

PRODUCTION OF CLUSTERS (UP TO $A=10$) DURING THE INTRANUCLEAR CASCADE PHASE OF SPALLATIONS REACTIONS

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Spallation reactions are generally considered to proceed in two stages: a cascade stage followed by an evaporation stage. Light charged particle (lcp) spectra indicate that these particles are produced in both stages. The mechanism of production in the cascade stage is still not fully understood. We have recently shown, using the improved versions of the Liège Intra-Nuclear Cascade model and of the ABLA evaporation model, that light clusters (up to alpha particles) are likely produced by some kind of dynamical coalescence process, by which a fast particle of the cascade drags out a few other nucleons. In the very recent years, precise measurements of the production of heavier clusters have been performed. We improved and generalized our production models for heavier clusters, up to $A=10$. Some typical results are presented here and show good agreement with experiment, strongly suggesting that the dynamical coalescence mechanism also applies to these heavier clusters. The importance of these results for spallation neutron sources and accelerator-driven systems is underlined. As an example, we discuss the implications for the production of astatine isotopes in lead-bismuth spallation sources.

I. INTRODUCTION

Spallation reactions are high-energy reactions induced by nucleons and/or light nuclei. A renewed interest has emerged in the recent years for proton-induced reactions in the GeV incident energy range, mainly triggered by the contemplation of realizing transmutation nuclear waste in Accelerator-Driven Systems (ADS). In such devices, a high-energy proton beam bombards a spallation target inside a subcritical nuclear reactor core. Neutrons are copiously produced by spallation reactions and are subsequently multiplied in the reactor core and finally used for transmuting nuclear waste, mainly by fission (see Ref. 1 for a review). The first project of this kind, the MYRRHA project, has

recently been launched.² Accordingly, we will limit ourselves in this paper to proton-induced reactions below 2 GeV. Spallation reactions are also of application in spallation neutron sources³, hadrontherapy⁴ and protection against radiation in space missions.⁵

In spallation reactions, the abundant emission of neutrons is accompanied by the much smaller (by one order of magnitude, typically) production of lcp's (defined here for convenience as $A \leq 4$ particles) and, with still a lesser importance, by the emission of so-called intermediate mass fragments (IMF, loosely defined as clusters with $4 < A < \sim 30$). However, the emission of lcp's and IMF's, which is the central topic of this paper, presents both scientific and practical importance. Indeed, the mechanism of production is under debate, and their production in spallation targets poses problems for protection against radiation.

There is a general agreement among physicists on the idea that spallation reactions proceed in two stages: a fast stage, characterized by the emission of fast particles, followed by a slower stage, where an excited and more or less equilibrated remnant de-excites by emitting slow particles through evaporation-like processes. Likewise, the basic theoretical tool consists in the coupling of an intranuclear cascade (INC) model, for the first stage, to an evaporation/fission model, for the second stage. In this basic approach, nucleons are emitted in both stages, and light clusters, like alpha particles, are emitted only in the evaporation stage. On the other hand, measurements clearly indicate that lcp's can be emitted with a kinetic energy of 100 MeV or more, much above typical evaporation energies ($\leq \sim 20$ MeV), very likely during the cascade stage. To cure the deficiency of this basic approach, emission of lcp's, up to α 's, has been introduced in the cascade stage, during the recent years. In Ref. 6, some of the authors of this paper have implemented a kind of dynamical coalescence model in the Liège Intra-Nuclear Cascade (INCL) model: an unbound nucleon crossing the nuclear surface can carry

away other nucleons to form a cluster, provided these other nucleons are sufficiently close in phase space. This model is “dynamical” in the sense that the emission of a cluster is determined by the evolution of the cascade itself. It is quite successful in reproducing the gross features of the double differential cross sections for emissions of lcp’s.⁶ In the latter reference, old versions of the INCL and ABLA codes were used. Recently, calculations have been remade with the latest versions of the models (see later for detail) and results were improved.⁷ Especially, the excitation functions of the total production cross sections of tritium and helium isotopes are well reproduced on a broad domain of incident energy.⁸ In the meantime, new measurements, especially by the PISA collaboration⁹, completing older measurements,¹⁰⁻¹³ have shown that heavier clusters can be emitted with a large kinetic energy. For convenience, in this paper, we will call “light clusters”, those which are not heavier than the alpha particle, and “heavy clusters”, those which are heavier, up to $A=10$.

