Collisional destruction of anionic carbon and silicon clusters by helium, neon, and argon atoms at intermediate velocities

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(Received 24 November 2000; published 31 July 2001)

We measured the total destruction cross sections of small X_n^- clusters (X = Si and C; $n = 2$, 3, and 4) in noble gas targets (Y = He, Ne, and Ar), with relative velocities v in the 0.10–0.95 a.u interval for silicon, and in the 0.20–1.20 a.u. interval for carbon. The measured values increase monotonically with target and projectile sizes, as expected on a geometrical basis. In particular the dependence of the cross sections on n is linear for carbon clusters, agreeing with a quantitative model based on the linear structure of these clusters. Ratios of the measured cross sections for the several X_n^- clusters, $\sigma_{X,n}^{Ne}(v)/\sigma_{X,n}^{He}(v)$, $\sigma_{X,n}^{Ar}(v)/\sigma_{X,n}^{He}(v)$, and $\sigma_{X,n}^{Ar}(v)/\sigma_{X,n}^{Ne}(v)$, were verified to fall into universal curves, i.e., each cross section is the product of target- and projectile-dependent factors.

DOI: 10.1103/PhysRevA.64.032701 PACS number(s): 34.50.Gb

I. INTRODUCTION

The study of neutral and cationic clusters of group-IV elements, such as carbon, silicon, and germanium, has been an extremely active research area during the past decade [1]. This is particularly noticeable for carbon, which presents technological and scientific challenges leading to great advances in materials science and other areas [2], but is also the case, although on a smaller extent, for silicon and other semiconductor clusters. Small- and medium-sized neutral silicon clusters, for instance, have been the object of many works [3] (and references therein), the main drive being the current trend of miniaturization in microelectronics. Besides these applied research areas, as clusters lie between atoms and condensed matter, their study is an important basic research area, essential for understanding condensed-matter phenomena.

Anionic clusters, although less studied than their neutral and cationic counterparts, also pose important basic problems. Among these, one may notice that anion structures present strong electron correlation, requiring refined many-body calculations, and, concerning collisions, single electron loss from anionic species may provide a better understanding of the role played by the long-range Coulomb interaction when comparing with similar processes for neutral projectiles. On the applied side, negative molecular ion collisions also are essential for areas such as upper-atmosphere chemistry. Carbon again is the most studied case, small carbon anionic clusters having their production, photodetachment, electron-impact dissociation, and other collision phenomena studied [4] and their ground-state structure calculated [5]. Silicon anionic clusters were also studied with, for instance, photoelectron spectroscopy measurements of Si_2 and Si_2^- [6], collision measurements on NO_2 [7], and *ab initio* calculations of their structure [8]. Recently, efforts have also been made in comparing properties of C, Si, and Ge small anionic clusters, such as their different growth patterns [9].

Notwithstanding these efforts, much remains to be done to understand the structure and the collision processes of anionic silicon and carbon clusters. For instance, even considering a recent measurement of the lifetimes of C_2^- vibrational levels [10], few works present experimental or theoretical results for the lifetimes of excited states (either bound ones or Feshbach resonances) of anionic clusters or even discuss their presence [11]. Concerning collisions of anionic clusters with atomic targets, there are, as far as the authors are aware, no available calculations. Although some measurements were recently done for small anionic clusters colliding at low velocities (below 0.1 a.u.) with atomic or molecular targets [12–14], they are scarcer than, for instance, the ones available for low-energy collisions of neutral and cationic C_{60} with noble-gas targets (see Ref. [15] and references therein). The need for experimental data at intermediate collision velocities is clear, as most works are concerned with the near-threshold region [16] while high-velocity collisions (much larger than 1 a.u.) are expected to be described by approaches such as the first-order Born approximation (BA), free-collision model (FCM), or sudden aproximation (SA).

