

# Morphology and evolution of size-selected metallic clusters deposited on a metal surface: $\text{Ag}_{19}^+/\text{Pd}(100)$

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We study size-selected deposition of  $\text{Ag}_{19}^+$  clusters on Pd(100) at total kinetic energies of 20 and 95 eV using thermal energy atom scattering and molecular-dynamics simulations. Contrary to the case of  $\text{Ag}_7$  where fragmentation is crucial to explain the data, the deposition leads at low temperature to noncompact structures localized around the impact point. We propose a model in which morphology changes take place between 200 and 300 K resulting in well-separated compact structures. [S0163-1829(98)06907-0]

## I. INTRODUCTION

During the last ten years a wealth of information has been obtained on the properties of free clusters as a function of their size, and characteristic properties different from those of the bulk or atoms have been obtained.<sup>1</sup> Over the past few years renewed effort on the preparation and understanding of the properties of clusters deposited in a matrix<sup>2</sup> or on a surface<sup>3</sup> has been undertaken, in the hope of forming nanostructures with a well-defined shape and number of atoms. Deposition at low energy of size-selected clusters on a solid substrate is well suited to achieve this goal. Understanding the landing process and the subsequent behavior of clusters on a surface in order to reach “soft-landing” conditions is from this perspective an essential first step. It is also interesting in itself and related to several recent studies,<sup>4,5</sup> which have examined the growth kinetic and diffusion behavior of clusters formed on a surface by thermal evaporation of atoms.

Several previous experiments have studied size-selected and non-size-selected clusters deposited on insulating<sup>6</sup> or metal<sup>7</sup> substrates using photoemission. Recent work has used scanning tunneling microscopy to investigate the deposition of size-selected metal clusters on highly oriented pyrolytic graphite (HOPG),<sup>8,9</sup> and the cluster morphology has been explored as a function of impact energy. In this paper we present thermal energy atom scattering (TEAS) results on the system  $\text{Ag}_{19}/\text{Pd}(100)$ . Our data are the first to describe the morphology of a cluster/substrate metal-on-metal system for a range of impact energies as low as 1 eV/atom and its evolution with temperature. The method of analysis is based on the large cross section  $\Sigma$  for diffuse scattering of adsorbates seen by the He beam. Since  $\Sigma$  is typically ten times larger than the surface unit cell, it decreases when the adatoms form clusters due to overlap of cross sections. This yields information on cluster size distribution and on its dynamics on the surface.<sup>10</sup> Our experimental evidence is corroborated by state-of-the-art molecular dynamics (MD) and static cal-

culations performed on the same system, which provide an atomic scale view of the collision outcome.

Previous results<sup>11</sup> using TEAS have been reported on the deposition of  $\text{Ag}_1^+$  ions and  $\text{Ag}_7^+$  cluster ions on Pd(100) at total incident kinetic energies of 20 and 95 eV. They have shown that both fragmentation and implantation are crucial to explain that for  $\text{Ag}_7$  the effective cross section at low temperature (80 K) is larger at 20 than at 95 eV, while at higher temperature (400 K) the reverse is true. We report in this paper results obtained by depositing  $\text{Ag}_{19}^+$  cluster ions on Pd(100). Although the total incident kinetic energies are the same as in the  $\text{Ag}_7^+$  experiment, the energies per atom are lower, equal to 1.05 and 5.0 eV. In particular, the lowest energy is smaller than both the cohesive energy of bulk Pd (3.9 eV) and  $\text{Ag}_{19}$  clusters (2.6 eV),<sup>12</sup> we are thus reaching a new regime where a soft cluster impinges at low energy on a hard substrate. We will show here that this new regime has the effect of reducing considerably the role played by fragmentation, while implantation of cluster atoms and chemical disorder in the upper substrate layer cannot be neglected. An important point is that by increasing the temperature up to 300 K the adatoms appear to remain localized around the impact point, while morphology changes toward more compact structures occur.

