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Diffusion studies in a nonequilibrium system with repulsive interactions

Gao-Xiang Ye,[1,2] Thomas Michely,[1] and Matthias Wuttig[1]
[1]*I. Physikalisches Institut, RWTH Aachen, D-52056 Aachen, Germany*
[2]*Department of Physics, Zhejiang University, Hangzhou 310027, People's Republic of China*

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The diffusion behavior of silver islands on charged silicone oil surfaces has been studied. Compact Ag islands with an average diameter of 1.2 μm form after thermal deposition of silver on a charged silicone oil surface. In contrast to the behavior of the uncharged oil surface, no aggregation of the Ag islands is observed. Instead the average distance of the islands increases as the islands diffuse toward the edge of the oil substrate. This process is driven by the repulsive Coulomb interaction. An exponential decay of the island density n with time t results everywhere on the oil surface, and the corresponding time constant O_f ranges between 1.0 $\times 10^{-4}$ and 2.0×10^{-4} s^{-1}. In the central area of the substrate, the relative speed V between two islands increases linearly with their distance L according to $V_{fit}=HL$, where V_{fit} is the linear fit speed of V. The slope H is of the order of 2.0×10^{-4}–3.0×10^{-4} s^{-1} after the deposition, and decreases with time. Our theoretical analysis indicates that $O_f\approx H$, in agreement with the experimental finding. The experiment also shows that this nonequilibrium expansive diffusion already starts at the early stage of deposition. During their motion, the islands must carry a small fraction of the electron charge on the oil surface. After the islands reach the sample edge, they form a band-shaped silver film along the border. This results in the accumulation of charge, which slows down the speed of the remaining islands, which diffuse toward the edge.

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I. INTRODUCTION

The study of the diffusion and aggregation of particles, including atoms and atomic clusters, on various substrates has been of great interest in the last two decades, since it is crucial for the microstructure of various atomic aggregates and thin films.[1–4] During the growth period, the adatoms or clusters diffuse and aggregate on the substrate surface, and then form different kinds of aggregates such as compact clusters, atomic wires, cluster rings, branched islands, etc.[5–8] If more material is deposited on the substrate, the aggregates grow and percolation proceeds. Finally, continuous films form. It is generally supposed that this evolution process is mainly governed by the Brownian motion of the particles, the attractive interaction among the particles, as well as the interaction between the particles and the substrate.

Much of the recent interest in this field has focused on the nature of the particle-particle as well as particle-substrate interactions. Most computer simulations assume an attractive interaction between adjacent neighboring particles. They suppose that if two particles occupy neighboring sites, the attractive force will lead to an irreversible attachment.[9–13] Recent experiments revealed that particle-particle interactions can be considerably more complex. In a quantitative study of long-range interaction between single Cu adatoms on Cu(111), Repp *et al.*[14] showed that the long-range interaction is oscillatory with a period of half the Fermi wavelength, and decays for larger distances d as $1/d^2$. Such an oscillatory interaction significantly changes the growth of Cu/Cu(111) at very low temperatures, and could be employed to achieve self-organization of cluster arrays on surfaces.

On the other hand, many naturally occurring colloidal particles in solution are charged. The resulting electrostatic repulsions are strong enough to stabilize their suspensions against flocculating. However, sometimes the counterintuitive observation is made that the particles attract each other and form ramified aggregates.[15]

Here we follow a different approach to study the behavior of charged particles. We present results on the nonequilibrium diffusion of silver nanocrystal aggregates with repulsive Coulomb interactions on silicone oil surfaces. The oil substrate was charged by the emission current of a filament before deposition. Then silver of 99.99% purity was deposited by thermal evaporation. Upon deposition, Ag nanocrystals form at the interface which are mobile and aggregate to form nanocrystal aggregates of lateral dimension of μm size visible in the optical microscope.[8] In contrast to experiments on an uncharged surface, these nanocrystal aggregates do not aggregate further to large ramified aggregate structures. Instead, the μm-sized Ag nanocrystal aggregates—henceforth named ‘‘islands’’—move away from each other and diffuse toward the sample edge without further aggregation due to their repulsive interaction. This leads to the formation of a band-shaped silver film along the edge of the substrate.

II. EXPERIMENT

In order to observe and investigate the behavior of Ag nanocrystal aggregates on an oil surface *in situ*, a special high vacuum system with a dedicated observation window was designed and built (shown in Fig. 1). Commercial silicone oil (DOW CORNING 705 Diffusion Pump Fluid) with a vapor pressure below 10^{-10} mbar was carefully painted onto a polycarbonate slide surface which was slightly roughened to prevent the oil layer from dewetting the smooth plastic surface. The resulting oil substrate with a radius R_0 between 1.5 and 3.0 mm had a uniform thickness of ≈ 0.2 mm. The plastic slide was glued to an aluminum alloy disk, which was fixed on the vacuum chamber (see Fig. 1).

