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## Geometry of surface-mediated interactions

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**Abstract.** – Soft interfaces can mediate interactions between particles bound to them. The force transmitted through the surface geometry on a particle may be expressed as a closed line integral of the surface stress tensor around that particle. This contour may be deformed to exploit the symmetries present; for two identical particles, one obtains an exact expression for the force between them in terms of the local surface geometry of their mid-plane; in the case of a fluid membrane the sign of the interaction is often evident. The approach, by construction, is adapted directly to the surface and is independent of its parameterization. Furthermore, it is applicable for arbitrarily large deformations; in particular, it remains valid beyond the linear small-gradient regime.

*Introduction.* – Physical interactions between spatially separated particles are mediated by fields: matter interacts by curving spacetime itself; if it is charged it will interact through the electromagnetic field. Equally well, interactions of an indirect nature may be mediated by effective fields: for example, colloidal particles in suspension can interact by disturbing local ion densities (if they are charged) [1], by distorting the order of an embedding liquid crystal [2], or by locally phase-separating a binary mixture [3].

An important class of such interactions, purely geometrical in nature, are those between particles localized at an interface which are induced through the local deformation in its shape. The simplest example is provided by capillary interactions, originating in *surface tension* with an energy proportional to the excess area of the interface. These interactions play a key role in many technological processes, among them ore flotation and foam stabilization [4]. To describe the *compression* and *bending* of surfactant monolayers and lipid membranes [5–8], surface energies involving higher-order derivatives are required. For example, the interactions between membrane-bound proteins, which play an important role in cell biology [9], are described by an energy quadratic in the curvature of the membrane surface.

A major obstacle to providing a theoretical description of these surface-mediated interactions is that the corresponding field equations are nonlinear. It is easy to see why: while

the underlying surface free energy density may be simple —some quadratic invariant of generalized strains and thus “harmonic” —the description of the curved manifold it inhabits is intrinsically nonlinear. Even in the case of an essentially flat surface, where its description in terms of a height function (“Monge gauge”) keeping only the lowest order in gradients leads to approximate linear differential equations (and thus to Green functions), exact analytical solutions are difficult to obtain. This is because a straightforward superposition ansatz [10] will typically violate the correct boundary conditions. It remains entirely unclear how any linear result generalizes to the full nonlinear situation.

Despite this obstacle, substantial progress can still be made by recognizing that the determination of the interaction between two objects mediated by the surface involves solving two distinct problems: first, the particular way in which objects bind to the interface, together with its elastic specifications, determines its equilibrium (*i.e.* energy minimizing) shape —find that shape; second, equilibrium of the resulting complex generally requires additional external forces which constrain the bound objects at their positions —find these forces. Usually, one thinks of the second problem in a way which depends heavily on the successful implementation of the first: since the energy of the complex depends on the relative positions of the bound objects, appropriate derivatives of the energy with respect to their coordinates yield the forces one is looking for. Rarely, however, is it possible to write down this “potential” energy; as a result, this conceptually straightforward route to the forces is infeasible in practice. In other words, a solution to part 1 is generally impossible, leaving no further handle on part 2. This reasoning distracts from the fact that forces transmitted by an interface must be encoded directly in its shape; if the Hamiltonian describing the surface is geometrical, so also are the stresses underpinning this shape.

In this letter we reformulate the full nonlinear interaction problem in terms of surface geometry; the conserved covariant surface stress tensor [11] associated with the geometry will play a central role. We show how to express the surface-mediated force on a particle as a closed line integral of the stress tensor. By suitably deforming the contour to exploit the symmetries of the configuration, a remarkably simple and transparent expression for the force is obtained. In particular, the *sign* of the force may turn out to be evident using only very qualitative features of the geometry. We should mention that a stress tensor approach to membrane-mediated interactions was suggested by Kralchevsky *et al.* [6]. However, the full potential of a completely geometric description has not previously been exploited.

*Differential geometry, the energy, and the stress tensor.* – In this section we sketch the essential geometric background. While this will define our notation, it is most likely too concise to serve as a stand-alone introduction. For the geometry we therefore refer the reader to refs. [12]; the stress tensor is introduced at greater length in ref. [11, 13].

