Comments

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Comments on alternate formulations for preequilibrium decay*

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The physical and mathematical differences of several formulations for preequilibrium decay are discussed. Mathematical models and examples are presented or referred to in order to illustrate what the author believes to be errors in the exciton formulation as being due to improper inclusion of spectator effects. An earlier work of Gadioli *et al.* is reinterpreted, and quotations therein to work of the present author are corrected.

NUCLEAR REACTIONS Preequilibrium decay formulations; compositions and comments.

I. MOTIVATION AND GOALS

Two superficially similar approaches to the problem of preequilibrium nucleon emission are presently in use; however, the underlying phys-ical assumptions and consequences are very different. One formulation describes a direct multistep process¹⁻⁸; the other, a quasi-equilibrium process.⁹⁻¹³

The differences have never been clearly aired. The first goal of this comment is, therefore, to present one point of view on the subject in the hope that proponents of the alternate approach will respond in kind, thereby clarifying the issues involved. Secondly, I should like to clarify what I consider to be incorrect statements of conclusions of work published in Ref. 7 by the authors of Ref. 13; and, third, I would point out the reasons that I reach some conclusions opposite to those given in Ref. 13 upon close reading of this work.

II. DIFFERENCES IN FORMULATIONS

The hybrid model for preequilibrium decay has been formulated $as^{1, 6}$

$$\begin{pmatrix} \frac{d\sigma}{d\epsilon} \end{pmatrix} = \sigma_R \sum_{\substack{n=n_0}}^{n} \left[{}_{n} p_x \frac{N_{p,h}(U,E)}{N_{p,h}(E)} \right] \left[\frac{\lambda_c(\epsilon)}{\lambda_c(\epsilon) + \lambda_*(\epsilon)} \right] D_n$$

$$= \sigma_R \sum_{\substack{n=n_0}}^{\bar{n}} P_n(\epsilon)$$
(1)

representing a sum over spectral contributions from a monotonically increasing (in n) series of

configurations characterized by the number of excited particles (p) plus holes (h) or excitons (n = p + h).

The quantity in the first set of square brackets in Eq. (1) gives the number of neutrons or protons which have energies in the continuum between ϵ and $\epsilon + d\epsilon$. A quantity called a scattering distribution function, $N_{\mathbf{p},\mathbf{h}}(E)$, or a partial state density, $\rho_{\mathbf{p},\mathbf{h}}(E)$, has been used in evaluating the number of excited particles with the mathematical relationship

$$N_{p,h}(E) = \rho_{p,h}(E)/g = (gE)^{p+h-1}/p!h!(n-1)!.$$
 (2)

In the ratio of Eq. (1), the use of either $N_{p,h}$ or $\rho_{p,h}$ gives identical results mathematically; however, the difference in interpretation and application is great and leads to different results for the expression for the fraction of particles emitted which is given in the second set of square brackets in Eq. (1). It has been shown that the distribution given by Eq. (2) can be derived as a consequence of an intranuclear scattering cascade due to the isotropy of $d\sigma/d\epsilon$ for allowed nucleon-nucleon scattering in nuclear matter.⁸ The near equivalence of scattering cross sections for different initial nucleon energies leads to the equal a priori population of each partition. This is an intranuclear cascade interpretation where the energy distribution function of the scattered nucleons follows the kinematic predictions of the angular distribution of free N-N scattering modified by the Pauli exclusion principle.

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A physically different interpretation is implied if the function is used as a state density with the equal *a priori* population assumption. Under this interpretation, with detailed balance being applied in the derivation of the final rate equations, there is an implication of quasi-equilibrium between each hierarchy of states and the final state particle plus residual nucleus configuration. This carries with it the requirement that the transition rate within a given configuration be very much greater than the continuum decay rate, or greater than the intranuclear transition rate to other exciton hierarchies.

That the requirement for quasi-equilibrium is not met may be simply demonstrated. First, as was pointed out by Feshbach,¹⁴ guasi-equilibrium results in an angular distribution which is symmetric about 90°; experimental angular distributions for preequilibrium spectra are forwardpeaked,¹⁵ consistent with the interpretation as being due to a series of direct processes (i.e., an intranuclear cascade). This may also be seen from a trivial theoretical consideration⁷): If the initial configuration in a reaction consists of two particles and one hole, in, e.g., a mass 100 nucleus, each particle can interact with one excited particle (λ_0 transition rate) and ~100 particles in the ground state (λ_+ transition rate). Clearly the transition rate to more complicated configurations (λ_{\star}) is much greater than the rate within a given hierarchy (λ_0) , just the reverse of the requirement for quasi-equilibrium. The quasi-equilibrium assumption, therefore, appears to be invalid both on the basis of experimental evidence (angular distributions) and from simple theoretical considerations.

