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Relaxation kinetics of lipid membranes and its relation to the heat capacity
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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 \( G(H - \overline{H}) \) with \( H \) missing. In the subsequent equations. This yields incorrect units for the free energy. Correctly this equation should be:

\[
G(H - \overline{H}) = -RT \ln P(H - \overline{H}) + \text{const} \tag{4}
\]

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

\[
G(H - \overline{H}) = RT \frac{(H - \overline{H})^2}{2\sigma^2} + \text{const} \tag{5}
\]

\[
S(H - \overline{H}) = \frac{(H - \overline{H})}{T} - \frac{R(H - \overline{H})^2}{2\sigma^2} - \text{const} \tag{6}
\]

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

\[
X(H - \overline{H}) = \left( \frac{\partial^2 S(H - \overline{H})}{\partial (H - \overline{H})} \right)_0 (H - \overline{H}) = \frac{R(H - \overline{H})}{\sigma^2} \tag{9}
\]

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

\[
\frac{d(H - \overline{H})}{dt} = L \cdot X(H - \overline{H}) = -L \cdot \frac{R(H - \overline{H})}{\sigma^2} \tag{10}
\]

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

\[
(H - \overline{H})(t) = (H - \overline{H})(0) \cdot \exp \left( -\frac{R \cdot L}{\sigma^2} t \right) \equiv (H - \overline{H})(0) \cdot \exp \left( -\frac{t}{\tau} \right) \tag{11}
\]

introducing a relaxation time, \( \tau \). Because \( \sigma^2 = RT^2 c_p \), it follows for the relaxation time,

\[
\tau = \frac{T^2}{L} c_p \equiv \alpha c_p \tag{12}
\]

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, \( \alpha \)

<table>
<thead>
<tr>
<th>Lipid</th>
<th>Coefficient ( L ) (( 10^6 \text{J/K/mol} ))</th>
<th>( \alpha ) (( 10^{-4} \text{s mol.K/J} ))</th>
</tr>
</thead>
<tbody>
<tr>
<td>DMPC MLV</td>
<td>7.35</td>
<td>1.20</td>
</tr>
<tr>
<td>DPPC MLV</td>
<td>8.45, (13.4)</td>
<td>1.17, (0.74)</td>
</tr>
<tr>
<td>DPPC LUV</td>
<td>(13.9)</td>
<td>(0.71)</td>
</tr>
<tr>
<td>DPPC, Cholest.=99:1 MLV</td>
<td>8.37</td>
<td>1.18</td>
</tr>
</tbody>
</table>

The overall message of the paper, however, is not affected.

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.

Uncertain values are given in brackets.

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.

\( L \) and \( \alpha \) were determined at the heat capacity maximum.

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