# THE BOLTZMANN MASTER EQUATION THEORY, NUCLEAR THERMALIZATION AND PRE-EQUILIBRIUM REACTIONS

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The Boltzmann Master Equation theory and some results which have been recently obtained with it in the analysis of heavy ion interactions are discussed.

## 1. Introductory remarks

In mid sixties when the growing experimental evidence of nuclear mechanisms intermediate in character between direct reactions and compound nucleus processes forced nuclear theorists to propose new models to deal with such reaction mechanisms, two phenomenological models were almost simultaneously proposed, the exciton model by Griffin [1] and the Boltzmann Master Equation (BME) theory by Harp, Miller and Berne [2].

Both models assumed that particles could be emitted by the composite nucleus, created in the interaction of a projectile and a target, along its way to equilibration, i.e., in the course of the cascade of nucleon interactions by which the inequally distributed initial excitation energy transforms into statistically distributed thermal energy. The exciton model assumes that the equilibration proceeds through a sequence of states of increasing complexity and postulates that at any stage of this chain all the state of a given particle-hole configuration are equally probable as well as are their decay modes. The BME theory evaluates the variation with time of the occupation probability of the single nucleon states without introducing the assumption of the equiprobability of the states of a given configuration. As Jack Miller pointed out "the BME theory takes a long time to proof what the exciton model takes for granted". Since in the case of nucleon induced reactions, which were mainly studied in that times, the assumption of the equiprobability of the states of a given configuration works well, the BME theory had less initial success than the exciton model even if it could reproduce the data with almost the same accuracy.

However BME theory may be used when the equiprobability assumption fails as it happens, just to give an example, in the case of asymmetric heavy ion reactions. In fact, in nucleon induced reactions, to a good approximation one may assume that the incident nucleon interacts with the nucleons of a degenerate Fermi gas and, just from beginning, the dynamics of nucleon-nucleon (N-N) collisions does not favour any particular final state, validating the equiprobability assumption [3]. However, in the case of asymmetric heavy ion interactions, the initial excitation energy is not evenly distributed among the projectile and target nucleons. In addition, an heavy ion interaction may produce many vacancies in the inner states of the composite nucleus (deep holes) and the BME theory may easily take into account their role in the subsequent cascade of nucleon interactions [4]. Other very attractive features of this approach are the very natural way in which the emission of many particles from the excited nucleus may be evaluated and the possibility of reproducing with remarkable success the emission of clusters (ranging from light particles to quite large intermediate mass fragments (IMF)) [5–7].

Before briefly describing BME theory one must stress that it may be used also to describe the final stage of a reaction induced by a very energetic projectile which at first gives a large fraction of its energy to a few high energy ejectiles (in a way which may be more conveniently described by a different approach) creating a non equilibrated nucleus which may still emit several particles along its way to equilibration.

## 2. Assumption underlying BME theory

The BME theory, as formulated by our research group, is described in several papers [4–11], so in this Section we limit ourselves to a brief discussion of the underlying assumptions. It is a way to evaluate the variation with time of the distribution of the momenta of the nucleons of an excited nucleus as a result of their mutual interactions and their emission into the continuum as separate entities or as part of a cluster. To do that one divides the momentum space into bins $i$ of volume $\Delta V = 2\pi\, m\, \Delta\epsilon \cdot \Delta p_z$ (where m and $\epsilon$ are the nucleon mass and energy and $p_z$ the component of nucleon momentum along the beam axis) and one evaluates how the occupation probability $n(\epsilon, \theta, t)$ of the states of each bin varies with time. This may be done by integrating a set of coupled equations which evaluate the variation of $n(\epsilon, \theta, t)$ in the time interval between $t$ and $t + dt$ as a function of the occupation probabilities at time $t$ and the decay rates for nucleon-nucleon interactions and emissions into the continuum. Considering the nucleus a two nucleon gas the set of BME reads for the protons:

$$\frac{d(n_i g_i)^\pi}{dt} = \sum_{jlm}[\omega^{\pi\pi}_{lm\to ij} g^\pi_l n^\pi_l g^\pi_m n^\pi_m (1-n^\pi_i)(1-n^\pi_j) - \omega^{\pi\pi}_{ij\to lm} g^\pi_i n^\pi_i g^\pi_j n^\pi_j (1-n^\pi_l)(1-n^\pi_m)] + \sum_{jlm}[\omega^{\pi\nu}_{lm\to ij} g^\pi_l n^\pi_l g^\nu_m n^\nu_m (1-n^\pi_i)(1-n^\nu_j) - \omega^{\pi\nu}_{ij\to lm} g^\pi_i n^\pi_i g^\nu_j n^\nu_j (1-n^\pi_l)(1-n^\nu_m)] - n^\pi_i g^\pi_i \omega^\pi_{i\to i'} g^\pi_{i'} \delta(\epsilon^\pi_i - \epsilon^\pi_F - B^\pi_i - \epsilon^\pi_{i'}) - \frac{dD^\pi_i}{dt}\ , \quad (1)$$