Negative ions are the ideal system to study electron correlation effects. A recent review [17] presents a detailed discussion of these many-body effects in photodetachment at the low- and intermediate-energy range. One such effect, obtained when comparing theoretical and experimental photodetachment results for C^-, Si^-, and Ge^-, all having np^3 configurations, is a sharp interference on Si near 5 eV which is not present in the carbon data. Nevertheless, distinct from these photodetachment results, no such structure was seen in the collisional detachment cross sections of atomic silicon anions on noble-gas targets, recently measured by our group [18]. A study of collisions of noble-gas atoms with small clusters of different semiconductor species, where their structures differ the most, is then expected to shed some light on the hitherto uncalculated collisional detachment process, allowing comparisons to be made both with their respective atomic anion destruction data and between themselves. These measurements, besides being relevant, are also feasible, as negative ions may be easily produced in cesium sputtering ion sources, common in tandem-type accelerators. This is the case, for instance, of C_n^- and Si_n^- beams,

$n=1$ to 4, which were already produced by these ion sources [19,20].

Simple scaling rules for cross sections were recently presented for electron-impact detachment in the low-to-intermediate velocity range [21,22], as also for collisional destruction of molecules [23,24] and for electron-impact ionization of atoms and molecules [25]. Robicheaux [21] calculated electron-impact detachment cross sections in the CM of the two free electrons and obtained a scaling based only on the electron affinity. Rost [22] studied the same process using the Wannier approach and obtained scaled cross sections as well, although dependent also on other empirical parameters. Simple scaling rules were also obtained in a similar systematic study of the destruction of fast small molecules colliding with noble-gas atoms (H_2, H_2^+, and H_3^+ projectiles and velocities going from 3.0 to 7.0 a.u.) [23]. The ratios of the total destruction cross sections—$\sigma_D(\mathrm{Ne})/\sigma_D(\mathrm{He})$ and $\sigma_D(\mathrm{Ar})/\sigma_D(\mathrm{He})$—were then observed to be, essentially, projectile-independent, and to present a slight velocity dependence. Besides this, the projectile dependence of the destruction cross section was observed to scale as the number of quasifree (least-bound) projectile electrons divided by the dissociation energy, as expected from the simple FCM [24]. A semiempirical model for electron-impact ionization of molecule and noble gases has also been proposed, based on classical geometrical considerations and the Bethe-Born model. Using distinct empirical weights for the contributions of the *s* and *p* electrons, a very good analytical description of the experimental cross sections was then obtained [25]. Concerning anionic clusters, however, the possible presence of excited-state projectiles may affect such simple scalings. For instance, evidence of the production of metastable C_n^- anions in sputtering-type ion sources was given by Safvan *et al.* [13] and by Andersen *et al.* [26].

In this work, we present results for systematic measurement of the noble-gas collisions with Si_n^- and C_n^- clusters in the intermediate velocity range. The cluster sizes went from 2 to 4, and helium, neon, and argon were employed as targets. As far as the authors are aware, there are no measurements of collisional destruction by atomic and molecular targets at this velocity range, going from the orbital velocity of the anion least-bound electron to 1 a.u. It is instructive to know whether some scaling rules, such as the ones observed in Ref. [23], also hold for the presently studied collision systems. It is also important to verify whether the cross-section linear dependence on the cluster size, observed for fixed-low-energy anionic carbon clusters [12,14] (and, consequently, size-dependent velocities), still holds when comparing equivelocity carbon and silicon clusters. In order to clarify these points, we specifically discuss the dependence of the present cross-section data on the target and the cluster atomic species, and on the cluster size.

II. EXPERIMENTAL ARRANGEMENT

The overall experimental arrangement has already been described elsewhere [20], and we will give here only a short description. Carbon and silicon cluster anions (C_n^- and Si_n^-, $n=1,4$) were obtained from a cesium sputtering negative ion source (SNICS) at the Institute of Physics of the Federal University of Rio de Janeiro (IF-UFRJ). Ion beam currents typically ranged from tenths to hundreds of nanoamperes. After extraction, the negative ions were mass selected by a Wien filter and electrostatically focalized into the first stage of a 1.7 MV tandem accelerator (5SDH, from NEC). At the end of this stage, they traverse a gas stripper, placed at the high-voltage terminal. Particles of various charge states can then be produced and, in the second stage of the accelerator, the positive ones were accelerated again while the negative decelerated. The desired specie ions were then deflected 15° angle by a steering magnet, the beam current being finally measured with a Faraday cup.

