Adhesion selectivity by electrostatic complementarity. I. One-dimensional stripes of charge

Ying Bai,1 Congrui Jin,3 Anand Jagota,1,2,a) and C.-Y. Hui3

1Department of Chemical Engineering, Lehigh University, Bethlehem, Pennsylvania 18015, USA
2Bioengineering Program, Lehigh University, Bethlehem, Pennsylvania 18015, USA
3Field of Theoretical and Applied Mechanics, Sibley School of Mechanical and Aerospace Engineering, Cornell University, Ithaca, New York 14850, USA

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Highly enhanced and selective adhesion can be achieved between surfaces patterned with charges even when each one has no net charge. In this and a companion paper [C. Jin, Y. Bai, A. Jagota, and C.-Y. Hui, J. Appl. Phys. 110, 054903 (2011)] we analyze the possibility of adhesion selectivity between two flat surfaces patterned with stripes of surface charge chosen such that each surface initially carries no net charge. A few combinations, with appropriately matching strip widths, are predicted to adhere to each other. We also find that the deformability of the materials plays a critical role in defining the range of patterns that recognize each other, i.e., their selectivity. With increasing compliance, a significant enhancement of adhesion can be achieved by deformation that allows better matching between charge patterns. © 2011 American Institute of Physics. [doi:10.1063/1.3626432]

I. INTRODUCTION

Enhanced, selective, and controllable adhesion between surfaces is a highly desired property. At the macroscopic scale it manifests in a variety of fastening devices and designs based on shape complementarity. As the property of a material surface, it can endow the ability to reject any surface other than its complementary one. That is, adhesive and friction energy can be large between two complementary surfaces, and highly attenuated when they are not matched. Selectivity usually requires complementarity of some sort between surfaces. In nature, this idea of selectivity extends all the way from the organism to individual molecules. For example, several interlocking mesoscale structures are responsible for attachment of forewings to the thorax in beetles (Coleoptera),1 as well as in the dragonfly scale structures are responsible for attachment of forewings to the thorax in beetles (Coleoptera),1 as well as in the dragonfly.

The deliberate control of adhesion selectivity of material surfaces by complementarity has not been much studied. Examples include the celebrated case of loop-clasp designs that led to the development of Velcro®.16 In other examples, selectivity in adhesion has been accomplished by design of surface chemistry,11–14 using fibrillar structures,15 or between two complementary rippled surfaces. A study about surface pattern recognition by using hydrophobic complementarity was reported by Kokkoli and Zukoski.17

Electrostatic complementarity presents a promising approach. In this work we ask: Is it possible to achieve high selectivity, as measured via adhesion, using relatively smooth and flat extended surfaces patterned with charges? Although a number of measurements and explanations for electrostatic complementarity have been developed for biological systems, its use as a generic method for designing in selectivity has not been studied. We consider the interaction between two flat surfaces separated by water, with simple striped patterns of alternating positive and negative surface charges, summing to zero net charge on either one. We predict that such surfaces will have highly selective adhesion depending on the matching between charge patterns on the two surfaces. This selectivity is modulated strongly by deformability of the materials.

II. TWO UNIFORMLY CHARGED PLATES

We imagine an experimental realization in which the surfaces of two flat dielectric solids are patterned by self-assembled monolayers with surface groups that protonate or de-protonate in aqueous medium (e.g., NH2 to NH3+ or COOH to COO−), creating stripes of surface charges.
The interaction between these charges is modeled by the Debye-Hückel equation, which is a linearized form of the Poisson-Boltzmann equation and is strictly valid for low ionic concentrations and potential.

Anticipating that often the gap between surfaces will be much smaller than the lateral length scale of charge patterns, we expect to be able to represent the total interaction by a much smaller than the lateral length scale of charge patterns, we have analyzed the full two-dimensional interaction between two uniformly charged infinite plates in water in the presence of ions. In a companion paper, we have analyzed the full two-dimensional potential field. The two plates can have different charge densities \( \sigma_1 \) and \( \sigma_2 \), but the charge distribution on each plate is uniform. They are placed parallel to each other at a distance of \( \alpha \), as shown in Fig. 1. We begin by establishing the force and energy of interaction between two such surfaces.

### A. Electric potential between two uniformly charged plates under Debye-Hückel electrostatics

In one-dimension, the variation in electrostatic potential in the region between the two plates is governed by the Debye-Hückel equation:

\[
\frac{d^2 \phi}{dx^2} = \frac{\phi}{l_D^2},
\]

where \( l_D \) is the Debye screening length that, for a z-z electrolyte (e.g., 1-1 for NaCl) is

\[
l_D = \sqrt{\frac{\epsilon \epsilon_0 k_B T}{2q^2 a^2 \epsilon_0}}
\]

where \( \epsilon \) is the dielectric constant of water, \( \epsilon_0 \) is the permittivity of free space, \( q \) is the charge of an electron, \( a \) is the is the unsigned valence of each of the two ions, \( \epsilon_0 \) is the concentration of the ions, \( T \) is temperature, and \( k_B \) is Boltzmann’s constant.

The potential at either interface decays on the dielectric side over a length approximately equal to the sample thickness. On the water side it decays over the Debye screening length. Because the sample thickness (~nm) is much larger than the Debye screening length (~nm), the dielectric can be modeled as a conductor (constant potential) in comparison with the strongly decaying field on the water side. We have separately analyzed the electric potential distribution in an infinite open system with five regions: a narrow gap between the two charged plates, the plates themselves, and the exterior region on either side of the plates (results available from authors). We find that, under the conditions of interest to us, i.e., when the gap is on the order of Debye screening length (a few nm), the field inside the charged plates is negligible. The solution is then nearly identical to that of a single domain between the two charged plates with a jump in the electric field balanced by the charge density on the two surfaces.

As a result, the boundary conditions at the two interfaces that relate the field to surface charge density take the form

\[
a. \quad -\epsilon \epsilon_0 \frac{d\phi}{dx} = \sigma_1 \quad (x = 0),
\]

\[
b. \quad \epsilon \epsilon_0 \frac{d\phi}{dx} = \sigma_2 \quad (x = a).
\]

The general solution of Eq. (1) is

\[
\phi(x) = A \cosh \left( \frac{x}{l_D} \right) + B \sinh \left( \frac{x}{l_D} \right)
\]

and

\[
\frac{d\phi}{dx} = \frac{A}{l_D} \sinh \left( \frac{x}{l_D} \right) + B \frac{l_D}{l_D} \cosh \left( \frac{x}{l_D} \right).
\]

Applying the boundary conditions, we find

\[
\phi(x) = \frac{\left[ \sigma_1 \cosh \left( \frac{a}{l_D} \right) + \sigma_2 \right]}{\epsilon \epsilon_0} \frac{l_D \cosh \left( \frac{x}{l_D} \right)}{\sinh \left( \frac{a}{l_D} \right)} - \frac{\sigma_1 l_D}{\epsilon \epsilon_0} \sinh \left( \frac{x}{l_D} \right).
\]

