Experimental Determination of Nucleation Scaling Law for Small Charged Particles

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The formation and growth of droplets, clusters, and, more generally, very small particles is of considerable interest. Indeed, these phenomena are essential for instance in clouds formation or crystal nucleation. The formation and growth of small particles can occur in different ways. The formation can start from a seed (heterogenous nucleation), in the presence of a buffer gas or simply occur in a pure vapor (homogenous nucleation). The growth is, for the smallest sizes, essentially governed by successive attachment of single units whereas for bigger sizes coalescence dominates. A huge amount of papers have been devoted to homogeneous nucleation, with the aim of getting a correct quantitative theory of the phenomenon. This aim has not been reached yet. A large majority of these theories originates from the so-called “Classical Nucleation Theory” (CNT), introduced by Becker and Döring [1]. It was originally developed for neutral droplets but can be applied without fundamental changes to ions (it is called in this case Classical Ion-Induced Nucleation Theory, or CIINT) [2, 3]. Ion-induced nucleation often predominates in nature because ions have a higher sticking cross section (SCS) than neutral particles. It has recently been demonstrated in the case of cirrus clouds condensation [4]. One crucial parameter in nucleation is the growth rate, which depends on the size, defined as the probability for a particle to be devoted to homogeneous nucleation, with the aim of getting a correct quantitative theory of the phenomenon. This aim has not been reached yet. A large majority of these theories originates from the so-called “Classical Nucleation Theory” (CNT), introduced by Becker and Döring [1]. 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sion energy to the energy in the Center-of-mass frame). The density of atoms in the cell \( \rho \) is deduced from the cell temperature through the vapor pressure curve. We carefully measured the temperature at different positions in the cell in order to determine as accurately as possible the density of atoms from the lowest encountered temperature. The density of atoms in the cell remains nevertheless the main source of uncertainties. An error of 5 K on the cell temperature leads to a relative error of 25% on the determination of the cross section. These uncertainties can shift the curves collectively, but do not change the size dependence of the cross-section.

Note that under our experimental conditions evaporation is completely negligible. We checked for evaporation effects on a few sample masses: For \( \text{Na}^+ \) we varied \( T_i \) from 150 to 250 K and we did not see any evaporation effect. This is not surprising since the lifetime for the product \( \text{Na}_i^+ \) is still 450 ms at 250 K. For \( \text{Na}_i^1 \) we varied \( T_i \) from 150 to 200 K and found no change in the SCS; the lifetime for \( \text{Na}_i^2 \) is still 63 ms at 200 K. For \( \text{Na}_i^5 \) we varied \( T_i \) from 150 to 340 K and we did see a change in the SCS after a rather long plateau. Evaporation starts to affect the cross section at about 290 K. We find a lifetime of 300 \( \mu \)s for the product \( \text{Na}_i^5 \), at this temperature (\( E_c = 20 \) eV) whereas the time of flight is about 130 \( \mu \)s. It is for sizes smaller than \( n = 25 \) at \( E_k = 20 \) eV and \( T_i = 150 \) K that the lifetime becomes comparable to the time of flight. We can thus safely exclude the effect of evaporation for the size range presented in our results.

Let us emphasize that contrary to previous experiments we work in the single collision regime with very well defined initial conditions (size, temperature and collision energy) and that we explored a wide range of sizes \((n = 25 - 200)\). In the few studies where reaction cross section of mass selected particles are measured as a function of size or collision energy reactants underwent evaporation and/or the size range was quite small [20, 21, 22].