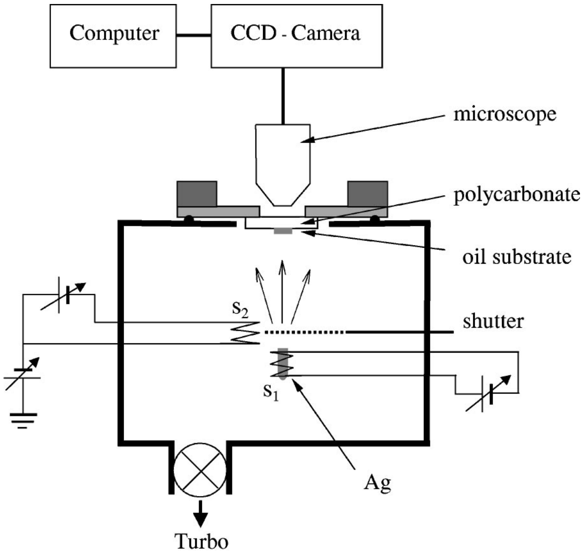

FIG. 1. Schematic setup of the experiment.

The insulating oil substrate was charged before deposition by a thermal emission current in a vacuum of 3×10^{-6} mbar. The emission filament was made of a tungsten wire with a diameter of 0.2 mm. The distance between the emission filament and the substrate was 100 mm (see Fig. 1). During the charging period, the filament was kept at a negative potential of -100 V with respect to ground and the heating current in the filament was about 4.75 A, resulting in a total emission current of 1 mA. The charging period was 480 s. We did not find any change in the experiment if the charging time was further increased. Therefore, we believe that the electrical potential on the oil surface should be approximately -100 V after the charging period of 480 s, and no further electrons could reach the oil surface afterwards.

Silver of 99.99% purity was deposited 1800 s after the charging period by thermal evaporation at room temperature. The deposition rate was determined to $f = 2.5 \times 10^{-3}$ nm/s by a quartz-crystal balance. The deposition time for all the samples is 480 s unless noted otherwise. All images for the surface morphologies of the samples were taken *in situ* through the observation window with an optical microscope, equipped with a CCD camera which was interfaced to a computer for data storage and data processing (see Fig. 1). The whole equipment is mounted on a vibration isolation table.

III. RESULTS

In order to compare the processes prevalent for charged and uncharged substrates, silver deposition on an uncharged oil substrate was performed first. Figure 2 shows the time dependence of the Ag island density n on an uncharged oil surface. The experiment shows that compact islands become clearly visible after a deposition period of 300 s, i.e., $t' \approx 300$ s. They grow and aggregate on the oil surface during deposition (see the insets in Fig. 2). As the time t' proceeds, the surface coverage increases and the Ag islands merge quickly. After about 1200 s percolation proceeds via the formation of interconnections between branched islands.

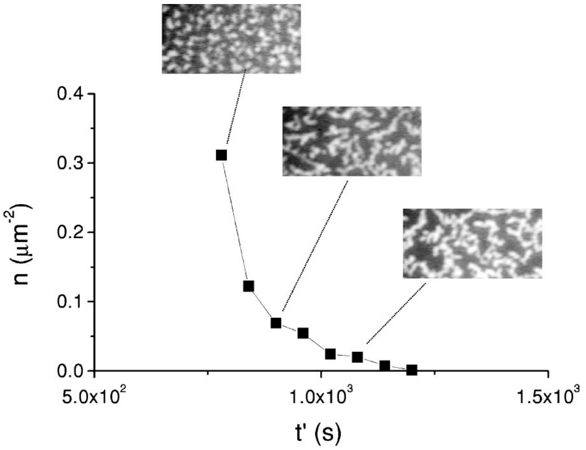

FIG. 2. Island density n vs deposition time t' on an uncharged oil surface. The three insets (22×11 μm^2) give examples of the three images from which the island densities are determined. Solid lines between data points are a guide to the eye.

If the deposition was stopped after $t' = 480$ s, the compact islands, as shown in Fig. 3(a), diffuse and aggregate on the oil surface with time t available for the diffusion. About 7200 s after deposition, i.e., $t \approx 7200$ s, branched aggregates could be observed [see Fig. 3(b)], in agreement with previous studies.[16,17]

For the silver atoms deposited on a charged oil surface, however, the diffusion and aggregation behaviors are quite

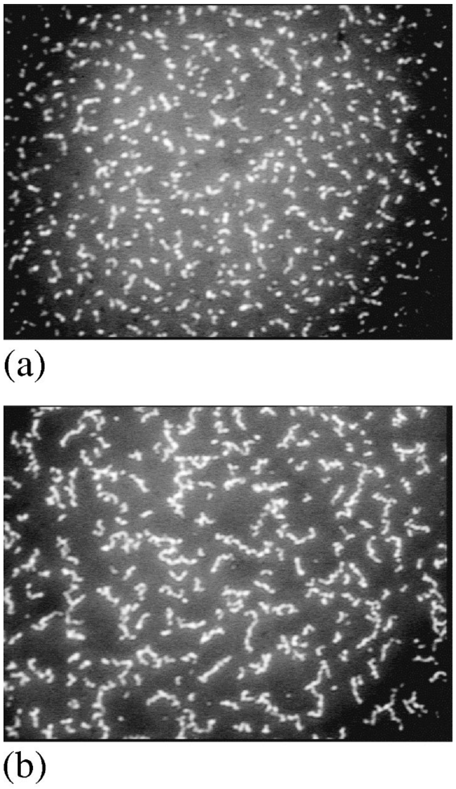

FIG. 3. Morphologies of the Ag islands on an uncharged oil surface. Ag islands aggregate on the uncharged oil surface. Image size 84×64 μm^2. (a) $t = 300$ s. (b) $t = 7200$ s.

