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Rapid Note

Comment on "Growth of Molecular Superlattice in Fully Hydrated Dipalmitoylphosphatidylcholine during Subgel Phase Formation Process"

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Abstract. This comment shows that data recently reported [1] as being seemingly in conflict with earlier data [2] are, in fact, in excellent agreement. Together, both studies confirm that the kinetics of the subgel phase transformation in dipalmitoylphosphatidylcholine (DPPC) lipid bilayers obeys Kolmogorov-Avrami (K-A) theory [3,4] with an anomalously low effective dimensionality.

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In the earlier study Yang and Nagle [2] reported volume data using a special thermal protocol to minimize small scale coexistence [5] and colliding domains [6] that sometimes complicate the kinetics of subgel phase transformation [7]. In this T-jump protocol temperature T is first reduced from above $T_{\mathrm{S}} = 13.5$ °C, which is the equilibrium subgel/gel phase transition temperature [8,9], to an incubation temperature $T_{\mathrm{I}} < 7$ °C for a short period of time. Then, temperature is jumped from T_{I} to T_{J} which is chosen in the range between 8 °C and T_{S}. At T_{J} the subgel nuclei formed at T_{I} continue to grow, but no new nuclei form; the evidence for this is that no subgel phase is formed if T is reduced from above T_{S} to an attempted incubation temperature T_{I} in the range 8 °C $< T_{\mathrm{I}} < T_{\mathrm{S}}$ [8]. For the kinetic state established at T_{J} for the T-jump protocol, K-A theory predicts that the (normalized) phase transformation kinetics of any property X should be given by

$$X(t) = 1 - \exp[-(t/\tau)^n] \tag{1}$$

where n is the effective dimensionality of the growing domains. Equation (1) fits the volumetric data very well with $1 < n < 1.3$ [2].

In the recent study Takahashi *et al.* [1] were able to avoid the complications of small scale coexistence by measuring the kinetics of the (11) X-ray peak; this peak is associated with headgroup ordering which has recently been shown to be a primary property that distinguishes the subgel phase from the gel phase [10]. The kinetics were fit satisfactorily to equation (1) and this supports the earlier findings [2,6–8] that the basic subgel transformation process is nucleation and growth. However, the fit to equation (1) yielded the value $n = 2.3 \pm 0.4$ and it was then emphasized that this disagreed with the earlier result [2]. It was suggested [1] that volume kinetics do not accurately reflect the kinetics of the subgel phase.

In this comment we point out that the reason for the difference in n values in the two studies [1,2] is due to the different temperature protocols. In the X-ray study [1] T was reduced from above T_{S} to $T_{\mathrm{I}} = 2.5$ °C and held there, so nucleation of new subgel domains proceeded continuously during the phase transformation. For this condition the K-A theory [3,4,11] predicts:

$$X(t) = 1 - \exp[-(t/\tau)^{n+1}], \tag{2}$$

in terms of the same effective dimensionality n as in equation (2). Therefore, the previously reported [1] value $n = 2.3$ obtained by fitting equation (1) should instead be reduced by 1 to $n = 1.3$ as given by equation (2).

Now that it is clear that the basic subgel transformation process is one of nucleation and growth and now that two independent studies obtain numerical agreement for the effective dimensionality of domains, the outstanding question remains, why is the effective dimensionality so low? If $n = 1$, then the process might be the one described by Cheng and Caffrey [12] for the main phase

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transformation in monoelaidin, namely, one-dimensional growth from one bilayer to the next in multilamellar vesicles. Of course, there may be different transformation processes with different effective dimensionalities depending upon which lipid or which transition is investigated. For example, in contrast to the main transition in monoelaidin, n for the main transition in DPPC has been reported to be closer to 2 [13]. For the subgel phase transformation, both quantitative studies [1,2] favor a non-integral effective dimensionality n somewhat larger than 1. A number of qualitative suggestions were discussed in [2]. However, quantitative theoretical understanding of this anomalous, but now more firmly established, experimental result for the effective dimensionality n has yet to be achieved.

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