The main purpose of this paper is to investigate whether the production of the heavy clusters can be viewed as a smooth continuation of the production of light clusters and be describable by the same theoretical model. This question presents both theoretical and practical interests. If it is clear that light clusters can be produced by both cascade and evaporation stages, it may be not so for heavier clusters like ${}^9\text{Be}$ or even less so for clusters like ${}^{22}\text{Ne}$, an issue which has been repeatedly raised by R. Michel.¹⁴ On the practical side, many of these (both light and heavy) isotopes are radioactive (like ${}^3\text{H}$, ${}^7\text{Be}$, ${}^{10}\text{Be}$, ${}^{22}\text{Na}$) or volatile (like H , He), and may pose problems of protection against radiation and/or damages of materials around spallation sources and ADS’s.

II. THE NEW VERSION OF THE INCL+ABLA MODEL

II.A. General description

The most recent versions of the Liège INC model,¹⁵ denoted as INCL4.5, and of the ABLA de-excitation model,^{16,17} known as ABLA07, have not been published yet, but good accounts are provided by Refs. 18-19, respectively. It is sufficient for our purpose here to remind the main features of the models. The INCL4.5 model is a time-like INC model, which follows the fate of all particles in space-time. Particles follow straight-line trajectories, until either two of them reach a sufficiently small minimum distance of approach, in which case a collision is realized, or until a particle hits the nuclear surface, in which case it is either reflected or transmitted according to transmission probability on the nuclear potential surface, or until a particle (a Delta resonance) decays. Collisions can be elastic or inelastic. Nucleon-

nucleon inelastic collisions are modeled by explicitly introducing pion and Delta degrees of freedom. Target nucleons are moving in a nuclear potential well and collisions are subject to Pauli blocking. Special care is exercised for soft collisions, which allows the model to work well even down to 50 MeV (see Ref. 14 for detail). As we said, in INCL4.5 a new module is introduced for the emission of clusters, which is described in some detail below. When the cascade is stopped (at a time which is determined self-consistently, a unique feature of INCL¹⁵), the main parameters of the remnant nucleus are transferred to the ABLA07 model, which treats the subsequent de-excitation. In this model, neutron, proton and any stable nucleus up to half of the mass of the remnant can be emitted, on the basis of the Weisskopf-Ewing formalism, even if simple estimates of the emission widths are used for the largest clusters ($Z>3$). On the other hand, parameterizations of inverse cross sections and of the level density, especially concerning the pairing effects, have been improved. Similarly, a simplified procedure has been used in order to take account of the emission of angular momentum in course of the successive emissions. The ABLA code is known for its sophisticated fission module: fission width is determined by the Bohr-Wheeler formula using realistic level densities at the barrier, mass partition is based on microscopically calculated energy surfaces, including realistic shell effects, and time delay is introduced to take account of viscosity in the collective motion toward scission. In ABLA07, the latter is replaced by a time-dependent fission width, the dependence being parametrized on solutions of detailed Fokker-Planck equations. In this new version, if the temperature of the remnant exceeds a certain value, which depends upon the mass, another exit channel, namely multifragmentation, is accommodated. Finally, thermal expansion of the nucleus is also introduced. These last two features are of minor importance in the context of this paper. A recent intercomparison of various numerical codes for spallation reactions, organized by the IAEA,²⁰ has “validated” the INCL4.5+ABLA07 code: the latter has been recognized as one of the best codes, for all kind of observables: double differential cross sections, multiplicities, mass and charge spectra for residues, excitation functions, etc.

II.B. The cluster formation model in INCL4.5

The description of the cluster production in INCL4.5 relies on the idea that an outgoing nucleon hitting the surface can drag along particles which are sufficiently close to it, elaborating on an idea, originally proposed by Butler and Pearson.²¹ We give some details about the procedure.