## II. EXPERIMENTAL RESULTS AND MD SIMULATIONS

The experimental setup has already been described in detail elsewhere.<sup>13</sup> The source of mass selected clusters, of the sputtering type, is connected to a UHV molecular-beam experiment. The axis of the cluster beam is within  $15^\circ$  of the normal to the surface of the sample, whose temperature can be varied between 80 and 1300 K. The He beam is emitted by a nozzle source cooled at 77 K, and the specular reflected intensity is recorded in real time during deposition. Values of the total current and deposition rate on the He probing spot are typically 0.3 nA corresponding to  $5 \times 10^{-6}$  impacts per unit cell and second. Ultraclean deposition conditions in or-

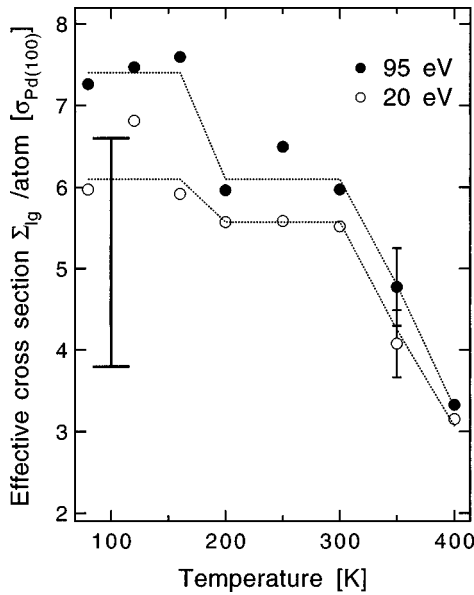


FIG. 1. Effective cross sections  $\Sigma$  per atom for deposition of  $\text{Ag}_{19}^+$  on Pd(100) as a function of substrate temperature. Lines are guides to the eye. The heavy vertical bar shows the range of possible cross sections for an unfragmented two-dimensional cluster of 19 Ag atoms. The  $\Sigma$  values have been obtained in fitting the lg model to the initial slope of  $I/I_0$  vs coverage in impact points.

der to avoid contamination are ensured by cryopumping, resulting in a low residual vacuum limit of  $2 \times 10^{-11}$  mbar.

We give in Fig. 1 the effective cross sections per atom for substrate temperatures  $T_s$  comprised between 80 and 400 K and for deposition energies equal to 20 and 95 eV. They have been obtained by analyzing the normalized He intensity  $I/I_0$  vs impact point density<sup>14</sup> with the so-called lattice-gas (lg) formula<sup>10</sup> for a maximum atomic coverage of 1% ML. Notice that the effective cross section depends on the initial slope of  $I/I_0$ , it thus corresponds to the behavior of the system when the density of clusters is very small, i.e., at most  $5 \times 10^{-4}$  cluster impacts per unit cell. In contrast to the behavior observed for  $\text{Ag}_7^+$  cluster ions,<sup>11</sup> the cross sections have qualitatively the same behavior at both deposition energies, with a more pronounced drop between  $T_s = 160$  and 200 K at 95 eV. Measurements of the cross section  $\Sigma$  of atoms deposited by thermal evaporation<sup>15</sup> indicate that  $\Sigma$  remains constant ( $\Sigma = 14.4\sigma_{\text{Pd}}$ ) up to 160 K and then decreases monotonously toward  $\Sigma = 1\sigma_{\text{Pd}}$  at 350 K where  $\sigma_{\text{Pd}}$  is the area of a Pd(100) unit cell equal to  $7.56 \text{ \AA}^2$ . By means of a diffusion and aggregation model we could show that isolated Ag atoms do not move on the surface during the measurement time (typically a hundred seconds) up to 160 K when they start to diffuse ( $E_{\text{diff}} = 0.37 \text{ eV}$ ).<sup>16</sup>

The evolution of  $I/I_0$  vs deposition time or coverage gives information on the dynamics of the processes.  $I/I_0$  vs time is shown in Fig. 2 at two different substrate temperatures. The minimal value of each curve corresponds to the stopping of the deposit, beyond which the system is thus evolving at constant coverage. On the same graph we have reported for each curve the lg behavior. The figure shows that our data closely match the lg behavior at 200 K; there are clear deviations at 300 K together with an increasing specularly reflected intensity when the deposition of clusters is stopped.