When the two plates have the same charge density, \( \sigma \), the result is

\[
\phi(x) = \frac{\sigma l_D}{\epsilon \epsilon_0} \frac{\cosh \left( \frac{a/2 - x}{l_D} \right)}{\sinh \left( \frac{a/2}{l_D} \right)}.
\]

Note that, as expected, this function is even about \( x = a/2 \). When both surfaces have the same magnitude of charge density, but of opposite sign \( \sigma_2 = \sigma; \sigma_1 = -\sigma \), then the electric potential is

\[
\phi(x) = \frac{\sigma l_D}{\epsilon \epsilon_0} \frac{\sinh \left( \frac{a/2 - x}{l_D} \right)}{\cosh \left( \frac{a/2}{l_D} \right)}.
\]

As expected, this function is odd about \( x = a/2 \).
B. Force and interaction energy between two uniformly charged plates

Using the Maxwell stress, the force, \( f \), (per unit area) on the plate at \( x = a \) can be expressed by (here we assume that each surface has a unit thickness out of plane):

\[
f = -\frac{\varepsilon_0}{2} \left( \frac{d\phi}{dx} \right)^2 + \frac{\varepsilon_0 \sigma^2}{2 \ell_D^2},
\]

(7)

which can be evaluated at any point \( 0 \leq x \leq a \). Introduce the normalization

\[
\phi = \frac{\varepsilon_0 \sigma}{\ell_D}, \quad \tilde{x} = \frac{x}{\ell_D}, \quad \tilde{a} = \frac{a}{\ell_D}, \quad \tilde{d}\phi = \frac{\varepsilon_0 d\sigma}{\sigma} dx, \quad \tilde{f} = \frac{\varepsilon_0 f}{\sigma^2},
\]

\[
\hat{E} = \frac{\varepsilon_0 E}{\sigma^2 \ell_D},
\]

(8)

where \( E \) is the interaction energy per unit area. The potential between plates with the same charge density, Eq. (5), becomes

\[
\phi(\tilde{x}) = \frac{\cosh(\tilde{a}/2 - \tilde{x})}{\sinh(\tilde{a}/2)};
\]

(5a)

and the force per unit area, Eq. (7), becomes

\[
\tilde{f} = \frac{\tilde{\phi}^2}{2} - \frac{1}{2} \left( \frac{d\tilde{\phi}}{d\tilde{x}} \right)^2.
\]

(9)

For plates with equal charge densities, we use Eq. (5a) in Eq. (9), evaluated at \( \tilde{x} = \tilde{a}/2 \), where, by symmetry, \( (d\tilde{\phi})/(d\tilde{x}) = 0 \). Then,

\[
\tilde{f}_R = \frac{\tilde{\phi}^2}{2} = \frac{1}{2 \sinh^2(\tilde{a}/2)} = \frac{1}{2} \text{csch}^2(\tilde{a}/2).
\]

(10)

where the subscript “\( R \)” denotes “repulsion,” since the force on the plate at \( x = a \) is always positive, i.e., the interaction is always repulsive. The interaction energy can be computed by calculating the work done (per unit area) to move one of the two plates from a very long distance to the position \( x = a \):

\[
E \equiv \frac{\text{Work}}{\text{Area}} = -\int_0^a \tilde{f} d\tilde{a}'.
\]

(11)

Also, in normalized form:

\[
\hat{E} = \frac{\varepsilon_0 E}{\sigma^2 \ell_D} = -\int_0^{\tilde{a}} \tilde{f} d\tilde{a}'.
\]

(12)

Therefore, the repulsive energy per unit area can be calculated by

\[
\hat{E}_R = \frac{1}{2} \int_\infty^{\tilde{a}} \text{csch}^2\left(\frac{\tilde{a}}{2}\right) d\tilde{a}' = \frac{2}{e^{\tilde{a}^2} - 1},
\]

(13)

For equal and opposite charge densities, \( \sigma_2 = \sigma; \sigma_1 = -\sigma \). Following the same procedure, we find that at \( (x = a)/2 \), the potential is zero so that,

\[
\hat{f}_A = -\frac{1}{2} \left( \frac{d\tilde{\phi}}{d\tilde{x}} \right)^2 = -\frac{1}{2} \text{sech}^2\left(\frac{\tilde{a}}{2}\right),
\]

(14)

where the subscript “\( A \)” denotes “attraction,” since the interaction between opposite-charged surfaces is attractive. The normalized work of bringing the surfaces together is

\[
\hat{E}_A = \frac{1}{2} \varepsilon_0 \int_0^{\tilde{a}} \text{sech}^2\left(\frac{\tilde{a}}{2}\right) d\tilde{a}' = -\frac{2}{e^{\tilde{a}^2} + 1}.
\]

(15)

where \( \hat{E}_A \) is the attractive energy per unit area.

III. INTERACTION BETWEEN RIGID SURFACES WITH STRIPED PATTERNS OF CHARGE

As a first model for complementary surfaces we consider two surfaces, each with striped patterns of alternating positive and negative surface charge, and each with zero net charge (Fig. 3). These two surfaces are set some distance, \( a \), apart from each other. The width of each charged strip on the upper surface is \( b \); that of strips on the lower surface is \( c \). In this paper, we assume both \( b \) and \( c \) to be much larger than the separation, \( a \). In an experimental realization, the characteristic length scale of the gap, \( a \), is expected to be the Debye screening length that is on the order of a few nm. On the other hand, length scales \( b \) and \( c \), if generated by

![FIG. 2. Force and interaction energy per unit area between two uniformly charged surfaces as a function of distance between them. For small separation distance, both force and energy of repulsion diverge to infinity whereas the force and energy of attraction remain finite. With increasing separation distance repulsion and attraction both decrease in magnitude, becoming equal and opposite for large separation.](image)
microcontact printing, will have characteristic dimensions in the microns. Also, we imagine patterning an extended surface such that the length of the striped surface will be in centimeters, much larger than length scales \( b \) and \( c \). If the lateral length scales, \( b \) and \( c \) are not much larger than the gap, \( a \), the problem of determining electrostatic interactions is two-dimensional and is considered in the companion manuscript.\(^{31}\)

If each charged strip faces a strip with charge of opposite sign, we have a perfectly matched pair of complementary surfaces that we expect would adhere well to each other. In another limit, let us say that the lateral size of the strips on one of the surfaces is very different from the other, e.g., \( a \ll b \ll c \). Then, each strip of width “\( c \)” on the lower surface is opposed by a surface with net zero charge. As we will show, in such a case we expect overall repulsion between the two surfaces. These two simple cases suggest that this family of surfaces should exhibit high selectivity in adhesion.

