Communications in Physics, Vol. 29, No. 3SI (2019), pp. 313-324
DOI:10.15625/0868-3166/29/3SI/14333

MICROCANONICAL STATISTICAL MODEL FOR FRAGMENTATION OF SMALL NEUTRAL CARBON CLUSTERS

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Received 22 August 2019
Accepted for publication 10 October 2019
Published 18 October 2019

Abstract. *We present the microcanonical statistical model to study fragmentation of small neutral carbon clusters C_n ($n \leq 9$). This model describes, at a given energy, the phase space associated with all the degrees of freedom accessible to the system (partition of the mass, translation and rotation, spin and angular momentum of the fragments). The basic ingredients of the model (cluster geometries, dissociation energies, harmonic frequencies) are obtained, for both the parent cluster and the fragments, by an ab initio calculation. The fragmentation channels probabilities obtained as a function of the excitation energy, were compared with the experimental data at the Orsay Tandem. The deposited energy distributions were adjusted so that the experimental measurements were optimally reproduced. Two algorithms were used: Non-Negative Least Squares and Bayesian backtracing. The comparison of the theoretical and experimental probabilities shows a good global agreement. Both algorithms result in deposited energy distributions showing peaks. These peaks could be the signatures of specific molecular states which may play a role in the cluster fragmentation.*

Keywords: fragmentation; small neutral carbon clusters; partition; partition probabilities.

Classification numbers: 36.40.Qv.

I. INTRODUCTION

The small neutral carbon clusters C_n ($n \leq 9$) are the subject of intense researches in both theory and experiment. They play an important role in the chemistry of the universe. The small neutral carbon clusters are observed in planetary environments, interstellar and circumstellar media [1] as well as in the comets [2]. They also present in flames. Their role is dominating in cold plasmas at low pressure used for decontamination of smoke. Fragmentation is the dominant dissociation process of excited carbon clusters [3, 4]. Therefore, the knowledge of fragmentation of carbon clusters can provide information on the stability of these clusters as well as on the dynamic of the excitation process [5]. In addition, understanding of physico-chemical characteristics of these clusters is an important issue especially for the protection of the environment. Presently, there is a lack of fragmentation data in astrochemical codes for most of the introduced species, including carbon clusters. Indeed, although numerous works have been devoted to carbon clusters [6, 7], they mostly rely on spectroscopic studies and very few on fragmentation, especially for neutral and multi-charged clusters.

Experimentally, the information of fragmentation of neutral carbon clusters is scarce. The Tandem accelerator in Orsay (France) and the detector AGAT have a leading role in the world for the experimental study of the fragmentation of the carbon clusters. Very recently, fragmentation of neutral carbon clusters C_n has been performed by Chabot et al. [8] at the Tandem accelerator. In these experiments, the neutral clusters C_n were produced by high velocity collision on helium gas. Clusters are accelerated by the Tandem accelerator and their fragmentation is analyzed by the 4π, 100% efficient detector, AGAT. Thanks to a shape analysis of the current signal from the silicon detector, branching ratios for all possible fragmentation channels have been measured.

Theoretically, the most studies concerning the fragmentation of carbon clusters have been conducted within a statistical framework. In this one, it is assumed that the energy of the cluster is concentrated on the electronic ground state and is shared between vibrational and rotational excitations. Amongst statistical approaches the Phase Space Theory (PST) was used for extracting, from metastable dissociation of C_n^+, dissociation energies in these species [9]. The simulation of kinetic energy distributions of fragments in the photodissociation of C_n clusters was obtained in a satisfactory way using the PST theory by Choi et al. [10]. Nevertheless, the most complete statistical fragmentation study of neutral carbon clusters was carried out by Diaz-Tendero et al. [11] within the Weisskopf and MMMC (Microcanocical Metropolis Monte Carlo) [12, 13] models through many aspects: consideration of all possible dissociative channels, introduction of a large number of isomers, inclusion of rotational energy, examination of kinetics. The MMMC model has been compared to experiment [11].

In this paper, we have improved and developed MMMC method to investigate fragmentation of small neutral carbon clusters. Instead of using the Metropolis algorithm towards the region of maximum weight in phase space of the MMMC method, all possible microcanonical states in phase space are taken into account in our calculations. Several improvements have been done in our calculations of microcanonical weight of fragmentation channels. We have compared our calculated branching ratios for all possible fragmentation channels with the experimental results. The agreement between theory and experiments is reasonably good. This combination of experimental mesurements with simulation allowed us to extract the deposited energy distributions of the neutral cluster just after the collision that would be extremely difficult to obtain from experiments.