where $\pi$ and $\nu$ indicate the protons and the neutrons and $n_i$ stands for $n_i(\epsilon,\theta,t)$. The quantities $g_i$ are the total number of states in bin $i$; $\omega_{ij\to lm}$, $\omega_{i\to i'}$ and $dD_i/dt$, whose expression may be found in [10], are, respectively, the decay rate of a two-nucleon interaction, of emission of single protons of the $i^{th}$ bin into the continuum, and a depletion term which accounts for the emission of protons of the $i^{th}$ bin which are part of a cluster. These aggregates may be created, during the cascade of $N$-$N$ interactions, by coalescence of nucleons whose momenta fall into a sphere of radius $p_{c,F}$ in momentum space. The $(1-n_i)$ are Pauli blocking factors which forbid the scattering of a nucleon into a completely occupied bin.

With an obvious notation, the multiplicity spectrum of emitted nucleons is given by

$$\frac{d^2M(\epsilon',\theta)}{d\epsilon' d\Omega} = \frac{1}{2\pi \sin\theta}\int n(\epsilon,\theta,t)\frac{\sigma_{inv} v}{V}\rho(\epsilon',\theta)dt\ , \quad (2)$$

where $\epsilon$ and $\epsilon'$ are, respectively the nucleon energy inside and outside the nucleus, $\sigma_{inv}$ is the cross section for the inverse process, $v$ the nucleon-residue relative velocity and $\rho(\epsilon',\theta)$ the nucleon continuum state density. $V$ is the laboratory volume which cancels out with an analogous term appearing in the expression of $\rho(\epsilon',\theta)$.

For calculating the multiplicity spectrum of a cluster $c$, one has to evaluate the probability that the momenta of ($Z_c$ + $N_c$) nucleons be correlated in such a way to move together as a cluster with centre of mass energy $E_c$ inside the composite nucleus. This quantity, which we call the occupation probability of cluster states, is a function of the occupation probabilities of the states of the nucleons constituting the cluster. and it is given by the joint probability that the momenta of nucleons of the required type fall within the coalescence sphere. For a cluster made of $Z_c$ protons and $N_c$ neutrons, it is given by [5, 6]:

$$N_c(E_c,\theta_c,t) = \prod_i (n^\pi_i(\epsilon,\theta,t))^{P_i(E_c,\theta_c)Z_c} \cdot \prod_i (n^\nu_i(\epsilon,\theta,t))^{P_i(E_c,\theta_c)N_c}\ , \quad (3)$$

where $i$ runs over all the bins in which the momenta of the protons and the neutrons of the cluster may be found and $P_i(E_c,\theta_c)$ is the fraction of bin $i$ within the cluster Fermi sphere of radius $p_{c,F}$.

The cluster multiplicity spectrum is given by an analogous expression to (2):

$$\frac{d^2M_c(E'_c,\theta_c)}{dE'_c d\Omega} = \frac{R_c}{2\pi \sin\theta}\int N_c(E_c,\theta_c,t)\frac{\sigma_{inv,c} v_c}{V} \cdot \rho_c(E'_c,\theta_c)\,dt\ , \quad (4)$$

except for a numerical factor $R_c$ $(\leq 1)$ which represents the probability that the cluster once formed be emitted before dissolving again into its constituents.

To integrate the set of BME one must know the occupation probabilities at an initial time $t = 0$, which depends on the dynamics of the two ion interaction. They are integrated up to a time $t_{eq}$ after which the emission of further pre-equilibrium particles is negligible and the yield of the more energetic ejectiles does not change anymore. This time is usually smaller than about 5 $10^{-22}$ s.

In this form the BME theory is of limited applicability for two reasons:

(a) it provides an inclusive information by averaging the contributions of many different reaction paths. In other words it may only evaluate the mean multiplicities of emitted particles which is not sufficient for exclusive processes or for evaluating other measurable quantities like, for instance, the cross section for residue's formation. Even the calculated evaporative contribution to inclusive particle spectra is that of the particles evaporated by the nucleus with average energy, mass and charge which is left when one stops the integration of the BME, while the actual one is that of the particles evaporated by many different nuclei which may be produced with different excitation energies.