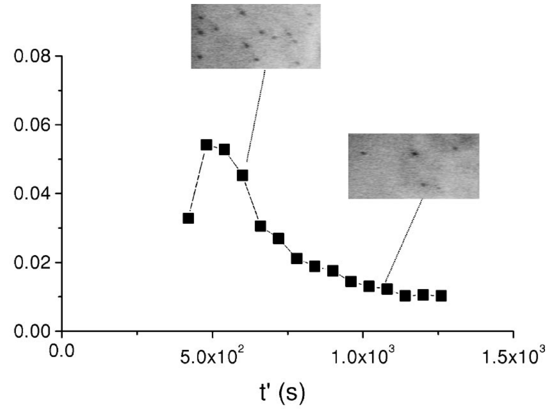

FIG. 4. Island density n vs deposition time t' on a charged oil surface. No aggregation phenomenon is observed. The two insets (22×11 μm^2) give examples of the two images from which the island densities are measured. Solid lines between data points are a guide to the eye.

different. The most interesting result is shown in Fig. 4. In contrast to the behavior observed on the uncharged oil surface, no island aggregation is observed on the charged oil surface (here we assume that the formation mechanism and the crystal structure of the compact Ag aggregates on the charged oil surfaces are similar to those of the compact Ag islands on uncharged oil surfaces,[8] see the explanation in Sec. IV C below). Rather we observe that the size of the islands is almost independent of deposition time once the islands are visible after a deposition period of 240 s (see the black dots in the insets in Fig. 4). With increasing deposition time the island density first increases and then decreases steadily. Hence the total amount of Ag on the oil surface decreases with increasing deposition time. This puzzling observation immediately raises a question where the additional amount of Ag disappears. Two possibilities have been considered: Ag could either diffuse into the oil substrate or diffuse toward the edge of the oil substrate. To clarify this situation, the deposition process was stopped after $t'=480$ s, and the subsequent diffusion of the Ag islands was studied. This experiment shows that the island density n decreases with the diffusion time t, even though no aggregation events among the compact islands are observed during this evolution process, as shown in Fig. 5. Furthermore, all measurements show that, to a good approximation, n decays exponentially with t everywhere on the oil surface (see Fig. 6), which can be expressed as

$$n=n_0 e^{-O_f t}, \tag{1}$$

where n_0 is the island density at $t=0$. The measured time constant O_f, i.e., the slopes of the fit lines in Fig. 6, ranges between 1.0×10^{-4} and 2.0×10^{-4} s^{-1}.

To unravel the puzzle described above, two images for the sample edge were taken and are shown in Fig. 7. We find that the compact Ag islands formed on the charged oil surface move toward the sample edge [Fig. 7(a)]. About 7200 s after the deposition, most of the islands have reached the edge and contribute to the formation of a band-shaped silver film along the border of the oil substrate [Fig. 7(b)]. This motion toward the sample boundary already qualitatively explains the phenomena shown in Figs. 4–6.

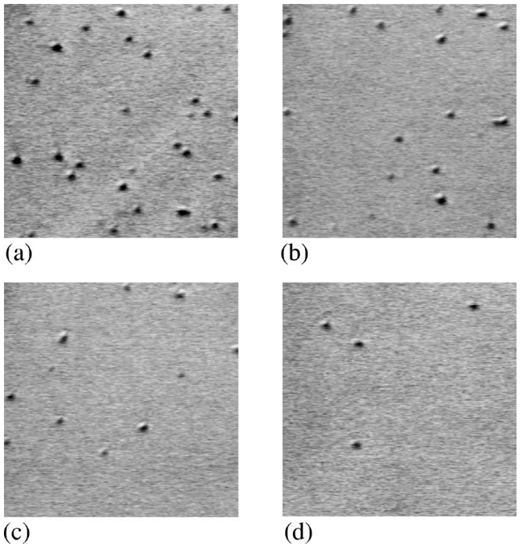
(a) (b) (c) (d)

FIG. 5. Morphologies of the Ag islands on a charged oil surface. The island density decays exponentially with time. The image sizes 30×30 μm^2 (middle area of the oil substrate). (a) $t=600$ s. (b) $t=2400$ s. (c) $t=4200$ s. (d) $t=6000$ s.