One should next explore the consequences of the application of equilibrium physics to a nonequilibrium system. The second term in Eq. (1) in the hybrid model formulation gives the fraction of the particles in the ϵ to $\epsilon + d\epsilon$ energy range which is emitted into the continuum. This is calculated as the ratio of the rate of particle emission into the continuum $[\lambda_{c}(\epsilon)]$ to the total nucleon transition rate where $\lambda_{+}(\epsilon)$ represents the rate at which that particle makes intranuclear transitions to n+2particle-hole states. By deriving an analogous expression based on detailed balance within each hierarchy of state, the $\lambda_{\epsilon}(\epsilon)$ and $\lambda_{\star}(\epsilon)$ of the denominator are replaced by average values over all particles and holes of the *n*-exciton state.⁹⁻¹³ At this point it can be seen that the philosophical difference in formulations leads to mathematical differences in results. These in turn require large differences in a mean-free path parameter.

The use of an average transition rate in (1) means that if one particle in, e.g., a two-particle,

one-hole state, is at a given energy, and the spectator particle makes a transition, then the first particle can no longer be emitted at the initial energy even though its energy is unchanged. This point can be illustrated by means of an example⁸:

Consider a reaction on a target of mass 70 induced by an incident particle at 50 MeV, which has a 10 MeV binding energy in the nucleus. The excitation energy of the composite nucleus is 60 MeV. Consider next the emission of a nucleon from the 2p-1h and 3p-2h states with 40 MeV of energy, leaving a residual nucleus with 10 MeV of excitation. Application of the partition function from Eq. (1), $N_3(10, 40)/N_3(60)$ and $N_3(10, 40)/N_3(60)$ gives the number of particles in the 40 ± 0.5 MeV range as 0.011 for the three quasiparticle configuration and 0.00093 for the five quasiparticle configuration.

Next, one should consider the relevant lifetimes which enter the two formulations. All lifetimes for these examples have been taken from the work of Gadioli et al.¹¹ The estimated lifetime for a particle at 50 MeV (40 MeV in the continuum) for intranuclear transitions is 0.45×10^{-22} sec).¹¹ This is the same for a particle in a 3p-2h configuration, as it is the single particle and not the exciton state lifetime. In the hybrid formulation, this determines the mean time that the particle remains at 50 MeV with a chance for emission into the continuum [assuming, to simplify the example, that $\lambda_c(\epsilon) \ll \lambda_{+}(\epsilon)$]. But in the exciton formulation the lifetimes are $\tau_{2p-1h} = 0.19 \times 10^{-22}$ sec and $\tau_{3p-2h} = 0.16 \times 10^{-22}$ sec.¹¹ It can be seen that the spectator transitions reduce the chance of emission of the particle at 50 MeV by a factor of 2.5 in this example. The fact that the chance of finding a particle at 50 MeV in the 3p-2h configuration versus the chance in the 2p-1h configuration is reduced by a factor of 12 assures that the emission chance lost by spectator transitions is never regained in higher order (in n) terms of Eq. (1).

The spectator bookkeeping error has been demonstrated in yet another way. The Harp-Miller-Berne (HMB) master equation¹⁶ was solved for a test case to give the time-weighted population of particles in each excitation range during equilibration. Because the HMB approach uses no scattering distribution function, it is free of the question of single particle versus average state transition rates. The same test case (⁵⁴Fe +39 MeV p) was calculated using the two approaches under discussion.⁶ The hybrid model gave a time-weighted population in agreement with the HMB model result to within 10% over the entire excitation range considered and over a dynamic range of 50. The use of average transition rates, on the other hand, gave weighted

populations which were low by factors of 2 to 2.5, illustrating the consequence of the "spectator" transition in the exciton formulation.