II. MICROCANONICAL STATISTICAL MODEL FOR FRAGMENTATION

In this paper, we investigate only the small neutral carbon clusters. All fragments are neutral, the Coulomb interaction energy between the fragments is thus zero. This model treats the system in internal thermodynamic equilibrium and, therefore, it allows to explore all possible microcanonical states of phase space. In our simulation, for a given excitation energy of parent cluster containing N_C carbon atoms, each phase space point $\mathbf{X}$ (also called a fragmentation configuration or a microcanonical state) is characterized by the physical parameters of the fragments which is composed by

$$\mathbf{X} = \left\{ N_f; \{n_{Cj}, S_{ej}, O_{ej}, G_j\}_{j=1}^{N_f}; \{\mathbf{r}_j\}_{j=1}^{N_f}; \{\mathbf{p}_j\}_{j=1}^{N_f}; \{\Phi_j\}_{j=1}^{N_f}; \{\mathbf{L}_j\}_{j=1}^{N_f}; \{E^*_{vj}\}_{j=1}^{N_f} \right\},$$

where N_f is the number of fragments; $\{n_{Cj}, S_{ej}, O_{ej}, G_j\}$ is the mass, the electronic spin, the electronic orbital degeneracy and geometry (atomic, linear or cyclic); $\mathbf{r}_j$ is the position (chosen such that fragments do not overlap each other); $\mathbf{p}_j$ is the linear momentum; Φ_j are the rotational angles that determine the space orientation (2 for a linear molecule and 3 for non-linear fragments); $\mathbf{L}_j$ is the angular momentum and E^*_{vj} is the internal vibrational excitation energy of the fragment labeled j. All accessible configurations of phase space must satisfy the constraints of conservation of mass ($\sum_{j=1}^{N_f} n_{Cj} = N_C$), total energy ($E_0$), total linear momentum ($\mathbf{P}_0$), and total angular momentum ($\mathbf{L}_0$). The total energy of the system is fixed which is equal to the sum of the fundamental electronic energy E_{gs} and the deposited excitation energy E^* of parent cluster. This energy E^* is distributed between fragments under the form:

$$\begin{aligned}
E^* =& E_b + E^*_v + K_t + K_r, \\
E_b =& \sum_{j=1}^{N_f} E_{gsj} - E_{gs}, \\
E^*_v =& \sum_{j=1}^{N_f} E^*_{vj}, \\
K_t =& \sum_{j=1}^{N_f} \frac{\mathbf{p}_j^2}{2m_j}, \\
K_r =& \sum_{j=1}^{N_f} \left(\sum_{\nu=1}^{f_{rj}} \frac{L^2_{\nu j}}{2I_{\nu j}} \right),
\end{aligned} \tag{1}$$

where E_b is the total electronic energy, E^*_v is the total internal vibrational excitation energy, K_t the total translational energy , K_r the total rotational kinetic energy, m_j the mass, f_{rj} the number of rotational degrees of freedom and $I_{\nu j}$ the principal moment of inertia of fragment j.
Each phase space point $\mathbf{X}$ is associated with a microcanonical weight given by [13]:

$$w(\mathbf{X})\, d\mathbf{X} = \delta(E - E_0)\, \delta(\mathbf{P} - \mathbf{P}_0)\, \delta(\mathbf{L} - \mathbf{L}_0)\, \delta(N - N_C) d\mathbf{X}. \tag{2}$$

Following the definition of $\mathbf{X}$, the volume element of the phase space is expressed as

$$d\mathbf{X} = \left(\prod_{j=1}^{N_f} \frac{d\mathbf{r}_j\, d\mathbf{p}_j}{(2\pi\hbar)^3} \right) \left(\prod_{j=1}^{N_f} \frac{d^{f_{rj}}\phi_j\, d^{f_{rj}} L_j}{(2\pi\hbar)^{f_{rj}}\, \sigma_{rj}} \right) \left(\prod_{j=1}^{N_f} \rho_{vj}(E^*_{vj})\, dE^*_{vj} \right), \tag{3}$$

where σ_{rj} is the symmetry number of the fragment j and $\rho_{vj}(E^*_{vj})$ is the density of vibrational states of fragment j at energy E^*_{vj}.

To be able to make these calculations, our model needs informations of physical characteristics of all possible fragments in their ground states and for all their possible isomers, that is the various multiplicities of spin and the various possible geometries.

II.1. The microcanonical weights of fragmentation partition

In our model, a fragmentation partition (fragmentation channel) of neutral carbon cluster of N_C atoms [14] is represented by a vector $\mathbf{n}$ of N_C components, whose component n_i is the number of fragments with i carbon atoms. The sum of components n_i is the number of fragments $N_f = \sum_{i=1}^{N_C} n_i$, and the mass conservation: $\sum_i i n_i = N_C$. Each fragmentation partition can exist under several configurations because it is necessary to consider all isomeric forms (linear and cyclic geometries and singlet and triplet multiplicities) for C_n ($n = 2-9$). The microcanonical weight of a partition $\mathbf{n}$ for a given excitation energy E^*, is the sum of the weights of all the possible configurations. If the partition $\mathbf{n}$ possesses N_{CF} possible configurations, the microcanonical weights are calculated by the following expression:

$$w(\mathbf{n}, E^*) = \sum_{i=1}^{N_{CF}} w(\mathbf{X}_i, E^*) = w_{\text{comb}}(\mathbf{n}) \sum_{i=1}^{N_{CF}} w_{ei}\, w_{\phi i}\, w_{ri}\, w_{qpli}. \tag{4}$$