The gas stripper is employed as a differentially pumped gas target [20]. The gas is introduced from outside through a pressurized insulating tube, the target pressure being regulated by an externally controllable mechanical valve. The target gases employed have nominal purities of 99.9%. The density of target particles is obtained from a pressure gauge placed at the high-energy end of the accelerator, linearly related to the stripper pressure. The absolute value of the stripper pressure for each gas target is then obtained by comparing the neutral fraction, measured on an auxiliary H^- beam experiment, to the hydrogen neutral fraction calculated employing the well-known cross sections of collisional charge change of hydrogen [27].

The destruction cross section of a given negative specie was extracted, in the usual way, by exponential fits to the transmitted current *versus* target pressure curves:

$$I = I_0 e^{-\sigma_D \pi},$$

where π is the area density of target particles, I_0 is the current at the target entrance, I is the transmitted current, and σ_D is the sum of the cross sections for all the inelastic channels, i.e. single and multiple ionization, fragmentation, etc. Although beam current may be lost in both accelerator stages due to the focalization effect of the terminal voltage, which is assumed to be constant for each beam energy, or in the extraction and detection regions, these effects are pressure-independent and, consequently, do not affect the measured cross-section values.

A major advantage of this method is that the transmitted anions have identical initial and final energies, with the initial energy, gained on the extraction stage, having a value around 20 keV. This fact enables the study of collisional detachment of fast (~MeV) and heavy (masses up to ~600 a.m.u.) particles with the use of a magnetic analyzer of small mass-energy product (in the present case, 12 a.m.u. MeV at 15°).

There are, however, limitations on the maximum and minimum velocities attained. In order to avoid electrical breakdown inside the gas tube, as the gas comes from a grounded high-pressure bottle to the gas target placed at the tandem high voltage, this voltage must have an upper limit, which depends on the gas. One also has the defocalizing effect of the terminal voltage, whether it be too high (close to the machine's maximum) or too low (close to the extracting potential). This effect, reducing the transmission of the clus-

ters, is more important for larger clusters with weaker beam currents. All this taken in account, we end up with size- and target-dependent maximum workable projectile velocities, as may be seen on our experimental cross-section results. For Si_2^- ($m=56$ a.m.u.), these limiting velocities are 0.75 for He, 0.6 for Ne, and 0.95 a.u. for Ar. For Si_3^- ($m=84$ a.m.u.), they are 0.6 for He and 0.7 a.u. for Ar. For Si_4^- ions ($m=112$ a.m.u.), both the He and the Ar limits are equal to 0.3 a.u. For C_2^- ($m=24$ a.m.u.), they are 1.0, 1.2, and 1.1 a.u., respectively, for He, Ne, and Ar. For C_3^- ($m=36$ a.m.u.), they are 0.7, 0.9, and 0.9 a.u. for He, Ne, and Ar. And, finally, for C_4^- ($m=48$ a.m.u.), they are 0.7, 0.8, and 0.8 a.u. for He, Ne, and Ar.

The cross-section standard deviation was experimentally evaluated by repeating the same measurement several times, and also by changing the pressure range for the exponential fit. One of the most important sources of error is the fluctuation of the beam current, so the total beam current was measured, for a given target pressure, immediately before and after measuring the transmitted current, and runs with a fluctuation greater than 10% were discarded. Considering all these factors, the standard deviation was, in the worst case, equal to 7%.

III. EXPERIMENTAL RESULTS AND DISCUSSION

The measured destruction cross sections for C_n^- clusters ($n=2$, 3, and 4) are shown in Figs. 1(a), 1(b), and 1(c), respectively, for helium, neon, and argon targets. In Figs. 2(a), 2(b), and 2(c), we present the measured destruction cross sections for Si_n^- clusters for helium, neon, and argon, with $n=2$, 3, and 4 for He, $n=2$ for Ne, and $n=2$, 3, and 4 for Ar. As expected on a geometrical basis, for a given velocity the measured values increase monotonically with the cluster number n and the target atomic numbers. Concerning the projectile atomic species, Si_n^- cross sections exceed the corresponding C_n^- ones.

