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Relaxation kinetics of lipid membranes and its relation to the heat capacity

Peter Grabitz, Vesselka P. Ivanova, and Thomas Heimburg

Membrane Biophysics and Thermodynamics Group, Max-Planck-Institute for Biophysical Chemistry, 37077 Göttingen, Germany

In a recent paper (Grabitz et al. 2002. Biophys.J.82:299-309) we found that the relaxation behavior of lipid membranes shows a close relation to the heat capacity. We calculated a proportional factor between heat capacity and the relaxation times on the basis of linear nonequilibrium thermodynamics concepts. In the theory section we made a minor mistake, which we will outline below. It yields incorrect units and numerical values for a phenomenological coefficient, L. The overall message of the paper, however, is not affected.

The error originates from eq. (4) where a factor of RT is missing. In the original manuscript the free energy, G, was given as $G(H-\overline{H}) = -\ln P(H-\overline{H}) + \text{const.}$. This yields incorrect units for the free energy. Correctly this equation should be:

$$G(H-\overline{H}) = -RT\ln P(H-\overline{H}) + \text{const.} \qquad (4)$$

The factor RT is also missing in the subsequent equations. The equations below are now corrected.

$$G(H-\overline{H}) = RT\frac{(H-\overline{H})^2}{2\sigma^2} + \text{const.} \qquad (5)$$

$$\begin{aligned} S(H-\overline{H}) &= \frac{(H-\overline{H})}{T} - \frac{R\,(H-\overline{H})^2}{2\sigma^2} - \frac{\text{const.}}{T} \\ &\approx -\frac{R(H-\overline{H})^2}{2\sigma^2} \end{aligned} \qquad (6)$$

Later in the theory section we calculate the thermodynamic force resulting from a fluctuation in the enthalpy from the entropy:

$$\begin{aligned} X(H-\overline{H}) &= \left(\frac{\partial^2 S(H-\overline{H})}{\partial(H-\overline{H})}\right)_0 (H-\overline{H}) \\ &= \frac{R\,(H-\overline{H})}{\sigma^2} \end{aligned} \qquad (9)$$

The flux of enthalpy back to equilibrium is given by the phenomenological equation

$$\frac{d(H-\overline{H})}{dt} = L\cdot X(H-\overline{H}) = -L\cdot\frac{R\,(H-\overline{H})}{\sigma^2} \qquad (10)$$

and thus the time dependence of the relaxation is given by the single exponential function

$$\begin{aligned} (H-\overline{H})(t) &= (H-\overline{H})(0)\cdot\exp\left(-\frac{R\cdot L}{\sigma^2}t\right) \\ &\equiv (H-\overline{H})(0)\cdot\exp\left(-\frac{t}{\tau}\right) \end{aligned} \quad , (11)$$

introducing a relaxation time, τ. Because $\sigma^2 = RT^2\,c_P$, it follows for the relaxation time,

$$\tau = \frac{T^2}{L}c_p \equiv \alpha c_p \qquad , \qquad (12)$$

Table 1: **The relaxation time, $\tau = (T^2/L)c_p \equiv \alpha c_p$, for four different lipid preparations, the phenomenological coefficient L, and proportionality constant, α**

Lipid	Phenomenological Coefficient L ($10^8 J\cdot K/s\cdot mol$)	α ($10^{-4} s\cdot mol\cdot K/J$)
DMPC MLV	7.35	1.20
DPPC MLV	8.45; (13.4)	1.17;(0.74)
DPPC LUV	(13.9)	(0.71)
DPPC:Cholest.=99 : 1 MLV	8.37	1.18

L and α were determined at the heat capacity maximum.
The difference of the two values for DPPC, which differ by $\sim$ 35%, probably arises from time dependent changes in the heat capacity profiles, which broaden by up to 30% after one week due to slow swelling of the sample.
Uncertain values are given in brackets.

and the relaxation time close to the chain melting transition of lipids becomes a proportional function of the heat capacity with a proportionality constant $\alpha = T^2/L$.
The values for L in Table 1 are consequently also changed.

Concluding, the main correction consists of the change of the numerical values and the units of the phenomenological coefficient, L from the phenomenological equations. The main result from our studies that the heat capacity is proportional to the relaxation times is unaffected.

correspondence to T.Heimburg: theimbu@gwdg.de, WWW: www.gwdg.de/∼theimbu