### A. Electrostatics between two rigid surfaces

We begin by regarding the two surfaces as rigid and asking how the interaction force and energy between two surfaces with striped patterns of charge depends on the geometrical parameters such as stripe dimensions and separation between the surfaces. Since \( a \) is assumed to be much smaller than either \( b \) or \( c \), we can assume that the interaction force and energy is dominated by 1D interactions computed in the previous section. Specifically, we assume that the 2D state near the region where charge density switches carries negligible contributions to the overall force and interaction energy. Therefore, in regions where similar charges face each other, the force and interaction energy per unit area are computed using Eqs. (10) and (13), respectively. Where opposite charges face each other the force and interaction energy per unit area are computed using Eqs. (14) and (15), respectively.

Suppose that the charged strips have been distributed as shown in Fig. 3 with zero net charge on both surfaces. Consider a family of surfaces such that the lengths \( b \) and \( c \) are related as

\[
mb = nc, \quad (16)
\]

where \( m \) and \( n \) are both positive integers and have no common factors other than unity. Assume that the surfaces are extended indefinitely. The surface then comprises repeats of a periodic unit cell with length \( L_c = 2mb = 2nc \).

Let \( z \) be the fraction of area within a unit cell where like charges face each other. In the remaining fraction of the unit cell, \( 1-z \), charges are equal and opposite on two sides of the gap. Then, the net force and electrostatic interaction energy per unit area are

\[
\begin{align}
\hat{f}_T &= \alpha \cdot \hat{f}_R + (1 - \alpha) \cdot \hat{f}_A, \\
\hat{E}_I &= \alpha \cdot \hat{E}_R + (1 - \alpha) \cdot \hat{E}_A. \quad (17a)
\end{align}
\]

Let us consider the situation where the two surfaces can adjust in the plane of the interface to find the configuration most favorable for adhesion (minimization of interaction energy). In the Appendix,\(^{32}\) we demonstrate that this favorable configuration is one in which the unit cell starts with segments on the opposite surfaces aligned with opposite charges. For example, if \( m = n = 1 \), the surfaces adjust so that \( z = 0 \), i.e., charges are equal and opposite throughout the interface. Again, Fig. 3 shows the configuration that minimizes interaction energy for \( 3b = 5c \). For arbitrary combinations of \( m \) and \( n \), we find:

1. If either \( m \) or \( n \) is even, \( \alpha = 1/2 \). This is because in the second half of the unit cell the charge pattern on the side with even divisions is repeated whereas on the side with odd divisions it is reversed. Therefore, the minimum interaction energy and the corresponding force are

\[
\begin{align}
\hat{E}_I &= \frac{1}{2} \hat{E}_A + \frac{1}{2} \hat{E}_R = \frac{2}{e^{2\alpha} - 1}, \quad (18a)
\hat{f}_T &= \frac{1}{2} \hat{f}_A + \frac{1}{2} \hat{f}_R = 1 / \sinh^2(\alpha). \quad (18b)
\end{align}
\]

Since \( \{1/\sinh^2(\alpha)\} > 0 \), the overall interaction between such two surfaces is always repulsive.

2. If \( m \) and \( n \) are both odd, interaction energy is minimized if each unit cell begins with aligned regions of opposite charge, for example, as shown in Fig. 3. Also, we demonstrate in Appendix that \( \alpha = (mn - 1)/(2mn) \). Thus, \( \alpha \) is always less than 0.5 for this case. The interaction energy and force are

\[
\begin{align}
\hat{E}_I &= \alpha \cdot \hat{E}_R + (1 - \alpha) \cdot \hat{E}_A = \frac{mn + 1}{2mn} \hat{E}_A + \frac{mn - 1}{2mn} \hat{E}_R \\
&= \frac{2(mn - e^{\alpha})}{mn(e^{2\alpha} - 1)}, \quad (19)
\hat{f}_T &= \alpha \cdot \hat{f}_R + (1 - \alpha) \cdot \hat{f}_A = \frac{mn + 1}{2mn} \hat{f}_A + \frac{mn - 1}{2mn} \hat{f}_R \\
&= \frac{1}{mn} \left[ \frac{mn - \cosh(\alpha)}{\cosh(2\alpha) - 1} \right]. \quad (20)
\end{align}
\]

It is clear from Eq. (19) that at large distances \( \hat{E}_I = (-2e^{-\alpha})/(mn) \); the interaction is attractive and decreases in magnitude exponentially with distance. As distance \( \alpha \to 0 \) energy and force both diverge to +\( \infty \), and there
is always repulsion, unless \( m = n = 1 \), in which case normalized interaction energy and the interaction force go to a limiting value of \(-1\) and \(-1/2\), respectively. Therefore, except when \( m = n = 1 \), there is always an equilibrium distance \( \hat{a}_e \) at which force is zero and where the two surfaces will come to rest naturally. Using Eq. (20), this condition is

\[
\hat{f}_T = x \cdot \hat{f}_A(\hat{a}_e) + (1 - x) \cdot \hat{f}_A(\hat{a}_e) = x \cdot \frac{1}{2} \cdot \cosh^2\left(\frac{\hat{a}_e}{2}\right) - (1 - x) \cdot \frac{1}{2} \cdot \text{sech}^2\left(\frac{\hat{a}_e}{2}\right) = 0,
\]

where we have used Eqs. (10) and (14). The dimensionless distance \( \hat{a}_e \) can be obtained by solving Eq. (21) (Fig. 4):

\[
\hat{a}_e = 2 \text{csch}^{-1}\left(\sqrt{\frac{1 - 2x}{x}}\right).
\]  

Equation (22) provides the general relation between equilibrium separation \( \hat{a}_e \) and fraction, \( x \). For the special case under consideration in this section, \( x = (mn - 1)/(2mn) \) and

\[
\hat{a}_e = \cosh^{-1}(mn).
\]

Substituting Eq. (22) or (23) into Eq. (19) give the interaction energy at equilibrium, \( \hat{E}_e \). For two rigid surfaces, this quantity (if negative) is defined as the adhesion energy.

For the special case of \( m = n = 1 \), \( \hat{a}_e = 0 \), i.e., the force is always attractive, and within this model the surfaces come to rest in contact. Equation (23) shows that, as the product \( mn \) increases, so does the equilibrium distance. Figure 5 plots the interaction force and energy versus \( \hat{a}_e \) for the case of \( m = 3; n = 1 \).

When both \( m \) and \( n \) are odd, the pattern of surface charges in the second half of a unit cell is the reverse of that in the first half on both surfaces. Consider, for example, Fig. 3 where \( m = 3, n = 5 \). Therefore for each region in the first half of the unit cell where the charges have the same sign, the corresponding region in the second half also has the same charge on the two sides.

Similarly, for each region in the first half with oppositely charged surfaces, the corresponding region in the second half also has oppositely signed charges. Due to this symmetry of electrostatic interactions, we only need to calculate the force and energy within half of one such unit cell. In addition, when later we allow the surfaces to deform, the node at the middle of the unit cell must remain fixed by symmetry.