Consider a surface  $\Sigma$  embedded in three-dimensional Euclidean space  $\mathbb{R}^3$ , which is described locally by its position  $\mathbf{X}(\xi^1, \xi^2) \in \mathbb{R}^3$ , where the  $\xi^a$  are a suitable set of (curvilinear) local coordinates on the surface. Given the two tangent vectors  $\mathbf{e}_a = \partial\mathbf{X}/\partial\xi^a = \partial_a\mathbf{X}$  and the (unit) normal vector  $\mathbf{n} = \mathbf{e}_1 \times \mathbf{e}_2 / |\mathbf{e}_1 \times \mathbf{e}_2|$ , the surface geometry is described completely in terms of the induced metric  $g_{ab} = \mathbf{e}_a \cdot \mathbf{e}_b$  and the extrinsic curvature  $K_{ab} = -\mathbf{n} \cdot \partial_a \mathbf{e}_b$  [14]. We denote the metric-compatible covariant derivative by  $\nabla_a$  and the corresponding Laplacian as  $\Delta = \nabla_a \nabla^a$ . As usual, repeated indices —one up one down— imply a summation. The total curvature  $K$  is the trace of the extrinsic curvature,  $K = g^{ab} K_{ab}$  [15].

We now associate with the surface an energy which can be written as a surface integral over a scalar Hamiltonian density  $\mathcal{H}$  constructed out of local geometric invariants,

$$H_\Sigma[\mathbf{X}] = \int_\Sigma dA \mathcal{H}(g_{ab}, K_{ab}, \nabla_a K_{bc}, \dots), \quad (1)$$

TABLE I – Euler-Lagrange derivative  $\mathcal{E}(\mathcal{H})$  and components of the stress tensor  $\mathbf{f}^a = f^{ab}\mathbf{e}_b + f^a\mathbf{n}$  for several simple scalar surface Hamiltonian densities  $\mathcal{H}$ . Projecting  $\nabla_a\mathbf{f}^a = \mathcal{E}\mathbf{n}$  implies the identities  $\nabla_a f^a - K_{ab}f^{ab} = \mathcal{E}$  and  $\nabla_a f^{ab} - K_a^b f^a = 0$ .  $R$  is the intrinsic scalar curvature [15]. Notice that  $K^2$  and  $K^{ab}K_{ab}$  yield identical  $\mathcal{E}$  and  $\mathbf{f}^a$  (a consequence of the Gauss-Bonnet theorem [12]). These results may be derived using techniques developed in refs. [11, 13].

$\mathcal{H}$	$\mathcal{E}$	$f^a$	$f^{ab}$
1	$K$	0	$-g^{ab}$
$K^n$	$[R - (1 - \frac{1}{n})K^2 - \Delta]nK^{n-1}$	$-n\nabla^a K^{n-1}$	$(nK^{ab} - Kg^{ab})K^{n-1}$
$K^{ab}K_{ab}$	$[R - \frac{1}{2}K^2 - \Delta]2K$	$-2\nabla^a K$	$(2K^{ab} - Kg^{ab})K$
$\frac{1}{2}(\nabla K)^2$ $\equiv \frac{1}{2}(\nabla_c K)(\nabla^c K)$	$(\Delta + K^2 - R)\Delta K$ $-K^{ab}[(\nabla_a K)(\nabla_b K) - \frac{1}{2}g_{ab}(\nabla K)^2]$	$\nabla^a \Delta K$	$(\nabla^a K)(\nabla^b K)$ $-\frac{1}{2}g^{ab}(\nabla K)^2 - K^{ab}\Delta K$

where the infinitesimal surface element is  $dA = d^2\xi\sqrt{g}$  with  $g = \det(g_{ab})$ . Let us perform a variation  $\mathbf{X} \rightarrow \mathbf{X} + \delta\mathbf{X}$  of the embedding functions. The concomitant first variation of the functional (1) can be cast as a bulk part plus a pure divergence:

$$\delta H_\Sigma = \int_\Sigma dA \mathcal{E}(\mathcal{H}) \mathbf{n} \cdot \delta\mathbf{X} + \int_\Sigma dA \nabla_a Q^a. \quad (2)$$