To obtain the microcanonical weight of each partition as a function of deposited excitation energy of parent cluster, the first step of our calculation is the generation of all the possible fragmentation channels $\mathbf{n}$. Then for a given partition, our program generates all the possible distributions of isomers of the fragments. For each distribution, the program calculates the various weights of Eq. (4). These weights then will be served to calculate the probability of fragmentation partition as a function of excitation energy. We present the calculations of the reduced weights corresponding to a configuration and properties related to each weight used in our model.

II.1.1. *The combinatorial factor* w_{comb}

The combinatorial factor accounts for the number of ways to allocate N_C carbon atoms to the fragments. There are $N_C!$ ways to arrange the atoms. However, the permutation of atoms inside a fragment does not change the partition nor does the permutation of equal size fragments. Thus this factor is given by

$$w_{\text{comb}}(\mathbf{n}) = \frac{N_C!}{\prod_{i=1}^{N_C} i!^{n_i}\, n_i!}. \tag{5}$$

We remark that this factor depends on the partition while that only depends on the number of fragments in the MMMC model [11].

II.1.2. *The weight* w_e

This weight factor is related to the degeneracy of the electronic ground state. It is determined by the electronic spin and the electronic orbital degeneracy of fragments. This weight can be expressed as

$$w_e = \prod_{j=1}^{N_f} (2S_{ej} + 1) O_{ej}, \tag{6}$$

where S_{ej} is the electronic spin and $O_{ej} = (2l_{ej}+1)$ is the electronic orbital multiplicity of fragment j.

II.1.3. ***The weight*** w_ϕ

This weight counts the possible orientations due to the eigen-rotation of the fragments in the space. It depends on the symmetry group to which they belong and their geometry. This factor is determined via the rotation angles of fragments by the following expression:

$$w_\phi = \prod_{j=1}^{N_f} \int \frac{d^{f_{rj}}\phi_j}{(2\pi\hbar)^{f_{rj}}\,\sigma_{rj}}, \tag{7}$$

where f_{rj} is the number of rotational degrees of freedom of fragment j. In this calculation, the monomers (single atomic fragments) are not included because the atoms are considered as a particle without intenal rotational structure. We consider fragments with linear ($f_{rj} = 2$) and non-linear geometry($f_{rj} = 3$) that can be of cyclic geometry or another geometry. We have

$$w_\phi = \prod_{j=1}^{N_l} \int \frac{d^2\phi_j}{(2\pi\hbar)^2\,\sigma_{rj}} \prod_{i=1}^{N_{nl}} \int \frac{d^3\phi_i}{(2\pi\hbar)^3\,\sigma_{ri}}, \tag{8}$$

where N_l is the number of linear fragments and N_{nl} is the number of non-linear fragments, σ_{rj} is the symmetry number of fragment j. The integration of equation (8) leads to

$$w_\phi = \left(\frac{1}{\sigma_r^l}\right)^{N_l} \left\{\prod_{i=1}^{N_{nl}} \left(\frac{1}{\sigma_{ri}}\right)\right\} \left(\frac{1}{\pi}\right)^{N_l+N_{nl}} \left(\frac{1}{\hbar}\right)^{2N_l+3N_{nl}}. \tag{9}$$

The symmetry number for non-linear fragments σ_{ri} is obtained by quantum chemical calculations. For the linear fragments of $D_{\infty h}$ symmetry, the symmetry number $\sigma_r^l = 2$ because they are invariant by rotation of 180 degrees.

II.1.4. ***The weight*** w_r

The weight w_r represents the spatial part of the volume of the phase space occupied by fragments. It is calculated so that there is no overlapping between fragments. It is defined as the accessible volume for each fragment and can be expressed as

$$w_r = \prod_{j=1}^{N_f} \int_{V_j} \eta(\mathbf{r}_1, \mathbf{r}_2, \cdots, \mathbf{r}_{N_f}) \frac{1}{(2\pi\hbar)^3}\, d\mathbf{r}_j, \tag{10}$$

where

$$\eta(\mathbf{r}_1, \mathbf{r}_2, \cdots, \mathbf{r}_{N_f}) = \begin{cases} 1, & r_{lk} = |\mathbf{r}_l - \mathbf{r}_k| \geq R_l + R_k, l \neq k, \text{ (non overlapping)} \\ 0, & \text{otherwise.} \end{cases} \tag{11}$$

The factor η is introduced in order to avoid the overlapping between two fragments. The fragment's occupation radius R_k is defined as half the largest distance between two carbon atoms for the linear fragment and the smallest radius of the sphere which includes all cluster atoms for the cyclic fragments.

