SELECTED ASPECTS OF PREEQUILIBRIUM PARTICLE EMISSION IN NUCLEAR REACTIONS

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The investigation of nonequilibrium phenomena in physical systems has been one of the main fields of research during the past two decades. In this paper we review the results of such investigations on a small-size many-body system with a variable number of particles, i. e. on atomic nuclei. Particular attention is paid to the achievements of the Zagreb group in the development and application of the preequilibrium model of nuclear reactions from its very beginning.

1. Introduction

Until the mid-sixties only two reaction mechanisms were considered when analyzing nuclear reactions, i. e. the formation and decay of the compound nucleus and direct processes.

The compound-nucleus concept was proposed by N. Bohr$^{1)}$. It supposes that the formation and decay of the composite (projectile and target) system are independent steps of a nuclear reaction except for conservation laws. A subsequent development of this concept was the Weisskopf-Ewing$^{2)}$ statistical theory of the spectra of particles evaporated from the compound nucleus. The theory was later improved by Hauser and Feshbach$^{3)}$ by including angular-momentum conservation. These theories predict an approximately Maxwellian energy distribution for evaporated particles with an angular dependence symmetric around 90°.

On the other hand, direct reactions$^{4,5)}$ connect the initial and final states in a nuclear-collision process without formation of an intermediate compound system. Contrary to compound-nucleus reactions, direct processes favour the emission of high-energy particles with the excitation of low-energy states of the residual nucleus with usually forward-peaked angular distributions of diffractive character.

Direct and compound-nucleus processes also take place on a very different timescale. While direct reactions occur approximately in the time taken by the projectile to traverse the target nucleus ($\approx 10^{-22}$ s), compound-nucleus reactions last much longer (10^{-15}—10^{-19} s, depending on incident energy). Our knowledge of the timescale of nuclear reactions comes partly from theoretical considerations (Heisenberg uncertainty principle) and partly from special experimental techniques (blocking method$^{6)}$).

The accumulation of data in the fifties and the early sixties indicated the existence of reaction mechanisms intermediate between one-step direct processes (involving only a few degrees of freedom) and compound-nucleus reactions (with the incoming energy shared by all nucleons in a completely statistical way). For obvious reasons, these intermediate processes are named preequilibrium or precompound reactions$^{7-9)}$.

In this paper we review the results of the Zagreb group in studying preequilibrium reaction mechanisms.

2. *Preequilibrium models*

2.1. *Master-equation approach*

The equilibration process following the initial project-target interaction in a nucleus is commonly envisaged as proceeding via a chain of intermediate states characterized by a number of excited particles and corresponding holes, together called excitons. Successive two-body residual interactions give rise to an intranuclear cascade which eventually leads, through a sequence of exciton states, to a fully equilibrated nucleus. Supposition of the two-body character of residual interactions leads to the $\Delta n = 0, \pm 2$ selection rule concerning the possible variation of n, the number of excitons. At each stage of this equilibration there is a competition among the transition to the more complex $\Delta n = +2$ state or back to the $\Delta n = -2$ state or to the emission of particles. Thus, the probability of finding the composite nucleus with excitation energy E in the n-exciton configuration $P(n, E, t)$ at the time t may be obtained by solving a set of equations$^{10,11)}$

$$\frac{\mathrm{d}P(n, E, t)}{\mathrm{d}t} = P(n-2, E, t)\,\lambda_+(n-2, E) + P(n+2, E, t)\,\lambda_-(n+2, E) - \\ - P(n, E, t)\,[\lambda_+(E) + \lambda_-(E) + W_c(n, E)], \qquad (1)$$

where λ_+ is the transition rate for $\Delta n = +2$, λ_- the transition rate for $\Delta n = -2$ and $W_c(n, E)$ the total decay rate into the continuum of whatever particle with whatever energy from an n-exciton state (of course, allowed by the conservation of energy). The set of master equations may be solved using numerical and analytical methods once the value of $P(n, E, t)$ at $t = 0$ is given. For light-particle induced reactions, one usually takes $P(n, E, 0) = \delta_{nn_0}$.