Here,  $\mathcal{E}\mathbf{n}$  is the *bulk* Euler-Lagrange derivative of  $H_\Sigma$ , which is evidently purely normal. The second integral (which is identical to the *boundary integral* of  $Q^a$  over  $\partial\Sigma$ ) originates in the tangential variations as well as the derivatives of normal variations:

$$Q^a = -\mathbf{f}^a \cdot \delta\mathbf{X} + \text{possible terms containing derivatives of } \delta\mathbf{X}. \quad (3)$$

The object  $\mathbf{f}^a$  is the *surface stress tensor*. Its components in the local frame  $\{\mathbf{e}_1, \mathbf{e}_2, \mathbf{n}\}$ , as well as the Euler-Lagrange derivative  $\mathcal{E}(\mathcal{H})$ , are listed for a few simple Hamiltonian densities in table I.

Suppose  $\delta\mathbf{X}$  is simply a constant translation, which of course leaves the Hamiltonian invariant. We thus have  $\delta H_\Sigma = 0$ , and with the help of eqs. (2), (3),  $\nabla_a \mathbf{f}^a = \mathcal{E}(\mathcal{H})\mathbf{n}$ . But a true equilibrium surface is stationary with respect to *arbitrary* variations. Thus the Euler-Lagrange (“shape”) equation  $\mathcal{E} = 0$  also holds, and we get the conservation law

$$\nabla_a \mathbf{f}^a = 0. \quad (4)$$

Its existence is simply a consequence of Noether’s theorem: a continuous symmetry implies a conservation law on shell. Note that if the surface encloses a fixed volume  $V$ , a term  $-PV = -\frac{1}{3}P \int_\Sigma dA \mathbf{n} \cdot \mathbf{X}$  involving the Lagrange multiplier  $P$  needs to be included in the functional, yielding the shape equation  $\mathcal{E} = P$ , and eq. (4) is replaced by  $\nabla_a \mathbf{f}^a = P\mathbf{n}$ . The same holds if there exists a pressure drop  $P$  across the two sides of the interface.

*Forces via the stress tensor.* – In elasticity theory the divergence of the stress tensor equals the external force per unit volume of a strained material [16]. Likewise, the divergence of the surface stress tensor defined above equals the force per unit area of the strained surface. For instance, the equation  $\nabla_a \mathbf{f}^a = P\mathbf{n}$  then states that a pressure  $P$  across the surface is a source of stress. Using Stokes’ theorem, the total force  $\mathbf{F}_{\Sigma_0}$  acting “within” any patch  $\Sigma_0$  is

$$\mathbf{F}_{\Sigma_0} = \int_{\Sigma_0} dA \nabla_a \mathbf{f}^a = \oint_{\partial\Sigma_0} ds l_a \mathbf{f}^a, \quad (5)$$

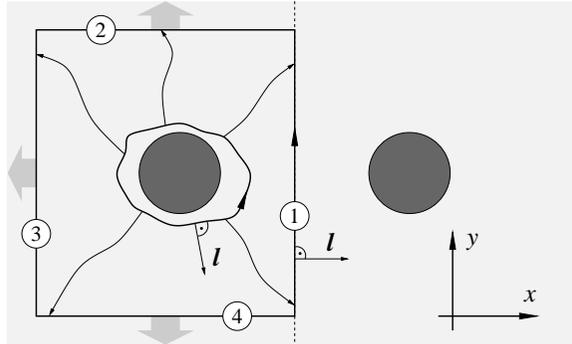


Fig. 1 – Illustration (view from the top) of how the force on one of a pair of objects bound to an interface can be calculated as a closed loop integral of the surface stress tensor. The contour can subsequently be deformed to conform to the symmetry of the situation.