In order to describe the expansive diffusion behavior quantitatively, we measured the relative speed V between two islands with their relative distance L at different diffusion times t for the islands in the central area of the oil substrate. The corresponding results are shown in Fig. 8. The

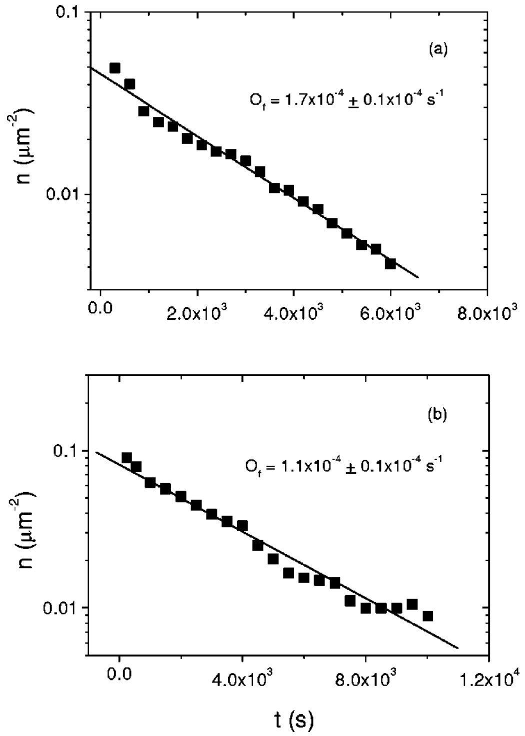

FIG. 6. Time dependence of the island density. The island density decays exponentially with time everywhere. (a) In the central area. (b) Near the edge of the oil substrate.

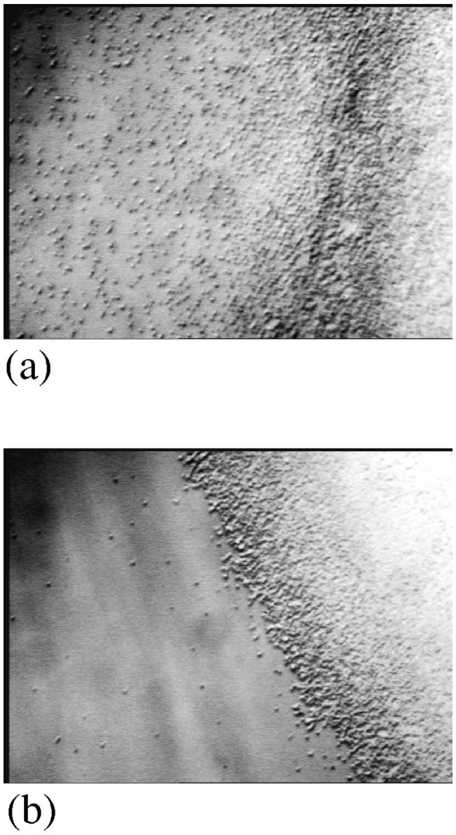

FIG. 7. Morphologies of the sample edge at different times. Ag islands on a charged oil surface diffuse toward the edge. The image size is 84×64 μm^2. (a) $t=300$ s. (b) $t=7200$ s.

speed V is defined as $\Delta L/\Delta t$, where ΔL is the increment of the distance between two islands during the time interval Δt. During the measurement, our optical microscope focused on and followed a group of the islands in the central area of the substrate and took two images at time t and $t+\Delta t$, respectively. Then we measured the increment of the distance between two islands from the images and calculated the speed V. At least 34 pairs of the islands were measured from each of the images, as shown in Figs. 8(a), 8(b) and 8(c). It can be seen in Fig. 8 that the linear fit speed V_{fit} increases linearly with L, i.e.,

$$V_{fit}=HL. \tag{2}$$

The linear fit slope H is $2.9\times10^{-4}\pm0.5\times10^{-4}$ s^{-1} after deposition and decreases with time t. About 7200 s after the deposition, H reaches nearly zero [see Fig. 8(c)]. The linear fits in Fig. 8, in a good approximation, go through the origin of the coordinates. These results indicate that, besides their Brownian motion, the islands perform a drift motion toward the edge and move away from each other.

In our experiments, the measurement for the expansive motion could only be performed after visible islands appear on our computer screen. However, it can be shown that the expansive diffusion starts already at the early stage of deposition. Figure 9 displays that the band-shaped Ag film starts to grow before the visible islands appear. This indicates that, as soon as the Ag nanocrystals and nanocrystal aggregates of different sizes form on the oil surface, they immediately start to move toward the sample edge. As a consequence the Ag band at the border of the oil substrate already grows before we can see discernible Ag islands on the oil surface. This leads to the conclusion that Ag islands which are below the diffraction limit of the optical microscope already diffuse toward the edge. Subsequently the band-shaped Ag film grows gradually along the border of the oil substrate during and after the deposition. In Fig. 9, from $t'=180$ to 210 s, i.e., from Fig. 9(d) to Fig. 9(e), there is a sudden shrinkage of the white band (i.e., the silver film) width, which we believe is an agglomeration or coalescence process of the silver islands and eventually would lead to a continuous silver film. After the shrinkage, the width of the band-shaped film decreases slowly but continuously. This relaxation process can be seen from Fig. 9(e) to Fig. 9(g).

