Stokes Formula and Density Perturbances for Driven Tracer Diffusion in an Adsorbed Monolayer

O. Bénichou,[1] A. M. Cazabat,[2] J. De Coninck,[3] M. Moreau,[1] and G. Oshanin[1]

[1]*Laboratoire de Physique Théorique des Liquides (CNRS-UMR 7600), Université Pierre et Marie Curie, 4 place Jussieu, 75252 Paris Cedex 05, France*

[2]*Laboratoire de Physique de la Matière Condensée, Collège de France, 11 place M. Berthelot, 75231 Paris Cedex 05, France*

[3]*Centre de Recherche en Modélisation Moléculaire, Université de Mons-Hainaut, 20 Place du Parc, 7000 Mons, Belgium*

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We study the intrinsic friction of monolayers adsorbed on solid surfaces from a gas phase or vapor. Within the framework of the Langmuir model of delocalized adsorption, we calculate the resistance offered by the mobile adsorbate's particles to some impure tracer molecule, whose diffusive random motion is biased by a constant external force. We find that for sufficiently small driving forces the force exerted on the tracer shows viscouslike behavior. We derive then the analog of the Stokes formula for two-dimensional adsorbates, calculate the corresponding friction coefficient, and determine the stationary particle distribution in the monolayer as seen from the driven impurity.

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When an ambient gas phase or vapor is brought in contact with a solid, some portion of it becomes reversibly attached to the solid surface in the form of an adsorbed layer. Such layers are important for various technological and material processing operations, including, for instance, coating, gluing, or lubrication.

Following the work of Langmuir, equilibrium properties of the adsorbates have been extensively studied and a number of significant developments have been made [1]. As well, some approximate results have been obtained for both dynamics of an isolated adatom on a corrugated surface and collective diffusion, describing spreading of the macroscopic density fluctuations in interacting adsorbates being in contact with the vapor [2,3].

Another important aspect of dynamical behavior concerns tracer diffusion in adsorbates, which is observed experimentally in scanning tunneling microscopy or field ion measurements and provides a useful information about adsorbate's intrinsic friction. In this regard, analysis of tracer diffusion is not only a challenging question in its own right but is also crucial for the understanding of various dynamical processes taking place on solid surfaces. To name but a few, we mention spreading of molecular films on solid surfaces [4], spontaneous or forced dewetting of monolayers [5], or island formation [6]. However, apart from a slightly artificial 1D model [7], available studies of tracer diffusion in adsorbed layers focus on strictly two-dimensional (2D) situations (see, e.g., Refs. [8–10]), excluding the possibility of particle exchanges with the vapor.

In this Letter we present first results on intrinsic friction in 2D adsorbed monolayers composed of mobile particles undergoing continuous exchanges with the vapor phase. The system we consider here corresponds to the generalized Langmuir model of adsorption with adsorbate lateral diffusion [1] and consists of three key ingredients: (i) a solid surface containing some concentration of adsorption sites, (ii) a vapor phase, and (iii) a monolayer of adsorbed hard-core particles, which perform activated random motion between the adsorption sites subject to the hard-core exclusion interactions (one particle per adsorption site at most), and undergo continuous exchanges (desorption/adsorption) with the vapor phase (Fig. 1).

To determine the intrinsic friction of such an adsorbate we wish to probe the resistance offered by the monolayer particles to some external perturbance. To this purpose, we add an extra hard-core particle—a tracer, which may move along the surface only and cannot desorb. Next, we suppose that the tracer is subject to an external force F, which favors its motion in the preferential direction such that it ultimately attains a constant velocity V. Our aim is then to evaluate the velocity-force relation $V(F)$ in the limit of small F and to determine the stationary particle distribution in the perturbed monolayer. General results for arbitrary F will be presented elsewhere.