**B. Numerical results and discussion**

In Fig. 6 we show results for the adhesion energy per unit area, \( \hat{E}_f \), for different combinations of \( m, n \). Recall that when \( m,n \) are even numbers the surfaces repel so there is no adhesion. Normalized adhesion lies in the range [-1, 0]; greater adhesion is represented by a more negative number. Each square represents a single \( m,n \) combination and is colored using the non-linear scale shown to the right of the figure. The results of the case where the two surfaces are rigid and \( m > n \) are shown below the diagonal of this matrix. The results shown above the diagonal will be discussed later.

From Fig. 6, it is clear that \( m=n \) represents the highest adhesion. Other combinations that show adhesion are \( 3n = m, \) \( 5n = m, \) \( 7n = m, \ldots \), and \( 5n = 3m, \) \( 7n = 3m, \) and so on. Note that configurations very “close” to each other in this space can have very different adhesion energy. For example, the combination \( (15, 5) \) is adhesive but \( (15, 4) \) is repulsive. Even more dramatically, the combination \( (20, 20) \) is highly adhesive but \( (20, 19) \) is repulsive. (More strictly, the equilibrium separation \( \hat{a}_e \) increases to infinity for cases such as \( (20,19) \) that are repulsive. Therefore, the corresponding “adhesion” in these cases vanishes.) Therefore, this simple motif of two rigid surfaces patterned with alternating stripes of equal and opposite charge is predicted to show strong selectivity of adhesion to surfaces with certain specific characteristic length, and to show overall repulsion against most other surfaces.
IV. Interaction Between Elastic Surfaces with Striped Patterns of Charge

So far we have considered rigid charged surfaces. This led to a conclusion that surfaces with patterned charges will show high selectivity. However, one might ask: how is it possible that two surfaces with, say \( m=1000 \), \( n=999 \), should have strong repulsion from each other while another two with \( m=n=1000 \) attract each other strongly? A related, practical question is, what use is such selectivity if the slightest misalignment or difference in dimensions will destroy adhesion? We notice that with just a small amount of deformation, \((1000,999)\) can be transformed into \((1000,1000)\), and this suggests that there exists a strong electrostatic driving force for such a deformation. That is, selectivity is bound to be limited and conditioned by deformability. To explore this idea, we have created a simple model described below.

A. One-dimensional model of elastically deformable surfaces patterned with stripes of charge

In this 1D problem, we start with the two surfaces introduced above, each with striped patterns of alternating positive and negative surface charges summing to zero net charge and aligned to be in phase, as defined previously. In addition, we allow nodes on both surfaces to move, but only sideways in the \( y \) direction, thus preserving the flatness of the dielectric layer. The movement of these nodes is driven by reduction in electrostatic free energy, and restrained by increase in elastic strain energy, until the system reaches a state of equilibrium. We assume that, although the materials on either side can deform (change the lengths on each side within every segment), the charge on each segment remains proportional to the current length, which means that the amount of surface charge can be changed by deformation. A physical realization of this assumption could be an elastomer in which additional surface groups would be exposed if the area of the surface were increased.

Suppose we have two-plane surfaces (Fig. 7). The upper surface has \( m \) segments with alternating positive and negative charges and \((m+1)\) nodes. Similarly, the lower surface has \( n \) segments with alternating charge, a different segment length, and \((n+1)\) nodes. We mark each segment by a Roman number and each node in an Arabic number. Let \( y(i) \) and \( u(i) \) be the location and displacement of node \( i \), respectively \((1 \leq i \leq m+n+2)\). We define displacement \( u(i) \) to be positive if oriented in the positive \( y \) direction. We analyze the family of surfaces given by Eq. (16). We generally analyze a half of a unit cell arranged such that the first regions on opposite sides of the gap, starting at the left, have opposite charge. As discussed earlier, this ensures that the starting configuration has the lowest interaction energy and greatest adhesion energy for rigid surfaces. Therefore, as shown in Fig. 7, \( y(1) = y(m+2) \) and \( y(m+1) = y(m+n+2) \). Moreover, we set \( u(1) = u(m+1) = u(m+2) = 0 \), i.e., while we allow deformations within a unit cell we do not allow any overall change in area.

FIG. 6. (Color online) Plots of interaction energy between two patterned surfaces for different \( m \) and \( n \). (Negative values represent adhesion.) Each square represents a single \((m,n)\) combination and is colored using the (non-linear) scale shown to the right of the figure. Results below the diagonal, i.e., for \( m>n \), are for two rigid surfaces; those above the diagonal \((m<n)\) are for a deformable surface with normalized stiffness.

Note that the best combination is for \( m=n \). Combinations of \( m \) and \( n \) on some other rays emanating from the origin, such as \( m/n=3/1 \) or \( 1/3 \), \( m/n=3/5 \) or \( 5/3 \), etc. also give rise to net attraction. Note that for rigid surfaces even the slightest variation from one of these combinations results in strong repulsion. For example, while \((20,20)\) results in the highest adhesion, \((20,19)\) results in net repulsion. There is therefore strong selectivity of one surface for another. The effect of deformability, shown above the diagonal reduces strong selectivity.

To estimate the sort of adhesion energies predicted by our model, let’s consider an example of two rigid surfaces with a realistic charge density of one elementary charge per square nanometer, which is about 0.16 Cs per square meter. Suppose the surfaces are placed in an aqueous solution at 300 K and one mM concentration of a 1-1 electrolyte (e.g., NaCl), for which Debye screening length \( l_D \) is about 10 nm. According to Fig. 6 and the normalization [Eq. (8)], the highest energy (adhesion) is achieved when \( (m,n) = (1,1) \), with a value that is approximately 361 mJ/m². For the case of \((3,1)\) combination, the equilibrium distance between the surfaces is found to be about 18 nm, and the total energy (adhesion) is only about 42.5 mJ/m², which is much smaller than for the \((1,1)\) combination. For case \((5,1)\), the equilibrium distance increases to 23 nm and the adhesion energy decreases to about 34.6 mJ/m².

FIG. 7. (Color online) Schematic diagram of a 1D elastic surface with patterned charge (half unit cell: \( 3b = 5c \)).
It turns out to be important to handle potentially large deformations of the surfaces to prevent collapse of two neighboring nodes. For this reason, we model the surfaces as one-dimensional hyperelastic neo-Hookean materials.\textsuperscript{27,28} For small deformations, the behavior is linearly elastic.

\section{Model and algorithm}

\subsection{Electrostatic energy}

Consider a general node \( i \), say on the upper surface. Holding all other nodes fixed, motion of this node \( i \) by \( u(i) \) changes the electrostatic energy by a magnitude \( U_{\text{ele}} = |(-E_R + E_A)u(i)| \) (for unit out-of-plane width), as long as the motion is small enough so that the node does not cross another node on the lower surface. Define dimensionless displacement as \( \tilde{u}(i) = u(i)/l_D \). The sign of the electrostatic energy change (positive for increasing repulsion; negative for increasing attraction) can be obtained by examining the sign of charge density on the two sides in the segments adjacent to node \( i \) and the segment opposite to node \( i \).