Primarily due to inclusion of the spectator transition rate in the exciton formulation, the authors⁹⁻¹³ have found it necessary to compensate by increasing mean-free paths to values of 3.3 to 5 times the values predicted by Pauli corrected nucleon-nucleon scattering cross sections. This in turn gives nucleon mean-free paths in nuclear matter of 15 to 30 fm. By comparison, the hybrid and geometry dependent hybrid models use the same Pauli corrected nucleon-nucleon mean-free paths as are used in the intranuclear cascade calculations. Alternatively, $\lambda_{\star}(\epsilon)$ is computed in the hybrid model⁵ using the imaginary optical potential parameters independently determined by Becchetti and Greenlees.¹⁷ Both values of $\lambda_{+}(\epsilon)$ give similar results, both are in good agreement with experimental spectra, and neither set of calculations requires modification of the independently calculated mean-free paths. 5-8, 18

One should consider the consequences if the intranuclear mean-free paths were truly 3-5 times the values derived from the optical potential or N-N scattering: (1) Calculated angular distributions with such mean-free paths would be nearly isotropic if orbiting trajectories were assumed, whereas experimental results are strongly forward-peaked.¹⁵ (2) Cascade calculations which reproduce angular distributions reasonably well for preequilibrium reactions would fail¹⁹ due to a reduction in multiple collision contributions. (3) Related calculations by Weidenmüller and coworkers,^{20,21} which use hybrid or exciton approaches to reproduce experimental angular distributions, would probably be in error; the higher order collisions necessary to reproduce the large angle yields would take place outside of the nucleus, i.e., be physically unrealistic to include. (4) Optical model calculations using *realistic* form factors and parameters would probably give values of σ_R for nucleon-induced reactions which were too low due to transparency.

To summarize, in my opinion it can be demonstrated by mathematical examples that the exciton model equivalent of Eq. (1) uses an improper lifetime dependence due to improper application of detailed balance in its derivation. This leads to unrealistic parameter values for mean-free paths in order to compensate for the first error.

It should be emphasized that the hybrid formulation of Eq. (1) is by no means exact and rigorous. However, it has been shown (in particular in the geometry dependent form) to be a good approximation to a more rigorous calculation,^{2,4} to be highly flexible (e.g., in including the density dependence) and to be one which uses commonly accepted parameters for *a priori* calculation of experimental spectra.

III. COMMENTS ON INTERPRETATIONS OF GADIOLI et al.

In this section, I should like to correct what I consider to be an incorrect interpretation and quotation of results of Ref. 7 by Gadioli *et al.*,¹³ and further to suggest that I would give an interpretation to their results which is rather opposite to their own. The points at issue can best be discussed by reviewing the geometry dependent hybrid model³:

$$\frac{d\sigma}{d\epsilon} = \pi \lambda^2 \sum_{l=0}^{\infty} (2l+1) T_l \sum_{n=n_0} P_n(\epsilon) , \qquad (3)$$

where the transmission coefficients are optical model results for the particle in the entrance channel. For each partial wave of the incident particle, the hybrid model terms (given by the summation over n) are computed for nuclear matter or for a potential averaged over the particle trajectory.³⁻⁵ If N-N scattering results are used for computing $\lambda_{+}(\epsilon)$, a Fermi density distribution is used for the nucleus with constants derived from electron scattering. For the average density along each trajectory, an average value of the Fermi energy is calculated in a local density approximation. An alternative option⁵ (which gives similar results^{5, 4, 18}) is to compute $\lambda_{+}(\epsilon)$ along the trajectory by averaging over the imaginary potential and form factors of Becchetti and Greenlees.17

In response to the questions raised in the reply to this comment concerning the large contributions from surface reactions in this approach, I would emphasize the following: The relatively large partial reaction cross sections in the surface region are a result of the nuclear optical model, not the GDH model. Use of T_i values from the optical model and densities from electron scattering reflect a personal conviction that these give a more realistic representation of nuclear properties than the square well distribution used in the exciton or hybrid models.

In applying Eq. (3) scattering distribution functions (or partial state densities) are used in which the hole excitation is restricted to the Fermi energy for each T; this is done for the first term in the series which, for nucleon-induced reactions, is assumed to be 2p-1h in nature, and which is the predominant contributor to the higher energy portion of the preequilibrium spectrum.³

Let me now emphasize that, contrary to the statement in Ref. 13, we always use an initial 2p-1h configuration for nucleon-induced reactions. not a 2p-0h configuration; the question of density variation is treated in a smooth, consistent, and continuous fashion as given by Eq. (3); we do not have a distinction between "surface and volume interactions based mainly on the hypothesis that in a surface process practically no hole states should be excited."¹³ The equations for averaging over the Fermi density distribution are given in Ref. 5 and are available in computer codes which have been given wide circulation and use.^{22,23}

Next, I would summarize the main conclusions from Ref. 3:

(1) It was shown that a calculation with densities averaged over trajectory, but permitting excitation of infinitely deep holes, was roughly equivalent to a hybrid model with mean-free paths increased by a factor of 2, due to longer average mean-free paths in the region of the nuclear surface; however, a poor spectral shape resulted at higher bombarding energies.