where  $\mathbf{l} = l^a \mathbf{e}_a$  is the *outward*-pointing unit normal to the boundary curve  $\partial\Sigma_0$  (which is by construction tangential to the surface), and  $s$  is the arc-length on  $\partial\Sigma_0$ . If  $\Sigma_0$  is a *free* equilibrium patch (*i.e.*, no external stresses), eq. (4) shows that  $\mathbf{F}_{\Sigma_0} = 0$ . Generally, however, the patch will contain regions where external stresses act and  $\mathbf{F}_{\Sigma_0}$  will be nonzero. Observe now that for  $P = 0$  the total force equals the line integral of the stress tensor along *any* curve enclosing these sources. This is because  $\nabla_a \mathbf{f}^a = 0$  permits us to deform the contour of integration —provided we do not cross any of these sources of stress in doing so. The case of  $P \neq 0$  will be treated elsewhere.

Consider now two identical particles bound to some asymptotically flat surface, as is schematically sketched in fig. 1. Due to surface-mediated interactions, such a situation can only be stationary if external constraining forces fix the particle positions. These forces transmit stresses onto the surface, which are thus picked up by a line integral over the surface stress tensor around either particle. In the absence of a pressure difference, the contour of the line integral can be deformed so as to take advantage of the available symmetry, see again fig. 1. Once the contour is pulled open wide enough, the surface will be flat at branches 2, 3, and 4, and the stress tensor will be very simple. In fact, the contributions from branch 2 and 4 will then cancel each other. The only nontrivial contribution stems from branch 1, and its evaluation is greatly simplified by the symmetry.

This outlines the basic strategy, which can evidently be tailored towards many other situations. We will now demonstrate its application to a few important standard cases.

*Force in fluid membranes.* – The example of a surface we would like to focus on is an elastic symmetric fluid membrane, described by the surface Hamiltonian [17]

$$\mathcal{H} = \frac{1}{2} \kappa K^2 + \sigma. \tag{6}$$

Here,  $\kappa$  is the bending stiffness and  $\sigma$  the surface tension. For the special case  $\kappa = 0$  this reduces to the problem of a surface with surface tension only, and describes a soap film on large enough length scales or a water surface on length scales smaller than the capillary length. From table I we find that the associated stress tensor is given by

$$\mathbf{f}^a = \left[ \kappa \left( K^{ab} - \frac{1}{2} K g^{ab} \right) K - \sigma g^{ab} \right] \mathbf{e}_b - \kappa (\nabla^a K) \mathbf{n}. \tag{7}$$

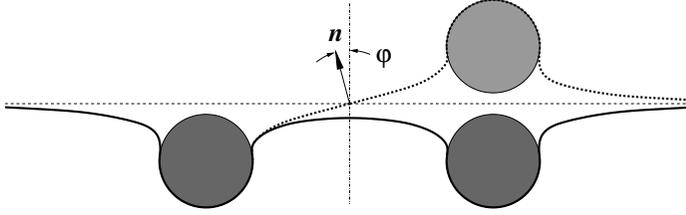


Fig. 2 – Illustration of the geometry of a symmetric (solid line) and an antisymmetric (dotted line) two-particle attachment. The angle between surface normal at the symmetry point and vertical is  $\varphi$ .

We now introduce (orthonormal) tangent vectors  $\{\mathbf{t}, \mathbf{l}\}$  along branch 1:  $\mathbf{t}$  points along the integration line ( $\mathbf{t} = t^a \mathbf{e}_a$ ), and (recall)  $\mathbf{l}$  points normally outward. A short calculation then shows that the force stemming from branch 1 is given by

$$\mathbf{F}_1 = - \int_1 ds \left\{ \left[ \frac{1}{2} \kappa (K_\perp^2 - K_\parallel^2) - \sigma \right] \mathbf{l} - \kappa (\nabla_\perp K) \mathbf{n} \right\}, \quad (8)$$

where we have defined the principal curvatures  $K_\perp = l^a l^b K_{ab}$  and  $K_\parallel = t^a t^b K_{ab}$ , as well as the derivative along  $\mathbf{l}$ ,  $\nabla_\perp = l^a \nabla_a = \partial/\partial l$ . By symmetry there is no contribution to  $\mathbf{F}_1$  along  $\mathbf{t}$ . The minus sign out front stems from the fact that the membrane-mediated force on the particle is *opposite* to the external force necessary to counterbalance it.

