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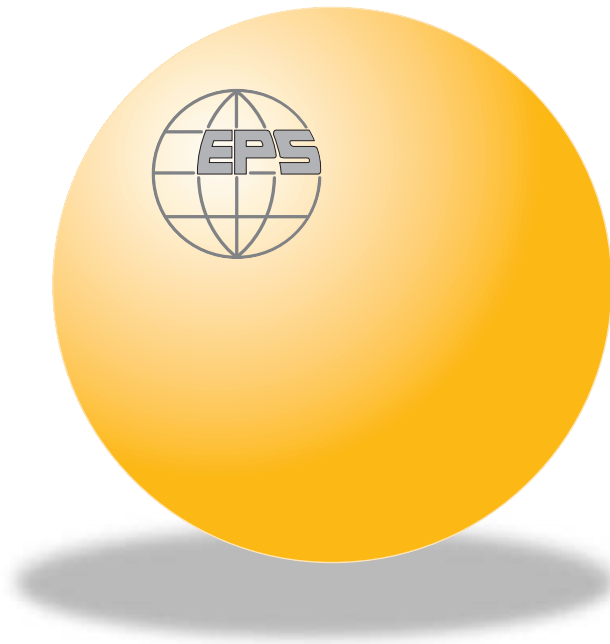
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Membrane pinning on a disordered substrate

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Abstract. – We investigate interactions between an elastic membrane and a substrate characterized by quenched positional disorder in the height function. We show that the positional disorder transforms the standard secondary DLVO minimum into two separate states: the hovering state characterized by a planar membrane at a finite separation from the interface and a pinned state where the membrane follows closely the asperities of the substrate and is, as a consequence, quite corrugated. The transition between the two states is continuous and depends on the parameters of the underlying DLVO potential as well as the parameters describing the quenched height-height correlation function of the substrate.

In the DLVO theory one usually assumes that when a membrane or, in general, an elastic manifold interacts with a rigid substrate, the membrane is modelled as flat and the substrate is envisioned as featureless [1]. This leads in a straightforward manner to the secondary minimum of the DLVO theory and to an equilibrium spacing between the membrane and the substrate. Taking into account the elastic degrees of freedom of a membrane can alter this picture drastically, leading to the emergence of Helfrich interaction which may induce an unbinding transition of the membrane. This transition has been clearly shown to be a consequence of the interplay between elastic fluctuations of the membrane and DLVO interactions between the membrane and the substrate [2].

In the present work we are motivated by recent experiments on deposited lipid multilayers on atomically smooth *vs.* rough surfaces [3]. These experiments reveal differences in equilibrium lamellar spacings of lipid multilayers in proximity of a substrate that seem to correlate with molecular roughness of the substrate [3]. In order to lay ground for understanding effects of this type, it is desirable to relax the model constraint of a featureless substrate and treat it supposedly more realistically as exhibiting (quenched) disorder in the height function. We will analyze the consequences of this new model of a substrate interacting with a membrane and show that it modifies the simplified DLVO conclusions in the sense that the original secondary DLVO minimum now splits into two separate states, characterized by the way the membrane

is (de)coupled to the substrate. We call these states the *hovering state* and the *pinned state*. The former one is characterized by a membrane in a flat configuration hovering a certain finite distance above the substrate and is directly related to the standard DLVO secondary minimum. The latter one is characterized by a corrugated membrane that basically follows the asperities of the quenched positional disorder of the substrate and is altogether missing from the standard DLVO theory. We stress that our description is unifying in the sense that it combines aspects of *mean-field* theory [4], which focuses on the nature of the pinned state; and a straightforward generalization [5] of Li and Kardar's [6] *Gaussian fluctuation theory*, which allows for a hovering state determined by the parameters of the DLVO interaction potential. Specifically, we find that for sufficiently strong disorder, the membrane indeed prefers the mean-field pinned state considered by Swain and Andelman [4]. Because of the increased role of substrate disorder fluctuations, in weakly disordered systems the properly self-averaged free energy can give way to a hovering state where the membrane is depinned, residing in a minimum related to but different from the usual DLVO minimum, as predicted by Gaussian fluctuation theory.