Define \( \text{sign(opp)} \) as the sign of the segment opposite the node \( i \), and \( \text{sign(local)} \) as the sign of the step in charge density moving from the segment to the left of node \( i \) to its right (see Table I for examples).

Based on these quantities, for each node \( i \), the change of dimensionless electrostatic energy after deformation is:

\[ \tilde{U}_{\text{ele}} = \frac{U_{\text{ele}} \epsilon_0}{\sigma^2 l_D} = (-\tilde{E}_R + \tilde{E}_A) \cdot \tilde{u}(i) \cdot \text{sign(opp, } i) \cdot \text{sign(local, } i). \quad (24) \]

The total change in electrostatic energy due to motion of nodes is obtained by summing up the contribution from each node:

\[ \tilde{U}_{\text{ele}} = \sum_{i=1}^{n+m+2} \Delta \tilde{E}(i) \cdot \tilde{u}(i) \cdot \text{sign(opp, } i) \cdot \text{sign(local, } i), \quad (25) \]

where \( \Delta \tilde{E}(i) = (-\tilde{E}_R + \tilde{E}_A) \).

\subsection{Elastic energy}

As an illustrative model to capture the penalty of imposing deformations, we model segments on both sides as springs under uniaxial strains imposed by differences in displacements on its two ends. The entire system is therefore composed by two strips of linked springs (the upper one has \( n \) springs in series, and the lower one has \( m \) springs in series).

\begin{table}[h]
\centering
\caption{Examples of arrangements for a given node, \( i \), and definition of terms sign(opp) and sign(local) that determine the change in electrostatic energy per unit movement of node \( i \).}
\begin{tabular}{|c|c|}
\hline
Position of node \( i \) & Signs \\
\hline
\( \bullet \) & + \\
\hline
\( - \bullet \) & - \\
\hline
\( \bullet \) & + \\
\hline
\( - \bullet \) & - \\
\hline
\end{tabular}
\end{table}

This model suppresses other deformation modes such as out of plane deformation which will change the electrostatic interaction. Nevertheless, we believe that this simple model captures the beneficial effect of deformation on adhesion.

Let \( L_{0,i} \) and \( L_i \) be the initial and stretched lengths for a segment \( i \), respectively. The stretch ratio for this segment is,

\[ \lambda_i = \frac{L_i}{L_{0,i}} = \frac{L_{0,i} + u_+ - u_-}{L_{0,i}} = \frac{L_{0,i} + \tilde{u}_+ - \tilde{u}_-}{L_{0,i}}, \quad (26) \]

where \( u_+ \) is the displacement of the node at the right end of segment \( i \) and \( u_- \) is the displacement of the node at its left end. All \( u_+, u_- \) and \( L_{0,i} \) are normalized by the Debye screening length \( l_D \):

\[ \left( \tilde{u}_+ = \frac{u_+}{l_D}, \tilde{u}_- = \frac{u_-}{l_D} \text{ and } L_{0,i} = \frac{L_{0,i}}{l_D} \right). \]

We model the spring as comprised of a Neo-Hookean solid for which the strain energy density is given by\textsuperscript{27,28}

\[ \Phi = Y(l_1 - 3)/6, \quad (27) \]

\[ l_1 = \lambda^2 + \frac{2}{\kappa}, \quad (28) \]

where \( Y \) is the small strain Young’s modulus.\textsuperscript{27} Substituting Eq. (28) into Eq. (27), the expression for strain energy density becomes

\[ \Phi = Y \left( \lambda^2 + \frac{2}{\kappa} - 3 \right) / 6. \quad (29) \]

For small deformations, this model reduces to Hooke’s law for a linear spring. For large deformations, it is nonlinear. Eq. (29) shows that the spring stiffens indefinitely under compression, removing the unphysical behavior predicted by a linear model, in which neighboring nodes can collapse.

In this model, the spring cannot be compressed indefinitely (\( \lambda \rightarrow 0 \)) since the strain energy density becomes unbounded in this limit. For any segment \( i \), the elastic energy can be calculated by (in dimensionless form)

\[ \tilde{U}_{\text{elas,}} = \tilde{\Phi} \tilde{L}_{0,i} \tilde{A}_0 = \tilde{Y} \left( \tilde{\lambda}_l^2 + \frac{2}{\tilde{\lambda}_l} - 3 \right) \cdot \tilde{L}_{0,i} \tilde{A}_0 / 6, \quad (30) \]

where \( \tilde{U}_{\text{elas}} = [(\epsilon \epsilon_0)/(\sigma^2 l_D)]U_{\text{elas}} \) and \( \tilde{Y} = [(\epsilon \epsilon_0 l_D)/\sigma^2]Y \) is the normalized modulus. The initial area of cross-section, \( A_0 \), is normalized by \( l_D^2 \) \( A_0 = A_0 / l_D^2 \) and we assume that \( A_0 \) is the same for every segment.

The total elastic energy in dimensionless form is

\[ \tilde{U}_{\text{elas}} = \sum_{i=1}^{n+m+2} \tilde{Y} \left( \tilde{\lambda}_l^2 + \frac{2}{\tilde{\lambda}_l} - 3 \right) \tilde{L}_{0,i} \tilde{A}_0. \quad (31) \]

For small strains, since \( \tilde{\lambda}_l \approx 1 \) for each segment, the expression for elastic energy is approximately:

\[ \tilde{U}_{\text{elas}} \approx \sum_{i=1}^{n+m+2} \frac{1}{2} \left( \frac{\tilde{U}_{\text{elas}}}{\tilde{L}_{0,i}} \right) \left( \tilde{u}_+ - \tilde{u}_- \right)^2. \quad (32) \]

Compare this to the case of a linear spring.
where \( k_i \) is the spring constant for segment \( i \) \((i = I, II, \ldots, m + n)\), which has the dimensionless form as \( k_i = [(\varepsilon_0 \varepsilon_r)/\sigma^2]k_i \).