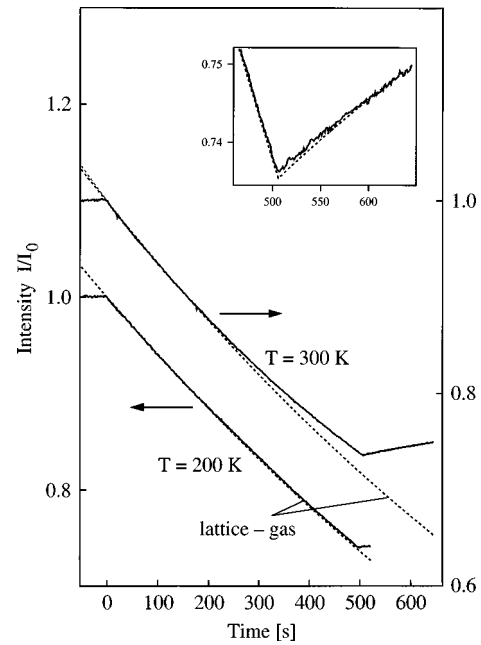


FIG. 2. He specular intensity  $I/I_0$  as a function of time, during and after deposition of  $\text{Ag}_{19}^+$ , for  $E_{\text{dep}} = 95 \text{ eV}$  and substrate temperatures equal to 200 and 300 K. The heavy dashed line corresponds to the lattice gas fit. At this scale the “two-population model” (see text) is indistinguishable from the experimental curves. The inset indicates for  $T = 300 \text{ K}$  the quality of the two-population model fit near the minimal value of the intensity curve.

We have found that  $I/I_0$  follows the lg model between 80 and 200 K. There are deviations between 200 and 350 K, with the system recovering a lg behavior at 400 K. Deviation from the lg model is evidence either for a nonrandom distribution of the particles, for instance, in the case of diffusion and coalescence of the deposited clusters, or for a modification of the local structure of the system on the experimental time scale.

Molecular-dynamics simulations are essential in order to get an insight into the collision process on an atomic scale. Several studies using effective potentials have been published<sup>17</sup> for energies ranging between less than 1 eV/atom up to 100 eV/atom or more. We have undertaken MD simulations, corresponding to our experimental conditions, using effective interaction potentials (EAM) modified as described in Ref. 18, to make them suitable to model energetic collisions. The (100) surface of Pd is simulated by slabs of different sizes, ranging from a 7-layer cell (1400 Pd atoms) to a 16-layer cell (12 800 Pd atoms), with periodic boundary conditions along the [001] and [010] directions. The impact of  $\text{Ag}_{19}$  occurs with total kinetic energies along the [100] direction equal to 20 or 95 eV, an angle of  $15^\circ$  being introduced in conformity with the experimental situation. To ensure a legitimate comparison with the experiments at 80 K a dynamical thermostat based on velocity rescaling is applied to the bottom layer.<sup>19</sup> The collision process is repeated a few hundred times starting from different initial conditions for statistical purposes. A temporal trajectory of about 10 ps proved sufficiently extended to reach thermodynamic equilibrium at  $T_s = 80 \text{ K}$ .

We give in Fig. 3 the final location of cluster and substrate atoms for typical events at 95 and 20 eV. Table I

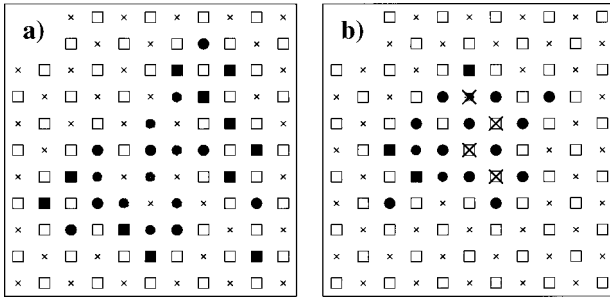


FIG. 3. Final location of cluster and substrate atoms after deposition of  $\text{Ag}_{19}^+$  on Pd(100) with an incidence  $15^\circ$  out of normal: (a)  $E_{\text{dep}}=95$  eV, (b)  $E_{\text{dep}}=20$  eV. Filled circles: Ag atoms in the adlayer; filled squares: Pd atoms in the adlayer; gray circles: Ag atoms in the first layer; white squares: Pd atoms in the first layer; white circles: Ag atoms in the second layer; large crosses: Ag atoms in the second layer above the substrate.

