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Ultrafast dynamics at lipid-water interfaces

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MATERIALS AND METHODS

Sample Preparation

Solutions of 1,2-dioleoyl-sn-glycero-3-phosphocholine (DOPC) and 1,2-dihexadecanoylsn-glycero-3-phosphocholine (DPPC) in chloroform were purchased from Avanti Polar Lipids and used without further purification. Deuterium oxide was purchased from Cambridge Isotope Laboratories, Inc. and used without further purification.

Aligned lipid multilayers were prepared according to an established procedure.^{1,2} First, lipids films were drop-cast on CaF₂ optical windows. Solvent was removed for at least an hour under vacuum. Lipid films were hydrated with deuterium oxide and allowed to equilibrate in a hydration chamber with 100% humidity above the lipid's gel-to-fluid phase transition temperature for an hour. After equilibration, lipid windows were rotated to align the multilayers and reduce scatter.

2D IR Spectroscopy

2D IR measurements were performed on a spectrometer which has previously been described in detail.³ Near-IR pulses (~100fs) were generated by a Ti:Sapphire laser (Astrella, Coherent Inc), then converted to mid-infrared frequencies using an optical parametric amplifier and a difference frequency generator. Pulses were split into pump and probe beams using a CaF_2

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beamsplitter. The probe pulse was converted into two sequential excitation pulses using a Gebased pulse shaper (PhaseTech Spectroscopy, Inc). The delay between pump and probe pulses at was controlled through a delay stage. The probe pulse was detected with a spectrometer and a 128x128 pixel magnesium cadmium telluride array detector (Teledyne). The detector array was used to resolve the detection frequencies and a numerical Fourier Transform of the excitation pairs from the pulse shaper was used to resolve the excitation frequencies.

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