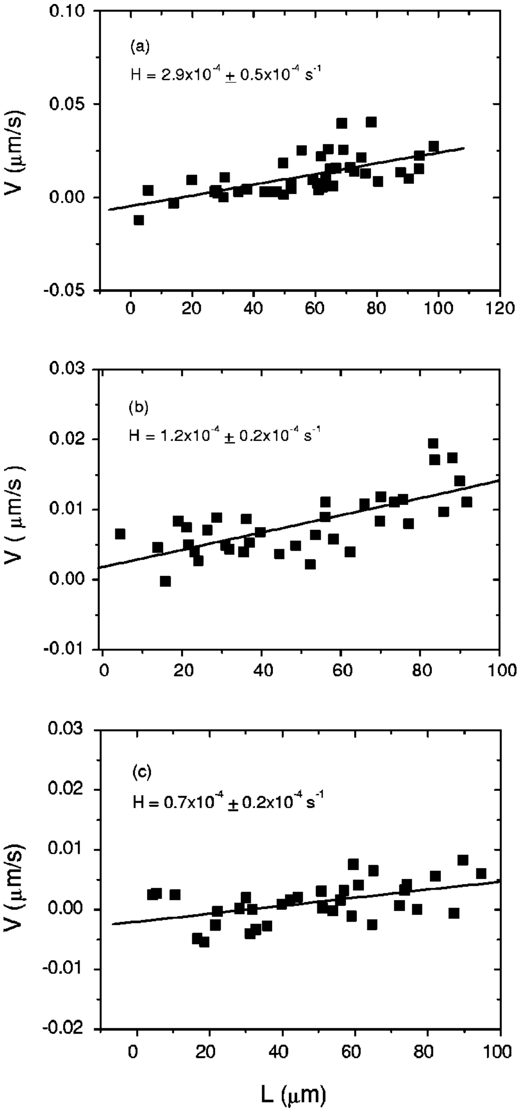

FIG. 8. Dependence between the relative speed V and distance L between the Ag compact islands in the central area of the substrate. The slope H decreases with time t. (a) $t=120$ s, $\Delta t=150$ s. (b) $t=1800$ s, $\Delta t=500$ s. (c) $t=7200$ s, $\Delta t=1000$ s .

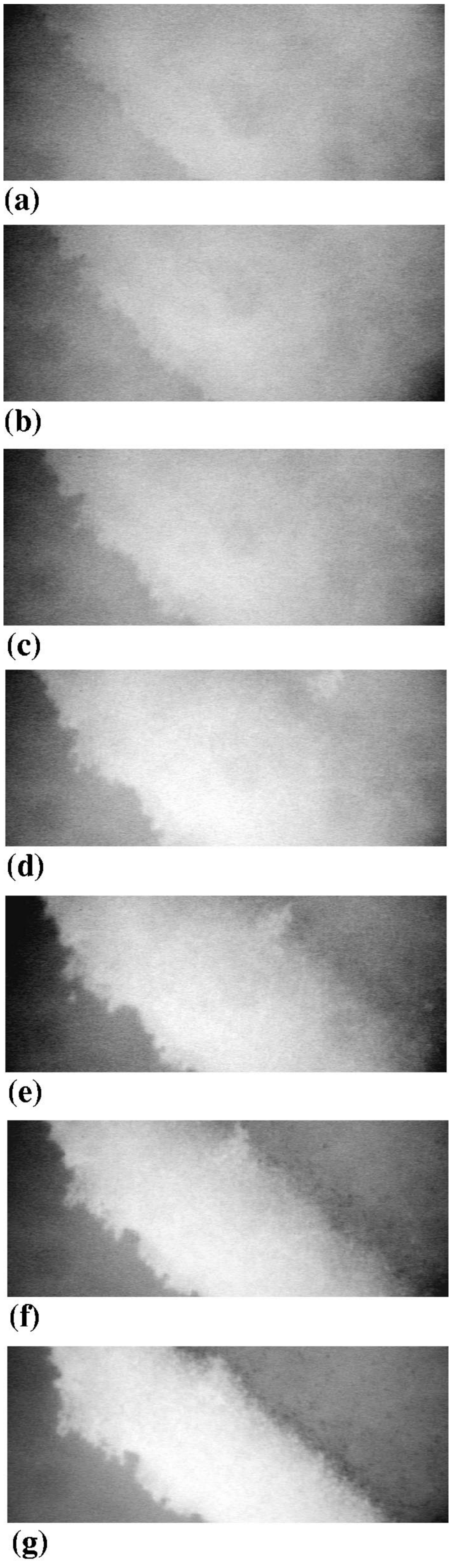

FIG. 9. Morphologies of the band-shaped silver film along the sample edge. The islands come from the top right corner and diffuse towards the sample edge at the bottom left side. The image size $84\times 33\ \mu\mathrm{m}^2$. (a) $t'=90$ s. (b) $t'=120$ s. (c) $t'=150$ s. (d) $t'=180$ s. (e) $t'=210$ s. (f) $t'=240$ s. (g) $t'=270$ s.

IV. DISCUSSION

A. Exponential decay law and the speed of expansive motion

As described above, the exponential decay of the island number density n with time t results from the expansive motion of the islands toward the sample edge. Therefore, a correlation between the exponential decay slope and the inhomogeneous diffusion speed distribution is expected. In our experiment, we found that all the islands diffuse radially and toward the round edge of the substrate. Therefore, let us now focus on an island C located in the central area of the oil substrate and assume approximately that the drift motion of the other islands with respect to the island C is radial symmetry, as shown in Fig. 10. The increment of the island number in the shadowed area is given by

$$r\,d\theta\,dr\,dn = n[v(r) - v(r+dr)]r\,d\theta\,dt, \tag{3}$$

where the first and second terms on the right side are the island numbers moving in and out of the shadowed area, respectively, and v is the island speed with respect to the oil. It should be noted that in Eq. (3) only the diffusion in radial direction is taken into account since we assume radial symmetry. Equation (3) can be simplified as[18]