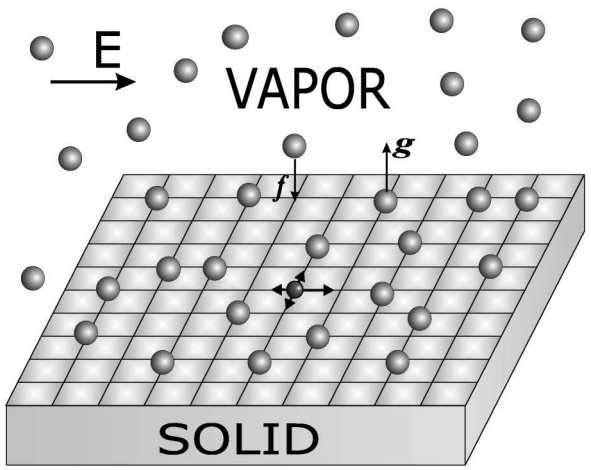

FIG. 1. Adsorbed monolayer in contact with a vapor. Grey spheres denote the monolayer (vapor) particles; the smaller black sphere stands for the driven tracer particle.

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More precisely, our model is defined as follows (Fig. 1): We suppose first that the adsorption sites form a regular 2D square lattice of spacing σ. Occupation of the lattice sites is characterized by the set of time-dependent variables $\eta_{\vec{r}}$, $\vec{r}$ being the lattice vector of the site with Cartesian coordinates (x, y); $\eta_{\vec{r}}$ equals 1 if the site $\vec{r}$ is occupied by an adsorbed particle and zero otherwise.

Next, we suppose that the particles from the vapor, (considered as a reservoir, which is maintained at a constant pressure), may adsorb onto any vacant lattice site at a fixed rate f/τ^*. Then, the adsorbed particles may move randomly by hopping with a rate $l/4\tau^*$ to any of four neighboring lattice sites, which process is constrained by hard-core exclusion preventing multiple occupancy of any site, or desorb from the lattice at a site- and environment-independent rate g/τ^*. For simplicity of exposition, we assumed here that typical adsorption, desorption, and jump times are equal to each other; these times, i.e., τ_{ad}, τ_{des}, and τ_{jump}, respectively, can be readily restored in our final results by the mere replacement $f \to \tau^* f/\tau_{\mathrm{ad}}$, $g \to \tau^* g/\tau_{\mathrm{des}}$, and $l \to l\tau^*/\tau_{\mathrm{jump}}$.

The tracer particle is initially put at the lattice origin. We stipulate that only this particle is not allowed to desorb and in addition to random thermal forces experiences an action of a uniform external force $\vec{F} = (F, 0)$. Dynamics of the tracer is then defined via standard rules [11]: the tracer, which is at the site $\vec{R}$, $\vec{R} = (X, Y)$, at time t, waits an exponentially distributed time with mean τ (which is in the general case different from τ^*), and then attempts to hop onto one of the neighboring sites, $\vec{R}'$, with probability $p(\vec{R} \,|\, \vec{R}')$, defined as

$$p(\vec{R} \,|\, \vec{R}') = \exp\left[\frac{\beta}{2}(\vec{F}\vec{e}_\nu)\right] \Big/ \sum_\mu \exp\left[\frac{\beta}{2}(\vec{F}\vec{e}_\mu)\right], \tag{1}$$

where $\vec{e}_\nu = \vec{R}' - \vec{R}$, $\nu, \mu = 1, 2, \ldots, z$, z being the coordination number of the lattice, while β is the reciprocal temperature. The hop is fulfilled only if the target site appears to be vacant; otherwise, the tracer remains at $\vec{R}$.

Now, several comments are in order. First, we note that particles diffusion does not matter in the case when the tracer and the interparticle attractive interactions are absent; here, the monolayer is homogeneous and the local occupation variables tend as $t \to \infty$ to the same value $\rho_L = 1/(1 + g/f)$, the latter relation being the Langmuir adsorption isotherm [1]. In the presence of an impure molecule, whose hopping rates differ from that of the monolayer particles, particles diffusion is significant and couples effectively the occupation variables of different lattice sites. This results, as we proceed to show, in the appearence of rather complex density profiles. Second, we remark that in the model under study the number of adsorbed particles is not explicitly conserved. The conserved particles number (CPN) limit can be achieved in the stationary regime by setting f and g to zero, while keeping their ratio fixed, $f/g = \rho_L/(1 - \rho_L)$. This limit corresponds to standard models of tracer diffusion in 2D hard-core lattice gases. Last, we remark that our model can be thought of as a certain generalization of a model for dynamic percolation proposed in [12].