To proceed further, we must look separately at the two different possible symmetries: either two particles adhere at the same side of the membrane (symmetric) or at different sides (antisymmetric), see fig. 2. In the *symmetric* case, the curvatures  $K_\perp$  and  $K_\parallel$  both have an extremum in the  $\mathbf{l}$  direction along branch 1, so  $\nabla_\perp K = 0$  and the normal force component vanishes there. Furthermore, since the profile is horizontal in the middle,  $\mathbf{l} = \mathbf{x}$  there (where  $\mathbf{x}$  is the unit vector pointing in the horizontal  $x$ -direction). Finally, on branch 3 the stress tensor simplifies to  $\mathbf{f}_{a,3} = -\sigma \mathbf{e}_a$ , since far enough away the curvature becomes zero. Therefore, the total force  $\mathbf{F}_1 + \mathbf{F}_3 = F_{\text{sym}} \mathbf{x}$  on the left particle is given by

$$F_{\text{sym}} = \sigma \Delta L - \frac{1}{2} \kappa \int_1 ds (K_\perp^2 - K_\parallel^2), \quad (9)$$

where  $\Delta L > 0$  is the excess length of branch 1 compared to branch 3. We immediately see that the contribution due to tension is attractive. The curvature contribution, on the other hand, is the integral over the difference between the squared principal curvatures along the mid-curve, and as such has no evident sign. However, if we had adsorbed two parallel *cylinders*, which are sufficiently long such as to neglect end effects, the contribution  $K_\parallel^2$  vanishes. Furthermore, the mid-curve becomes a line, and thus  $\Delta L = 0$ . In this case we find for the force per unit length of the cylinder

$$F_{\text{sym,cyl}}/L = -\frac{1}{2} \kappa K_\perp^2 \leq 0, \quad (10)$$

which is evidently always repulsive. Note that even though the tension  $\sigma$  does not occur explicitly, it will enter the force indirectly through its influence on the value of  $K_\perp$ . In Monge gauge, with height function  $h$ ,  $K_\perp = [h'/(1+h'^2)]'|_{x=0} = h''(0)$ , since  $h'(0) = 0$ . Using this, eq. (10) is then quantitatively corroborated (in the linear regime) by the calculations in ref. [8]. Details of this will be presented elsewhere.

In the *antisymmetric* case, branch 1 is a rotational symmetry axis of degree 2 and thus a line; hence both  $K_\parallel$  and  $K_\perp$  vanish. Since  $K_\perp$  changes sign from positive to negative,

$\nabla_{\perp} K_{\perp} < 0$ . Observe that the profile on the midline is always tilted in the direction indicated in fig. 2, because otherwise it would have *three* nodal points on the asymptotic horizontal line and not just one, and the energy is expected to be higher. If we conceive of the constraining external forces as fixing the *horizontal separation* but not the vertical position of the particles, the latter is equilibrated and the vertical force component vanishes. The force which remains on the left particle is thus again horizontal,  $\mathbf{F}_{\text{antisym}} = F_{\text{antisym}} \mathbf{x}$ , and given by

$$F_{\text{antisym}} = \int_1 ds [\sigma(\cos \varphi(s) - 1) - \kappa \sin \varphi(s) \nabla_{\perp} (K_{\perp} + K_{\parallel})]. \quad (11)$$

This time the tension contribution is repulsive. In the bending contribution the  $\nabla_{\perp} K_{\perp}$  is attractive, but unfortunately not much can be said about the sign of  $\nabla_{\perp} K_{\parallel}$ , hence the overall sign is not obvious. However, in the case of two cylinders bound on opposite sides, eq. (4) implies that  $|\mathbf{f}^a|$  is constant on each of the three membrane segments, and the force expression (per unit length) simplifies to [18]

$$F_{\text{antisym,cyl}}/L = |\mathbf{f}_{\text{midpoint}}^{\perp}| - \sigma = \sqrt{\sigma^2 + (\kappa \nabla_{\perp} K_{\perp})^2} - \sigma \geq 0, \quad (12)$$

which is manifestly positive, implying particle attraction. If we expand the square root to first order in the bending part, we arrive at the result for linear theory  $F_{\text{antisym,cyl},\varphi \ll 1}/L = \frac{1}{2} \kappa (\lambda \nabla_{\perp} K_{\perp})^2$  (with  $\lambda = \sqrt{\kappa/\sigma}$ ). This latter expression is again quantitatively confirmed by using profile and force as they are calculated in ref. [8].