We introduce our approach by first specifying the model of the membrane and the substrate that interact via a DLVO potential with (for matters of convenience) hydration and van der Waals terms. Formally, our analysis owes a lot to investigations of effects of the disorder on polymerized membranes that have been analyzed in a variety of contexts [7]. It is based on the assumption of a Gaussian disordered substrate height distribution function and the application of the $1/d$ expansion method [8] ([1]) to evaluate the complicated partition function stemming from the replicated Hamiltonian [9]. This approach allows us to evaluate the coupled effect of the fluctuations and disorder on the mean-field configuration itself, as opposed to the Gaussian fluctuation theory [6] which considers only fluctuations around a disorder-unperturbed mean-field.

The Hamiltonian of a flexible membrane described in the usual Monge parameterization $u(\boldsymbol{\rho})$, where $\boldsymbol{\rho} = (x, y)$ is a 2D coordinate vector, above a substrate characterized by the height function $\zeta(\boldsymbol{\rho})$, where $\zeta(\boldsymbol{\rho})$ is a quenched-disorder field describing the profile of the substrate, is composed of three contributions. First of all, we have the elastic energy of the membrane, then the interaction free energy per unit surface area between the membrane and the substrate $V(u(\boldsymbol{\rho}) - \zeta(\boldsymbol{\rho}))$ of the general DLVO form, and, finally, the free-energy contribution of an external force per unit surface area π pushing the membrane towards the substrate. The total free energy or, equivalently, the mesoscopic Hamiltonian thus assumes the form

$$\mathcal{H}[u(\boldsymbol{\rho})] = \frac{1}{2} \iint \mathrm{d}^2\boldsymbol{\rho}\,\mathrm{d}^2\boldsymbol{\rho}'\mathcal{K}(\boldsymbol{\rho}, \boldsymbol{\rho}')u(\boldsymbol{\rho})u(\boldsymbol{\rho}') + \int \mathrm{d}^2\boldsymbol{\rho} V\big(u(\boldsymbol{\rho}) - \zeta(\boldsymbol{\rho})\big) + \int \mathrm{d}^2\boldsymbol{\rho}\pi\big(u(\boldsymbol{\rho}) - \zeta(\boldsymbol{\rho})\big).$$

Usually, one takes for the elastic part the classical Canham-Helfrich-Evans *ansatz* $\mathcal{K}(\boldsymbol{\rho}, \boldsymbol{\rho}') = \boldsymbol{\nabla}^4 K_\mathrm{c}\delta^2(\boldsymbol{\rho} - \boldsymbol{\rho}')$, where K_c is the bending modulus of the membrane, while the interaction part is composed of the attractive and repulsive DLVO ingredients. Since the interaction part of the Hamiltonian is, in general, non-linear, it is convenient to introduce the following new variable: $B(\boldsymbol{\rho}) = (u(\boldsymbol{\rho}) - \zeta(\boldsymbol{\rho}))^2$ at every $\boldsymbol{\rho}$ via a functional constraint [10],

$$\delta\mathcal{H}[B(\boldsymbol{\rho}), g(\boldsymbol{\rho})] = \frac{1}{2} \int \mathrm{d}^2\boldsymbol{\rho} g(\boldsymbol{\rho})\big((u(\boldsymbol{\rho}) - \zeta(\boldsymbol{\rho}))^2 - B(\boldsymbol{\rho})\big).$$

Clearly, the auxiliary field $g(\boldsymbol{\rho})$ plays a role akin to the self-energy part of the Green function. The two auxiliary fields $B(\boldsymbol{\rho})$ and $g(\boldsymbol{\rho})$ just introduced play the role of fixing the local

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([1])Strictly speaking, what we use is actually a $1/(d-2)$ expansion, since the displacement field is one-dimensional.