Let $P(\vec{R}, \eta)$ denote the probability of finding at time t the tracer at the site $\vec{R}$ and all adsorbed particles in the configuration $\eta = \{\eta_{\vec{r}}\}$. Furthermore, let $\eta^{\vec{r},\nu}$ denote the configuration obtained from η by the Kawasaki-type exchange of the occupation variables of the two neighboring sites $\vec{r}$ and $\vec{r} + \vec{e}_\nu$, and let $\tilde{\eta}^{\vec{r}}$ be the configuration obtained from η by the replacement $\eta_{\vec{r}} \to 1 - \eta_{\vec{r}}$, which corresponds to the Glauber-type flip of the occupation variable due to the adsorption/desorption events. Then, counting up all events which can result in the configuration $(\vec{R}, \eta)$ at time t or modify it, we write down the following master equation:

$$\begin{aligned}\partial_t P(\vec{R}, \eta) = {} & \frac{1}{\tau}\sum_\nu [\, p(0 \,|\, \vec{e}_\nu) P(\vec{R} - \vec{e}_\nu, \eta)(1 - \eta_{\vec{R}}) - p(0 \,|\, \vec{e}_\nu) P(\vec{R}, \eta)(1 - \eta_{\vec{R}+\vec{e}_\nu})] \\ & + \frac{l}{4\tau^*}\sum_{\vec{r},\nu}[P(\vec{R}, \eta^{\vec{r},\nu}) - P(\vec{R}, \eta)] + \frac{g}{\tau^*}\sum_{\vec{r}}[(1 - \eta_{\vec{r}})P(\vec{R}, \tilde{\eta}^{\vec{r}}) - \eta_{\vec{r}} P(\vec{R}, \eta)] \\ & + \frac{f}{\tau^*}\sum_{\vec{r}}[\eta_{\vec{r}} P(\vec{R}, \tilde{\eta}^{\vec{r}}) - (1 - \eta_{\vec{r}})P(\vec{R}, \eta)],\end{aligned} \tag{2}$$

where the symbol $\vec{r}$ under the summation sign means that the sum runs over all the lattice sites.

Now, we are in position to obtain the X component of the tracer velocity V. Multiplying both sides of Eq. (2) by X and summing over all $(\vec{R}, \eta)$, we have

$$\begin{aligned}V_X = {} & \frac{\sigma}{\tau}\{\, p(0 \,|\, (\sigma, 0))[1 - k(\sigma, 0)] - p(0 \,|\, (-\sigma, 0)) \\ & \times [1 - k(-\sigma, 0)]\}, \qquad k(\vec{\lambda}) = \sum_{\vec{R},\eta} \eta_{\vec{R}+\vec{\lambda}} P(\vec{R}, \eta),\end{aligned} \tag{3}$$

where $k(\vec{\lambda})$ is the probability of having at time t an adsorbed particle at position $\vec{\lambda} = (\lambda_x, \lambda_y)$, defined in the frame of reference moving with the tracer particle. In other words, $k(\vec{\lambda})$ is the density profile as seen from the tracer.

Consequently, V_X depends on the particle density in the immediate vicinity of the tracer. Note that if the monolayer is perfectly stirred, i.e., $k(\vec{\lambda}) = \rho_L$ (which implies decoupling of the occupation variables), we would obtain from Eq. (3) a trivial mean-field result $V_X = V_0 = \sigma(p - q)(1 - \rho_L)/\tau$, which states that the tracer jump time τ gets merely renormalized by a factor $(1 - \rho_L)^{-1}$, i.e., the frequency of *successful* jump events. However, this is not the case and $k(\vec{\lambda}) \neq \rho_L$, except for $|\lambda| \to \infty$. Moreover, $k(\vec{\lambda})$ is a complicated function of V_X, and Eq. (3) is strongly nonlinear.

