



Physical mechanisms of the interaction between lipid membranes in the aqueous environment



Alexander Schlaich^a, Bartosz Kowalik^a, Matej Kanduč^{a,b}, Emanuel Schneck^c, Roland R. Netz^{a,*}

^a *Fachbereich Physik, Freie Universität Berlin, Arnimallee 14, 14195 Berlin, Germany*

^b *Department of Theoretical Physics, J. Stefan Institute, SI-1000 Ljubljana, Slovenia*

^c *Institut Laue-Langevin, 6 Rue Jules Horowitz, 38042 Grenoble, France*

HIGHLIGHTS

- Classical density functional theory explains basic features of the hydration repulsion.
- The overlap of ordered interfacial water layers constitutes one mechanism for repulsion.
- The choice of order parameters and boundary conditions is crucial in this description.
- Several alternative mechanisms are discussed as contributions to the total membrane–membrane interaction.

ARTICLE INFO

Article history:

Available online 23 July 2014

Keywords:

Membrane interactions
Hydration repulsion
Classical density functional theory
Electrostatic interactions
Order parameters
MD simulations

ABSTRACT

We review various mechanisms underlying the interaction between biological membranes in their aqueous environment. In the first part we describe van der Waals interactions, derive generally-applicable density functional expressions for solute-mediated interactions with a focus on electrostatic interactions across aqueous electrolytes, and discuss interaction mechanisms involving the overlap of structured interfacial water layers. We then turn our attention to the long-debated hydration repulsion and review its quantitative investigation using classical molecular dynamics simulations.

© 2014 Elsevier B.V. All rights reserved.

1. Introduction

Biomembranes play central roles in the structure and function of cells in prokaryotic as well as eukaryotic organisms. Forming their outermost boundary, they are responsible for the exchange of substances between the interior and the exterior of cells, which is regulated by embedded membrane proteins, and also for their interaction with the surrounding. Also inside cells, biomembranes of various compositions define different compartments, known as cell organelles, and are vital for their functioning.

Biomembranes are mainly composed of lipids, which offer important features that are crucial for their properties. Lipids are amphiphilic [1] and spontaneously form bilayers in the aqueous environment. The entropic forces that lead to the formation of such architectures arise from the hydrophobic effect [2]. Moreover, the lipid chains inside the bilayer are usually disordered and fluid-like, thus allowing for a high flexibility of the membrane. The polar head groups point towards the bilayer exterior, where they interact with surrounding water molecules (see Fig. 1 for a schematic illustration). This bilayer structure is remarkably stable in a wide range of thermodynamic conditions. At high lipid concentrations bilayers typically

* Corresponding author.

E-mail address: rnetz@physik.fu-berlin.de (R.R. Netz).

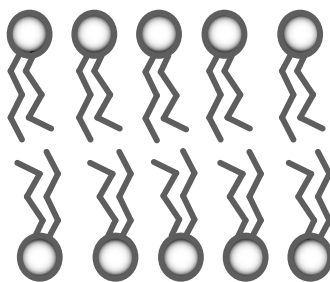


Fig. 1. Schematic picture of the aggregation of lipids into bilayers when exposed to water.

arrange in lamellar structures. Here, the water is evenly distributed between the head groups of the parallel-aligned bilayers. But even in excess of water, such stacked structures often are formed, which reflects that the interaction potential between the membranes exhibits a minimum at a finite distance. The interaction potential plays an important role in biological processes involving membranes [3]. Predominantly repulsive membrane–membrane interactions, for instance, prevent close membrane–membrane contacts and generate aqueous pathways for the diffusion of biomolecules between the membrane surfaces. Attractive interaction contributions, on the other hand, can create well-defined average membrane separations that facilitate membrane fusion or cell adhesion processes via specific binding sites.

Here, we review the various types of generic (non-specific) forces acting between lipid membranes in the aqueous environment and discuss the underlying physical mechanisms, with a particular focus on the competing roles of enthalpic and entropic contributions. The interaction free energy (or interaction potential) is typically the result of a subtle interplay of several, often antagonistic, contributions of comparable magnitude. First, we will introduce the underlying physics of various kinds of surface–surface interactions, starting with the theoretical description of van der Waals interactions and then covering electrostatics in aqueous electrolytes as well as interactions due to accumulation/depletion of solute molecules at the membrane surfaces and solvent-structural effects. We then focus our attention on a strong repulsive interaction at small membrane–membrane separations known as hydration repulsion. It has been under debate and investigation for decades and is not well captured by continuum approximations; thus, in the last part, we will mainly rely on atomistic simulation techniques.

2. Van der Waals interactions between membranes

The electric charge distribution (due to nuclei and electrons) of atoms and molecules manifests in the form of an interaction that acts among all matter, even when the constituting parts are charge-neutral. A simplified picture, which has been established in the 1930s, attributes this interaction to three kinds of atomic dipole–dipole interactions [4]:

- interactions between two permanent molecular dipoles (Keesom interactions),
- interactions between a permanent molecular dipole and a corresponding induced molecular dipole (Debye interactions),
- interactions between two spontaneously induced molecular dipoles (London dispersion interactions).

The unified concept of van der Waals (vdW) interactions accounts for all three cases, and in each case the interaction potential, i.e. the work needed to bring two molecules from infinite separation to a certain distance r , varies as the inverse-sixth power of distance,

$$u(r) = -\frac{C}{r^6}, \quad (1)$$

where C is a positive coefficient. Biomembranes are extensive bodies composed of atoms and molecules, which all interact via such vdW potentials. In a first approximation, we can assume pair-wise additivity of the individual interactions and sum them all up. In the case of two parallel semi-infinite media separated by distance D in vacuum, we obtain the free energy per unit area,

$$\mathcal{F}_{\text{vdW}}/\mathcal{A} = -\frac{H}{12\pi D^2}, \quad (2)$$

where $H = \pi^2 C n^2$ is known as the *Hamaker constant* [5] and n corresponds to the particle number density of the two media. In the case when the media are immersed into water, additional contributions appear,

$$H = \pi^2 C n^2 - 2\pi^2 C_{\text{cw}} n n_w + \pi^2 C_w n_w^2, \quad (3)$$

with n_w being the number density of water molecules, C_{cw} the interaction coefficient between water and the particles constituting the semi-infinite media, and C_w the coefficient for the water–water interactions. The vdW pressure between the membranes can then be obtained by derivation of the free energy, $p_{\text{vdW}} = -\partial(\mathcal{F}_{\text{vdW}}/\mathcal{A})/\partial D$, which yields

$$p_{\text{vdW}} = -\frac{H}{6\pi D^3}. \quad (4)$$

Download English Version:

<https://daneshyari.com/en/article/976995>

Download Persian Version:

<https://daneshyari.com/article/976995>

[Daneshyari.com](https://daneshyari.com)