constraint $(u(\boldsymbol{\rho}) - \zeta(\boldsymbol{\rho}))^2 = B(\boldsymbol{\rho})$. They also show up in the partition function, where one has to eventually take the trace over these auxiliary fields together with $u(\boldsymbol{\rho})$. The partition function can thus be written as

$$\mathcal{Z}[\zeta(\boldsymbol{\rho})] = \int \mathcal{D}u(\boldsymbol{\rho})\mathcal{D}B(\boldsymbol{\rho})\mathcal{D}g(\boldsymbol{\rho}) \exp\big[-\beta\big(\mathcal{H}[u(\boldsymbol{\rho})] + \delta\mathcal{H}[B(\boldsymbol{\rho}), g(\boldsymbol{\rho})]\big)\big].$$

The average over the quenched-disorder distribution, assuming the self-averaging property of the free energy, is defined to be of the form

$$\overline{(\ldots)} = \int \mathcal{D}\zeta(\boldsymbol{\rho})(\ldots)\mathcal{P}[\zeta(\boldsymbol{\rho})] = \int \mathcal{D}\zeta(\boldsymbol{\rho})(\ldots) \exp\left[-\frac{1}{2}\iint \mathrm{d}^2\boldsymbol{\rho}\,\mathrm{d}^2\boldsymbol{\rho}'\mathcal{G}(\boldsymbol{\rho}, \boldsymbol{\rho}')\zeta(\boldsymbol{\rho})\zeta(\boldsymbol{\rho}')\right],$$

where, by assumption, the disorder probability distribution function $\mathcal{P}(\zeta(\boldsymbol{\rho}))$ that characterizes the quenched disorder in the height function of the substrate is given by a Gaussian *ansatz*. The free energy, after being averaged also over the quenched-disorder distribution, is obtained finally as

$$\mathcal{F} = -kT\,\overline{\log \mathcal{Z}[\zeta(\boldsymbol{\rho})]} = -kT \lim_{n \to 0} \frac{\overline{\mathcal{Z}^n[\zeta(\boldsymbol{\rho})]} - 1}{n}. \tag{1}$$

With these preliminaries the free energy eq. (1) can be evaluated via the standard Edwards-Anderson replica trick [9], where the replicated Hamiltonian $\mathcal{H}_n$ is composed of the replicated elastic term

$$\frac{1}{2}\sum_{i=0}^{n} \int\int \mathrm{d}^2\boldsymbol{\rho}\,\mathrm{d}^2\boldsymbol{\rho}'\mathcal{K}(\boldsymbol{\rho}, \boldsymbol{\rho}')u_i(\boldsymbol{\rho})u_i(\boldsymbol{\rho}'),$$

where i is the index of the replica, the replicated constraint on the variable $B_i(\boldsymbol{\rho})$ that now reads

$$\frac{1}{2}\sum_{i=0}^{n} \int \mathrm{d}^2\boldsymbol{\rho}g_i(\boldsymbol{\rho})\big((u_i(\boldsymbol{\rho}) - \zeta(\boldsymbol{\rho}))^2 - B_i(\boldsymbol{\rho})\big),$$

and, finally, of the replicated interaction and external "source" terms

$$\sum_{i=0}^{n} \int \mathrm{d}^2\boldsymbol{\rho}V(B_i(\boldsymbol{\rho})) + \sum_{i=0}^{n} \int \mathrm{d}^2\boldsymbol{\rho}\,\pi\big(u_i(\boldsymbol{\rho}) - \zeta(\boldsymbol{\rho})\big).$$

Since we now have Gaussian integrals over the variables $u_i(\boldsymbol{\rho})$, $\zeta(\boldsymbol{\rho})$, we can evaluate them explicitly, while the functional integrals over the auxiliary fields $B_i(\boldsymbol{\rho})$, $g_i(\boldsymbol{\rho})$ can be evaluated on the saddle point level, with the *proviso* that there is no replica symmetry breaking. This constitutes the essence of the $1/d$ expansion in this context. We have obviously ignored the steric constraint between $u_i(\boldsymbol{\rho})$, $\zeta(\boldsymbol{\rho})$ which would lead straightforwardly to Helfrich interactions, since we are not able to evaluate the ensuing functional integral. We will nevertheless put in the steric interactions "by hand" at the end of our calculation.