(b) the residual nuclei are assumed to recoil always in the $z$-axis direction due to the azimuthal symmetry and the averaging over all the possible events. It is thus impossible to evaluate the residue's double differential spectra which can be measured and provide useful information on the reaction mechanisms [14, 16].

In order to eliminate such limitations, the multiplicity spectra evaluated with the BME may be used as input to a Monte Carlo calculation which gives the probability of any possible sequence of events and predicts the residue's spectra taking into account the composite nucleus recoils.

To do that one assumes that the differential multiplicities $d^3M_i(\epsilon,\theta,t)$ of particles $i$ emitted in the time interval [t,t+dt], with energy $[\epsilon,\epsilon + d\epsilon]$, and polar angle $[\theta,\theta + d\theta]$ are equal to their emission probabilities. This assumption is acceptable if the total differential multiplicity

$$\iint d^3M_T(\epsilon,\theta,t)\,d\epsilon\,d\theta = \iint \sum_i d^3M_i(\epsilon,\theta,t)\,d\epsilon\,d\theta \quad (5)$$

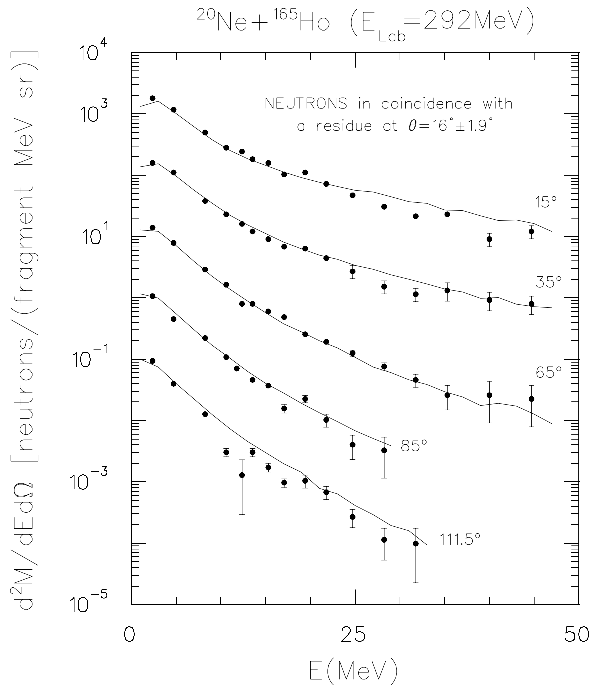


Figure 1: Spectra of neutrons emitted in the interaction of $^{20}$Ne with $^{165}$Ho at an incident energy of 14.6 MeV/n in coincidence with heavy residues emitted at a forward angle. The black circles give the experimental values [17], the lines the result of the calculation. Starting from bottom the spectra are progressively scaled up by a factor ten.

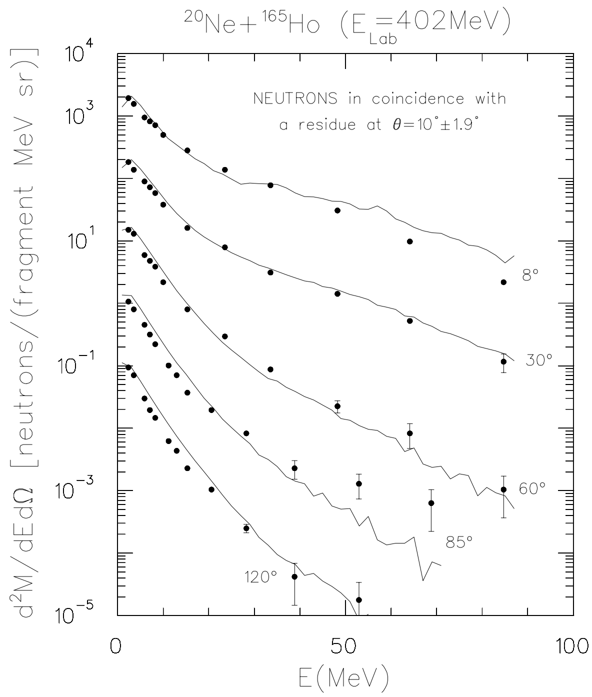


Figure 2: Spectra of neutrons emitted in the interaction of $^{20}$Ne with $^{165}$Ho at an incident energy of 20.1 MeV/n in coincidence with heavy residues emitted at a forward angle. The black circles give the experimental values [17], the lines the result of the calculation. Starting from bottom the spectra are progressively scaled up by a factor ten.