To determine this factor, we simulate the fragmentation in the finite spatial volume. This volume must be large enough to contain all isomeric forms of the parent cluster and all its fragments and mutual interaction (van der Waals forces and exchange of atoms) is negligible. This

volume is called the freeze-out volume. Thus we assimilate it to a spherical volume of radius $R_{sys} = r_f N_C$, where r_f is an adjustable parameter. It was shown that as from one certain value (2 Å per carbon) the freeze-out radius does not have influence anymore on the probability of the partitions. V_j is the volume that the j^{th} fragment can occupy without exceeding freeze-out volume, $V_j = \frac{4}{3}\pi(R_{sys} - R_j)^3$, R_j is the occupation radius of fragment j.
This weight factor measures the number of ways to distribute fragments inside the sphere without covering between them. In order to do this, we make a fixed number of attempts of distributions of fragments in the sphere. The probability of not covering is then given by $P_{\text{nr}} = \frac{n_{\text{nr}}}{n_{\text{tot}}}$ where n_{nr} is the number of attemps not giving covering.

II.1.5. ***The weight*** w_{qpl}

If the excitation energy E^* of the parent cluster is strictly superior to the dissociation energy of the partition, the remaining energy is distributed between fragments or by excited vibrational states of the fragments or kinetic energy of rotation and translation. The weights allow to represent the distribution of the available energy among the fragments of a configuration of the partition under shape respectively of vibrational excitation energy and of kinetic energy of rotation and translation. This weight represents the energy part of phase space, which is the dominant part for the fragmentation.

The volume of phase space concerning the energies of fragments is given by a convolution corresponding to density of states, which is determined by the following expression:

$$w_{qpl} = \int_{E^*_{v1}=0}^{\min(D_1,E')} \int_{E^*_{v2}=0}^{\min(D_2,E'-E^*_{v1})} \int_{E^*_{v3}=0}^{\min(D_3,E'-E^*_{v1}-E^*_{v2})} \cdots \int_{E^*_{vN_f}=0}^{\min(D_{N_f},E'-\sum_{i=1}^{N_f-1} E^*_{vi})} \prod_{j=1}^{N_f} \rho_{vj}(E^*_{vj}) \times \prod_{k=1}^{N_f-1} \prod_{\mu=1}^{f_{rk}+3} \left(\frac{2}{\lambda_{\mu k}}\right)^{1/2} \frac{f(\mathbf{E}^*_v)\,\pi^\alpha}{\Gamma(\alpha)} dE^*_{v1}\, dE^*_{v2} \cdots dE^*_{vN_f}, \tag{12}$$

where E' is the available energy for fragments resulting from the deposited excitation energy in the parent cluster decreased in the dissociation energy corresponding to the given configuration. The energies $\{D_1, D_2, \cdots, D_{N_f}\}$ are the lowest dissociation energy of fragments, they are obtained from the energy data of the ab initio quantum calculations. In this work, for each fragment, we consider only levels of excited vibration which are lower than the lowest dissociation energy. The repartition of the vibrational excitation energy of fragments is represented by a vector $\mathbf{E}^*_v$ of N_f dimensions : $\mathbf{E}^*_v = (E^*_{v1}, E^*_{v2}, \cdots, E^*_{vN_f})$ and $f(\mathbf{E}^*_v)$ is determined by:

$$f(\mathbf{E}^*_v) = \begin{cases} 0 & \text{if } \left(E' - \sum_{j=1}^{N_f} E^*_{vj}\right) < 0 \\ \left(E' - \sum_{j=1}^{N_f} E^*_{vj}\right)^{\alpha-1} & \text{if } \left(E' - \sum_{j=1}^{N_f} E^*_{vj}\right) > 0 \end{cases} \tag{13}$$

In the harmonic approximation, the vibrational level density of each fragment $\rho_{vj}(E^*_{vj})$ is given by the density of states of a f_{vj}-dimensional harmonic oscillator

$$\rho_{vj}(E^*_{vj}) = \frac{(E^*_{vj})^{f_{vj}-1}}{\Gamma(f_{vj}) \prod_{i=1}^{f_{vj}} (h\nu_{ij})}, \tag{14}$$

where f_{vj} is the number of vibrational degrees of freedom of fragment j, Γ is Euler's gamma function and ν_{ij} is the frequency of its i^{th} vibrational mode of fragment j. In the case of monomer (atom), the vibrational density of states does not exist. Thus vibrational density of states is equal to unit. In practice, the factor $\prod_{i=1}^{f_{vj}}(h\nu_{ij}) = \bar{\nu}_j^{f_{vj}}$ where $\bar{\nu}_j$ is the geometrical average of the vibrational frequencies of fragment j which is calculated from the vibrational frequencies obtained from *ab initio* quantum chemistry calculations. The factors $\lambda_{\mu k}$ and α are given by