A. Target dependence

A measure of the target dependence (target $Y=$ He, Ne, or Ar), for a given X_n^- projectile ($X=$ C or Si; $n=2$, 3, or 4), may be furnished by the ratios of the measured destruction cross sections as functions of the velocity. These ratios, plotted in Fig. 3 for both the carbon and the silicon cases, are smooth functions of the velocity and are essentially projectile-independent, evidencing that the cross sections may be factorized into target- and projectile-dependent factors. A similar phenomenon had already been found [23] for fast hydrogen molecules H_2, H_2^+, and H_3^+ (these results, also plotted on Fig. 3, present very similar values for the different projectiles and were averaged for the sake of clarity). Although these two scalings may be associated with distinct phenomena, these two sets of results may be fitted by a smooth curve for each pair of targets.

As a detailed description of the molecular hydrogen data and their analysis has already been presented [23], let us give here just a short review. Cross sections were measured and their ratios, for any given molecule, verified to fall into universal curves. As the relative velocities are larger than the average orbital velocities of the noble-gas valence electrons (1.1–1.9 a.u. [28]), a FCM description could be employed, where molecular fragmentation was associated to its electronic excitation into a dissociative state mostly by the impact of quasifree noble-gas valence electrons. A strong role should then be played by the outermost ns^2 electrons, present on all three noble gases, and by the np^6 electrons, present only on Ne and Ar, partially explaining the large values of the Ar/He and Ne/He ratios at large velocities and also, as Ne and Ar have similar s^2p^6 configurations, the observed small velocity dependence of the Ar/Ne ratio.

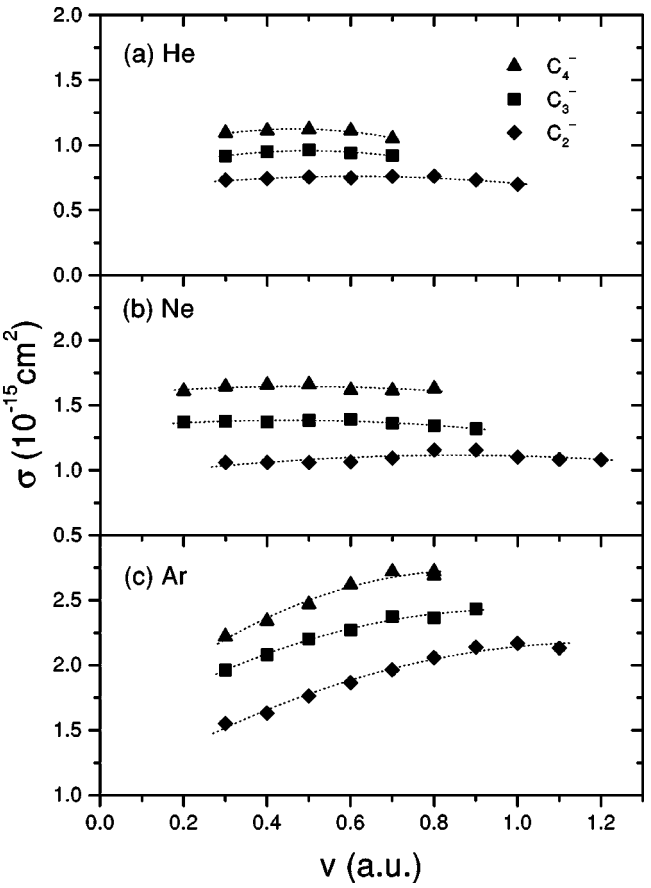

FIG. 1. C_2^-, C_3^-, and C_4^- destruction cross sections for He, Ne, and Ar targets, with dotted lines only to guide the eyes.

Considering now the presently studied anionic cluster destruction, arguments based on the FCM model are hardly applicable, as the relative velocities are smaller than or equal to 1.1 a.u. and consequently the noble-gas valence electrons may not be considered as free particles. A simple empirical observation, however, is that cross sections in general present smooth velocity dependence for relative energies greater than the energy threshold. Hence in the present case almost constant cross-section ratios, not only for the Ar/Ne case but also for Ne/He and Ar/He, are predictable for a narrow velocity range, as was experimentally obtained for the present 0.20–1.10-a.u. range.