of the emitted particles is small ($\leq$0.2) [9, 11]. This happens for time intervals of suitably small lenght. The joint probabilty of a sequence of events may be evaluated using these *elementary* probabilities. By simulating many sequences of events one can evaluate the fluctuations around the average which permit to reproduce the results of exclusive experiments. In the most recent version our Monte Carlo code considers also the emission of intermediate mass fragments in addition to the light particles with Z$\leq$2 which were considered in previous calculations [11]. The importance of this development is discussed in next Section.

## 3. Comparison of the theoretical predictions with the experimental data

The BME predictions have been compared with a large set of data comprising double differential spectra of nucleons and light particles [5, 6, 8, 11] and intermediate mass fragments [7, 15], cross sections for residue formation [12–14], residue's forward recoil distributions [14], Doppler shift and broadening of residue's $\gamma$ lines [16]. In all the cases they reproduced satisfactorily the data using parameters fixed *a priori* displaying a weak mass dependence. Here we show a few representative examples of our most recent results and discuss some perspectives which such studies open.

Figs. 1 and 2 show the double differential spectra of the neutrons emitted in central collisions of 14.6 and 20.1 MeV/nucleon $^{20}$Ne ions with $^{165}$Ho [17]. The experimental spectra are reproduced with a remarkable accuracy. These results are representative of the accuracy with which the theory is able to reproduce the double differential spectra of neutrons and protons emitted in a large number of heavy ion interactions at energies of some tens of MeV/nucleon. These calculations in addition to two body interactions (nucleon-nucleon collisions) take into account also mean field effects which may significantly affect the initial momentum distribution of the nucleons of the interacting ions [8, 18]. These effects, at incident energies not greatly exceeding the entrance channel Coulomb barrier, reduce the pre-equilibrium emissions because, as described in [18], the cascade of nucleon-nucleon interactions develops while a sizeable fraction of the two ion energy is still frozen as collective excitation energy. At the contrary, at considerably higher incident energies, when the two ions merge in a