Equations describing the time evolution of $k(\vec{\lambda})$ can be found from (2) by multiplying both sides of it by $\eta_{\vec{R}+\vec{\lambda}}$

and summing over all configurations $(\vec{R}, \eta)$. In doing so, we find that these equations are not closed with respect to $k(\vec{\lambda})$, but are coupled to the third-order correlations,

$$T(\vec{\lambda}, \vec{e}_\nu) = \sum_{\vec{R},\eta} \eta_{\vec{R}+\vec{\lambda}} \eta_{\vec{R}+\vec{e}_\nu} P(\vec{R}, \eta). \quad (4)$$

In turn, if we proceed further to the third-order correlations, we find that these are coupled, respectively, to the fourth-order correlations. Consequently, in order to compute V_X, one faces the problem of solving an infinite hierarchy of coupled equations for the correlation functions. Here we resort to the simplest nontrivial closure of the hierarchy in terms of $k(\vec{\lambda})$ representing $T(\vec{\lambda}, \vec{e}_\nu)$ as

$$T(\vec{\lambda}, \vec{e}_\nu) = \sum_{\vec{R},\eta} \eta_{\vec{R}+\vec{\lambda}} P(\vec{R}, \eta) \sum_{\vec{R},\eta} \eta_{\vec{e}_\nu} P(\vec{R}, \eta). \quad (5)$$

We hasten to remark that the decoupling in Eq. (5) provides exact results for the analogous 1D model in the CPN limit [13,14] and serves as a very good approximation for the 1D model with a reservoir [7]. We set out to show in what follows that in the CPN limit our results reproduce exactly the classic results of Nakazato and Kitahara [9], which are exact for $\rho_L \ll 1$ or $\rho_L \sim 1$, and serve as a very good approximation for any intermediate ρ_L [8]. Since adsorption/desorption processes are essentially linear, we expect that such a closure will provide an accurate description for arbitrary f and g.

Employing the approximation in Eq. (5), we obtain for $k(\vec{\lambda})$ the following closed evolution equations:

$$\partial_t k(\vec{\lambda}) = \tilde{L} k(\vec{\lambda}) + \frac{f}{\tau^*},$$

$$\tilde{L} \equiv \frac{l}{4\tau^*} \Delta - \frac{f+g}{\tau^*} + \frac{1}{\tau} \sum_\nu p(0 \mid \vec{e}_\nu)[1 - k(\vec{e}_\nu)] \nabla_{\vec{e}_\nu}, \quad (6)$$

$$\Delta k(\vec{\lambda}) \equiv \sum_\nu \nabla_{\vec{e}_\nu} k(\vec{\lambda}) \equiv \sum_\nu [k(\vec{\lambda} + \vec{e}_\nu) - k(\vec{\lambda})]. \quad (7)$$

Equation (6) holds for any site $\vec{\lambda}$, except for the four sites in the immediate vicinity of the tracer: $k(\vec{\lambda})$ at these four neighboring sites is essentially perturbed due to the asymmetric hopping rules of the tracer and obeys

$$\dot{k}(\vec{e}_\nu) = [\tilde{L} k(\vec{\lambda})]|_{\vec{\lambda}=\vec{e}_\nu} + \frac{1}{\tau} p(0 \mid \vec{e}_\nu) k(2\vec{e}_\nu)[1 - k(\vec{e}_\nu)] - \frac{1}{\tau} k(\vec{e}_\nu) p(\vec{e}_\nu \mid 0)[1 - k(-\vec{e}_\nu)] - \frac{l}{4\tau^*} \nabla_{-\vec{e}_\nu} k(0) + \frac{f}{\tau^*}. \quad (8)$$

Note that Eqs. (6)–(8) constitute a closed system of equations, which suffice in principle the computation of the density profiles and of the tracer velocity. However, these equations are nonlinear, since $k(\vec{e}_\nu)$ enters the prefactor before the gradient terms, which makes such a computation to be a nontrivial problem. Below we consider the stationary solution of Eqs. (6) and (8).