SCS’s have been measured at two kinetic energies. Figure 1 presents our experimental results for \( E_k = 10 \) eV and 20 eV. The most noticeable feature is the two curves are different for small sizes and merge at about \( n = 80 \). From this size on, they follow the geometrical scaling law in \( n^{2/3} \) predicted by the hard sphere model (Figure 2): \( \sigma_{\text{geo}} = \pi R^2 n^{2/3} \), with \( R = 2.4 \) Å (the Wigner-Seitz radius for sodium is 2.1 Å). For smaller sizes, the cross section departs from the hard sphere model. It drops below the geometric cross section for \( E_k = 20 \) eV whereas it increases for 10 eV. For sizes smaller than 80, the cross section depends on the collision kinetic energy: it decreases for increasing kinetic energy. Figure 1 also displays cross sections calculated in the frame of a Langevin model [23, 24]. The cluster-atom interaction is described by charge-induced dipole potentials. Two potentials are involved, corresponding respectively to \( \text{Na}_i^+ \) + Na and \( \text{Na}_i^+ + \text{Na} \) (see Fig. 3). These two states are asymptotically separated by the ionization potential difference \( \Delta IP \) between atom and cluster. In this frame, the sticking can be described as a harpooning mechanism: due to the much larger polarisability of the cluster compared to the atom, an electron transfer from the atom onto the cluster is energetically favored. The adiabatic potentials presented in Figure 2 are calculated using as a coupling the hopping integral in \( \text{Na}_i^+ \) [21]. From the Landau-Zener formula, the propagation is found to take place on the ground state potential curve. In the Langevin model, the largest reactive impact parameter \( b_{\text{max}} \) is reached when the maximum of the effective potential is equal to the collision energy. The cross-section is then given by \( \sigma_n = \pi b_{\text{max}}^2 \). It is taken as geometrical when the Langevin cross section becomes smaller than the geometrical one. This simple model gives satisfactory results for \( E_k = 10 \) eV. However, at \( E_k = 20 \) eV, calculated values disagree with the experiment: the model is unable to account for experimental cross sections being clearly below the geometrical ones. More elaborated models exist [23, 24, 25, 26, 27] but they would also give a cross-section larger than the geometrical one. Moreover we have a charge-induced dipole interaction whereas [23] deals with neutral particles and [24] consider molecules with permanent dipole.

One possible explanation for this behavior is the presence of non reactive excited states of the cluster. Indeed since the experiment is done at fixed kinetic energies in the laboratory frame, the collision energy \( E_c \) increases as the size \( n \) decreases: \( E_c = E_k - \frac{3k_B T}{n} \). Non reactive excited states that can be reached as the collision energy increases are likely to be responsible for the drop in the cross sections for small \( n \). Preliminary calculations based on a two-center jellium model show the appearance of excited states corresponding to the first excitations of \( \text{Na}_i^n \). These excited states lay between the two curves shown in

![FIG. 1: Log-Log plot of the SCS as a function of cluster size at two kinetic energies. Experimental results are compared to a Langevin model and a hard sphere model. The calculated mean cross sections in a vapor at 400 K are also shown.](image-url)
energies which can be considered as small in the sense de-
volution processes involve thermal range collision
non-reactive channels [24]. Our model thus applies only
Figure 2 and might be responsible for quenching towards
values for the
structures. We introduced in our model the experimental
due to electronic shells closure [33], no magic numbers
Although these values exhibit the well known variations
FIG. 2: Effective interaction potentials in the harpooning
action [17]. In this case,
tral particles interacting through a V an-Der-W aals inter-
really surprising. It has been experimentally observed in
experimentally demonstrated in the case of charged clus-
Nucleation in the frame of the CNT also depends in
principle on the SCS scaling law. Nucleation theories aim
to estimate the nucleation rate $J_i$ defined as the number of
droplets of size $i$ created from gas phase per unit time and
volume. Although a number of different versions have been
developed since the work of Becker and Döring, the
basic assumptions remains the same [6]. CNT is
derived from the population equations:
\[
\frac{\partial n_i}{\partial t} = \beta_{i-1}n_{i-1} - \gamma_i n_i - \beta_i n_i + \gamma_{i+1}n_{i+1} = J_{i-1} - J_i
\]
where $\gamma_n$ is the cluster decay rate, $\beta_{n-1}$ the sticking rate, $E_n$ the internal energy, $D_n$ the dissociation energy and $\varepsilon$ the kinetic energy of the evaporated atom. $\Omega_n$, $\Omega_{atom}$ and $\Omega_{n-1}$ are the densities of state respectively of the parent cluster, the evaporated atom and the product cluster. $\beta_{n-1}$ is related to the sticking cross section by the relation $\beta_{n-1} = \sigma_{n-1}pv$, where $p$ is the volumic density of atoms and $v$ the atom velocity. The point we lay the stress on here is that in this microreversible approach the decay rate $\gamma_n$ or the KERD [28, 29] are directly related to the sticking cross section $\sigma_{n-1}$.