contains the average number of occurrences of Ag and Pd atoms in the higher layers, together with the calculated effective cross section, obtained under the hypothesis of geometrical overlap of atomic cross sections centered at each defect on the surface.<sup>20</sup> As a first observation we notice that implantation and surface alloying occur at both 20 and 95 eV, with implantation of Ag atoms in the substrate layers essentially compensated by ejection of Pd atoms in the adlayer. While chemical disorder in the upper layers is important, structural defects are scarce or absent, with only a few collisions leading to the creation of vacancies. The calculations indicate that clusters have a predominant two-dimensional character, although at 20 eV an average of four atoms occupy stable sites in the second layer above the substrate. Comparison between calculated and experimental cross sections at  $T_s=80$  K is favorable for both impact energies and especially at 95 eV. As it can be deduced from Fig. 3, the large  $\Sigma$  value at 95 eV is related to a highly noncompact structure, exhibiting adatoms only fivefold coordinated, practically absent at 20 eV. A limited extent of fragmentation can be inferred from the moderate spreading on the surface of the adsorbate, whose average width is not

TABLE I. Average distribution of Ag and Pd atoms in different layers of Pd(100), average number of vacancies, and total cross section  $\Sigma_{\text{calc}}$  obtained over 100 collision events at  $E_{\text{dep}}=20$  eV and  $E_{\text{dep}}=95$  eV. The number of vacancies refers to the first (uppermost) layer of the substrates.  $\Sigma_{\text{meas}}$  is the total experimental cross section at low temperature, measured in units of the substrate unit cell area.

	95 eV	20 eV
Ag on the adlayer	0.11	3.95
Ag in the adlayer	7.54	10.97
Ag in first layer	7.30	4.00
Ag in second and third layer	4.06	0.08
Pd on the adlayer	0.08	0.08
Pd in the adlayer	11.84	4.00
Vacancies	0.60	0.00
$\Sigma_{\text{calc}}$	$133 \pm 5$	$88 \pm 5$
$\Sigma_{\text{meas}}$	$140 \pm 10$	$115 \pm 10$

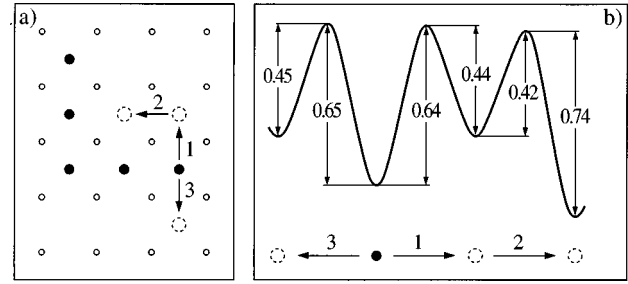


FIG. 4. EAM calculations of the static energy barriers of a Ag adatom moving along different paths in the vicinity of a four atom noncompact Ag structure on Pd(100). (a) Displacement of the adatom on Pd(100), (b) schematic representation of the energy barriers corresponding to the paths indicated in (a).

larger than  $20 \text{ \AA}$  at 95 eV. It is worth mentioning that even at the maximum coverage employed (0.04–0.06 ML) the average distance between the impact points is of the order of  $30 \text{ \AA}$ . Our sample can thus be viewed at low temperature as a collection of well-separated noncompact structures, lying on the substrate and having chemically heterogeneous character.