Solution of Eqs. (6)–(8) in the limit $t \to \infty$ can be readily obtained by applying the generating function approach and reads

$$k(\vec{\lambda}) = \rho_L + \left(\frac{\sigma}{2\pi\sqrt{\alpha_F}}\right)^2 \left\{ \sum_\nu A_{\vec{e}_\nu} [k(\vec{e}_\nu) - \rho_L] \nabla_{-\vec{e}_\nu} \mathcal{F}_{\vec{\lambda}} - \rho_L (A_{\sigma,0} - A_{-\sigma,0}) (\nabla_{\vec{e}_\nu} \mathcal{F}_{\vec{\lambda}} - \nabla_{-\vec{e}_\nu} \mathcal{F}_{\vec{\lambda}}) \right\}, \quad (9)$$

where $A_{\vec{e}_\nu} = 1 + 4\tau^* p(0 \mid \vec{e}_\nu)[1 - k(\vec{e}_\nu)]/l\tau$, $\alpha_F = \sum_\nu A_{\vec{e}_\nu} + 4g/l(1 - \rho_L)$, $\theta = 2(\sqrt{A_{\sigma,0} A_{-\sigma,0}} + A_{0,\sigma})/\alpha_F$,

$$\mathcal{F}_{\vec{\lambda}} = \left(\frac{A_{-\sigma,0}}{A_{\sigma,0}}\right)^{\lambda_x/2\sigma} \int_{-\pi}^{\pi} \int_{-\pi}^{\pi} d^2\vec{k} \, \frac{\exp[-i(\vec{k}\vec{\lambda})]}{1 - \theta \xi(\vec{k})}, \quad (10)$$

and

$$\xi(\vec{k}) = \frac{\sqrt{A_{\sigma,0} A_{-\sigma,0}} \cos(\sigma k_x) + A_{0,\sigma} \cos(\sigma k_y)}{\sqrt{A_{\sigma,0} A_{-\sigma,0}} + A_{0,\sigma}}. \quad (11)$$

Last, we have to determine four particular values $k(\vec{e}_\nu)$ which appear on the right-hand side of Eq. (9). This can be done by setting $\vec{\lambda} = \vec{e}_\nu$, $\nu = 1, 2, \ldots, z$, to the left-hand side of Eq. (9) and solving the resulting system of four linear equations. In doing so, we define $k(\vec{e}_\nu)$ explicitly, as functions of V_X and of the characteristic parameters.

Consider now the behavior of the tracer velocity V_X in the limit $\sigma\beta F \ll 1$. First, we have from Eq. (3) that

$$V_X \sim \frac{\sigma}{4\tau} [\beta\sigma F(1 - \rho_L) - \delta k], \quad (12)$$

where $\delta k \equiv k(\sigma, 0) - k(-\sigma, 0)$ denotes the density jump in the vicinity of the tracer. Note that since $\delta k > 0$, the tracer velocity V_X is always smaller than the mean-field prediction V_0. Next, from Eqs. (9)–(11) we find

$$\frac{\delta k}{\sigma\beta F} \sim 2\rho_L(1 - \rho_L) \Bigg/ \left[3\rho_L - 1 - \frac{l\tau}{\tau^*} + \frac{l\tau A_{\sigma,0}(F = 0)}{\tau^*} \Bigg/ \int_0^\infty \frac{dw}{w} \exp\left(- \frac{\alpha_0 w}{2A_{\sigma,0}(F = 0)} \right) I_1(w) I_0(w) \right], \quad (13)$$

$I_n(w)$ being the modified Bessel function of order n.

Consequently, we find from Eq. (12) that in the limit $\sigma\beta F \ll 1$ the force-velocity relation attains the physically revealing form of the Stokes formula, i.e., $F = \zeta V_X$, which signifies that the friction force exerted by the monolayer particles on the driven tracer is viscous. The corresponding friction coefficient ζ is given by

$$\zeta = \zeta_0 + \zeta_{\text{coop}}, \quad (14)$$

where $\zeta_0 = 4\tau/\sigma^2\beta(1 - \rho_L)$ and ζ_{coop} follows

$$\zeta_{\text{coop}} = \zeta_0 \frac{(\delta k/\sigma\beta F)}{1 - \rho_L - (\delta k/\sigma\beta F)}, \qquad (15)$$

$(\delta k/\sigma\beta F)$ being defined in Eq. (13). While ζ_0 is the typical mean-field-type result [see the discussion following Eq. (3)], the second term is associated with the cooperative behavior: dehomogenization of the monolayer by driven impure molecule and formation of stationary density profiles, whose characteristic parameters depend on the velocity V_X. This second term can be small when either $\rho_L \ll 1$, or $\tau^* \to 0$ (perfect mixing) but dominates the overall friction for any intermediate values of systems parameters. We finally remark that in the CPN limit Eq. (14) reduces to the classical result obtained in [9].