$$\frac{dn}{dt} = -n\frac{dv}{dr}. \tag{4}$$

From Fig. 8 and Eq. (2) we have determined the dependence of V_{fit} upon distance L, which is $V_{fit}=HL$. Hence $dv/dr = H$. Therefore, Eq. (4) can be written as

$$\frac{dn}{dt} = -Hn. \tag{5}$$

To the first order of approximation, we can assume that H is time independent. Then integrating Eq. (5) yields

$$n = n_0 e^{-Ht}, \tag{6}$$

where n_0 is the island density at $t=0$. Comparing Eqs. (1) and (6), we have

$$H = O_f. \tag{7}$$

From Figs. 6 and 8, we can see that the measurements for H and O_f agree well. This indicates that Eq. (5) is a good approximation for describing the phenomenon at the early diffusion stage in Figs. 5–9. Therefore, we conclude that it is the radial drift motion of the islands that results in the exponential decay law.

B. Interaction between the islands and the oil substrate

In a previous study,[16,17] we reported on the Brownian motion of Ag islands on silicone oil surfaces due to statistical fluctuations in their momentum by the collisions with the oil molecules. This random interaction still exists in the present system. However, since these random collisions average out and do not affect the radial drift motion, it can be omitted in the following discussion.

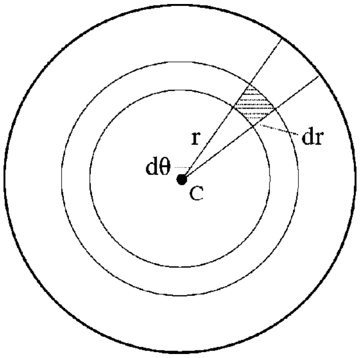

FIG. 10. Sketch of the expansive diffusion of the islands on the charged oil surface.

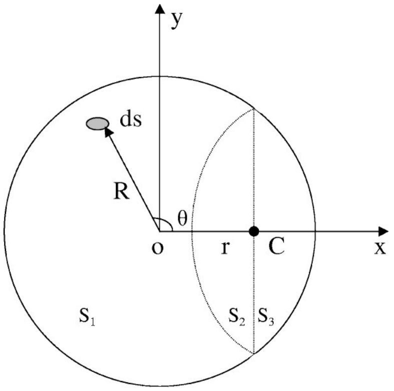

FIG. 11. Sketch of the interaction between a Ag island and the charged oil surface.

Based on the experimental observations, we propose that the expansive drift motion of the Ag islands described in Figs. 5–9 results from the charged oil substrate. The charge on the oil surface also gives raise to a repulsive Coulomb force between the islands, which drives the islands away from each other.

Suppose that the charge density σ on the oil surface, including the charges on both the oil molecules and the Ag islands, is homogeneous. Let us consider the electric field component $\mathbf{E}(\mathbf{r})$ parallel to the oil surface, as shown in Fig. 11. According to electromagnetic theory,[19] the electric field $E(r)$ at the location of island C is given by

$$E(r)=\frac{\sigma}{4\pi\varepsilon\varepsilon_0}\int\int_S\frac{[r-R\cos(\theta)]RdRd\theta}{[R^2+r^2-2rR\cos(\theta)]^{3/2}}, \qquad (8)$$

where ε_0 is the dielectric constant in free space, ε is the dielectric constant on the oil surface and r is the distance between the island C and the center O of the oil substrate. The integration in Eq. (8) extends over the whole oil surface $S=S_1+S_2+S_3$. For the sake of simplicity, here the contribution from the charges on the polycarbonate slide surface is not taken into account. From symmetry analysis, we know that the total contribution to the electric field from the charges in area S_2 and S_3 is zero (see Fig. 11). Therefore, the integration in Eq. (8) extends over S_1 only. Since $\sigma<0$, the vector $\mathbf{E}(\mathbf{r})$ points to the center of the oil substrate.

If the silver island C does not carry charges, then the electrical response of the island can be considered as an induced dipole. The interaction between the dipole and the field $\mathbf{E}(\mathbf{r})$ results in an attractive force,[19] which would pull the island to diffuse toward the oil center. This conclusion, however, is contrary to our experimental observation.

In order to explain the experimental results consistently, we have to assume that the Ag islands must carry some negative charge (electrons) during their diffusion and the interaction between the electric field $\mathbf{E}(\mathbf{r})$ and the charge is the main force which results in the expansive diffusion. Suppose each island carries a charge q. The interaction between island C and the electric field $\mathbf{E}(\mathbf{r})$ is then given by

$$\mathbf{F}=q\mathbf{E}(\mathbf{r}). \qquad (9)$$

Since both q and σ are negative, $\mathbf{F}$ is a repulsive force, resulting in the expansive diffusion of the islands toward the sample edge. Also, the negative charge q that each island carries will keep the islands diffusing away from each other. Therefore, they will not aggregate during their diffusion towards the sample edge.