A striking point, however, is the almost constant value for the Ar/Ne ratio, going from 1.5 to slightly over 2.0, in a wide velocity range encompassing these two velocity regions, i.e.,

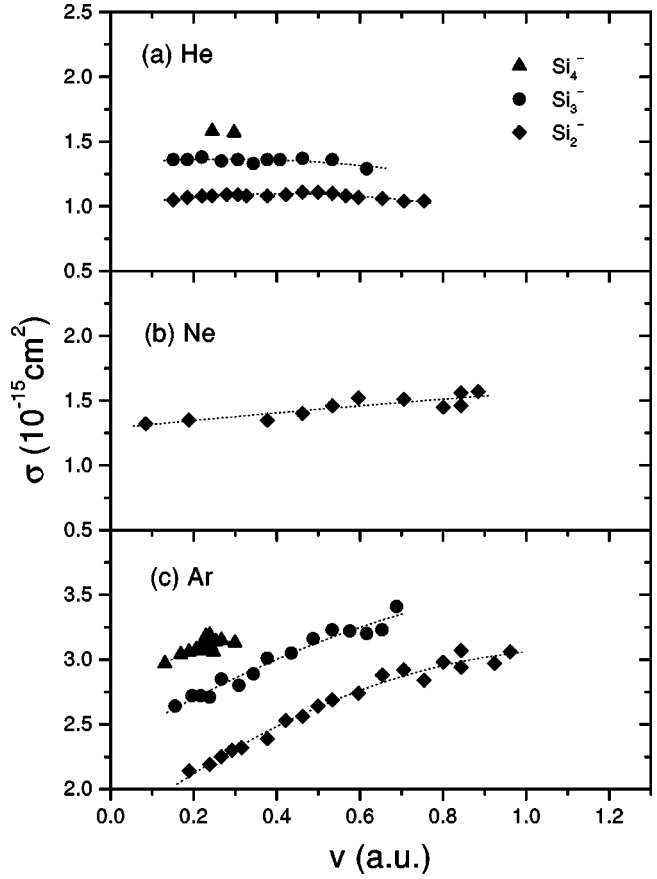

FIG. 2. Si_2^-, Si_3^- and Si_4^- destruction cross sections for He, Ne, and Ar targets, with dotted lines only to guide the eyes.

going from 0.2 to 7.0 a.u. This behavior contrasts with the marked velocity dependences of the Ar/He and the Ne/He ratios. Considering that six distinct anionic clusters and three hydrogen molecules were studied, this experimental result is hardly casual, although no theoretical explanation is as yet available.

B. Projectile dependence

As a second point, a near parallelism of the curves for silicon or carbon clusters, with different sizes and the same target, is noticeable at low velocities. This indicates that each cluster destruction cross section may be considered as the respective atomic anion destruction cross section plus a constant, this constant being monotonically dependent on the cluster size. Besides this parallelism, for a given velocity the differences among subsequent cross sections are similar to each other, particularly in the carbon case. One way of looking into these phenomena is trying to fit straight lines to the low-velocity cross sections as a function of cluster size, as done in Fig. 4 for a typical low velocity, $v=0.3$ a.u. (also shown are Si^- results [18] and preliminary C^- results). A linear dependence with cluster size is then clearly observed in the carbon case and, less evidently, in the silicon case.

Negative carbon clusters possess a linear structure and present nearly constant internuclear spacings [4], with electron affinities showing an alternating behavior with cluster size (clusters with even number of atoms have greater values than the ones with odd numbers [5]). A simple geometrical model [14] considers the anionic carbon cluster as a cylinder ended by two hemispheres, where the extra electron wave function is localized. Given an internuclear spacing L and the hemispheric radius R, we get a simple expression for the cross section, after averaging over the polar angles:

$$\sigma_{C,n}=\pi R^2+\frac{\pi}{2}(n-1)LR,$$

where πR^2 is the measured $\sigma_{C,1}$ cross section. This expression is also plotted in Fig. 4 for carbon clusters. A good agreement is obtained for carbon clusters colliding with all the targets, discrepancies being smaller than 10%. This is remarkable considering the simplicity of the model assumptions (only the atomic anion detachment cross section is an adjustable parameter). Also the presence of nonlinear geometries associated to excited states is neglected, although data