When an unbound nucleon, denoted as the leading nucleon, is leaving the target nucleus, all potential

clusters of mass $A=2$ to A_{max} are constructed, by locating the leading nucleon on its trajectory at a radial distance $r = R_0 + h$ (R_0 being the half-density radius) and selecting $A-1$ nucleons which, with the leading nucleon, are sufficiently “packed” in phase space. Actually, they have to satisfy the following criteria:

$$r_{i,[i-1]} p_{i,[i-1]} \leq \Pi, \quad i=1,2,\dots,A \quad (1)$$

where $r_{i,[i-1]}$ and $p_{i,[i-1]}$ are the Jacobian coordinates of the i -th nucleon, i.e. the relative spatial and momentum coordinates of this nucleon with respect to the subgroup of the first $[i-1]$ nucleons, the leading nucleon corresponding always to $i=1$. The quantity Π is a function of A .

The “most bound” cluster is selected. Let \sqrt{s} be the total c.m. energy of a constructed cluster and $B(A, Z)$, its nominal binding energy, the cluster corresponding to the lowest value of $(\sqrt{s} - A m_N - B(A, Z))/A$ is selected.

This cluster is emitted provided (i) its energy (in the target system) is above the threshold for emission, (ii) the test for transmission through the Coulomb barrier is positive, (iii) the direction of emission is not too tangential. Let θ be defined as the angle between the direction of emission (determined by the total momentum of the cluster) and the radial outward direction at the location of the leading nucleon. It is required that $\theta < 45^\circ$. The idea beyond this condition is that the longer a cluster stays in the surface region, the more it is expected to get dissolved. If one of these conditions is not met, the leading nucleon is emitted, provided it can tunnel through its corresponding Coulomb barrier, otherwise it is reflected.

All known clusters up to $A_{max}=10$ are considered here; larger clusters can also be handled, in principle, but their treatment becomes in practice heavily time-consuming; unstable clusters of very short lifetime, such as ${}^8\text{Be}$, are forced to decay at the end of the cascade stage.

The parameter h is equal to 1.0 fm. We could not obtain satisfactory results, especially at low energy, with a single “proximity” parameter, contrarily to what is obtained for light clusters, at high incident energy at least (see Ref. 6). We let Π depend upon the mass A and used (in $\text{MeV} \times \text{fm}/c$ units): $\Pi(d) = 424$, $\Pi(t) = 300$, $\Pi({}^3\text{He}) = 300$, $\Pi({}^4\text{He}) = 300$ and for $A > 4$, $\Pi(A) = 210A^{1/3}$. These values, which are quite reasonable from what is known about the phase space extension of light nuclei, have been obtained by fitting experimental data in a few illustrative cases.

Compared to our previous model for cluster production, which was restricted to light clusters,⁶ the main difference deals with the selection of the cluster: in the previous model, the largest possible cluster was

selected. We had also only one single proximity parameter for all clusters.

III. PRODUCTION OF LIGHT CHARGED PARTICLES

We want to mention first that, for light clusters (up to α 's), our new model generates, most of the time, better results than before. This applies to experimental data for proton incident energy ranging from 62 to 2500 MeV and targets ranging from Al to Bi . Results can be found in Refs. 8 and 20. We just show here the results for double differential cross sections for production of lcp's in a particular case, to give a taste of the results and for the sake of the discussion contained in Section V. They are presented in Fig. 1 below.

