Supporting Information

Understanding Liquid-Solid-Like Behavior of Tetrahydrofuran Adlayers at Room Temperature between Graphene and Mica: A Born-Oppenheimer Molecular Dynamics Study

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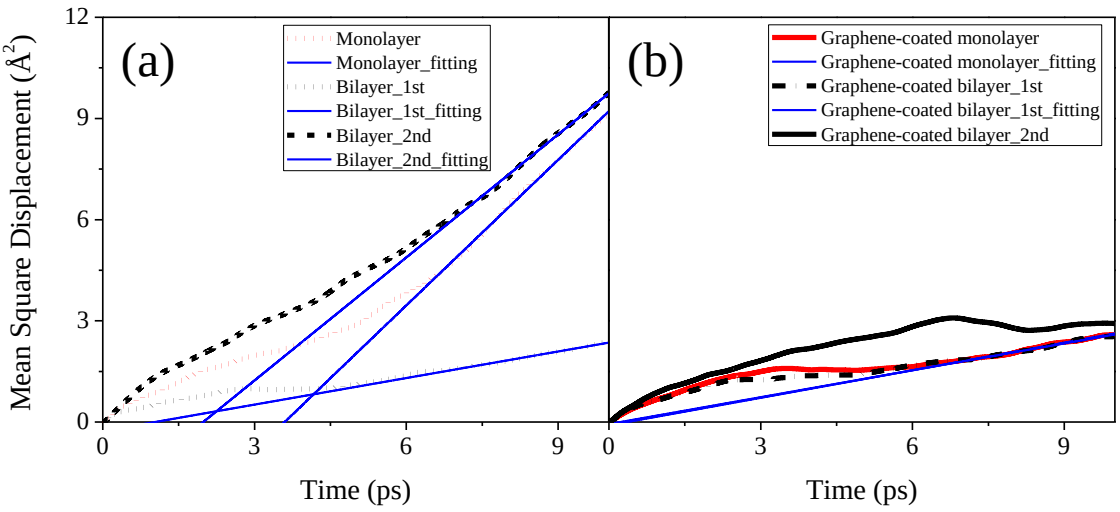

Figure S1. Variations of mean square displacement (MSD) with time for the monolayer, the first and second adlayers of the bilayer on mica (a) without and (b) with the graphene coating, based on BOMD simulations. The corresponding linear fitting of the MSD-time curves are present in blue.

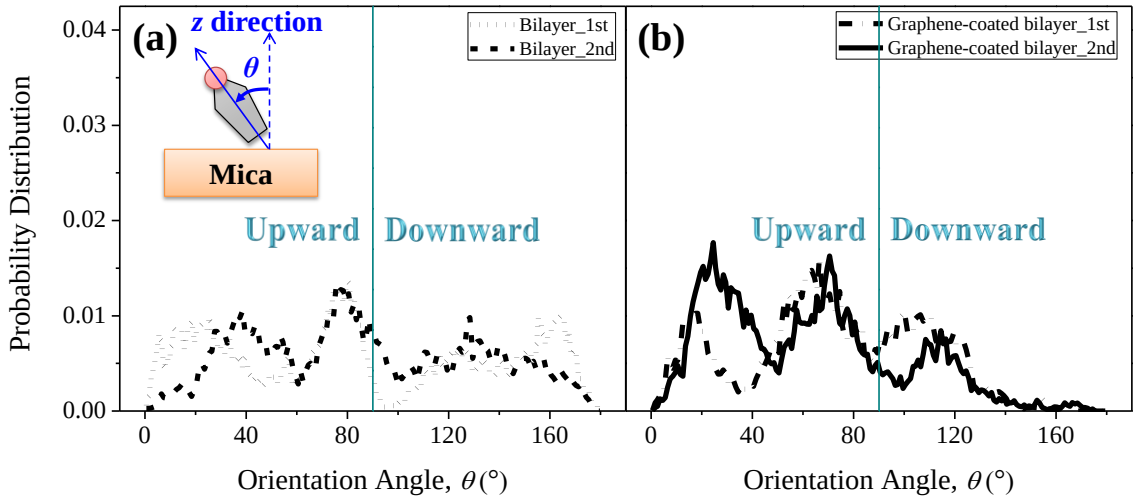

Figure S2. Probability distribution of the orientation angle of THF molecules in the first and second adlayers of the bilayer (a) without and (b) with the graphene coating, respectively. The molecular axis of the THF is defined as the vector from the center of the C-C bond to the O atom in THF ring. The orientation angle (θ) of THF molecules is also estimated as the azimuthal angle of molecular axis to the z direction (perpendicular to the mica surface). The upward and downward orientations of THF molecules are divided by the dark cyan line located at θ = 90°.

Table S1. Comparison of probability for the THF molecules in upward (θ < 90°) and in downward (θ > 90°) orientations and in the monolayer and bilayer without and with the graphene coating, respectively.

	Monolayer	Bilayer		Graphene-coated monolayer	Graphene-coated bilayer	
		1st	2nd		1st	2nd
Initial						
Upward	0.4	0.4	0.6	0.4	0.4	0.6
Downward	0.6	0.6	0.4	0.6	0.6	0.4
Statistically-averaged						
Upward	0.579	0.593	0.567	0.9	0.658	0.783
Downward	0.421	0.407	0.433	0.1	0.342	0.217

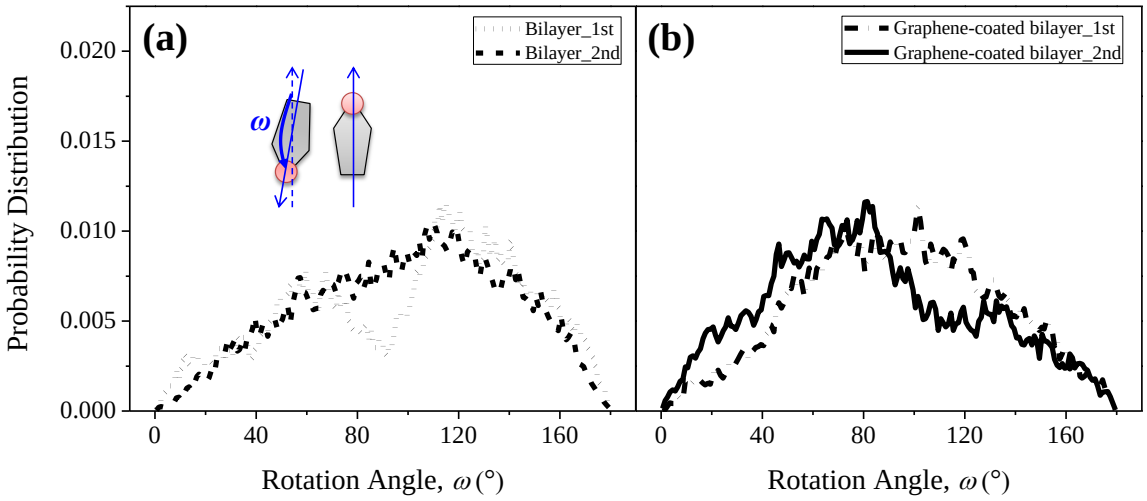

Figure S3. Probability distribution of the rotation angle of THF molecules in the first and second adlayers of the bilayer (a) without and (b) with the graphene coating, respectively. The rotation angle is defined as the angle of molecular axes between two THF molecules indicated in insets.