LETTER TO THE EDITOR

Comparison of Forces Measured Between Phosphatidylcholine Bilayers

Dear Sir:

We write to correct an unintended misrepresentation in two recent papers (1, 2) that compared results between bilayer membranes immobilized onto curved mica sheets with earlier force measurements (3–5) between bilayers in spontaneously forming planar multilayer arrays. To compare results obtained in the two different geometries, use was made of the Derjaguin approximation (6, 7) which states that the force between curved surfaces is proportional to the interaction energy between equivalent planar surfaces. Thus it was necessary to integrate the force measured between planar bilayers or to differentiate that between curved surfaces to compare results. It was in not integrating the long-range "tail" of the van der Waals force between planar bilayers that Horn (1) and Marra and Israelachvili (2) misrepresented the comparison. As a consequence, there appeared to be a greater difference than actually exists between the two kinds of data.

To show the actual good agreement, we have differentiated the force curves of reference 2 for the frozen chain phosphatidylcholines (PCs) (distearoylPC [DSPC], dipalmitoylPC [DPPC], and dimyristoylPC [DMPC]) and for the melted chain PCs (DMPC and dilauroylPC [DLPC]) and plotted them in Fig. 1, *a* and *b*, as a force per molecule F_R with the data from reference 5 for DLPC and DPPC. (All data from references 2 and 3 fall within the shaded bands.) The latter transformation avoids invoking explicitly the long-range van der Waals force. Multilayer data for other PCs show similar good agreement, as does an integration of the smoothed data from that planar system. In all cases, there is an especially pleasing similarity of slopes for $F_R > 10^{-8}$ dyn/molecule. This is the region of "hydration forces" known to dominate the interaction of these bilayers approaching contact.

There do remain some instructive differences, though, especially for $T > T_c$. There are real differences between the two methods of measurement to account for these details. One consideration is the difference in the definition of zero separation, necessarily different in the two systems. If, however, one shifts the limiting separation (*shaded arrow*) reported for $T < T_c$ in the coated mica system to coincide with the limiting spacing (*solid arrow*) apparent in multilayers, the curves coincide nicely also at the upper end.

Another important consideration, at $T > T_c$, is the contribution of repulsive forces due to undulations (8–10) which can presumably occur with free melted bilayers but not those adsorbed on a solid surface. One can extract the elastic force by subtracting these fluctuations (cf. Fig. 2 in reference 10) to obtain the dashed line in Fig. 1 *b* here. Then, superposing the limiting separations (*shaded and solid arrows*) again shows much better agreement between the coated-mica-surface data and the earlier repulsive force measurements in multilayers.

In may be noted that the depth of the energy minimum obtained from the mica-immobilized lipid measurements (2) is almost an order of magnitude higher than those inferred from the multilayer measurements (12) and obtained between single bilayer vesicles from which undulations have been limited by applied lateral tension (e.g., reference 13). Recent observations (14) on the increase of lecithin bilayer adhesion with increased lateral tension strongly suggest that weaker bilayer–bilayer energy minima can at least partly be ascribed to the action of undulations, but most of the difference in the depth of the minima given by the two methods is still not understood.

Received for publication 22 February 1988 and in final form 3 August 1988.

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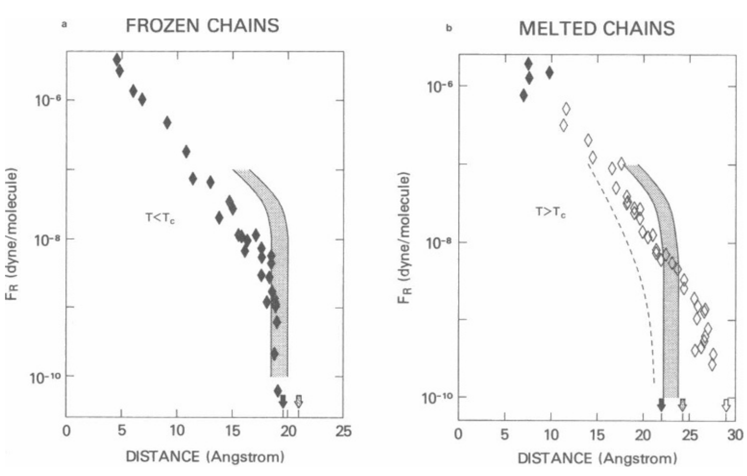

FIGURE 1 Forces between frozen ($T < T_c$) and melted ($T > T_c$) chain phosphatidylcholines. (*Solid and open diamonds*) Data on multilayers for (*a*) DPPC and (*b*) DLPC (4, 5). (*Shaded bands and arrows*) (traced from Fig. 16 *c* of reference 11) Differentiated force data and limiting spacings for mica surfaces coated with (*a*) DSPC, DPPC, or DMPC ($T < T_c$), and (*b*) DLPC or DMPC ($T > T_c$), respectively (2). The dashed line in *b* is the elastic force inferred after subtraction of undulatory steric contributions (Fig. 2 of reference 10) in the melted-chain multilayer system.

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