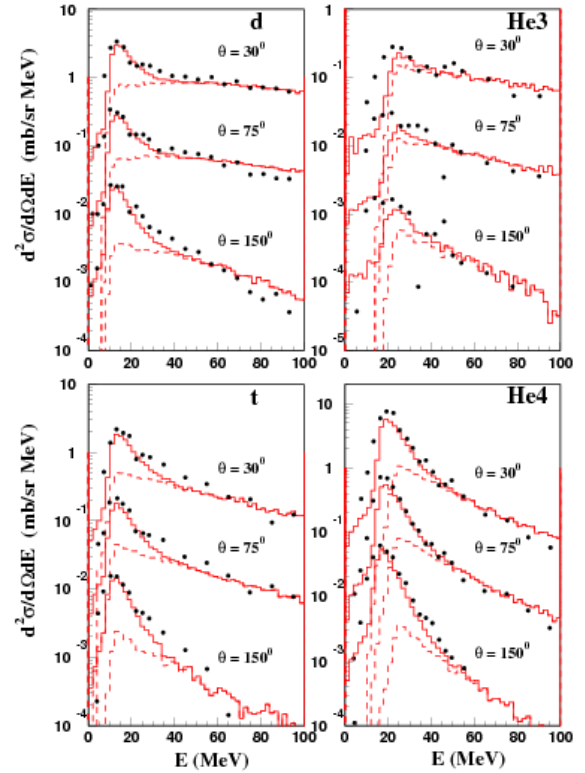


Fig. 1. Double differential cross sections for the production of lcp's (indicated in the respective panels) in $p(1.2\text{GeV})+Ta$ reactions, as functions of the kinetic energy E of the produced cluster. Comparison of our predictions, using the INCL4.5+ABLA07 model (red histograms), with the experimental data (dots) of Ref. 22. Experimental error bars (not shown) are of the order of a few percent. The cascade contribution is singled out by the dashed lines.

One can see that the model is able to reproduce the data quite well, especially the high-energy tails. It is worth to

notice that ${}^3\text{He}$ nuclei are produced predominantly by the cascade, although the evaporation component seems to be slightly underestimated. We want also to mention that Ref. 8 shows that the excitation functions for the production of He isotopes are very well reproduced by our calculations over the 30 MeV-2GeV range of incident energy.

IV. RESULTS FOR THE PRODUCTION OF “HEAVY” CLUSTERS

We remind that this expression refers here to clusters between $A=5$ and $A=10$, before possible decay for short-lived ones (${}^5\text{He}$ and ${}^5\text{Li}$, for instance). Results below correspond to yields after decay.

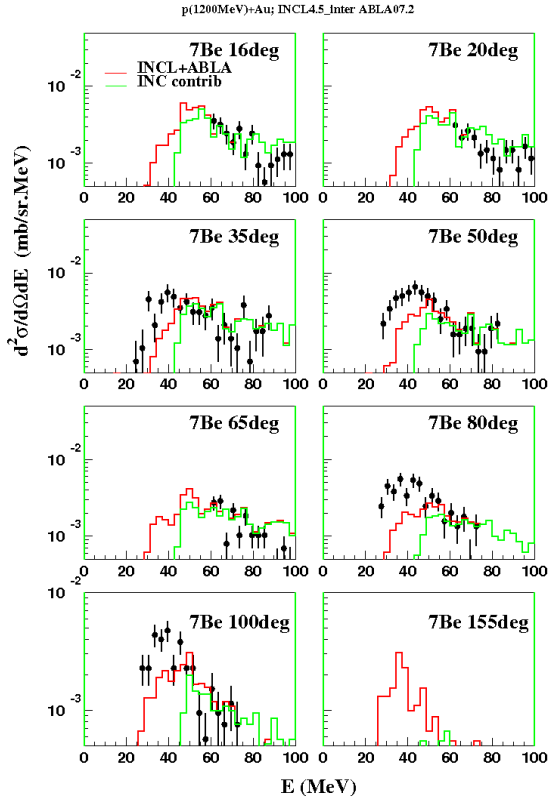


Fig. 2. Double differential production cross section for the production of ${}^7\text{Be}$ in $p(1200\text{MeV})+\text{Au}$ reactions, as function of the kinetic energy E of the produced cluster. The corresponding angles are given in the various subpanels. Comparison of our predictions, using the INCL4.5+ABLA07 model (red histograms), with the experimental data (dots) of Ref. 22. The cascade contribution is singled out by the green histograms.

In Fig. 2, we show typical results concerning the double differential cross section for production of ${}^7\text{Be}$

clusters in $p(1200\text{MeV})+\text{Au}$ reactions.²² Clearly, the high energy tails of the spectra, say above 50 MeV, are entirely due to the cascade and are well reproduced by our calculation. In Fig. 3, we show the results for the production of ${}^6\text{Li}$, for which our calculations are also in good agreement with experiment. It is interesting to note that if, in both cases, the high energy tails are dominated by the cascade, the total production yield (integrated spectra) of ${}^6\text{Li}$ is dominated by evaporation, whereas the situation is roughly reversed for ${}^7\text{Be}$, which is mostly produced in the cascade stage. We have performed calculations for the production of ${}^6\text{He}$, ${}^6\text{Li}$, ${}^7\text{Li}$, ${}^7\text{Be}$, ${}^8\text{Li}$, ${}^9\text{Li}$, ${}^9\text{Be}$, ${}^{10}\text{Be}$ and ${}^{10}\text{B}$ isotopes and compared with the experimental data of Ref. 22, for $p+\text{Au}$ reactions at 1.2, 1.9 and 2.5 GeV, with those of Ref. 23, for $p+\text{Au}$ reactions at 2.5 GeV, and with those of Ref. 24, for