### III. DISCUSSION

The properties of the islands formed on a solid surface by cluster deposition depend on the temperature of the substrate. This is reflected in the thermal behavior of  $\Sigma$  shown in Fig. 1. A comparison of the  $\text{Ag}_{19}$  cross sections with those resulting from thermal deposition<sup>15</sup> reveals that the cluster cross sections at both deposition energies, and more appreciably at 95 eV, decrease right at the onset of Ag adatom mobility ( $T_s=160$  K). However, instead of dropping monotonously as in the thermal case, they reach a plateau between  $T_s=200$  K and  $T_s=300$  K before decreasing at higher temperatures. The drop recorded at  $T_s=160$  K can be attributed to free Ag and Pd atoms, which migrate towards a neighboring noncompact structure or, more generally, a more stable adsorption site. The plateau found at larger temperatures provides a strong indication that the first rapid rearrangement is followed by a dynamical evolution, whose characteristic relaxation time is long enough so that it does not modify significantly the initial slope of  $I/I_0$  vs time or coverage. This dynamics is evidenced by the non-lg behavior of  $I/I_0$  and the increasing reflected intensity once the deposition is stopped, as shown in Fig. 2. We propose that the physical origin of the plateau at intermediate temperatures and of the underlying dynamical evolution can be understood in terms of morphology changes from noncompact structures into more compact ones, which does not alter the distribution of clusters on the substrate, but rather promotes shape changes in the individual units.

This idea is supported by static EAM calculations on the energetics of adatoms in the surroundings of supported clusters.<sup>21</sup> As an example we give in Fig. 4 our results for a five silver atom noncompact structure deposited on Pd(100). The qualitative behavior is clear, migration barriers are systematically lowered with respect to the isolated adatom diffusion barrier, equal in this EAM calculation to 0.66 eV (measured value 0.37 eV), when atoms at the border of small islands migrate to increase their coordination (see, e.g., path

TABLE II. Effective cross sections  $\Sigma_A$ ,  $\Sigma_B$  and relaxation time  $\tau$  in the two-population model (see text) for a 95 eV deposition energy. The cross sections are given in units of the substrate lattice cell area. For a given temperature the first line corresponds to the fit parameters obtained in letting free all parameters, in the second line the value of the final cross section  $\Sigma_B$  is set equal to the cross section of a compact Ag<sub>19</sub> cluster.

$T$ (K)	$\Sigma_A$	$\Sigma_B$	$\tau$ (sec)
200 K	112.8	80.6	$5.8 \times 10^3$
	113.4	72.8	$4.9 \times 10^3$
250 K	124.4	72.6	$1.2 \times 10^3$
	124.4	72.8	$1.2 \times 10^3$
300 K	115.0	69.4	$5.9 \times 10^2$
	115.4	72.8	$5.2 \times 10^2$

2 and reverse path 3). The same does not hold when the atoms move away from the islands, on the contrary, the barrier height increases with respect to the free adatom case when the coordination of the atom is high (see, e.g., reverse path 2). The probability that an atom diffuses away from an island is thus significantly lower than the probability that it moves to increase its coordination number. This simple argument, which neglects entropic effects, appears to be legitimate in the intermediate temperature range we are considering here. The morphology change from a noncompact toward a compact structure thus results from the movement of atoms, which gradually increase their coordination.

A detailed description of the continuous passage from a noncompact morphology toward a range of morphologies with increasing compactness needs elaborate simulations (e.g., kinetic Monte Carlo) that are not available. In order to obtain a qualitative idea of the dynamics, let us assume that the morphology changes can be modeled by two populations of clusters, one corresponding to the initial noncompact structure (noted  $A$ ) and the other to a compact structure (noted  $B$ ). The simplest idea is to introduce an average relaxation time  $\tau$  characterizing the evolution from  $A$  to  $B$  and write first-order rate equations for the population  $n_A$  and  $n_B$  of structures  $A$  and  $B$ , where  $n_{A,B}$  represents the number of impact points per unit cell. The resulting scattering intensity vs time can then be calculated<sup>22</sup> by introducing two different cross sections  $\Sigma_A$  and  $\Sigma_B$  for structures  $A$  and  $B$ . They can be obtained, together with  $\tau$ , as fit parameters from the experimental curves. We find that between 200 and 300 K this simple model reproduces well the experimental evolution of  $I/I_0$  as a function of time, and more particularly at 95 eV where the fit is excellent. To indicate the significance of the fit parameters we give in Table II the results obtained at 95 eV in letting free all three parameters  $\Sigma_A$ ,  $\Sigma_B$ , and  $\tau$ , as well as the fit parameters obtained in setting  $\Sigma_B$  equal to the cross section ( $\Sigma_B = 72.8\sigma_{\text{Pd}}$ ) of a compact structure formed of 19 atoms (Pd and Ag atoms). As an example of the quality of the fits we give in Fig. 2 (see in particular the inset) a comparison between the experimental and fitted evolution for two substrate temperatures ( $T_s = 200$  K and  $T_s = 300$  K), a total kinetic energy of 95 eV and  $\Sigma_B = 72.8\sigma_{\text{Pd}}$ .