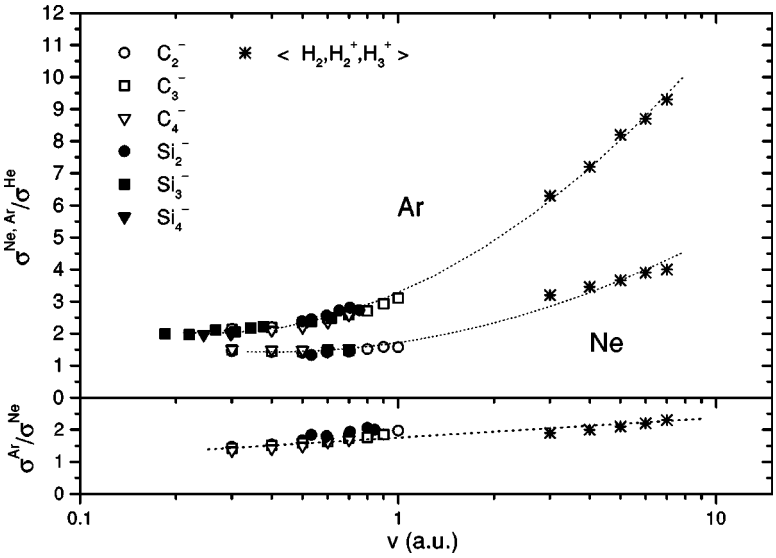

FIG. 3. Destruction cross-sections ratios as a function of velocity: $\sigma_n^{Ar}(v)/\sigma_n^{He}(v)$, $\sigma_n^{Ne}(v)/\sigma_n^{He}(v)$, and $\sigma_n^{Ar}(v)/\sigma_n^{Ne}(v)$. Data for the fragmentation of fast hydrogen molecules [23]—H_2, H_2^+, and H_3^+—are also shown, averaged for the sake of clarity, with dotted lines representing polynomial fits.

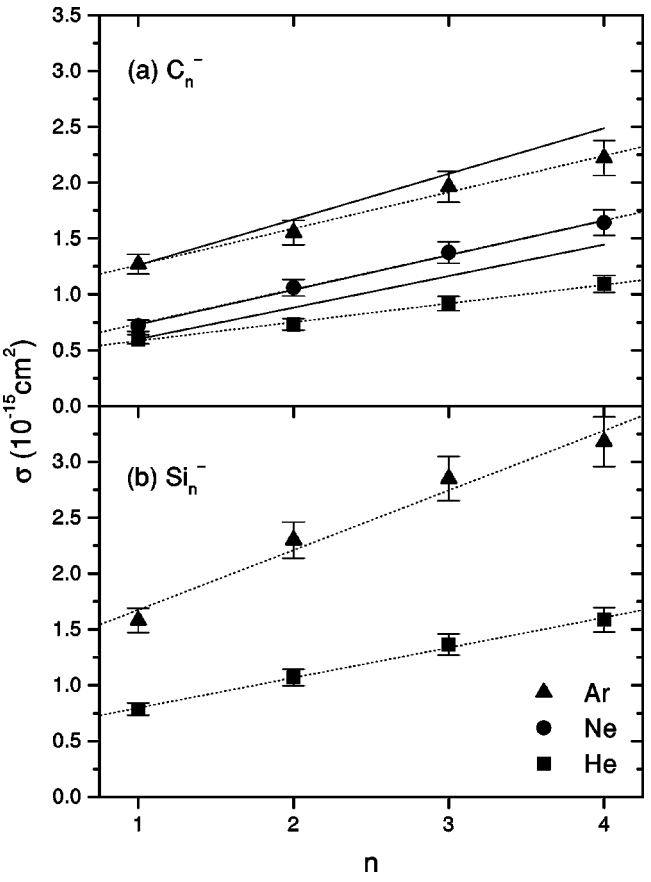

FIG. 4. Destruction cross sections as a function of the cluster size at $v=0.3$ a.u.: (a) C_n^- for He, Ne, and Ar targets (including preliminary results for C^- at this velocity) and (b) Si_n^- for He and Ar targets. The full lines are obtained from the geometrical model expression [14], while the dotted lines represent straight-line fits.

taken in sputtering-type ion sources point to C_4^- anions being on the ground state while C_2^- and C_3^- present long-lived excited states [26].