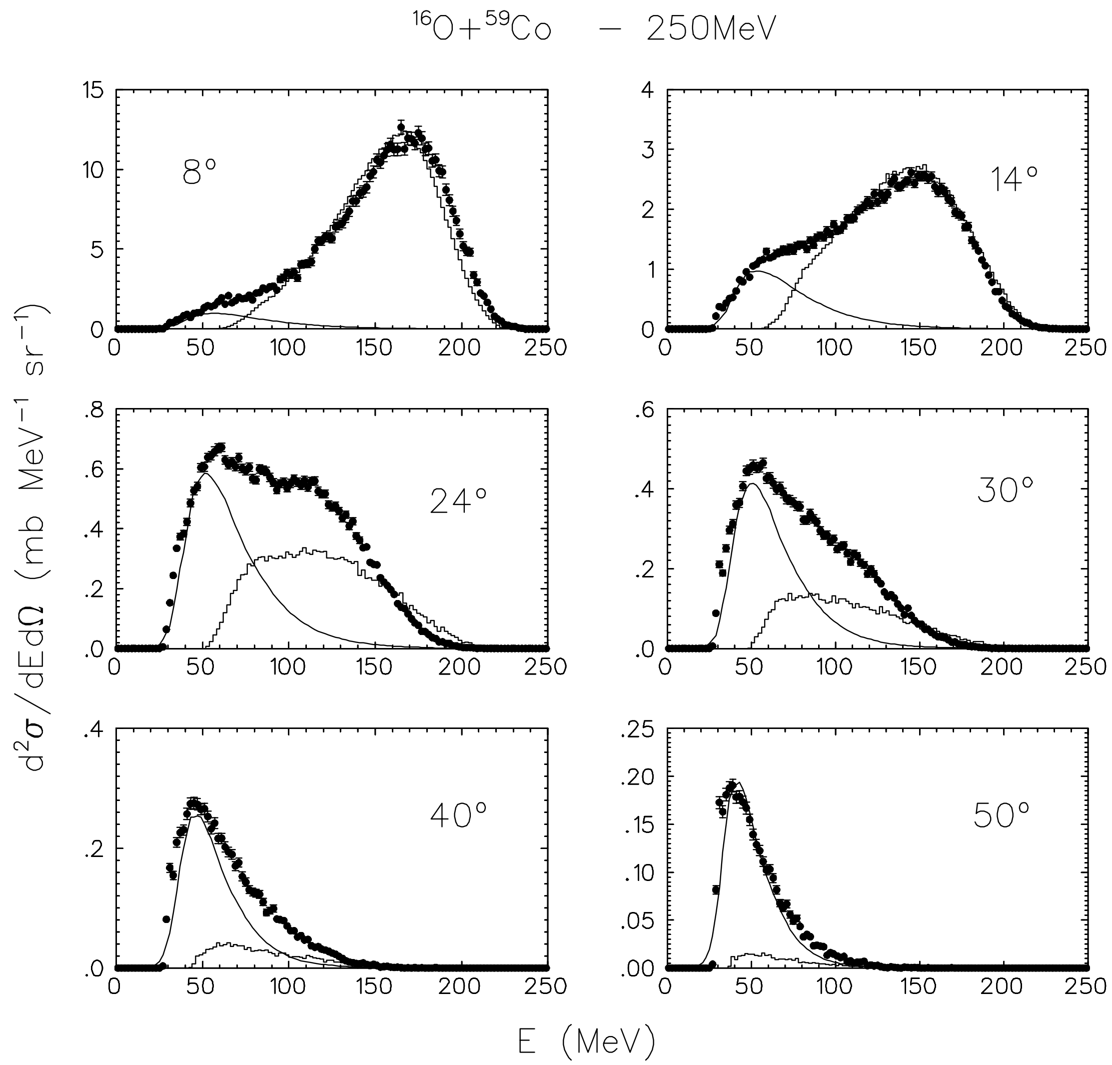


Figure 3: Spectra of carbon fragments emitted in the interaction of $^{16}$O with $^{59}$Co at incident energies of 250 MeV [7]. The experimental spectra are given by the black circles, the coalescence contribution by the full lines. At the most forward angles, in addition to the coalescence one observes the contribution of carbon fragments produced by a different mechanism, the binary fragmentation of the projectile. This contribution is given by the histograms.

common intermediate nucleus, their nucleons gain an extra energy falling in a deeper potential well and this enhances the emission of fast particles before the reaching of thermal equilibrium. Consideration of these effects is essential to correctly reproduce not only the yield and the spectra of the emitted particles, but also the cross section for formation of the evaporation residues [13, 18].

One of the most exciting new results obtained in calculations of this type is the reproduction of the double differential spectra of low energy intermediate mass fragments which are emitted even at quite low incident energies in heavy ion interactions. We have in particular studied this emission in the interaction of $^{12}$C and $^{16}$O ions with nuclei ranging from $^{59}$Co to $^{197}$Au at incident energies varying from about 6 to about 35 MeV/nucleon. In reactions induced by light heavy-ions the intermediate mass fragments are produced by two different mechanisms: the projectile's fragmentation which is mainly responsible of the observed yield at the most forward angles and the coalescence of nucleons [7, 15, 19, 20]. This last contribution dominates at large emission angles and is satisfactorily predicted by the BME theory. As an example of our results, Figs. 3 and 4 show the double differential spectra of C fragments produced in the interaction of 250 and 400 MeV $^{16}$O ions with $^{59}$Co [7]. Several isotopes, for each Z, contribute to the calculated yield of coalescence IMF with a relative amount which depends on their spin and isospin, atomic weight and mass excess and the isospin of the interacting ions, as shown, as an example, in Figs. 5