*Discussion.* – In the previous sections we have outlined a very general method to obtain exact results for surface-mediated interactions which sidesteps the need to solve the field equations explicitly. This approach is fully covariant: one is free to choose the parameterization which is most appropriate; it is also valid for large deformations and *not* limited to a linear approximation. To our knowledge, there are no analogues based on energy minimization of our results outside this regime. It is true that our formulas contain unknown quantities related to the shape of the surface. It was clear from the beginning, however, that we could not expect to solve the problem completely without determining this shape; what has been established is the connection between the *geometry* of the surface and the *forces* transmitted by it, something which does not come across when energy is differentiated—even in the linear regime. With sufficient ingenuity, it may be possible to extract the necessary information on the shape from the Euler-Lagrange equation (which so far we have not used at all!) without needing to solve it explicitly. Even without this input the functional form alone may identify the sign of the interaction [19]. Most importantly, this framework provides a new window onto an old and important problem. It can be combined with any approach, be it analytical or numerical, which determines the surface shape. At the very least, we have shown that it provides valuable nontrivial consistency conditions for analytical calculations. When such calculations are ruled out, the determination of the force between particles by numerically integrating the stress around one of them is a process which is not only more straightforward but also considerably more economical than calculating the energy as a function of distance and numerically differentiating it. A detailed description of the interaction between two identical particles using this approach will be presented elsewhere.

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- [14] Locally, a smooth surface can be represented by a differentiable height function  $h(x, y)$  above the local tangent plane. In these coordinates  $K_{ab}$  is simply the Hessian of  $h$  divided by the square root of the metric determinant:  $K_{ab} = -\partial_a \partial_b h / \sqrt{g}$  ( $= -\partial_a \partial_b h$  to lowest order in  $\nabla h$ ).
- [15] We recall that the intrinsic and extrinsic geometries are related by the Gauss-Codazzi-Mainardi equations  $R_{abcd} = K_{ac}K_{bd} - K_{ad}K_{bc}$  and  $\nabla_a K_{bc} = \nabla_b K_{ac}$ . Here  $R_{abcd}$  is the Riemann tensor, which quantifies the extent to which covariant derivatives fail to commute:  $[\nabla_a, \nabla_b]e_c = R_{abcd}e^d$ . These equations occur as the integrability conditions on the formulae of Gauss,  $\nabla_a e_b = -K_{ab}n$ , and Weingarten,  $\nabla_a n = K_{ab}e^b$ , obtained by taking a second derivative  $\nabla_c$ , switching the indices  $a$  and  $c$  and subtracting [12]. The intrinsic scalar curvature  $R = g^{ac}g^{bd}R_{abcd}$  thus satisfies  $R = K^2 - K^{ab}K_{ab} = 2K_G$ , where  $K_G = \det(K_a^b)$  is the Gaussian curvature.
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- [19] We mention, as an aside, that the same trick has recently rendered possible a proof of the (non-trivial!) fact that within mean-field electrostatics two identical like-charged colloids repel. Here, the Euler-Lagrange equation of the underlying field theory is the nonlinear Poisson-Boltzmann equation, which cannot be solved analytically in this geometry, but certain properties of the corresponding (Maxwell) stress tensor nevertheless permit the determination of the *sign* of the interaction. See: NEU J., *Phys. Rev. Lett.*, **82** (1999) 1072; SADER J. E. and CHAN D. Y. C., *J. Colloid Interface Sci.*, **213** (1999) 268; TRIZAC E., *Phys. Rev. E*, **62** (2000) R1465.