$$\lambda_{\mu j} = \begin{cases} m_j^{-1} + \left(m_{N_f} + \sum_{l=1}^{j-1} m_l\right)^{-1} & , \ \mu = 1,2,3 \\ I_{\mu-3,j}^{-1} + \left(I_{\mu-3,N_f} + \sum_{l=1}^{j-1} I_{\mu-3,l}\right)^{-1} & , \ \mu = 4,...,f_{rj}+3 \end{cases} \tag{15}$$

$$\alpha = \frac{1}{2}\left(3N_f - 3 + \sum_{i=1}^{N_f} f_{rj} - \max(f_{r1},\cdots,f_{rN_f})\right). \tag{16}$$

In this paper, to calculate the weight w_{qpl}, we created an algorithm which convolves in an exact way the available energy E' for fragments on all the degrees of freedom (vibration, rotation and translation), the remaining energy being the kinetic energy of fragments.

The convolution method

In this method, the integration (12) is effectuated in an exact following way:

$$w_{qpl} = \sum_{i_1=1}^{\frac{\min(D_1,E')}{\triangle E}} \sum_{i_2=1}^{\frac{\min(D_2,E'-(i_1-1/2)\triangle E)}{\triangle E}} \cdots \sum_{i_{N_f}=1}^{\frac{\min\left(D_{N_f},E'-\sum_{s=1}^{N_f-1}(i_s-1/2)\triangle E\right)}{\triangle E}} \prod_{j=1}^{N_f} \rho_{vj}\left((i_j-\frac{1}{2})\triangle E\right)$$
$$\times \prod_{k=1}^{N_f-1}\prod_{\mu=1}^{f_{rk}+3}\left(\frac{2}{\lambda_{\mu k}}\right)^{1/2} \frac{f(\mathbf{E}_v^*)\,\pi^\alpha}{\Gamma(\alpha)}(\triangle E)^{N_f}. \tag{17}$$

The integration step $\triangle E$ has to be much smaller than all the characteristic energies of the system. We showed that the results become stable, when the $\triangle E$ is smaller than 1/20 times of the smallest of the dissociation energies. When the number of fragments is large and the difference between the smallest and the biggest of the dissociation energies are large too, this calculation can be very long.

II.2. Partition probabilities

For a given excitation energy E^* of the carbon cluster, a possible fragmentation partition $\mathbf{n}$ possesses a microcanonical weight calculated by Eq.(4). Especially, Eq.(4) allows to determine the partition probabilities as a function of the initial excitation energy. It is calculated by:

$$P(\mathbf{n}|E^*) = \frac{w(\mathbf{n},E^*)}{\sum_{\mathbf{n}} w(\mathbf{n},E^*)}, \tag{18}$$

where the sum is over all the possible partitions.

III. RESULTS

We present the results for partition probabilities as a function of the excitation energy obtained from our simulations based on quantum chemistry calculations by using the density functional theory (DFT) with hybrid B3LYP functional for exchange and correlation [11]. In our simulations, all isomeric forms for C_n are taken into account. Thus all fragments can play an important role in fragmentation.

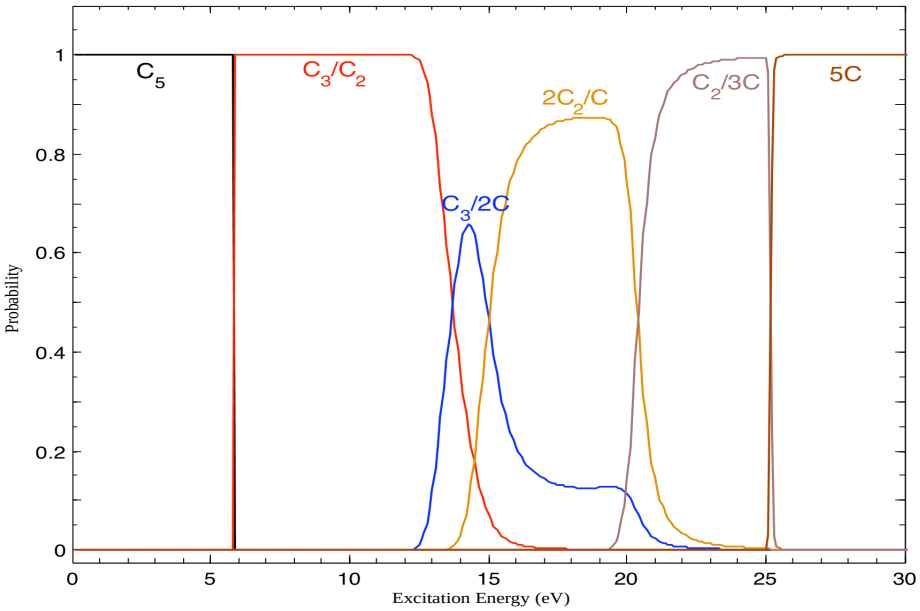

Fig. 1. Fragmentation channel probabilities as functions of excitation energy for neutral carbon cluster C_5.