3. Equilibrium

The total energy consists of electrostatic and elastic contributions,

\[
\bar{U}_{tot} = \bar{U}_{ele} + \bar{U}_{ela} .
\]

In equilibrium, for a given separation \( \hat{\text{a}} \), the first derivative of the total energy with respect to each degree of freedom should vanish:

\[
\frac{\partial \bar{U}_{tot}}{\partial \hat{u}_i} = \frac{\partial (\bar{U}_{ele} + \bar{U}_{ela})}{\partial \hat{u}_i} = \frac{\partial \bar{U}_{ele}}{\partial \hat{u}_i} + \frac{\partial \bar{U}_{ela}}{\partial \hat{u}_i} = 0 .
\]

According to Eq. (25),

\[
\frac{\partial \bar{U}_{ele}}{\partial \hat{u}_i} = \Delta \dot{E}(i) \cdot \text{sign(opp, } i \text{)} \cdot \text{sign(local, } i \text{)} .
\]

The contribution to Eq. (35) from elastic energy generally is from each of the elements adjoining the node \( i \). In our case, except for nodes at the ends that are handled by boundary conditions, each node has contributions from two terms in Eq. (31), which are \( \bar{U}_{ela,i-1} \) and \( \bar{U}_{ela,i} \) [see Eq. (30)]. So the derivative of elastic energy can be calculated by,

\[
\frac{\partial \bar{U}_{ela}}{\partial \hat{u}_i} = \frac{\partial \bar{U}_{ela,i-1}}{\partial \hat{u}_i} \frac{\partial \hat{u}_{i-1}}{\partial \hat{u}_i} + \frac{\partial \bar{U}_{ela,i}}{\partial \hat{u}_i} \frac{\partial \hat{u}_i}{\partial \hat{u}_i}
\]

\[
= \frac{\hat{Y} \hat{A}_0}{L_{0,i-1}} \left( \frac{1}{\lambda_{i-1}^2} - \lambda_{i-1} + \frac{1}{\lambda_i} \right) / 3 .
\]

For small strains, the linearized form of Eq. (37) is

\[
\frac{\partial \bar{U}_{ela}}{\partial \hat{u}_i} \approx \frac{\hat{Y} \hat{A}_0}{L_{0,i-1}} (\hat{u}_i - \hat{u}_{i-1}) - \frac{\hat{Y} \hat{A}_0}{L}\hat{u}_i + \hat{A}_0 (\hat{u}_{i+1} - \hat{u}_i)
\]

\[
= \hat{k}_i (\hat{u}_i - \hat{u}_{i-1}) - \hat{k}_i (\hat{u}_{i+1} - \hat{u}_i) .
\]

The right hand side of Eq. (38) is the same as the first derivative of the elastic energy for small displacements [Eq. (33)] with respect to \( \hat{u}_i \).

Substituting Eq. (36) and Eq. (37) into Eq. (35), we find that for each node \( i \)

\[
\Delta \dot{E}(i) \cdot \text{sign(opp, } i \text{)} \cdot \text{sign(local, } i \text{)}
\]

\[
+ \frac{\hat{Y} \hat{A}_0}{L_{0,i-1}} \left( \frac{1}{\lambda_{i-1}^2} - \lambda_{i-1} + \frac{1}{\lambda_i} \right) / 3 = 0,
\]

which represents a system of nonlinear equations for the unknown nodal displacements subject to boundary conditions \( \hat{u}_1 = \hat{u}_{m+1} = \hat{u}_{m+2} = \hat{u}_{m+n+2} = 0 \). Note that if we take the material on the two sides to be the same, the normalized set of Eqs. (39) depend on a single dimensionless parameter, \( \hat{k}_0 \equiv Y \hat{A}_0 \) since \( \Delta \dot{E}(i) \) is a function only of the gap \( \hat{a} \) through Eqs. (13) and (15).

We solve the system of Eq. (39) for fixed \( \hat{a} \) using a viscous relaxation technique. Define the left-hand side of Eq. (39) to be the residual, and introduce a fictitious viscous term in the equation such that the rate of change of displacements is proportional to the residual:

\[
\dot{\hat{u}}_i = \Delta \dot{E}(i, t) \cdot \text{sign(opp, } i \text{)} \cdot \text{sign(local, } i \text{)}
\]

\[
+ \hat{k}_0 \left( \frac{1}{\lambda_{i-1}^2} - \lambda_{i-1} + \frac{1}{\lambda_i} \right) / 3 .
\]

Approximating \( \dot{\hat{u}} = (\hat{u}_{i+\Delta t} - \hat{u}_i)/\Delta t \), (40) becomes

\[
\hat{u}_{i, i+\Delta t} = \Delta t \cdot \hat{\eta}^{-1} \left[ \Delta \dot{E}(i, t) \cdot \text{sign(opp, } i \text{)} \cdot \text{sign(local, } i \text{)}
\]

\[
+ \hat{k}_0 \left( \frac{1}{\lambda_{i-1}^2} - \lambda_{i-1} + \frac{1}{\lambda_i} \right) / 3 \right] + \hat{u}_i .
\]

Equation (41) is marched forward in time with a sufficiently small time step to ensure stability until we obtain a solution with residual smaller than a specified tolerance. Two sorts of events need special handling. The first occurs if the deformation causes a node on one surface to cross a node on the other surface. Because such an event results in a sudden change in sign (opp, \( i \)), often the solution will not converge. We handle such events by introducing a transition function that smoothly changes the value of sign (opp, \( i \)). The second event occurs if a linear elastic model is used to represent deformation and if the electrostatic driving forces dominate over the elastic resistance. Under such a circumstance, some springs can be compressed to zero length, i.e., nodes can cross on the same side. In the Neo-Hookean model such a situation does not arise but for the linearly elastic model it can, and we terminate the computation when it does since node crossing on the same side violates the condition of small deformations and is not a physically meaningful event.

Once we have obtained new displacements for every node, we compute the area fraction of repulsive interactions, \( \alpha \), and then find a new value of \( \hat{a} \) using Eq. (22). This procedure is repeated iteratively until we achieve a converged equilibrium solution in which the total energy is minimized with respect to all \( \hat{u}_i \) and separation, \( \hat{a} \).

C. Analysis of a simple case, \((m, n) = (1, 3)\)

We begin by examining a simple example in some detail. Consider the combination of \( m = 1 \) and \( n = 3 \) (Fig. 8) and let \( L = 1 \). Because the two ends are fixed for both sides, only two nodes on the lower surface are free to deform and, by symmetry, they must have equal and opposite deformations, defined as \( \delta \) (dimensionless).

In this case, the upper surface does not deform, so its elastic energy remains fixed at zero. The total energy is the summation of the electrostatic energy and elastic energy of the lower surface:

\[
\bar{U}_{tot} = \bar{U}_{ele} + \bar{U}_{ela} .
\]
available to propagate an interfacial crack. Figure 9 also shows how the total energy (electrostatic and elastic), identified here with the adhesion energy of the system, decreases as the deformation increases.

D. How deformability affects selectivity

We now examine the entire family of surfaces given by Eq. (16) and return to the questions raised at the beginning of Sec. IV. To explore how deformability affects the adhesion selectivity seen in the right lower half of Fig. 6 ($m > n$), we vary the elasticity by changing the parameter $\tilde{k}_0$. For convenience, in the following discussion, we take the total dimensionless length of each surface equal to 1, ($L_{\text{tot}} = 1$). As an example, Fig. 10 shows initial and final nodal positions for a $(3,5)$ surface for two different values of stiffness as predicted by the neo-Hookean model.

In Fig. 6 we show results of the total adhesion energy, $\tilde{E}_e$, for different $(m,n)$ after deformation using the neo-Hookean model with $\tilde{k}_0 = 10$. These results are presented above the diagonal of this matrix, i.e., for $m < n$. Again, $m = n$ represents the highest adhesion achievable. However, we notice that deformability has a profound influence on selectivity. It “smooths out” the sharp selectivity predicted for rigid surfaces. Specifically, “compositions” close to the selective ones such as $(1,3)$ and $(3,5)$ in the space of $m$ and $n$, find favorable modes of deformation to increase their adhesion. This is particularly true for compositions near $m = n$.