The cross section for the emission of particles in the channel c is easily calculated once $P(n, E, t)$ is known, i. e.

$$\sigma_c(E, \varepsilon_c)\,\mathrm{d}\varepsilon_c = \sigma_R \int_0^{t_{eq}} \sum_{n, \Delta n = 2} P(n, E, t)\, W_c(n, E, \varepsilon_c)\, \mathrm{d}t\mathrm{d}\varepsilon_c, \tag{2}$$

where σ_R is the reaction cross section for the incident channel.

In the early days of the above approach, a question arose as to whether and how the transitions $\lambda_0\,(n \to n)$ influence the preequilibrium emission of particles. Starting from a general master equation in which each specific particle-hole configuration is treated separately, we showed[12)] that the commonly used set of master equations with averaged transition and emission rates does not contain terms with λ_0.

2.2. *Closed-form exciton model*

The consideration of transition rates shows that λ_+, the transition rate to more complex exciton states, is larger than λ_-, the transition rate back to less complex states. This is the reason that the composite system evolves towards the equilibrium (which is then characterized by $\lambda_+ = \lambda_-$). On the other hand, the emission rate to the continuum λ_c decreases with increasing exciton number. Thus, in many practical situations it is possible to use the closed-form expressions instead of the master equation in performing preequilibrium calculations. The original closed-form expression of Griffin[13)] was, however, modified in order to take into account the Pauli principle, proton-neutron distinguishability, finite depth of the nuclear potential well, etc.

Here we present only the closed-form expression with a short explanation of symbols. Details can be found in Ref. 14, for example. The differential precompound cross section is given by

$$\frac{\mathrm{d}\sigma(a, \beta)}{\mathrm{d}\varepsilon_\beta} = \sigma_{CN}(a, E_i) \sum_{n = n_0,\, \Delta n = 2} W_\beta(n, E, \varepsilon_\beta)\, T(n, E). \tag{3}$$

According to Eq. (3), the emission of particles of type β with kinetic energy ε_β from an n-exciton state of the composite system at excitation energy E, is proportional to the emission rate $W_\beta(n, E, \varepsilon_\beta)$ multiplied by the time $T(n, E)$ the system spends in this particular state. $\sigma_{CN}(a, E_i)$ is the cross section for the formation of the composite system by an incoming particle a of energy E_i. In the absence of direct processes, $\sigma_{CN}(a, E_i)$ is equal to the reaction cross section σ_R. The emission rate $W_\beta(n, E, \varepsilon_\beta)$ of particles from an exciton state composed of p particles and h holes $(n = p + h)$ is given by[15)]

$$W_\beta(n, E, \varepsilon_\beta) = \frac{2s_\beta + 1}{\pi^2 \hbar^3}\, \mu_\beta\, \varepsilon_\beta\, \sigma_\beta(\varepsilon_\beta)\, Q_\beta(p)\, \frac{\omega^{EP}(p - p_\beta, h, U)}{\omega^{EP}(p, h, E)}. \tag{4}$$

Here ω^{EP} is the Ericson state density[16)] corrected for the Pauli principle, s_β and μ_β are the spin and the reduced mass of the emitted particle β, respectively; $\sigma_\beta(\varepsilon_\beta)$

is the inverse cross section, E and U are the excitation energy of the composite and residual nucleus, respectively. $Q_\beta(p)$ is the proton-neutron-distinguishability factor.

The time $T(n, E)$ depends essentially on the λ_+ transition rate, which is determined by the golden rule, i. e. λ_+ depends on the density of the accessible final states and on the average matrix element squared of the two-body interaction.

3. *Light-particle induced reactions*

3.1. *Early applications of the preequilibrium model*

After the pioneer paper by Griffin$^{13)}$, the simple closed-form expression of the preequilibrium model was extensively used to calculate spectra (and, less successfully, the absolute cross sections) of reactions with nucleons in both incoming and outgoing channels. Čaplar and Kulišić$^{17,18)}$ showed that the preequilibrium model could be successfully applied to (n, α) and (p, α) spectra at incoming energies above 10 MeV. (See, e. g. Fig. 3 in Ref. 18.)