Last, we discuss the characteristic features of the monolayer density profiles as seen from the stationary moving tracer, Eq. (9), at large distances in front of and past the tracer. It follows from Eq. (9) that in the limit $|\lambda_x| \to \infty$, $(\lambda_y = 0)$ the density profiles obey

$$k(\lambda_x, 0) \sim \rho_L \pm \frac{\delta k}{2}\left(\frac{\sigma}{|\lambda_x|}\right)^{1/2} \exp\left(\pm \frac{|\lambda_x|}{\sigma} \ln(z_\pm)\right), \qquad (16)$$

where the sign "+" ("−") corresponds to the domain $\lambda_x > 0$ ($\lambda_x < 0$), while $z_\pm$ are two of four eigenvalues of the operator $\tilde{L}$ in Eq. (6), which are given by

$$z_\pm = A^{-1}_{-\sigma,0}\left(\frac{\alpha_F}{2} - A_{0,\sigma}\right)\left[1 \pm \sqrt{1 - \frac{2A_{\sigma,0}A_{-\sigma,0}}{\alpha_F - 2A_{0,\sigma}}}\right]. \qquad (17)$$

Hence, in the domain $\lambda_x > 0$ the monolayer density is higher than ρ_L, which means that there is a "traffic jam"-type region in front of the tracer, which impedes its motion. This region decays exponentially with the distance. In turn, past the tracer there is an exponentially decaying depleted region in which the density is lower than ρ_L; since $z_- < z_+$, the depleted region is more extended in space than the condensed one in front of the tracer, such that the density profiles are asymmetric with respect to $\lambda_x = 0$. It is interesting to note that for $k(\vec{\lambda})$ as in Eq. (9) the sum $\sum_{\vec{\lambda}}[k(\vec{\lambda}) - \rho_L] \equiv 0$, which means that the tracer does not perturb the global balance between adsorption and desorption. This is not, however, an *a priori* evident result in view of the asymmetry of the density profiles.

The salient feature of the behavior past the tracer is that in the CPN limit $z_- \equiv 1$ at any fixed F, which signals that Eq. (16) is no longer valid and correlations get somewhat stronger. Indeed, we find that

$$k(\lambda_x, 0) - \rho_L \sim -\text{const} \times |\lambda_x|^{-3/2}, \qquad \lambda_x \to -\infty; \qquad (18)$$

i.e., the correlations fall off with the distance as a *power law*. Remarkably, this implies that in the CPN limit the mixing of the monolayer is not at all efficient and there are considerable memory effects—the host medium remembers the passage of the tracer on a large space and time scales. This situation can be realized experimentally for ultrathin liquid films confined in narrow slits between solid surfaces, e.g., in boundary lubrication. We note also that the power-law behavior in Eq. (18) can be observed as an intermediate scale decay for adsorbed layers exposed to a low vapor pressure.

To conclude, we have studied analytically the intrinsic frictional properties of 2D adsorbed monolayers, composed of mobile hard-core particles undergoing continuous exchange with the vapor. By analyzing the force-velocity relation of a driven impure molecule—a tracer, which is designed to probe the resistance offered by adsorbate particles to the external perturbance, we have derived the analog of the Stokes formula for 2D mobile adsorbates and calculated explicitly the corresponding friction coefficient. Besides, we have determined the stationary density profiles, which emerge in the adsorbate in response to the presence of a driven impurity, as well as obtained explicit results for both the density jump in the immediate vicinity of the tracer and the asymptotical density relaxation at large separations from it.

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