Dissociation energies are deduced from decay times $\tau_n$ at cluster temperature $T$ through approximations of relation [2] that can be written to a good accuracy as
\[
\tau_n^{-1} = \sigma_{n-1}A_n \exp\left(-D_n/(k_BT)\right)
\]
$A_n$ is a prefactor independent of $\sigma_n$ whose exact expression
depends on the model. Dissociation energies are
deduced from $\tau_n$ by inverting relation [4]. For sodium clus-
ters, the experimental temperature must be of the order
do sodium (see fig. 1). One can estimate the
order of magnitude of the error made by using a geomet-
trical sticking cross section $\sigma_{geo}$. Replacing $\sigma_{geo}$ by our
estimated value at 400K (see fig. 1) leads to $\Delta D = \pm 4\%$. This error is at least similar to experimental uncertainty
and should be corrected in future experiments on sodium. Unfortunately, a general form of the correction cannot be
inferred here since it is strongly system-dependent, essen-
tially through the energy shift and the coupling between
the two (or more) electronic states involved.

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principle on the SCS scaling law. Nucleation theories aim
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\]
where $\beta$ and $\gamma$ are respectively growth and decay rates.
The CNT considers the steady state solution such as the
fluct $J$ is constant for all $i$. This solution $J_M$ is valid in
a medium time range. Growth at very short delays will
briefly be examined later. For very large delay, the
nucleation phenomenon is qualitatively different, notably
involving coalescence. Moreover, gas depletion prevents
particles from growing indefinitely. All CNT-based theo-
ries arrive at an essentially common expression [6]:
\[
J_M = K\beta_i \exp\left(-\Delta G(i^*)/(k_BT)\right)
\]
where $k_B$ is the Boltzmann constant, $T$ the temperature, $\Delta G^\ast(i*)$ the formation free energy of the critical cluster of size $i^\ast$. The critical size $i^\ast$ is defined by $\frac{\partial^n \Delta G^\ast}{\partial i^n} = 0$, which is equivalent to $\frac{\partial^{n+1} \Delta G^\ast}{\partial i^{n+1}} = 1$. The prefactor $K$ does not depend on the cross section. $K$ includes the so-called Zeldovich factor \cite{38} which does not depend on the sticking scaling law provided that it is a smooth function of the size \cite{38}. Relation \cite{38} thus the micro reversibility principle, makes $i^\ast$ independent of the SCS scaling law. This had already been pointed out by Vasil'ev and Reiss \cite{38}. Finally, the nucleation rate in the constant flux regime is modified only linearly through $\beta_L$. In CNT, $\beta_L$ is always calculated under the assumption of hard sphere collisions. In CIINT, however, many authors consider that the stronger interaction between charged clusters and atoms leads to an enhancement of $\beta_L$. They introduce a so-called Enhancement Factor (EF) \cite{39,40}. If the vapor molecule has no permanent dipole, EF is generally considered as negligible, as far as clusters bear a single charge \cite{40}. However, when evaluating EF, only the interaction between the charged cluster and the neutral particle is considered. It is demonstrated here that, at least in the case of sodium, the harpooning mechanism strongly modify the SCS’s. An example of EF($n$) at 400 K is readily deduced from fig. 1: it is the ratio $\frac{\sigma_{400} n}{\sigma_{geo}}$. EF depends on the system, the temperature, the coupling between the electronic states. So we cannot derive any general rule for estimating EF. Nevertheless, we have shown that more attention should be paid to this factor.

Let us focus now briefly on short times. There is a so-called lag time $t_l$ that characterizes the time spent before the steady state is reached. The nucleation rate $J$ follows approximately the law $J(t) \approx J_M(1 - \exp(-B(\beta_L t)))$, where the prefactor $B$ do not depend on the SCS \cite{41}. Here the change could be more significant; however, the effect on the steady-state nucleation rates is not significant since the lag time by itself is very short in vapor-to-liquid transformations \cite{42}. Far from equilibrium phenomena may therefore be more influenced by a change in the sticking scaling law. Cluster nucleation in a supersonic expansion is an example of such phenomena. As mentioned above, Vigué et al already studied the case of neutral clusters \cite{43}. The final average cluster size $n_f$ can be represented by empirical scaling laws of the type $n_f \approx \Gamma \frac{1}{\sqrt{\beta_L}}$ for a sticking law in $n^\alpha$. The scaling parameter $\Gamma$ introduced by Hagenia only depends on the geometry, the pressure and the temperature in the source \cite{15}. The example calculated at thermal energy on figure \cite{44} shows that $\alpha$ can be close to 0. The change in $n_f$ is obviously not negligible.

In summary, absolute SCS of mass selected sodium clusters have been measured for the first time. The non geometrical sticking law that depends on the collision energy is shown to influence not only nucleation but also cluster evaporation measurement results.

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