Figures 1 and 2 present the diagram for fragmentation channel probabilities of C_5 and C_9 clusters, respectively. These figures show the thresholds of appearance of the fragmentation channels as well as the dominant partition corresponding to a domain of excitation energy. The highly excited C_5 cluster can break up according to seven fragmentation channels (partitions): C_5, C_4/C, C_3/C_2, C_3/C/C, C_2/C_2/C, C_2/C/C/C and C/C/C/C/C. The C_5 cluster does not dissociate up to 6 eV. We observe that appearance of fragmentation is sudden. In the range of excitation energy 6-14 eV, the C_3/C_2 partition is dominant, while the other competing channel leading two fragment C_4/C is at very low level. Because of that, the dissociation energy of C_3/C_2 is smaller than that of C_4/C channel. The channels leading to three fragments play a significant role in the region of excitation energy 14-21 eV. The channel of four fragments C_2/C/C/C appears in the domain of energy 20-25 eV. The C_5 is completely broken up from 26 eV. We note that the partitions having the same number of fragments cover approximately the same range of excitation energy. In the case of C_9 cluster, the excited C_9 can follow thirty fragmentation channels. The fragmentation of C_9 begins from 6 eV. The C_9 is completely dissociated from 51 eV in our calculations but from 57

eV in MMMC model [11]. This can be explained by the phase space being expanded faster in our calculations.

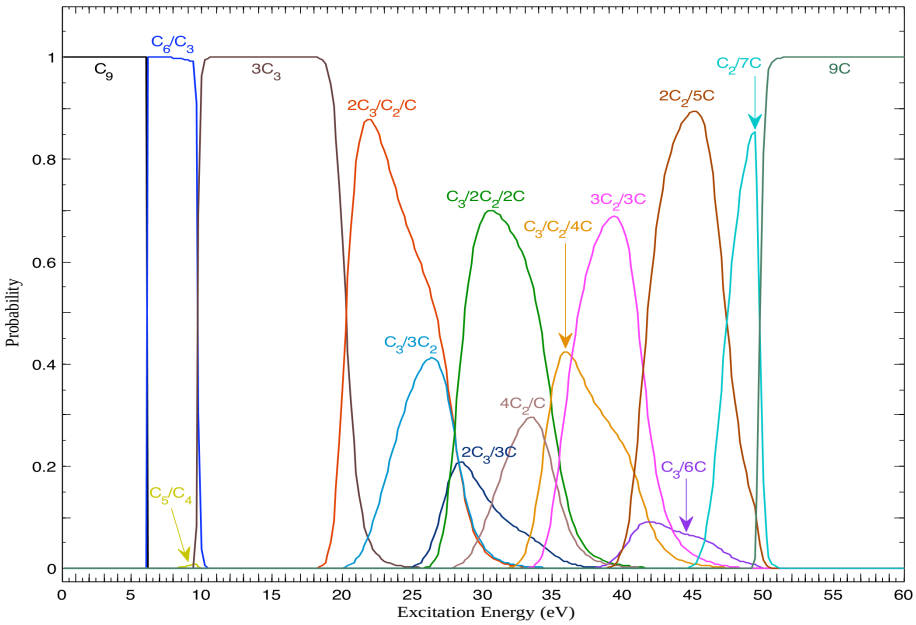

Fig. 2. Fragmentation channel probabilities as functions of excitation energy for neutral carbon cluster C_9.

IV. CONFRONTATION TO EXPERIMENT

The objective of this section is to compare the experimental branching ratios with the results obtained from our simulations. Our calculations give the probability to obtain a fragmentation channel for a given excitation energy. The deposited energy distributions just after the collision were adjusted so that the experimental measurements were optimally reproduced. This adjustment is obtained by solving the system of discrete equations:

$$\forall \mathbf{n}, \qquad P_{\text{exp}}(\mathbf{n}) = \sum_{E^*=0}^{E^*_{\text{max}}} D(E^*) P_{\text{model}}(\mathbf{n}|E^*), \tag{19}$$

where $P_{\text{exp}}(\mathbf{n})$ is the experimental branching ratios of fragmentation channels $\mathbf{n}$, $D(E^*)$ is the excitation energy distribution of the clusters and $P_{\text{model}}(\mathbf{n}|E^*)$ is the probability of fragmentation partition $\mathbf{n}$ obtained from our calculations for a given excitation energy E^*. To solve these equations, two algorithms were used: Non-Negative Least Squares (NNLS) [15] and Bayesian backtracing (BKT) [16]. The objective is to study the uniqueness of the solution by comparing the excitation energy distributions obtained by these two algorithms.