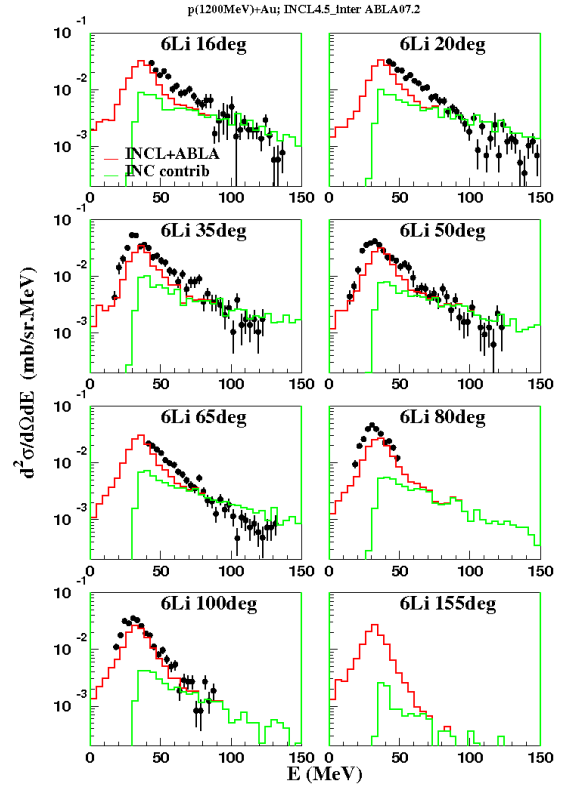


Fig. 3. Same as Fig.2 for the production of ${}^6\text{Li}$ clusters.

reactions of 1.2 GeV protons on 13 targets ranging from Al to Th . Other interesting data are contained in Refs. 10-13. We did not compare with these works because the data are less systematic than or are more or less similar to those of Refs. 22-24. We cannot give the results of all our calculations, for evident lack of space (this will be the

object of a future publication²⁵), but we can summarize the most important results obtained up to now. Our model generates an overall good agreement for all calculated clusters and all calculated reactions. It is worth mentioning here that the integrated production cross sections spread over many orders of magnitude, from about $1b$ for deuteron down to $1mb$ for the heaviest clusters. For most of the heavy clusters, the quality of the agreement is comparable to the one shown in Figs. 2 and 3. The cross section for the production of ${}^6\text{He}$ is surprisingly good, in view of the complex structure of this isotope. They are however some systematic deviations: the production of ${}^9\text{Be}$ is underestimated in the cascade (by a factor 2 or 3) as well as in the evaporation, the production of Li isotopes is somehow underestimated at the largest incident energy. Finally, the cascade contribution, in comparison with the evaporation one, is fading out at $A=9-10$. Heavier clusters are hardly produced by coalescence. We noticed also that the ABLA07 contribution for C, N, O isotopes in $p(2.5\text{GeV}) + \text{Au}$ reactions²³ is particularly good.

We have to warn that, even if our results are very encouraging, they have to be taken with some care for the heaviest clusters. We have presented here results including up to $A=10$ clusters in the calculations. Moving the limit from $A=8$ to $A=10$ have changed the predictions for $A \leq 8$ clusters. Of course, predictions for light clusters are very stable, but the yield for the isotopes close to the limit may change by a few percent to, for some of them, a few tens of percent. Actually, we cannot raise the limit very much further up, because of increasing computation time, due to the huge combinatorics implied in the construction of the clusters. There is also some uncertainty concerning evaporation of large clusters, for which Coulomb barrier properties are not well known. To give an idea, we have also performed calculations (not shown here) with the GEM evaporation code.^{26,27} Comparison to the calculations with ABL07 shows discrepancies of factors 2 or 3 for special isotopes.