As described above, the electric field $\mathbf{E}$ is a function of r. In the extreme case of $r=0$, i.e., in the center of the oil substrate, $\mathbf{F}$ must be zero due to the symmetry constraints. The electric field acting on the islands is clearly location dependent [see Eq. (8)]. This is the main reason that leads to the inhomogeneous speed distribution of the islands in Fig. 8. In another extreme case, i.e., $r=R_0$, Eqs. (8) and (9) show that $\mathbf{F}$ reaches its maximum value if the charge density σ is constant everywhere on the oil surface.

If the description above is correct and the islands do carry charge during their diffusion, then, after the islands reach the sample edge, a charge accumulation near the edge is expected. Figure 12 shows that the speed v of the islands, located at distance l from the edge, is slowed down and the accumulated charges at the edge impede the attachment of islands near the edge. It appears from Fig. 12 that there is approximately a region of 50 μm width close to the edge where the speed v is considerably reduced. This is indicative for an interaction between the islands and the edge (see Fig. 12), and, therefore, the charge density σ should be location dependent at the sample edge.

Now let us focus on the diffusion behavior of the islands near the sample edge and located between $l=40$ and 60 μm, where the average drift speed of the islands with respect to the edge (or to the oil), i.e., v, does not change with l sensitively (see Fig. 12). Figure 13 shows that, after the deposition, the speed v is reduced by the charge accumulated on the sample edge. This indicates that the charge on the sample edge increases during the first 1800 s because of the fast accumulation of charged islands in this period [see Fig. 7(a)]. About 1800 s later, v approaches a stable value and becomes nearly time independent (see Fig. 13). This suggests that the speed of the charge accumulation near the edge is very slow for the time $t>1800$ s, i.e. the charge q that each island carries should be a small quantity since at that time the island density near the edge is still not very

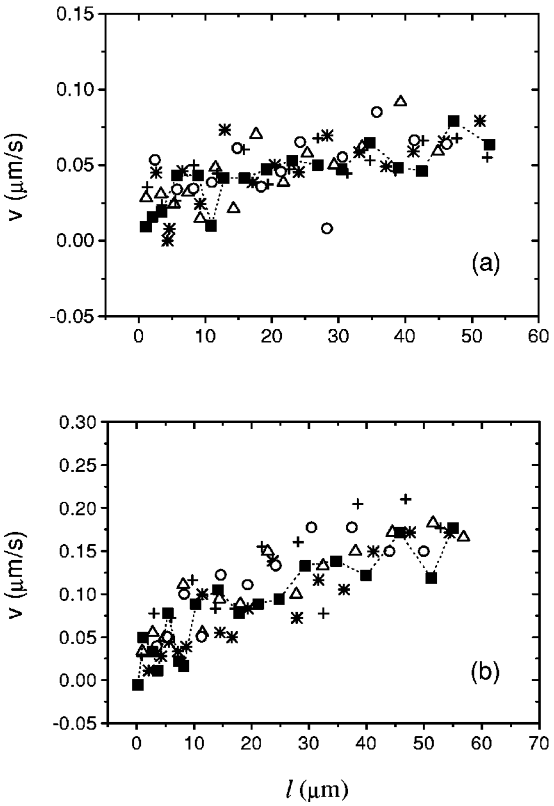

FIG. 12. Location dependence of the speed v. The speed of the Ag islands is slowed down near the edge. Different signs represent the data for different islands measured. Dashed lines between the data points for two islands are a guide to the eye. (a) $t=1800$ s, $\Delta t=75$ s. (b) $t=9000$ s, $\Delta t=40$ s.

small (see Fig. 6). We can conclude from Fig. 13 that most of the charges on the sample edge are accumulated at the early stage of the diffusion period.

C. Charge on the Ag islands

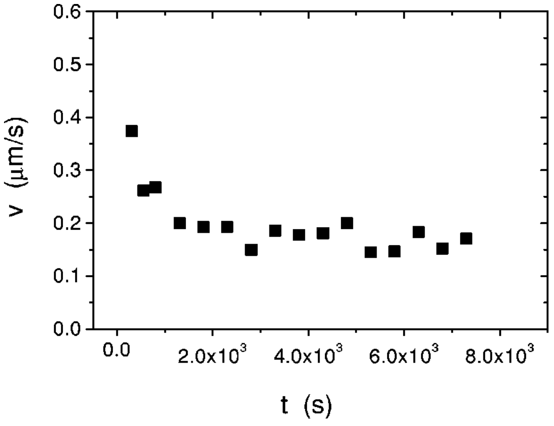

FIG. 13. Island speed v vs time t. The speed of the islands near the edge, i.e., in the area between $l=40$ to 60 μm, decreases first and then reaches a stable value. Each data point represents an average value of over 17 islands. $\Delta t=10$ s.

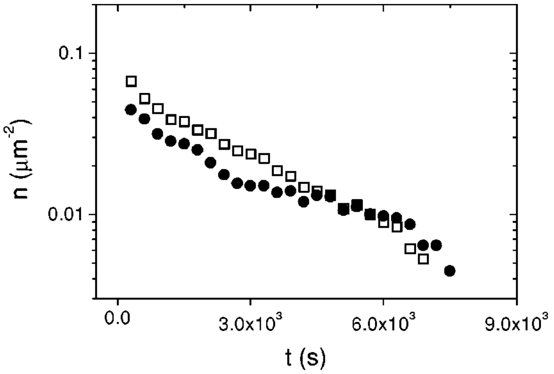

FIG. 14. Time dependence of the island density after two subsequent depositions. Three hours after the first deposition, almost all the islands arrive at the edge and n reaches almost zero. Then the second deposition on the original oil substrate starts. Open squares: data measured after the first deposition; Solid dots: data measured after the second deposition.