After performing all the indicated integrations and taking the $n \to 0$ limit, the free energy can be obtained as a sum of the mean-field part (^{2}) and a fluctuation part. The mean-field part is

$$\mathcal{F}_0 = \int \mathrm{d}^2\boldsymbol{\rho}\,\pi u_0(\boldsymbol{\rho}) + \frac{1}{2}\int\int \mathrm{d}^2\boldsymbol{\rho}\,\mathrm{d}^2\boldsymbol{\rho}'\chi(\boldsymbol{\rho}, \boldsymbol{\rho}')u_0(\boldsymbol{\rho})u_0(\boldsymbol{\rho}'),$$

where the mean field $u_0(\boldsymbol{\rho})$ is obtained via minimization of $\mathcal{F}_0$ with

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(^{2})Not to be confused with the mean-field approach introduced in [4]. The term mean field is used here strictly as it pertains to the $1/d$ expansion.

$$\chi(\boldsymbol{\rho}, \boldsymbol{\rho}') = \mathcal{K}(\boldsymbol{\rho}, \boldsymbol{\rho}') + \delta^2(\boldsymbol{\rho} - \boldsymbol{\rho}')g(\boldsymbol{\rho}).$$

Again we note that on the $1/d$ expansion level the mean-field part includes the effect of the disorder, the $g(\boldsymbol{\rho})$ term in the above equation, contrary to the Gaussian fluctuation approach [6]. The fluctuation part of the free energy is concurrently obtained as

$$\mathcal{F} = \frac{kT}{2} \int \mathrm{d}^2\boldsymbol{\rho}\,\mathcal{G}^{-1}(\boldsymbol{\rho}, \boldsymbol{\rho})g(\boldsymbol{\rho}) - \frac{kT}{2} \int\int \mathrm{d}^2\boldsymbol{\rho}\,\mathrm{d}^2\boldsymbol{\rho}'\chi^{-1}(\boldsymbol{\rho}, \boldsymbol{\rho}')\mathcal{G}^{-1}(\boldsymbol{\rho}', \boldsymbol{\rho})g(\boldsymbol{\rho})g(\boldsymbol{\rho}') -$$
$$- \frac{1}{2} \int \mathrm{d}^2\boldsymbol{\rho}g(\boldsymbol{\rho})B(\boldsymbol{\rho}) + \int \mathrm{d}^2\boldsymbol{\rho}V(B(\boldsymbol{\rho})). \tag{2}$$

On the $1/d$ expansion level the auxiliary fields contribute only at the saddle point. The saddle point equations are now obtained simply by minimizing eq. (2) with respect to $B(\boldsymbol{\rho})$ and $g(\boldsymbol{\rho})$. We will not reproduce the general rather awkward form of these equations, but will concentrate on a rather particular solution characterized by $u_0 = \mathrm{const}$ and $B = \mathrm{const}$, implying also $g = \mathrm{const}$. In addition to this, we will limit ourselves to the conceptually most interesting case of vanishing external confining force, *i.e.* $\pi = 0$.

Assuming that the system is homogeneous in the $\boldsymbol{\rho}$ plane, we can introduce the Fourier transforms of all the relevant quantities that allow us to write the mean-field equations in a rather simple form:

$$u_0\chi(\boldsymbol{Q} = 0) = u_0g = 0. \tag{3}$$

The saddle points for B and g are obtained, respectively, as

$$\frac{\partial V(B)}{\partial B} = \frac{1}{2}g, \qquad \text{and} \qquad B = u_0^2 + kT\sum_{\boldsymbol{Q}} \frac{\mathcal{K}(\boldsymbol{Q})^2}{\mathcal{G}(\boldsymbol{Q})(\mathcal{K}(\boldsymbol{Q}) + g)^2}\,.$$

This set of equations has two fundamentally different solutions describing the state of the elastic membrane interacting with a disordered substrate.