(2) By using $N_{p,h}(E)$ with hole excitations limited to the local density Fermi value, the proper shape and cross section resulted. Later works reported spectra calculated with the hybrid formulation but with hole depth limited to 20 MeV²; good spectral shapes resulted but absolute cross sections were low.

In view of these conclusions concerning the geometry dependent model, I would point out the following with respect to Ref. 13: (1) At bombarding energies above 30 MeV the calculations of Ref. 13 use a partial state density expression with a limit to the depth of hole excitations of 20 MeV, a change from earlier calculations and consistent with the example of Ref. 2. (2) They use, as discussed, a mean-free path which is four times the value calculated from N-Nscattering (or optical model) results. It was demonstrated in Eq. (3) that a factor of 2 in the mean-free path approximately compensates the hybrid model to the geometry dependent model result. The other factor of 2 can be ascribed to compensation due to the spectator transition rate effect discussed above.

It was shown in Ref. 2 that a hybrid model [Eq. (1)] calculation with a hole depth limit to 20 MeV gives a reasonable spectral shape (but low absolute cross sections) whereas the same calculation without limited hole depth does not (for projectile energies significantly above 20 MeV). The fact that the same is found in the calculation of Ref. 13 is consistent with this finding; I would interpret this result as confirmation of the necessity of the geometry dependent approach, specifically the necessity of limiting the hole degree of freedom, and find this aspect of the results of Ref. 13 consistent with our own earlier findings.^{2,3} Our conclusions on the failure of treatments using infinitely deep wells, ⁷ as in earlier calculations with the exciton model, ⁹⁻¹¹ stands. By changing the method of calculation to one utilizing a hole depth limit, Ref. 13 does not disprove this conclusion; rather a crude confirmation is provided. I am confident that if a step further was taken and the sum over impact parameter done as in the GDH model [Eq. (3)], the results would improve still further.

Finally, I should like to comment on a few aspects of the reply²⁴ to this "Comment." This mainly centers on the section in which arguments are given, with reference to an article²⁵ in which it is claimed that abundant evidence is presented to show that the longer mean-free path (mfp) values (4 times normal) are supported by the cascade model as well as the hybrid model.

Reference is made²⁵ to cascade calculations by Bertini in which the calculated (p, p') precompound spectra are low by an average of 20% (Tabel VII of Ref. 26) with respect to the experimental yields. Due to this discrepancy, it is concluded that the cascade calculations support the contention that the mean-free path should be increased fourfold. On the other hand, in comparing their own calculation with experimental results (Ref. 13, p. 581) the authors state: "The curve" (exciton model calculation) "runs some 30% lower than the average derived from the measured data, but this can be considered in our opinion as good an agreement as might be reasonably expected."

In reply to the lack of quantitative grounds for my statements concerning mfp values, a simple transmission model calculation was presented in Ref. 27, showing that calculated reaction cross sections in, e.g., $^{89}\mathrm{Y},$ are decreased to $\sim 40\%$ (419 mb) of the experimental value by such long mfp values. Reference 27 incidentally responds to each point made in Ref. 25 and finds no basis for the claim that the longer mfp values are model independent. Further quantitative comparisons have been made by actually comparing cascade calculations with a fourfold mfp increase with standard results.²⁸ Suffice it to say the reaction cross sections with the longer mfp values were 467 and 302 mb for ⁹⁰Y and ⁵⁴Fe targets, compared with experimental yields of 1059 and 733 mb,²⁹ and calculated yields of 928 and 678 mb using standard mfp values. The precompound spectra showed either no improvement or poorer agreement versus experimental yields, and obviously the compound spectra would be disastrous. These considerations also show why the comparison made in the reply to this comment to a result of Miller's (in which the mfp values were increased fourfold, but without recalculating the reaction cross section in a consistent fashion) is at best inconclusive.

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