Figure 14 shows the time dependence of the island densities after two subsequent depositions. The open squares represent the data measured after the first deposition, which show the expected exponential drop of the island density n with time t. Three hours later, almost all the islands arrive at the edge and n reaches approximately zero (see Figs. 5–7). Subsequently, a second deposition on the original oil substrate with similar deposition conditions was performed. The dots in Fig. 14 denote the data measured after the second deposition. Again an exponential decay of island density with time is observed. This implies that even after the second deposition, there is still enough charge on the oil surface for the second expansive diffusion. This result suggests that, during their diffusion, the silver islands carry only a small part of the charges on the oil surface, which is in agreement with the conclusion from Fig. 13 (see the discussion above). On the other hand, it is also possible that the charge distribution on the oil surface may rearrange after enough islands reach the sample edge because of the repulsive interaction among the electrons accumulated at the edge.

As we have shown in this paper, on a charged oil surface Ag islands do not aggregate due to Coulomb repulsion. As the charge on the Ag islands stops their aggregation, one may

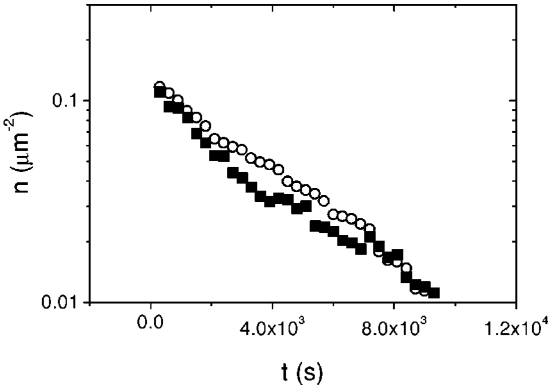

FIG. 15. Island density n vs time t. The decay behavior of the two samples is similar. Solid squares: $R_0=1.5$ mm; Open circles: $R_0=3.0$ mm.

ask why the charge did not stop the formation of Ag islands themselves by Coulomb repulsion. Indeed, a rough estimate shows that already two singly charged Ag atoms are unlikely to form a dimer due to a repulsive Coulomb barrier, which amounts to several eV, and exceeds their thermal energy by far. One speculation that would allow to understand island formation in the presence of charge relies on the size dependence of the electron affinity for Ag clusters. While for an Ag atom an energy gain of 1.3 eV results by adding an extra electron, for a large Ag cluster the energy gain will be the average work function of about 4.6 eV.[20] If the oil molecule electron affinity would be somewhere between these two numbers Ag atoms and small cluster of Ag atoms would stay uncharged, as their electron affinity is below the one of the oil molecules, but larger Ag clusters or Ag nanocrystals would charge up gradually as their electron affinity increases beyond the one of the oil molecules. The nucleation and growth of the Ag nanocrystals would then hardly be influenced by the presence of the charge on the oil surface, but the charging of the Ag nanocrystals would eventually limit their aggregation, thus limiting the size of the observable islands and their aggregation. To clarify the situation and to confirm or reject the speculations above further experiments related to the initial growth and nucleation stage are necessary with a higher resolution microscopic technique.

D. Substrate size effect

In our experiment, the thickness of the oil layer may vary from 0.1 to 0.3 mm and the radius of the oil substrates may change from $R_0 = 1.5$ to 3.0 mm. No obvious difference of the island density $n(t)$ can be observed within these size ranges. Figure 15 shows the decay behaviors of two different samples with $R_0 = 1.5$ and 3.0 mm, respectively. One finds that the behaviors are quite similar. According to Eq. (8), however, the electric force acting on the islands located in the central area of the oil substrate approaches zero if $R_0 \to \infty$. Therefore, we believe that, for very large substrate sizes, the islands near the edge would be accelerated first and diffuse toward the substrate edge immediately after they form and the expansive diffusion of the islands in the central area of the substrate would be delayed and weakened. The nonuniform speed distributions described in Fig. 8 support this assumption. However, to see the substrate size effect quantitatively, more precise experiments are needed.

V. CONCLUSION

In summary, we have described the drift behavior of Ag islands on charged oil surfaces. Compact Ag islands form during the thermal deposition of silver on the charged liquid substrate. They move away from each other and drift toward the sample edge with an inhomogeneous speed distribution driven by the repulsive Coulomb force. No aggregation phenomenon among the islands is observed during the whole diffusion period. We show that the Ag islands must carry a small part of the charge but not all the charge on the oil surface. When an island reaches the sample edge, both the silver atoms and electrons accumulate on the edge, which will slow down the speed of the following islands diffusing toward the edge. Finally, a band-shaped Ag film forms along the edge of the liquid substrate.

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[18] A similar analysis shows that Eq. (4) is also true for three-dimensional expansive systems. Therefore, we believe the method described in this section can also be used for other analogous systems.

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