Figures 3 and 4 show the comparison between the experimental branching ratios and the results obtained from our simulations with adjusted energy distributions obtained by NNLS and BKT algorithms for C_5 and C_9 clusters, respectively. The probability distribution of the partitions

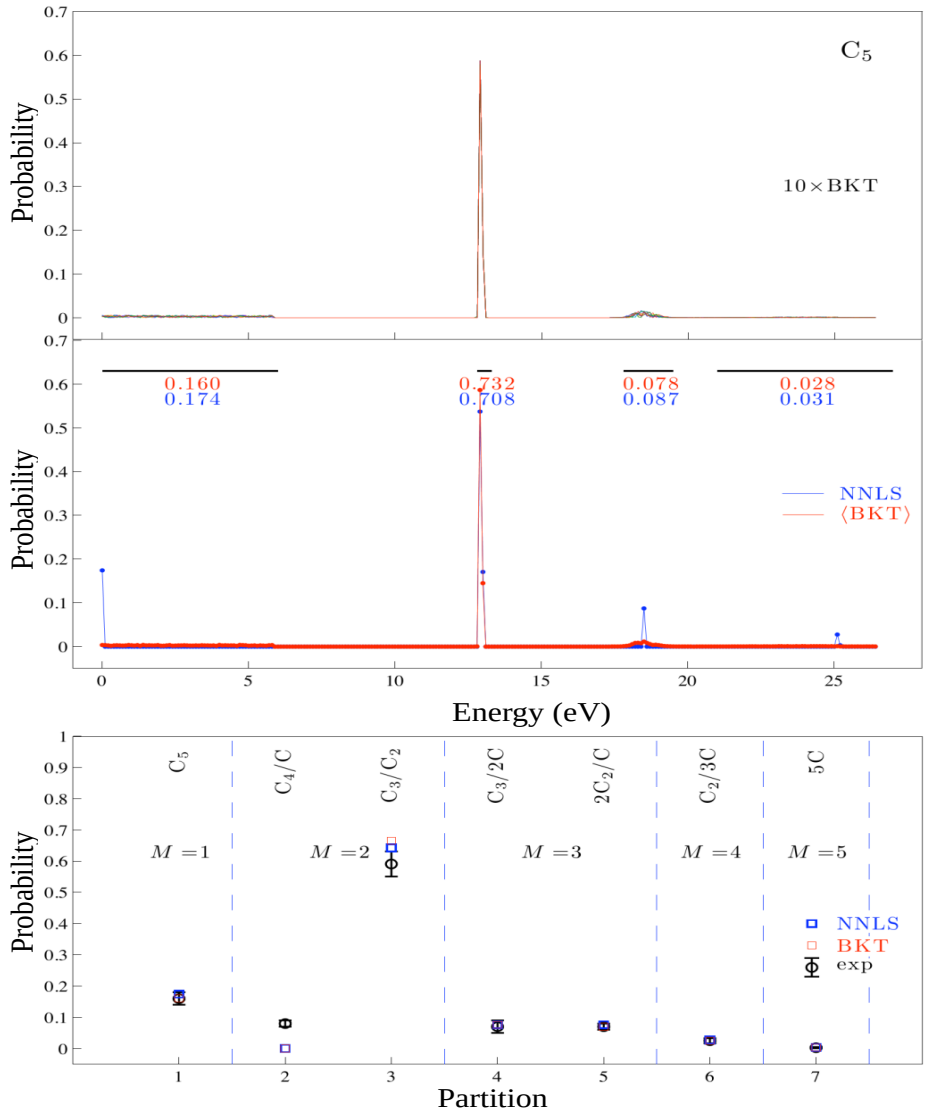

Fig. 3. Results for C_5. Top Figure: Superposition of the $D(E^*)$ excitation energy distributions obtained by the backtracing adjustment method for 10 random initial distributions. Middle figure: energy distribution obtained by NNLS (blue dots) and average distribution of 10 backtracing (red dots). The integrals of distributions in the domains indicated by the black dash are indicated for NNLS (blue values) and backtracing (red values). Bottom figure: comparison of the branching ratios of the partitions: experiment [17] (black circles) and our simulation with the energy distributions adjusted by the BKT (red squares) and NNLS (blue squares), respectively.

is generally well reproduced. In agreement with our theoretical findings, fragmentation channels leading to C_3 are strongly favored. The only problem is related to the prediction of C_4/C channel in