At the end of Sec. III, we showed that for realistic charge densities in a $1 \text{ mM } 1\text{-I}$ electrolyte the $(1,1)$ combination is predicted to have significant adhesion but that this reduces significantly for other combinations; the second best $(3,1)$ is nearly a factor of ten lower in adhesion. To illustrate the beneficial effect of deformability, let us consider the same example where now the patterned charge is on a deformable elastomer instead of a rigid surface. Young’s modulus of a representative elastomer, e.g., poly-dimethylsiloxane (PDMS), is about 5 MPa. If the dielectric has a thickness of 200 microns and width of about 3.6 mm, the parameter $\tilde{k}_0 \approx 10$.

From the region of the Fig. 6 above the diagonal, we observe that the maximum adhesion is still given by case $(1,1)$, and its value is the same as for rigid surfaces. However, for other combinations, the deformability allows greater adhesion. For example, for the case $(1,3)$, the total energy increases a little, to about $65 \text{ mJ/m}^2$. Much greater changes in adhesion are predicted when either $m$ or $n$ [Eq. (16)] is even, especially for combinations “near” the...
diagonal in Fig. 6. Recall that for the rigid case, if either \( m \) or \( n \) are even, the net force between the surfaces is repulsive. The originally repulsive case (4, 5) becomes adhesive and the adhesion energy is about 59 mJ/m²; the case (19, 20) has adhesion energy of 216 mJ/m², which is a great enhancement of adhesion and approaches that of (1, 1).

V. SUMMARY AND CONCLUSIONS

To explore whether and how adhesion selectivity can be designed using electrostatic complementarity, we have analyzed the interactions between two surfaces patterned with stripes of charge. We have shown that strong adhesion can be achieved by exact complementarity between two such pattern-charged surfaces. If the materials are rigid, we predict strong selectivity, which can be rather more a bane than a benefit because even a slight difference in dimensions or misalignment can drastically reduce adhesion. We show that deformability of the materials, modeled here by non-linear (neo-Hookean) elasticity, strongly alters adhesion selectivity. Specifically, by allowing the surfaces to deform, compositions similar to each other are found to have similar adhesion. The kind of striped charges we have analyzed can be achieved experimentally by microcontact printing of appropriate molecules onto an elastomer. We have specifically analyzed the case where charges result from ionization of terminal groups on such molecules in aqueous medium. As such, we expect that the predictions of our work are eminently realizable experimentally.

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APPENDIX: ELECTROSTATIC INTERACTION BETWEEN TWO SURFACES WITH STRIPES OF CHARGE

Equations (17) in the main text show that the total electrostatic interaction energy and force (per unit area) between two surfaces patterned with stripes of charge are known once we know \( a \), the fraction of area within a unit cell where like charges face each other. Here we consider the family of surfaces defined by \( nb = mc \), where \( n \) and \( m \) are both odd positive integers and \( n > m \), \( b \) and \( c \) are lengths of unit elements on the two sides, and \( b < c \). We demonstrate that (a) the strongest electrostatic interaction occurs when the top and bottom surfaces begin with aligned opposite charged regions, and (b) the fraction of area with oppositely charged surfaces is

\[
\alpha = \frac{mn - 1}{2mn}.
\]

(A1)

We have already shown that if either \( m \) or \( n \) is even, \( \alpha = 1/2 \).

Consider a unit cell of length \( L \) in which, without loss of generality, the left end is aligned with the start of a negatively charged region on the lower surface. The lower region has \( m \) units of length \( c \) each. The upper surface is shifted by \( \delta \) in the positive “y” direction compared to a configuration in which it begins with a positively charged unit region. We will show that electrostatic attraction is strongest for \( \delta = 0 \). (We need to consider only \( \delta < b \)).

Let a negatively charged region be assigned the number “1” and a positively charged region the number “0.” The charge pattern on the lower surface can be represented by the following function:

\[
\phi_l(x) = H(x) - H(x - c) + \ldots - H[x - (2m - 1)c]
\]

\[
= \sum_{k=1}^{2m} (-1)^{k-1} H[x - (k - 1)c],
\]

(A2)

where

\[
H(x) = \begin{cases} 
1 & x \geq 0 \\
0 & x < 0
\end{cases}
\]

(A3)

This represents a function of the form:

\[
\text{FIG. 10. Initial and equilibrium nodal positions for a (3,5) surface. (a) The prediction of the neo-Hookean model for } k_0 = 15. \text{ For this relatively large stiffness, the deformation is small, and the associated reduction in energy is slight. (b) The nodal displacements when } k_0 = 3; \text{ the deformation is significantly larger.}
\]
The total length for which the two surfaces have identical charge is therefore

\[ L_A = L - L_A; \quad x = 1 - L_A/L. \] (A7)

By choosing \( L = 1 \) we can compute \( x \) by calculating \( L_A \). Now,

\[
L_A = \int_0^L [\phi_1(x) - \phi_2(x)]^2 \, dx
= \int_0^L [\phi_1^2(x) - \phi_2^2(x) - 2\phi_1(x)\phi_2(x)] \, dx. \] (A8)

However, it is clear from the graph that

\[
\int_0^L \phi_i^2(x) \, dx = \int_0^L \phi_i(x) \, dx = L/2 \quad i = 1, 2. \] (A9)

Equations (A8) and (A9) imply that

\[
L_A = L - 2 \int_0^L \phi_1(x)\phi_2(x) \, dx. \] (A10)

Using Eqs. (A2) and (A4), the integrand in Eq. (A10) can be written as

\[
\phi_1(x)\phi_2(x) = \{H(x) - H(x - c) + \cdots - H[x - (2m - 1)c]\} \{H(x) + H(x - b - \delta) - H(x - 2b - \delta)
+ \cdots + H[x - (2m - 1)b - \delta] - H(x - 2nb)\}
= H(x) \sum_{k=1}^{2m} (-1)^{k-1} H[x - (k - 1)c]
+ \left\{ \sum_{j=0}^{2n-1} (-1)^{j-1} H(x - jb - \delta) \right\} - H(x - c)
\times \left( \sum_{j=0}^{2n-1} (-1)^{j-1} H(x - jb - \delta) \right) + \cdots - H[x - (2m - 1)c]
\times \left( \sum_{j=0}^{2n-1} (-1)^{j-1} H(x - jb - \delta) \right) - H(x - 2nb) \left( \sum_{k=1}^{2m} (-1)^{k-1} H[x - (k - 1)c] \right). \] (A11)

The following integrals in Eq. (A11) are simple

\[
\int_0^L H(x) \sum_{k=1}^{2m} (-1)^{k-1} H[x - (k - 1)c] \, dx = L/2, \quad (A12a)
\]

\[
\int_0^L H(x - 2nb) \sum_{k=1}^{2m} (-1)^{k-1} H[x - (k - 1)c] \, dx = 0. \quad (A12b)
\]

From Eqs. (A10) and (A11), we need to evaluate integrals of the form

\[
I_k = \int_0^L H(x - kc) \sum_{j=0}^{2n-1} (-1)^{j-1} H(x - jb - \delta) \, dx,
\]

Note that

\[ k = 0, 1, 2, \ldots 2m - 1. \] (A13)
\[
\int_{0}^{L} H(x - \alpha)H(x - \beta)dx = \int_{0}^{L} H(x - \alpha)H(x - \beta)dx = \int_{\beta}^{L} H(x - \beta)dx = (L - \beta), \quad (A14)
\]
where \( L \geq B \geq \alpha \geq 0 \).