Silicon clusters present more complex geometries [7]. Si_2^- has $^2\pi_u$ ground and $^2\Sigma_g^+$ excited states, the latter lying 0.1 eV higher than the former; Si_3^- has two bound states, both with C_{2v} geometries, $^2A_1$ and $^2B_2$, with electron affinities differing by 0.03 eV; the Si_4^- cluster is nonlinear, with a ground D_h $^2B_{2g}$ state and an excited C_{2v} $^2B_{1g}$ state 0.48 eV higher. The ground-state affinities were similar for these three silicon clusters. The distinct geometries presented in the silicon case therefore do not allow a geometrical model as simple as the one for carbon, but, even so, there is a reasonable linearity of the cross sections in the He and Ar cases, displayed at Fig. 4. In order to better understand these results, a simple model may be made, where these clusters, averaged over the polar angles, were considered as spheres of radii R_n, with the $n^{2/3}$ size dependence given by the liquid-drop model. The destruction cross section σ_n will then be

$$\sigma_{\mathrm{Si},n}=\pi(R_n+R_t)^2,$$

where R_t are the atomic target radii estimated using the average radius values of the ns^2 noble-gas electrons [28]. Using these target radii values, the cluster radii obtained are equal to 1.02 ± 0.16, 1.49 ± 0.22, 1.72 ± 0.36, and 1.64 Å, when n goes from 1 to 4. These values present an increase with the cluster size n, which is somewhat slower than the $n^{2/3}$ liquid-drop model prediction and give only an order-of-magnitude agreement with the cluster dimensions, but, considering the roughness of the model assumptions—spherical silicon clusters and liquid-drop model—even a qualitative agreement is surprising.

IV. CONCLUSIONS

Destruction cross sections for intermediate-velocity small anionic carbon and silicon clusters were measured. As a first point for each element, the cross sections present remarkable similarities, which is also verified when comparing with the C^- and Si^- cross sections, their differences being nearly constant over the studied velocity range. A second important point, coming from the experimental results, was that at low velocities and for all three targets the carbon cross sections present a linear dependence on the cluster size n. This linear behavior may be quantitatively described using a purely geometrical model, with an unexpectedly good agreement. The complexity of this problem, however, is indicated by the fact that nonlinear silicon clusters present cross sections that are also linearly-dependent on n in the He case and are roughly linear in the Ar case. These two ponts show the need for a quasimolecular description of these collision processes.

The most striking finding, however, was the behavior of the $\sigma_{X,n}^{\mathrm{Ne}}(v)/\sigma_{X,n}^{\mathrm{He}}(v)$ and the $\sigma_{X,n}^{\mathrm{Ar}}(v)/\sigma_{X,n}^{\mathrm{He}}(v)$ cross-section ratios. These ratios were calculated for different X_n^- clusters and were verified to fall into universal curves, i.e., each cross section could be factored into a product of target and projectile-dependent factors. A similar finding had also been previously obtained for the dissociation of faster hydrogen molecules H_2, H_2^+, and H_3^+ [23]. The existence of these universal curves is qualitatively explainable on the basis of simple arguments, but the small velocity dependence of their $\sigma_{X,n}^{\mathrm{Ar}}(v)/\sigma_{X,n}^{\mathrm{Ne}}(v)$ ratio remains an open question. Clearly further measurements with anionic semiconductor clusters of larger sizes and at larger velocities, as well as intermediate-velocity hydrogen molecules, are needed, particularly in the 1.0–3.0 a.u. velocity range, to clarify both the range of validity for the present experimental scalings and what causes them.

In order to interpret the target and projectile dependences of these results, there is also a clear need for theoretical analysis of collisions at intermediate velocities, where PWBA does not apply, the molecular orbital/coupled channel approaches are numerically prohibitive, and the frequently used classical trajectory Monte Carlo is not expected to fully describe a quantum many-body problem [29]. These difficulties increase particularly when considering the strong electron correlation for negative ions. The fact that, even so, experimental scalings of a wide variety of projectile-target systems are possible strongly suggests an underlying simplicity of these intermediate-velocity collisions.

ACKNOWLEDGMENTS

We gratefully acknowledge partial financial support from the following Brazilian agencies: Fundação Universitária José Bonifácio (FUJB), Fundação de Apoio à Pesquisa do Estado do Rio de Janeiro (FAPERJ), Coordenação de Aperfeiçoamento de Pessoal do Ensino Superior (CAPES), and Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq).

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