## SUPPLEMENTARY MATERIAL

Figures of the 2D-ELDOR spectra, shown as contours, of 16PC in a model membrane composed of different compositions of DPPC and cholesterol. The spectra are shown in the full  $S_{c}$ - display with the real signal in the upper part and the imaginary signal in the lower part. The plots contain positive and negative values in intensity. The color map varies from red (, which corresponds to the largest positive value) to yellow, light blue (zero), and dark blue (, which corresponds to the smallest negative value). Note that we show here only the  $T_m$ =50 ns case, but the spectra of all the different mixing times are simultaneously fit using the same set of dynamic parameters.

- FIGURE S1 The full Sc- fits for the 2D-ELDOR spectra of the [Chol] 20% at 35 °C. They are fit with one spectral component with the dynamic parameters characterized of the gel phase. (a) ELDOR mode; (b) SECSY mode.
- FIGURE S2 The full Sc- fits for the 2D-ELDOR spectra of the [Chol] 40% at 35 °C. They are fit with one spectral component with the dynamic parameters characterized of the L<sub>o</sub> phase. (a) ELDOR mode; (b) SECSY mode.
- FIGURE S3 The full Sc- fits for the 2D-ELDOR spectra of the [Chol] 20% at 48  $^{\circ}$ C. They are fit with two spectral components, indicating the coexistence of the L<sub>d</sub> and L<sub>o</sub> phases. (a) ELDOR mode; (b) SECSY mode.
- FIGURE S4 The full Sc- fits for the 2D-ELDOR spectra of the [Chol] 20% at 57  $^{\circ}$ C. They are fit with two spectral components, indicating the coexistence of the L<sub>d</sub> and L<sub>o</sub> phases. (a) ELDOR mode; (b) SECSY mode.







EXPERIMENT (SECSY) vs. full Sc- fits, Tm=50ns, 35°C, [Chol] 40%





Figure S3(a)

EXPERIMENT (SECSY) vs. full Sc- fits, Tm=50ns, 48°C, [Chol] 20%







EXPERIMENT (SECSY) vs. full Sc- fits, Tm=50ns, 57°C, [Chol] 20%

Figure S4(b)