowski *et al.*, 1995).

Fig. 11. The energy spectrum of neutrons inelastically scattered by ^{58}Ni at 14.1 MeV (Takahashi *et al.*, 1992) compared with MSC, MSD and collective cross-sections (Demetriou *et al.*, 1995).

These results show that analysis of inelastic scattering that omit the collective excitations to the continuum give anomalously large values of the effective interaction strength V_0 . It may be noted that including these collective excitations would not affect the conclusions from Fig.2 concerning the presence of P to Q chain transitions, since they would simply be absorbed by the MSD contribution.

The parameters used in these multistep calculations can all be obtained, with more or less accuracy, from previous work. Thus the optical potentials that give the incident and outgoing distorted waves are obtained from elastic scattering data and the level density parameters from standard compilations. The remaining parameter is the strength V_0 of the effective nucleon-nucleon interaction (taken to have the Yukawa form with range 1 fm) that determines the overall normalisation of the MSD cross-section. The value of V_0 is usually determined by normalising the calculations to the experimental data, and it is then found that they agree well with those obtained from the real part of the nucleon optical potential using the folding model (Austin, 1980), as shown in Fig.12. If these values are used, the calculation becomes essentially parameter-free. The higher values of V_0 found in some (p, p') analyses at low energies are at least partly attributable to the neglect of collective contributions to the continuum.

Fig. 12 The effective nucleon-nucleon interaction strength V_0 as a function of incident nucleon energy with a Yukawa form factor of range 1 fm. The full curve shows the energy dependence of the nucleon optical potential normalised to the value at low energies obtained from the real part of the nucleon optical potential using the folding model.

The values of V_0 found by Watanabe *et al* (1995) from their analyses of proton inelastic scattering from 12 to 26 MeV by several nuclei are substantially greater than those given in Fig.12. To see if this can be attributed to collective excitations in the continuum, calculations were made with the set of parameters obtained by Marcinkowski *et al* (1995a) from their analysis of (p, n) cross-sections. With the addition of the

collective excitations, these parameters fit the corresponding (n, n') data, but the cross-sections were still too low to fit the (p, p') data. The reason for this discrepancy is not understood.

A major difficulty with analysis of nuclear reactions with several competing processes is that it is often rather easy to fit the experimental data by adjusting parameters, so that attainment of a good fit cannot be taken as a validation of the correctness of the calculation. The first requirement is to establish the theory on a sound physical basis, and significant progress in this direction has been made in recent years. To test a theory, it is desirable to fix all or nearly all the parameters from other studies and to analyse in a unified way as wide a range of data as possible. It is particularly desirable to include angular distributions in the analysis, as these are usually the most sensitive to the reaction mechanism. Even when all this is done, there are other more subtle ways of ensuring a good fit. For example, the cross-sections of many reactions are very sensitive to the optical potentials used, even when they fit the elastic scattering cross-section, as shown in Fig.13. This is because the cross-sections for reactions depends on the wavefunctions in the nuclear interior, whereas the elastic scattering depends only on their asymptotic behaviour. Many potentials may therefore have the same asymptotic behaviour but differ inside the nucleus (Hodgson, 1994). It is thus possible to make calculations with several potentials, select the one that best fits the data, and then claim that the analysis is parameter-free.

The MSD is able to give a good account of the (p, p') inelastic scattering data at high incident and outgoing energies where the collective and MSC contributions are negligible (see Fig.14). At still higher energies it is possible for more than one particle to be emitted from the multistep chain. The MSD formalism has been extended by Chadwick *et al* (1994) to include such multiple emission, and one of their results is compared in Fig.15 with the angle-integrated differential cross-section for the $^{90}\text{Zr}(p, n)$ reaction at 160 MeV.

The multiple emission of particles may also be calculated by the quantum molecular dynamics (QMD) theory, which follows the path of each nucleon through the target nucleus using the Newtonian equations of motion in the self-consistent mean field, with two-body collisions. The cross-sections for multiple emission have been calculated for both FKK and QMD by Chadwick *et al* (1995) and the results are in good agreement, as shown in Fig.16.

These and other calculations show that two-step processes become significant ($> 5\%$) for incident energies above about 20 MeV and three-step processes above 100 MeV.

The FKK theory has recently been extended to include alpha-particle emission (Olaniyi *et al*, 1995) using the knock-out model. It is assumed that the incident proton collides with a pre-formed alpha-particle in the target nucleus and knocks it out, the proton being captured into an orbit in the residual nucleus. The transition matrix is expressed in terms of the wavefunctions of the initial and final states and the proton-alpha particle effective interaction, and is multiplied by the probability of finding the alpha-particle in the target nucleus. Double differential cross-sections for the (p, α) reaction at 30 and 44.3 MeV on several nuclei were analysed using the subtraction method, and some results are shown in Fig.17. The angle-integrated cross-sections show a strong peak at lower energies attributable to compound nucleus emission, and this was evaluated using the Hauser-Feshbach theory. The total cross-section is well described by the sum of compound nucleus and MSD processes, as shown in Fig.18.

I am grateful to all those colleagues who kindly permitted me to reproduce figures from their papers.

Fig. 13. Angle-integrated spectra for the inelastic scattering of 18 MeV protons by ^{98}Mo and 14.1 MeV neutrons by ^{93}Nb showing the sensitivity to the optical potentials (Watanabe *et al.* 1995).

Fig. 14 The cross-section of the $^{90}\text{Zr}(p,p')$ reaction at 80 MeV compared with multistep direct calculations (Cowley *et al.* 1991).

Fig. 15 Angle-integrated cross-section for the $^{90}\text{Zr}(p, xn)$ reaction at 160 MeV compared with FKK calculations including multiple emission. The contributions of the individual steps to the first and second emissions are shown separately (Chadwick *et al*, 1994).

Fig. 16 Comparison of primary and multiparticle emission cross-sections for the $^{90}\text{Zr}(p, xp')$ reaction at 160 MeV. The upper section shows the results of FKK calculations and the lower those of the QMD theory. At the lower emission energies the FKK theory was supplemented by compound nucleus calculations using the program GNASH, while for the QMD theory the statistical theory was used (Chadwick *et al*, 1995).

Fig. 17 Subtracted double-differential cross sections for the (p, α) reactions at 30 MeV (Ferrero *et al* 1978) for ^{118}Sn compared with similarly subtracted zero-range MSD FKK calculations using the alpha-particle knock-out model (Olaniyi, Demetriou and Hodgson, 1995).

Fig. 18 Angle-integrated cross sections for the (p, α) reaction at 44.3 MeV (Ferrero *et al* 1978) compared with zero-range MSD FKK and compound nucleus calculations (Olaniyi, Demetriou and Hodgson, 1995).

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