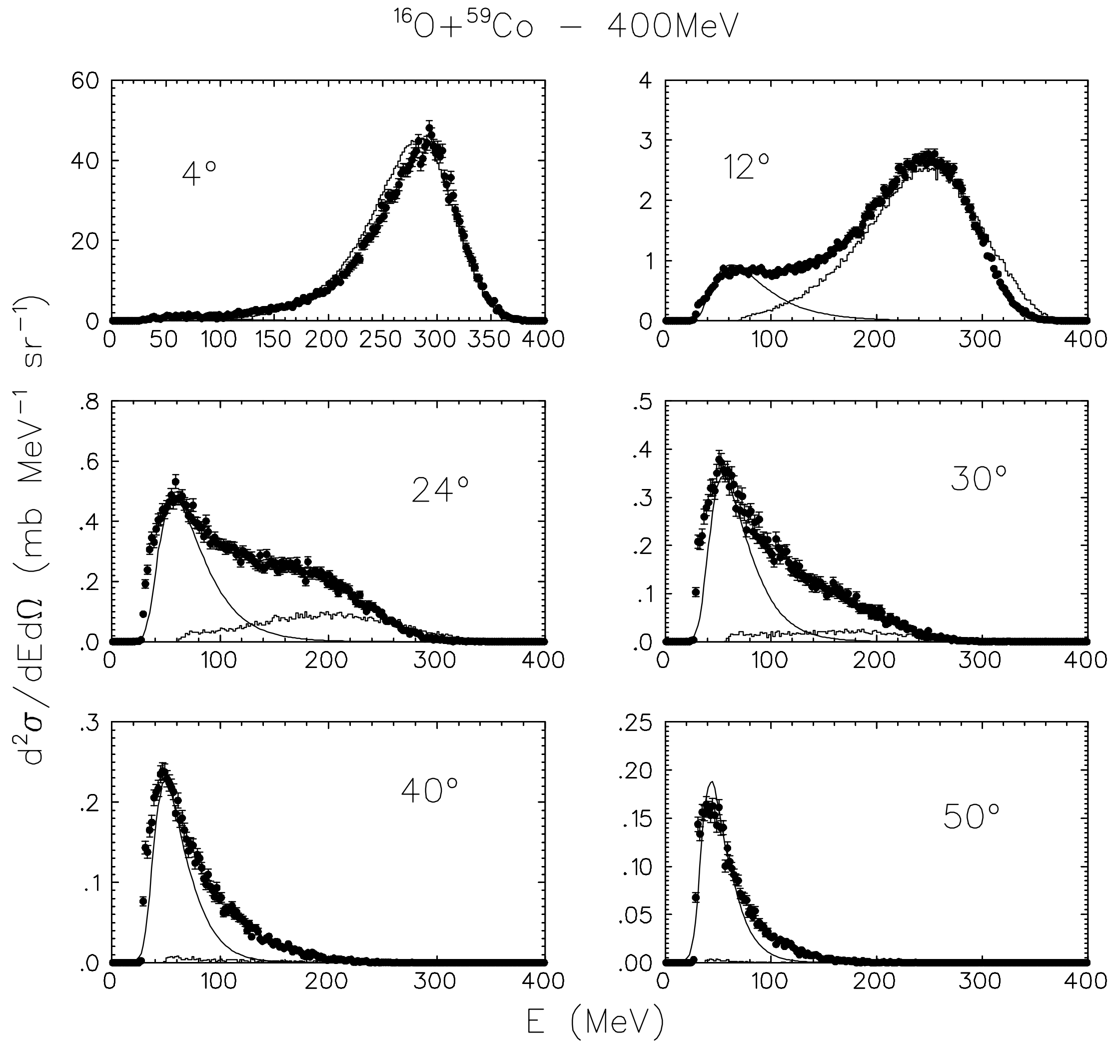


Figure 4: Spectra of carbon fragments emitted in the interaction of $^{16}O$ with $^{59}Co$ at incident energies of 400 MeV [7]. The experimental spectra are given by the black circles, the coalescence contribution by the full lines. At the most forward angles, in addition to the coalescence one observes the contribution of carbon fragments produced by a different mechanism, the binary fragmentation of the projectile. This contribution is given by the histograms.

for the lithium isotopes produced in the interaction of $^{58}Ni$ with $^{40}Ca$ and $^{62}Ni$ with $^{48}Ca$ at an incident energy of 25 MeV/n. These theoretical predictions cannot be compared with the experimental findings because in most of the experiments described in literature, the IMF are well discriminated in Z but not in A. The measurement of isotope distributions could not only provide a more accurate test of the theory we propose, but might also help in fixing much more accurately the values of some of the parameters which enter in our calculations, for which we use tentative values.

Preliminary calculations suggest that the emission of IMFs during the equilibration cascade must be taken into account in the study of entrance channel effects, to assess correctly the excitation energy and the mass and charge distribution of the equilibrated nuclei in phase transition studies and for evaluating the mass and charge distribution of the lowest mass residues which are produced in heavy ion interactions.