3.2. *(n, 2n) reactions*

$(n, 2n)$ reactions induced by 14 MeV neutrons were considered for a long time as a par excellence example of compound-nucleus reaction mechanisms. This consideration was based on the fact that even the simplest closed-form evaporation expressions could account for the total cross sections in a wide range of nuclei within better than 20 to 30% of the experiment. However, a systematic study performed by Holub and Cindro$^{19,20)}$ showed that the calculations based on the compound-nucleus model (even with the angular momentum included) in general yield higher values of $(n, 2n)$ cross sections than observed in the experiments. They interpreted the above observation as due to the presence of preequilibrium-emission mechanisms. Preequilibrium emission hardens the spectrum of primary neutrons and in this way reduces the fraction of neutrons capable of giving rise to the emission of secondary neutrons; thus the reduced calculated values are in agreement with experimental data. The above conclusion was confirmed in a series of kinematically complete measurements of $(n, 2n)$ cross sections performed at the Hamburg cyclotron$^{21,22)}$.

3.3. *Systematic investigation of neutron-induced reactions*

A nucleon with an energy higher than, say, 10 MeV, when colliding with a target nucleus, gives rise to the emission of various particles: neutrons, protons, α-particles, tritons, ^{3}He and combinations thereof; the cross sections for the various emissions differ by orders of magnitude, depending on the mass and charge of the target nuclei. Furthermore, many of the reactions in this energy range are threshold reactions and consequently display a strong dependence on the incoming energy.

Holub et al.$^{14)}$ performed a systematic study of precompound (and compound-nucleus) emission. The analysis was performed for twelve nuclei in the range $A = 45$—209. The calculated spectra and the excitation functions for (n, n'),

(n, p), $(n, 2n)$ and $(n, 3n)$ reactions at $E_n = 4$—24 MeV were compared with experimental data. The main conclusion was that all these data could be reproduced simultaneously and in a consistent way when preequilibrium emission was taken into account in addition to the compound-nucleus and direct processes.

3.4. *(n, p) isotopic trend*

The systematic dependence of various (n, particle(s)) cross sections on the neutron number of isotopes (isotopic effects) in reactions with fast neutrons was noticed in the early days of nuclear physics and has since represented a challenging and fruitful field of investigation. Pioneer papers on the subject tried to establish phenomenological and/or semiphenomenological expressions in order to describe the observed isotopic trends. The semiempirical approaches were of course based on the compound-nucleus model. However, more precise measurements with modern experimental techniques yielded data including low (≈ 1 mb) cross sections on a number of low-abundance isotopes. The data showed that the statistical evaporation model is not able to describe isotopic effects all over the periodic table.

The (n, p) isotopic effect in reactions induced by fast (14 MeV) neutrons is of particular interest. The (n, p) cross sections for a given element generally decrease with increasing neutron number of an isotope for all the elements. However, the data for intermediate-mass and heavy elements show a markedly less steep slope than for lighter nuclei. Čaplar et al.[23] showed that the preequilibrium model (together with an appropriate contribution from compound-nucleus emission which is important for neutron-deficient intermediate-mass isotopes) describes both the absolute values and the observed slope of (n, p) cross sections quite well (see, e. g. Figs. 1 and 3 in Ref. 23). In this way it was demonstrated that (n, p) reactions on heavy target-nuclei are preequilibrium processes par excellence. This can be easily understood by noting that when going from lighter to heavy targets, the increasing Coulomb barrier hinders the emission of low-energy evaporation protons more and more strongly. Finally, the Q-values were identified in Ref. 23 as the basic parameter responsible for the observed isotopic behaviour for heavier targets.

3.5. *Preequilibrium model and the structure of nuclei*

3.5.1. *Transition rates*

A systematic study of neutron reactions[14] showed that the data could be reproduced only by calculations performed using the structure-independent transition rate, i. e. the rate which does not depend on the specific target nucleus. The study was performed on a wide range of targets with one stable isotope only; the level density parameter a varied along the valley of stability. The same conclusion was drawn in our study of (n, p) isotopic effects[23] on heavier nuclei, i. e. on a completely different sample of data where the level density parameter a varied across the valley of stability. The two analyses mentioned above have thus considerably contributed to answering the question of the difference between compound-nucleus emission (with the structure-dependent level-density parameter a) and preequilibrium emission (the structure-independent transition rate λ_+ closely related to the structure-independent parameter a).