1) a *hovering state*, with $u_0 \neq 0$, $g = 0$, characterized by

$$\frac{\partial V(B)}{\partial B} = 0, \qquad \text{with} \qquad B = u_0^2 + kT\sum_{\boldsymbol{Q}} \mathcal{G}^{-1}(\boldsymbol{Q}). \tag{4}$$

2) and a *pinned state*, with $u_0 = 0$, $g \neq 0$, characterized by

$$\frac{\partial V(B)}{\partial B} = \frac{1}{2}g, \qquad \text{with} \qquad B = kT\sum_{\boldsymbol{Q}} \frac{\mathcal{K}(\boldsymbol{Q})^2}{\mathcal{G}(\boldsymbol{Q})(\mathcal{K}(\boldsymbol{Q}) + g)^2}\,. \tag{5}$$

Obviously, in order to progress we have to assume a certain form for the quenched-disorder correlation function. As a simplest approximation we take a spatially short-range coupling $\mathcal{G}(\boldsymbol{\rho}, \boldsymbol{\rho}') = G\delta^2(\boldsymbol{\rho} - \boldsymbol{\rho}')$, implying $\mathcal{G}(\boldsymbol{Q}) = G$. Thus, we have in the hovering state

$$\frac{\partial V(B)}{\partial B} = 0, \qquad \text{with} \qquad u_0^2 = B - B_\mathrm{c}, \tag{6}$$

where we introduced $B_\mathrm{c} = kT\sum_{\boldsymbol{Q}} \mathcal{G}^{-1}(\boldsymbol{Q}) = \frac{kT}{G}\frac{Q_{\max}^2}{4\pi}$, that obviously depends on the upper wave vector cutoff in the Fourier space. The first of the above equations determines B as a function of the parameters of the DLVO potential (*e.g.*, Hamaker constant, hydration interaction strength, etc.). The second one gives the dependence of u_0 on these parameters.

In order to understand the physical nature of the two phases, we evaluate the average of the separation between the substrate and the membrane that can be obtained as

$$S^{-1} \int \mathrm{d}^2\boldsymbol{\rho}\,\overline{\langle(u(\boldsymbol{\rho}) - \zeta(\boldsymbol{\rho}))\rangle} = u_0.$$
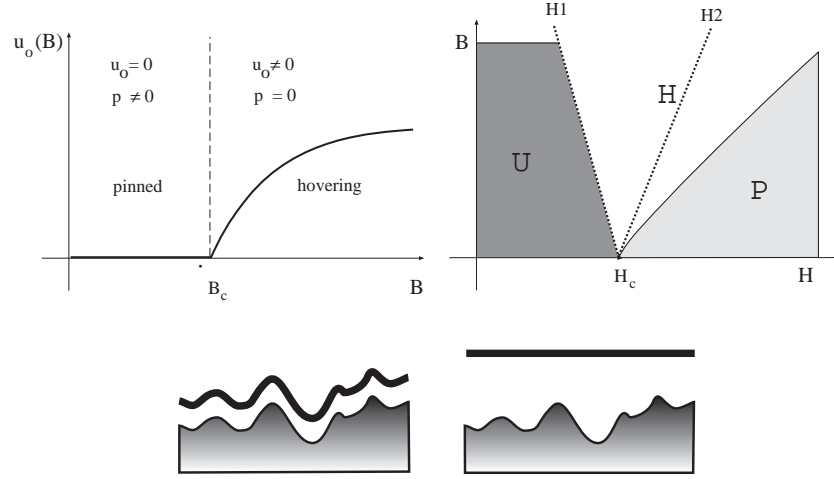
Fig. 1 – Upper left: schematic dependence of u_0 on the auxiliary field B. The hovering state exists only for $B > B_c$. The critical value B_c depends on the disorder characteristics of the substrate and the microscopic wave vector cutoff. The hovering-pinned transition is obviously second order in B, which thus plays the role of the temperature. Upper right: Schematic phase diagram of a membrane as a function of the Hamaker coefficient; $V(u)$ is assumed to be equal to the sum of hydration, van der Waals and Helfrich terms eq. (7). The substrate disorder introduces a hovering (H) state window into the phase diagram, located between the pinned (P) and the unbound (U) phases. Two possible (H-U) boundaries are indicated by the H1 and H2 lines. The size of this window depends on the strength of the Hamaker coefficient and the value of G, characterizing the disorder effects. In the case of no disorder ($B \to 0$) the critical value of the Hamaker coefficient H_c marks the unbinding transition of the membrane. Bottom: a schematic representation of the hovering (H) and the pinned (P) state of the membrane in proximity of a disordered substrate. The hovering state is characterized by a slowly varying average separation between the membrane and the substrate that does not follow closely the local corrugations of the substrate. In the pinned state, however, the membrane follows closely the asperities of the substrate.