In Fig. 6 are compared the experimental and calculated excitation functions for production of Sb, Sn and In isotopes in the interaction of $^{12}C$ ions with $^{103}Rh$ [12]. These residues are produced in a complete fusion process and their excitation functions display after the maximun a much more gentle decrease than that predicted for the evaporation decay of an equilibrated compound nucleus. This is analogous to what is observed in light particle induced reactions and is due to the pre-equilibrium emission of particles with energy considerably exceeding that of evaporated particles. These emissions increase very significantly the probability of producing a given residue at incident energies above the threshold

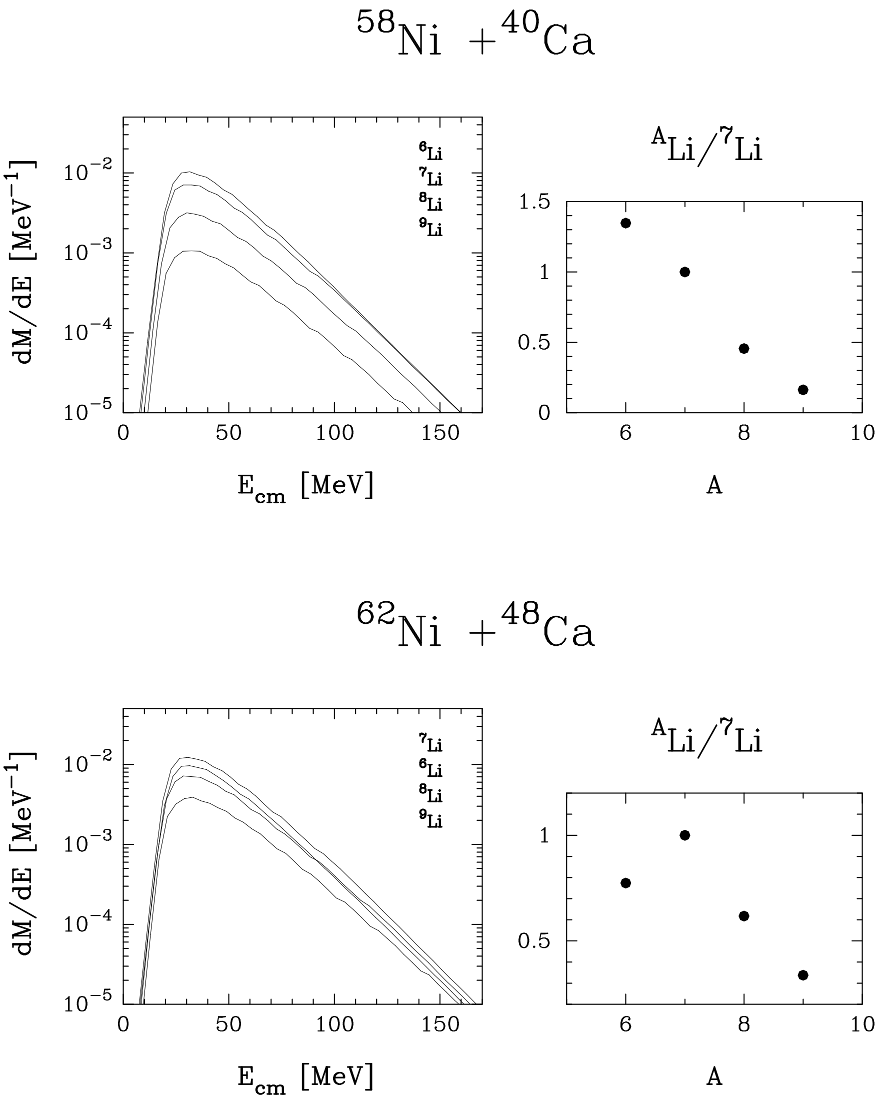


Figure 5: The figure shows in its upper- and lower-left part the predicted angle integrated multiplicity spectra of Li isotopes produced by nucleon coalescence in the interaction of $^{58}$Ni with $^{40}$Ca and $^{62}$Ni with $^{48}$Ca at an incident energy of 25 MeV/n and, in its upper- and lower-right part, the angle and energy integrated lithium isotope's yields normalized to that of $^{7}$Li.These results are an example of predicted entrance channel dependence of isotope yields.

for production of lower mass nuclei. The Monte Carlo calculations reproduce quite accurately these excitation functions accounting satisfactorily for the pre-equilibrium emissions which are found to be non-negligible even at incident energies only slightly greater than the interacting ion Coulomb barrier.