Let us mention that although in most preequilibrium calculations it is not necessary to take care of details of nuclear structure, there are situations where nuclear structure is of importance (see next subsection).

3.5.2. *High-energy part of preequilibrium spectra*

Semiclassical preequilibrium models which are based on intranuclear nucleon-nucleon transition rates, and incorporate the equidistant-level-spacing assumption, successfully reproduce the smooth structureless part of the experimental preequilibrium spectra contributing to the major part of the integrated preequilibrium cross section. However, it is obvious that, because of this assumption, these models cannot describe the gross structure observed at high spectral energies (corresponding to low excitation energies in the residual nucleus). In the experimental preequilibrium spectra, this structure is most pronounced in reactions with target nuclei in the vicinity of the closed neutron and/or proton shells suggesting its close connection with the structure of nuclei. The measurements and the subsequent analysis of (p, xn) reactions performed by the Hamburg group[24,25)] and recently by the Hamburg—Zagreb group[26)] have shown that this is indeed the case. Furthermore, in Ref. 26, the influence of deformation on preequilibrium spectra in (p, xn) reactions has been demonstrated on a series of increasingly deformed Pd isotopes.

4. *Heavy-ion induced reactions*

As shown above, light-ion induced reactions have considerably contributed to the present-day knowledge of nonequilibrium phenomena in atomic nuclei. However, these reactions are by their very nature limited to asymmetric projectile-target combinations and relatively low angular momenta. The development of heavy-ion accelerators in the eighties provided beams of various heavy projectiles with energies per nucleon corresponding to light-ion energies at which preequilibrium phenomena were observed. And, indeed, at these energies the preequilibrium emission of particles was also observed in heavy-ion[27)] reactions.

4.1. *Spectra of light particles from heavy-ion collisions*

The proper assignment of the preequilibrium contribution is, however, more involved in the experimental spectra of particles emitted in heavy-ion than in light-ion induced reactions. This is because the evaporation from, e. g., the fast projectile-like fragment can also contribute to the high-energy part of the spectra competing with true preequilibrium particles. In addition, evaporated particles from fast fragments are forward peaked for kinematical reasons. Thus, neither the relatively high-energy nor the forward-peaked angular distributions guarantee that the observed particles stem from preequilibrium emission. The preequilibrium-emission contribution can be extracted from the data using multisource analysis. The method tries to fit the spectra at as many angles as possible by taking into account the emission of particles from a few (physical) sources (composite system, projectile- and target-like fragments, ...).

The Zagreb group[28,29] introduced the so-called refined multisource analysis into the field. The main point of this refinement is a built-in asymmetry factor which explicitly takes into account the anisotropy of the emitted preequilibrium particles already in the source system (c. m. system). The degree of anisotropy is taken as depending on the relative energy of preequilibrium particles, in full accordance[30] with the observed behaviour in light-particle induced reactions. In this way it is possible to reliably disentangle the preequilibrium contribution in the light-particle (nucleon) spectra of heavy-ion collisions.

4.2. *Equilibration of a nuclear system; initial number of degrees of freedom*

An important quantity in preequilibrium calculations is the initial number of degrees of freedom n_0. The analysis of a large body of preequilibrium data has shown that $n_0 = 3$ in nucleon-induced reactions (incoming nucleon-particle and excited particle-hole pair). In heavy-ion reactions the situation is more complicated. This can be readily understood by writing down the master equation for heavy-ion collisions[31]. Schematically, it reads

$$\mathrm{d}n/\mathrm{d}t = \text{gain term} + \text{loss term} + \text{escape term} + \text{mixing term (fusion)}.$$

This equation, as compared with that for light-ion induced reactions, contains a new term: it is the mixing term describing the gradual fusion of two heavy ions. Comparison of the calculated contribution obtained using the above equation with the experimental preequilibrium contribution enables us to extract n_0. We performed[32,33] analyses of exclusive neutron spectra from ^{20}Ne + ^{165}Ho and of inclusive proton spectra from ^{16}O, ^{32}S and ^{58}Ni on a series of targets from ^{27}Al to ^{197}Au in the energy range 10—25 MeV per nucleon. The main feature of the best-fit values of n_0 is their dependence on the entrance channel: the values are grouped around the mass number of the projectile (A_P), viz. for collisions induced by the heavy projectile ^{58}Ni, they are grouped around the mass number of the lighter collision partner. Furthermore, the values of n_0 show an increase with increasing mass of the system (i. e. with the target mass (A_T) for a given projectile).