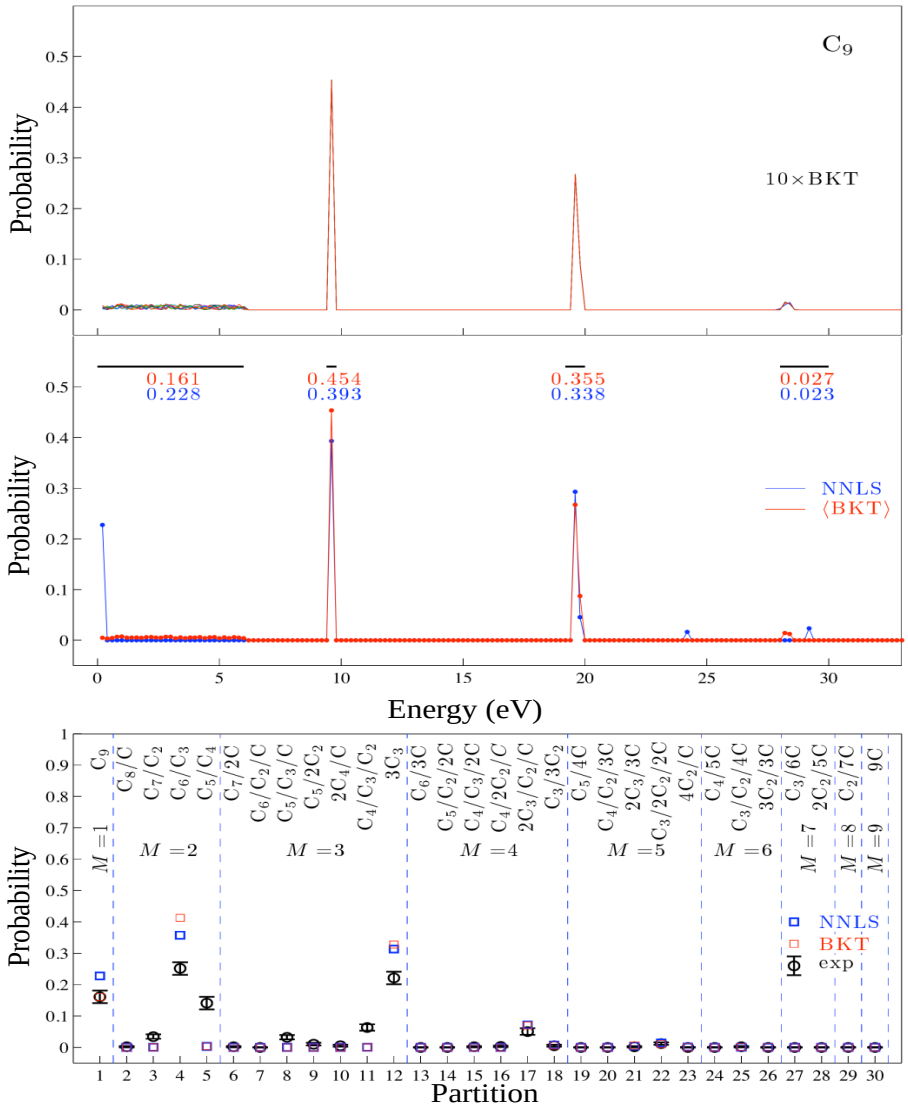

Fig. 4. Results for C_9. Top Figure: Superposition of the $D(E^*)$ excitation energy distributions obtained by the backtracing adjustment method for 10 random initial distributions. Middle figure: energy distribution obtained by NNLS (blue dots) and average distribution of 10 backtracing (red dots). The integrals of distributions in the domains indicated by the black dash are indicated for NNLS (blue values) and backtracing (red values). Bottom figure: comparison of the branching ratios of the partitions: experiment [18] (black circles) and our simulation with the energy distributions adjusted by the BKT (red squares) and NNLS (blue squares), respectively.

the case of C_5. The maximal probability of this channel obtained by our method is 1.5 10^{-4} while experimentally it was about 8 10^{-2}. For energy distribution, it is thus not possible to reproduce its

intensity. As we see, the underestimate of the channel C_{n-1}/C is also present for C_9. For the C_9, the important order of the partitions is well predicted by our simulation, but the global agreement is worse than the fragmentation channels of 2 and 3 fragments. The partitions containing the cluster C_3 are overestimated by our calculation while those containing C_5 are underestimated. A particularly interesting result extracted by the theory/experience confrontation is the appearance of the peaks of energy distributions. However, this result needs to be further studied to validate these peaks.

V. CONCLUSIONS

In this paper, we have presented results concerning the fragmentation of small neutral carbon clusters C_n (n = 5 and 9) obtained by using a microcanonical statistical model. We have found that several fragmentation channels are efficiently populated, but the most probable one always corresponds to C_{n-3}/C_3. Branching ratios for C_n (n = 5 and 9) fragmentation were compared to experimental results. The agreement between theory and experiment is good. We conclude that our statistical fragmentation simulations provide a reasonable estimation of the cluster energy distribution just after the collision. Despite the successful application of microcanonical statistical model to understand fragmentation of small carbon clusters, several improvements can still be done such as introduction of vibrational anharmonicities which might be important at high excitation energy. Particularly, it must continue investigations to conclude the validation of the peaks of excitation energy distributions.

ACKNOWLEDGMENT

We acknowledge the financial support of the International Centre of Physics at the Institute of Physics, Vietnam Academy of Science and Technology.

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