We also paid attention to the excitation functions. Two examples are given in Fig. 4. One can see that our calculations predict the cascade contribution to the production of ${}^7\text{Be}$ clusters to be overwhelming at low energy and to reduce to about 50% above 1 GeV. The well-documented case of ${}^{22}\text{Ne}$ is quite interesting, even if it is somehow outside the scope of this paper. The cascade contribution is vanishing; as explained above, our coalescence model does not accommodate emission of clusters with $A>10$, for the moment, but simple extrapolation of our results up to $A=22$ is expected to give negligible cross sections, on the scale of Fig. 4. So, it is reasonable to consider that, in reality, ${}^{22}\text{Ne}$ is overwhelmingly produced in the de-excitation stage. The

comparison of our present results with those obtained with the previous version INCL4.2+ABLA v 3p, indicated by the green curve, is interesting. In this version, there is no evaporation of IMF and ${}^{22}\text{Ne}$ is produced as an evaporation residue (presumably not from the target, but more likely from fission fragments). The fact that our new results so nicely reproduce the experimental data offers a viable answer to Michel's question indicated in the Introduction: it is very likely that ${}^{22}\text{Ne}$ is an evaporation product in this kind of reactions.

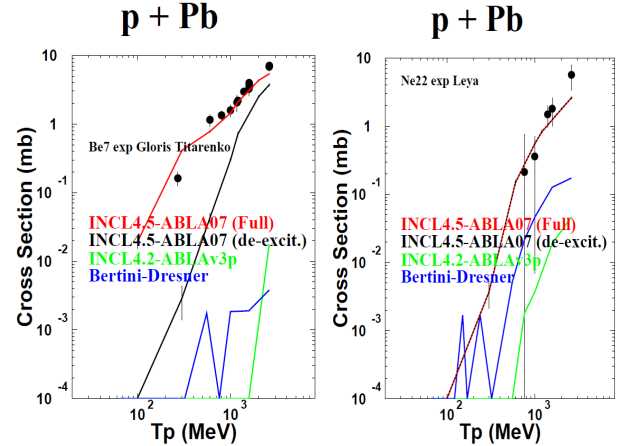


Fig. 4. Total ${}^7\text{Be}$ (left panel) and ${}^{22}\text{Ne}$ (right panel) production cross sections in $p+\text{Pb}$ reactions, as functions of the incident proton kinetic energy T_p . Comparison of our predictions, using the INCL4.5+ABLA07 model (red curves), with the experimental data (black dots) of Refs. 28-30. The de-excitation contribution is given by the black curves (for the ${}^{22}\text{Ne}$ case, the black curve is identical to the red curve). The predictions of the previous version of our model are given by the green curves. The blue curves give the predictions of the Bertini-Dresner model.^{31,32}

V. IMPLICATIONS FOR PRODUCTION OF $Z+2$ ISOTOPES IN THICK TARGETS.

We want to comment on the implications of cluster production for a case where they are not obviously expected, namely the production of $Z+2$ isotopes in a thick target made of ${}^A\text{Z}$ nuclei. In a thin target, this production is possible only through a (p, π^-xn) process. In other words, the incident proton is absorbed and a single negative pion (besides neutrals) is emitted in order to increase the charge by two units. In a thick target, another channel is possible: clusters issued from the primary reactions can make specific secondary reactions to populate the $Z+2$ residues. An experiment performed at ISOLDE has measured the yields of At isotopes issued from a molten Pb-Bi target bombarded by 1.4 GeV protons.³³ We have investigated this problem with our improved version INCL4.5+ABLA07 embedded in the

transport code MCNPX2.7.b (See Ref. 34). The main contributions to the production of *At* isotopes are the $^{209}\text{Bi}(p,\pi xn)$ reactions (hereafter, the “pion channel”) and the secondary reactions $^{209}\text{Bi}(\alpha, xn)$ and $^{209}\text{Bi}(^3\text{He}, xn)$ (the “helium channel”). Lighter lcp’s cannot transfer two charges and the contribution of heavier clusters is roughly one order of magnitude smaller. Our preliminary results are shown in Fig. 5. Since the properties of the release of astatine by a liquid Pb-Bi target not well known, there are some serious uncertainties about the absolute normalization of the data.³⁵ Therefore, we deliberately make the comparison with the data by normalizing the latter on the theoretical value for $A=205$ (where the pion channel contribution is expected to be dominant). Independently of the normalization, the shape of the data curve is definitely not reproduced by our calculation. This remark applies also to calculations performed with other transport codes, as mentioned in Ref. 33.