We find that the relaxation times obtained within this simple model have an Arrhenius-type behavior. The Arrhenius plot gives a preexponential factor comprised between 0.1

and 0.6 Hz and an activation energy of  $0.12 \pm 0.02$  eV. These values are completely different from those typical of single atomic processes ( $10^{12}$  Hz and 0.5 eV). In order to interpret them it must be remembered that the effective rate is the result of several correlated motions, which lead from noncompact to compact morphologies. It is thus doubtful that a single relaxation time describes correctly the process; the calculated  $\tau$  values represent at best an average relaxation time. Models of hierarchically constrained dynamics<sup>23</sup> have been proposed in the literature to explain glassy-type behavior. Although our physical situation is certainly different, it represents a model in which the relaxation time depends on the evolution of the system. This leads to effective relaxation times that are several orders of magnitude larger than the relaxation time of individual events. Despite its limitations, we believe that our model is useful, as it well allows quantifying the time evolution and its temperature dependence.

The effective cross section  $\Sigma_A$  remains constant up to 400 K within our two-population model, contrary to the experimental results. This implies that other mechanisms have to play a role. Further on, the measured effective cross sections become smaller than the cross section of a 19 atom compact cluster with increasing temperature, which in itself also implies that other mechanisms occur. Numerous experiments on the properties of islands formed on a surface suggest that at intermediate temperatures the atoms move around the island's edge, which leads to regular island shapes. At higher temperature, individual atoms start leaving the island and move on the substrate. This two-dimensional evaporation results in the exchange of atoms between clusters and contributes to the migration of the aggregates. These processes have been recently studied both experimentally<sup>24</sup> and theoretically,<sup>25</sup> in particular Shao, Weakliem, and Metiu<sup>25</sup> have shown for large islands that the evaporation of atoms is a first-order process with a rate constant characterized by an activation energy that includes not only the evaporation energy from an island edge but also energies needed to promote an atom on the island edge. One plausible explanation is to associate the drop in cross section beyond 300 K to the onset of two-dimensional evaporation processes. They will significantly affect the initial cross sections reported in Fig. 1 if the rate constant for evaporation corresponds to times of the order of 0.1 sec or shorter. At 300 K the corresponding energy is equal to 0.65 eV with a preexponential factor equal to  $10^{12}$  Hz; it has the right order of magnitude. Notice also that the  $\Sigma$  values characterize the system at the beginning of the deposition where the density of clusters is small and their average distance is larger than 100 Å. The evaporated atoms can thus contribute to form larger aggregates or condense on the steps, where the cross section per atom is equal to  $1\sigma_{\text{Pd}}$ .

#### IV. CONCLUSION

We have shown in this paper that the TEAS technique is well adapted to study cluster deposition on a well-characterized substrate. It is sensitive, nondestructive, and gives access to the dynamics of the deposition processes. This technique has been applied to the deposition of Ag<sub>19</sub> clusters on Pd(100) in a new regime where a soft cluster impinges at low energy on a hard substrate. MD calculations and experiments indicate that at low temperature the adatoms

(both Ag and Pd adatoms resulting from the impact) remain localized around the impact point, where they form a noncompact structure. This behavior is different from that reported for Ag<sub>7</sub> aggregates where the dominant mechanism was related to implantation and fragmentation of the cluster. We propose that between 200 and 300 K morphological changes take place, leading from a noncompact toward a compact structure. At higher temperature, the decrease of the

effective cross section below that of a compact Ag<sub>19</sub> cluster indicates that other phenomena take place; we suggest that they correspond to the onset of two-dimensional evaporation.

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