Clearly, u_0 quantifies the disorder-averaged separation between the membrane and the substrate. For a finite u_0 the membrane hovers a finite separation away from the substrate. In order to characterize the hovering state further, we evaluate the average square of the deviation from a unit normal to the membrane in the z-direction given to the lowest order as $\delta \boldsymbol{n}(\boldsymbol{\rho}) \approx \boldsymbol{\nabla}_\perp \cdot u(\boldsymbol{\rho})$,

$$ p^2 = S^{-1} \int \mathrm{d}^2 \boldsymbol{\rho} \, \overline{\langle \delta \boldsymbol{n}^2(\boldsymbol{\rho}) \rangle} = \frac{kTg^2}{2G} \sum_{\boldsymbol{Q}} \frac{Q^2}{(\mathcal{K}(\boldsymbol{Q}) + g)^2} = \frac{kT}{16\pi} \frac{g}{GK_c} \, . $$

Thus, in the hovering state with $g = 0$ not only is the membrane decoupled from the substrate and hovers above it, but also appears to be flat *on the average*, since the mean disorder-averaged squared deviation from the normal, p^2, equals zero at a finite separation u_0 (see fig. 1). One should not forget here that all the averages are also with respect to the quenched surface disorder and thus the membrane appears to be "flat" only in the sense that it does not follow the disordered corrugations of the substrate. Its elastic fluctuations, however, before the disorder average, are not zero! This statement is true of course only to the lowest order in the $1/d$ expansion. In order to analyze the fluctuations around this state, one should go to higher-order terms.

Obviously, the hovering solution exists only for $B > B_{\mathrm{c}}$. The hovering line in the "phase diagram" thus ends at the value of the interaction parameters where the solution of $\frac{\partial V(B)}{\partial B} = 0$ also satisfies $B = B_{\mathrm{c}}$. u_0 thus behaves as an order parameter of a second-order phase transition, and B behaves as the temperature. Furthermore, the surface density of the adhesion free energy (U) in the hovering state is given by

$$U = \min\{\mathcal{F}/S\} = V(B) = V\big(u_0^2 + B_{\mathrm{c}}\big).$$

This obviously differs from the standard secondary minimum of the DLVO theory, determined from $\frac{\partial V(u_0)}{\partial u_0} = 0$, with $U = V(u_0)$. The free energy in the hovering state is thus not given by the value of the DLVO potential at the minimum $V(u_0)$, but at a (much smaller) value of $V(u_0^2 + B_{\mathrm{c}})$, since V is a decreasing function of its argument above its minimum.

In the hovering state the membrane is obviously in close proximity ("contact") of the substrate only for a fraction of its total surface area. This effective contact area (S_{c}) can be estimated [4] from $S_{\mathrm{c}}/S = V(u_0^2 + B_{\mathrm{c}})/V(u_0^2)$. In view of the discussion presented above, this ratio is smaller than one if V is a decreasing function of its argument above its minimum value.