We analyze a little bit our results. First, one can see that the sum of the pion and helium channels practically exhausts the full contribution. Our calculation of the pion channel seems reliable in view of Fig. 6, which compares our results for *At* production on a ^{209}Bi thin target. Of course, the agreement is not perfect, but we are dealing

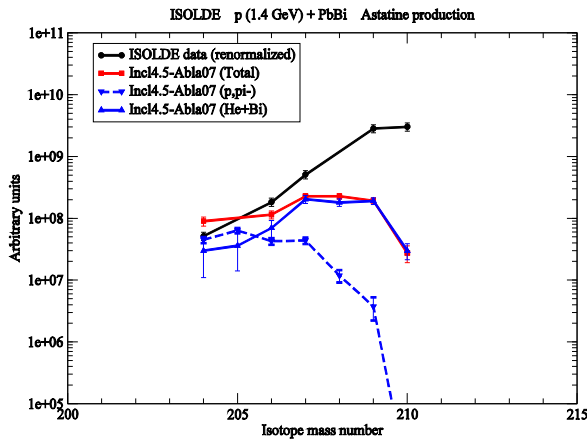


Fig. 5. Yield of Astatine isotopes released from a *Pb-Bi* target irradiated by a beam of 1.4 GeV protons.³² Comparison of production in a MCNPX2.7.b calculation using INCL4.5+ABLA07 (red lines) with the data (full black lines), normalized differently in the two panels (see text for detail). The dashed blue line gives the contribution of the pion channel and the full blue line, the one of the helium channel.

here with factors ~ 3 and not with orders of magnitude as in Fig. 5. We are also rather confident in our predictions for the production of helium clusters, as explained in this work (see Fig. 1) and in Refs. 8 and 20. Therefore, it is rather our calculation of the cross sections for the reactions induced by ^3He and ^4He clusters that should be

blamed. One has to realize that mainly low-energy helium clusters are contributing to the *At* production (on the average, the kinetic energy of these *He* clusters is of the order of 10-20 *MeV*). In this energy range, both in the cascade and in the evaporation, one is sensitive to details of the structure of nuclei, like the *Q*-values for (α, xn) or $(^3\text{He}, xn)$ reactions. These quantities are rather crude in our cascade model and catch only the average variations with charge and mass of the nuclei. We are currently improving this point, by using criteria for emitting particles based on experimental *Q*-values. We envisage to do the same in ABLA07, although the existing *Q*-values are more realistic than in INCL4.5. We are also paying attention to the Coulomb effects in the entrance channel, which largely determines the threshold for (α, xn) or $(^3\text{He}, xn)$ reactions.

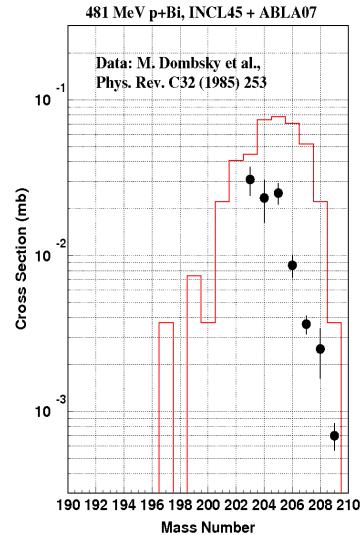


Fig. 6. Production cross section of Astatine isotopes by 481 *MeV* protons on a thin target of ^{209}Bi , as a function of the mass number. Comparison of our predictions with the experimental data of Ref. 36.

VI. CONCLUSIONS.

We have presented the new version of our cascade+evaporation model INCL4.5+ABLA07 and applied it to the production of heavy clusters. The latter can be produced in the cascade stage through dynamical coalescence and in the de-excitation stage by evaporation. Preliminary and promising results have been shown. In general, the shape and the amplitude of the spectra of produced clusters are well described. We noticed that some isotopes are more produced in the cascade and some other ones are essentially emitted by evaporation.

The reasons for this apparently complex behavior will be analyzed in a forthcoming publication. The importance of the cluster production for the generation of Z+2 isotopes in thick spallation targets has been underlined and an account of our work related to the ISOLDE experiment concerning a Pb-Bi target has been given. The puzzling results concerning the production of At isotopes has forced us to reconsider some details of the cascade, to which little attention had been paid up to now.

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