4.3. *Sharing of the excitation energy in the initial stages of nucleus-nucleus collisions: preequilibrium-temperature concept*

The above results differ somewhat from the prescription $n_0 = A_P$ as used by Blann[34], which states that n_0 is independent of the target. However, as shown by Korolija et al.[35], another important physical quantity turns out to be independent of the target (system) properties, i. e. E^*/n_0, with E^* the excitation energy of the composite system formed in a central collision (see Fig. 2 in Ref. 35). This result led us[35,36] to introduce the so-called preequilibrium temperature parameter when describing the early stages of a nucleus-nucleus collision. Namely, the quantity $E^*/\overline{n}$ ($\overline{n}$ being the number of degrees of freedom in equilibrium) has a precise physical meaning in equilibrium thermodynamics, i. e. it is related to the temperature by $E^*/\overline{n} = kT/2$, with k the Boltzmann constant. Based on the similarity between $E^*/\overline{n}$ and E^*/n_0 (equal sharing of energy among various degrees of freedom), one can introduce a quantity T_{PE}, i. e. an analogue for nonequilibrated systems of what »normal« temperature is for equilibrated systems.

In analogy to the equilibrium temperature related to the average energy per equilibrated degree of freedom, the newly introduced preequilibrium-temperature parameter T_{PE} is simply related to the average energy per initial degree of freedom. One should, however, keep in mind that by the theoretical derivation of the »preequilibrium temperature« T_{PE}, the validity of some propositions is not so obvious for the nonequilibrated spot-heated system as for the fully equilibrated system. In this respect, the »preequilibrium temperature« T_{PE}, although quite analogous to the thermodynamic temperature T, appears rather as a parameter serving to describe the behaviour of the system in its very early stages. However, being essentially determined only by the size of the projectile and the total available energy and thus independent of the microscopic properties of the system, the quantity T_{PE} is a very useful parameter.

5. *Summary and conclusions*

The preequilibrium model of nuclear reactions has reached its mature stage twenty-five years after the pioneer paper by Griffin[13]. On the theoretical side, the model is now firmly founded[37], although some problems are still open. On the practical side, it allows quite reliable calculations of particle emission over a time span of many orders of magnitude (from 10^{-22} to 10^{-16} s) both in light-ion[38] and heavy-ion[39] induced reactions.

In this paper we have mainly stressed the contributions of the Zagreb group. These contributions are in the following fields:

— improvements of the preequilibrium model (role of λ_0),

— first evidence for the preequilibrium contribution in $(n, 2n)$ reactions and one of the first applications of the model to the reactions with composite particles in the exit channel,

— explanation of the isotopic trend in (n, p) reactions on heavy nuclei,

— definite evidence that the preequilibrium model (coupled with compound-nucleus and direct processes) can simultaneously and consistently account for various cross sections differing by orders of magnitude,

— development of the method for proper separation of preequilibrium particles from fast particles of different origin in heavy-ion collisions,

— introduction of the »preequilibrium temperature« concept, which at least represents a successful and convenient parametrization of nonequilibrium particle emission in energetic heavy-ion collisions.

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PREDRAVNOTEŽNA EMISIJA ČESTICA U NUKLEARNIM REAKCIJAMA

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Istraživanje neravnotežnih fenomena u fizikalnim sustavima predstavlja jedno od glavnih područja istraživanja tijekom proteklih dvadeset godina. Ovaj rad daje pregled rezultata takvih istraživanja na malim sustavima s promjenljivim brojem čestica, tj. na atomskim jezgrama. Posebice su prikazana dostignuća što su ih ostvarili istraživači iz Zagreba u razvoju i primjeni predravnotežnog modela nuklearnih reakcija od njegovog samog početka.