For the pinned state, with $\mathcal{K}(\boldsymbol{Q}) = Q^4 K_{\mathrm{c}}$, we obtain the following expression to the lowest order in the wave vector cutoff Q_{max}:

$$\frac{\partial V(B)}{\partial B} = \frac{1}{2}g \qquad \text{with} \qquad B = B_{\mathrm{c}} - \frac{3kT}{16G}\sqrt{\frac{g}{K_{\mathrm{c}}}}\,,$$

where B_{c} is defined in the same way as in the hovering state. The adhesion free energy in the pinned state, defined in complete analogy with the hovering state, is given by

$$U = V(B) + \frac{kTK_{\mathrm{c}}}{4\pi G}\left(\frac{g}{K_{\mathrm{c}}}\right)^{\frac{3}{2}} = V(B) + \frac{16^3}{4\pi 3^3}\frac{K_{\mathrm{c}}G^2}{(kT)^2}(B_{\mathrm{c}} - B)^3.$$

In the pinned state the membrane is thus coupled to the substrate and follows it closely, being always in its close proximity since $u_0 = 0$ (see fig. 1). It thus exhibits a very corrugated configuration since in the pinned state $p^2 \neq 0$. We note that for non-zero external driving force π the system is always in the pinned state.

The adhesion energy in both states is nowhere in general equal to its DLVO counterpart. Both the hovering as well as the pinned states carry in the free energy the signature of the substrate disorder in the height-height correlations. Only in the limit of vanishing disorder, or in the language of our model as $G \to \infty$, does the hovering state approach the DLVO secondary minimum in a continuous fashion, while the pinned state simply disappears.

In order to gain further insight into the nature of the hovering and the pinned states, we investigate the phase diagram for a particular typical choice of the DLVO interaction potential augmented by the Helfrich undulation interaction. The latter has to be put in "by hand" [4] because the steric constraint it introduces into the functional integral for the partition function is extremely difficult to handle. For the sake of simplicity, we assume that the DLVO part is given by the sum of the hydration and van der Waals interactions while the Helfrich interaction is assumed to have the same form as between a flexible membrane and a flat substrate [1, 2],

$$V(u) = Ae^{-u/\xi} - \frac{H}{12\pi u^2} + \frac{6\pi^2 (kT)^2}{256 K_{\mathrm{c}} u^2}\,, \tag{7}$$

where A is the magnitude and ξ the range of the hydration interaction, and treat the Hamaker constant H as a variable tuning parameter. The above form of the total interaction energy

between a membrane and a substrate would be strictly valid only for a membrane fluctuating near a flat substrate. However, Swain and Andelman note [4] that the form of the Helfrich interaction should not be far off from the one in the above expression even for a rough substrate. We assume this is the case when we analyze the phase diagram for a membrane near a disordered substrate. This assumption would however have to be tested via a more sophisticated and hopefully more accurate approach.

Without the disorder the interaction potential eq. (7) leads to an unbinding transition at the critical value of the Hamaker constant equal to $H_c = \frac{72\pi^3(kT)^2}{256K_c}$. When H approaches this value, the secondary DLVO minimum is displaced towards infinity and we have a continuous unbinding of the membrane. Adding disorder to this scenario we instead obtain a modified phase diagram as presented in fig. 1. There is now a window corresponding to the hovering state (the former DLVO secondary minimum) in between the pinned and the unbound state of the membrane. The dimensions of this window depend on the value of $\mathcal{G}$ that characterizes the intensity of the substrate disorder. In the limit of no disorder $\mathcal{G} \to \infty$ with $B_c \to 0$ the hovering window is expanded to the whole axis $H > H_c$ and is transformed back into the standard DLVO secondary minimum.

The disorder usually does not figure in the theories of membrane substrate interactions. The present work is basically a plea for a change of this perspective. There obviously exist phenomena, where ignoring the disordered nature of the substrate does not, even qualitatively, lead to the correct physical picture. In this sense the DLVO theory has to be amended.

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