## 4. Concluding remarks

After about forty years from its first appearance the nuclear BME theory enters in its maturity and appears to be a very useful mean to describe heavy ion interactions up to incident energies of about 50

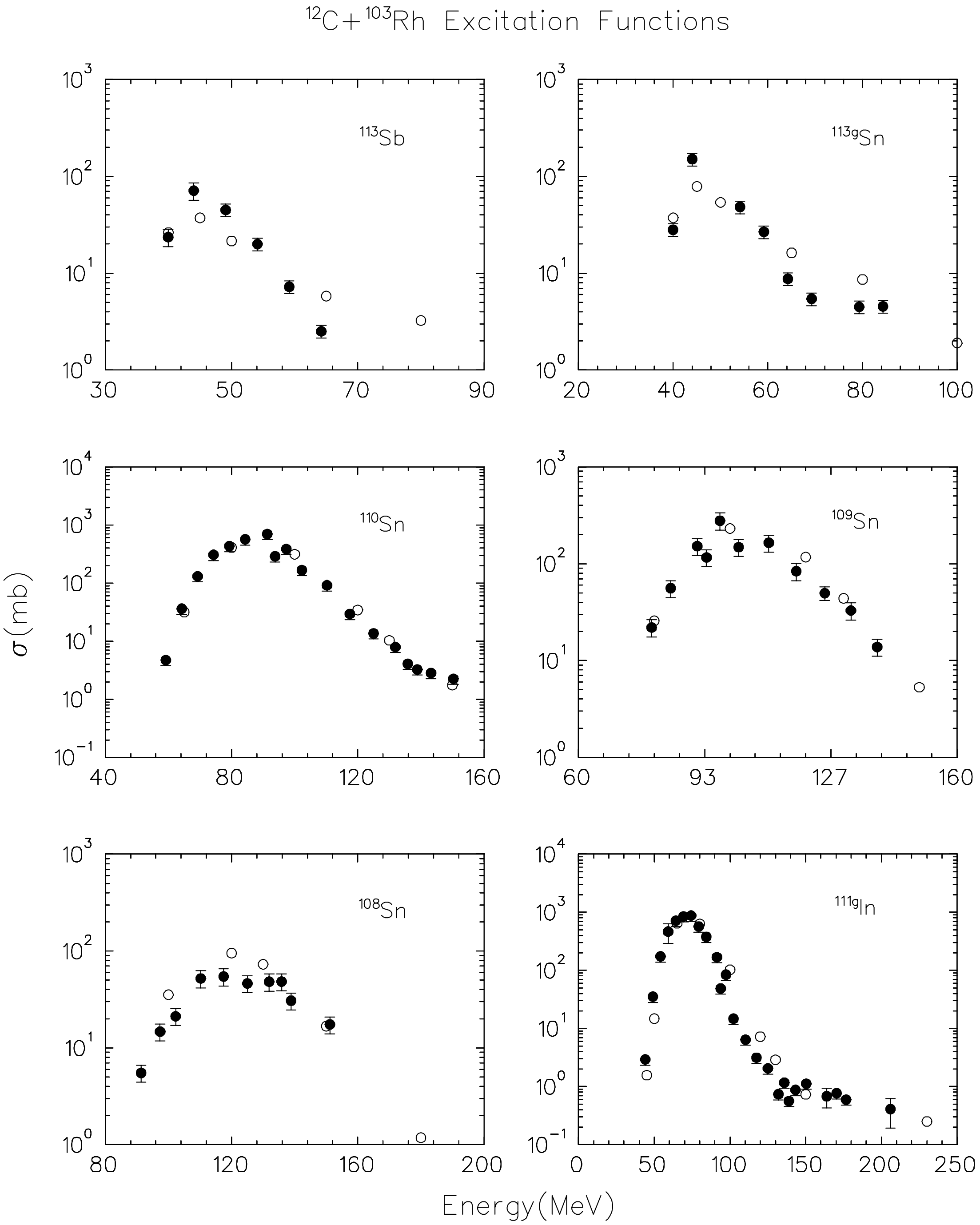


Figure 6: Excitation functions for production of Sn, Sn and In isotopes in the complete fusion of $^{12}$C with $^{103}$Rh. The experimental values [12] are given by the black points, the calculated values by the open circles.

MeV/nucleon. As mentioned at the beginning of the paper, it might be used also in the simulations of much more energetic interactions in which, after an initial stage, which may be better described by other models, non-equilibrated intermediate nuclei are created with considerable excitation energies. The matching of the BME theory with high energies codes, such as for instance FLUKA which has been discussed by Dr. Ferrari in a previous talk at this Conference [21], is at present under study. Hopefully at the next Varenna Conference we will be able to discuss these new developments.

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