We have already assumed that
\[
c > b \Leftrightarrow \frac{n}{m} > 1.
\quad (A15)
\]

This means that
\[
kc = (p_k + \alpha b) \quad p_k = [kn/m] \quad 0 \leq \alpha_k < 1 \quad k = 1, 2, \ldots
\quad (A16)
\]
where \([ \cdot ]\) is the greatest integer function, e.g., \([18] = 1\).

Now,
\[
I_k = \sum_{j=0}^{2n-1} (-1)^{j-1} \int_{0}^{L} H(x - jb - \delta)H(x - (p_k + \alpha_k)b)dx
\]
\[
I_k = \sum_{j=0}^{p_k} (-1)^{j-1}[L - (p_k + \alpha_k)b] + \sum_{j=p_k+1}^{2n-1} (-1)^{j-1} \int_{0}^{L} H(x - jb - \delta)H(x - (p_k + \alpha_k)b)dx
\]
\[
= (L - kc) \sum_{j=0}^{p_k} (-1)^{j-1} + \sum_{j=p_k+1}^{2n-1} (-1)^{j-1}(L - jb - \delta).
\quad (A17)
\]
when \((\delta \leq \alpha_k b)\)

\[
I_k = \sum_{j=0}^{p_k} (-1)^{j-1}[L - (p_k + \alpha_k)b] + \sum_{j=p_k+1}^{2n-1} (-1)^{j-1} \int_{0}^{L} H(x - jb - \delta)H(x - (p_k + \alpha_k)b)dx
\]
\[
= (L - kc) \sum_{j=0}^{p_k} (-1)^{j-1} + \sum_{j=p_k+1}^{2n-1} (-1)^{j-1}(L - jb - \delta).
\quad (A18)
\]
when \((\delta > \alpha_k b)\)

Similarly,
\[
\sum_{j=p_k}^{2n-1} (-1)^{j-1}(L - jb - \delta)
\]
\[
= (L - \delta) \sum_{j=p_k}^{2n-1} (-1)^{j-1} + b \sum_{j=p_k+1}^{2n-1} (-1)^{j}.
\quad (A22)
\]

Now,
\[
(L - \delta) \sum_{j=p_k}^{2n-1} (-1)^{j-1} = \begin{cases} 0 & p_k \text{ odd} \\ (L - \delta) & p_k \text{ even} \end{cases},
\quad (A23)
\]
\[
(L - \delta) \sum_{j=p_k}^{2n-1} (-1)^{j-1} = \begin{cases} (L - \delta) & p_k \text{ odd} \\ 0 & p_k \text{ even} \end{cases}.
\quad (A24)
\]

Note that
\[
\sum_{j=1}^{2n-1} (-1)^{j} = -1 + (2 - 3) + (4 - 5) + \cdots
\]
\[
= + (2n - 2 - 2n + 1) = -n,
\quad (A25)
\]
\[
\sum_{j=1}^{p_k} (-1)^{j} = (p_k - 1)/2 - p_k = -(p_k + 1)/2 \quad \text{if } p_k \text{ is odd},
\quad (A26)
\]
\[
\sum_{j=1}^{p_k} (-1)^{j} = p_k/2 \quad \text{if } p_k \text{ is even},
\quad (A27)
\]
\[
\sum_{j=1}^{p_k} (-1)^{j} = -p_k/2 \quad \text{if } p_k \text{ is even},
\quad (A28)
\]

So
\[
\sum_{j=1}^{2n-1} (-1)^{j} - \sum_{j=1}^{p_k} (-1)^{j} = -n + (p_k + 1)/2 \quad \text{if } p_k \text{ is odd},
\quad (A30)
\]
\[
\sum_{j=1}^{2n-1} (-1)^{j} - \sum_{j=1}^{p_k} (-1)^{j} = -n - p_k/2 \quad \text{if } p_k \text{ is even},
\quad (A31)
\]
\[
\sum_{j=1}^{2n-1} (-1)^{j} - \sum_{j=1}^{p_k} (-1)^{j} = -n -(p_k - 1)/2 \quad \text{if } p_k \text{ is odd},
\quad (A32)
\]
\[
\sum_{j=1}^{2n-1} (-1)^{j} - \sum_{j=1}^{p_k} (-1)^{j} = -n + p_k/2 \quad \text{if } p_k \text{ is even}. \quad (A33)
\]

Thus,
\[
\sum_{j=p_k+1}^{2n-1} (-1)^{j} = -n + p_k/2 \quad \text{if } p_k \text{ is odd},
\quad (A34)
\]
FIG. 11. $(mn - 1)/(2mn)$ vs fraction of repulsive part for different $m$ and $n$ combinations.

$\frac{I_k}{2} = \frac{L}{2} + kc - b \cdot \frac{p_k - 1}{2} - \delta$ if $p_k$ is odd, 

$\frac{I_k}{2} = \frac{L}{2} + k c - b \cdot \frac{p_k - 1}{2} - \delta$ if $p_k$ is even, 

Combining (A10), (A11), and (A13) leads to the formula

$L_A = 2 \sum_{k=0}^{2m-1} (-1)^{k+1} I_k$. 

Figure 11 compares the value of $a$ computed by this method (with $\delta = 0$) with the relation $a = (mn - 1)/(2mn)$, showing the latter to be correct.

Figures 12 and 13 shows how $x$ varies with shift $\delta$. When $m = 1$, the area fraction $x$ is minimized when $\delta = 0$; this corresponds to strongest attraction. Figure 13 shows that there are several minima with varying $\delta$ when $m$ is larger than one, e.g., $m = 3, 5, 7, \ldots$. The fraction of $x$ becomes a periodic function of the shift $\delta$, and the minimum value of $x$ is achieved when

$\delta = 2k/mb$ if $k = 0, 1, \ldots, (m - 1)/2$. In the main text, we have picked $k = 0$.

27. R. Ogden, Non-linear Elastic Deformations (Dover, New York, 1984).
30. In the Appendix we show how to calculate the electrostatic interaction between two rigid surfaces with stripes of charge for arbitrary alignment between them.