Supplementary Information:- Similar Structure but Different Thermodynamic, Dielectric, and Frictional Properties of Confined Water in Twisted 2D Materials: MoS_2 vs. Graphene

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Order Parameter Values

Table 1S: The Ψ_4 and $\diamond$ order parameter values for MoS_2 systems with respect to twist angle

Twist [degree]	Ψ_4	pcd($\diamond$)
0.0	0.76 ± 0.01	0.85 ± 0.01
9.4	0.75 ± 0.01	0.86 ± 0.01
21.8	0.72 ± 0.01	0.87 ± 0.01
29.4	0.73 ± 0.02	0.81 ± 0.03

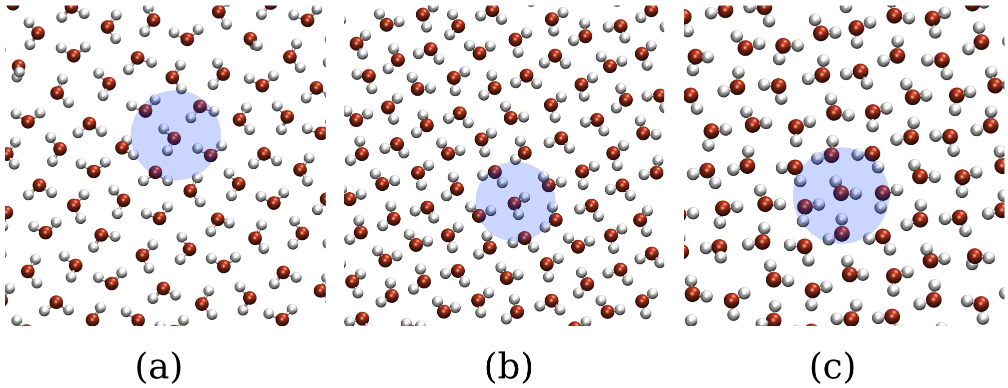

Figure 1S: The confined water structure inside (a) 0.0°, (b) 9.4°, (c) 21.8° twisted bilayer graphene system. These final structures show a regularly ordered water structure, which is described as square like ordering. (d) Shaded region denotes the radius of first nearest neighbors that determine the quartic order parameter.

Table 2S: The Ψ_4 and $\diamond$ order parameter values for graphene systems with respect to twist angle

Twist [degree]	Ψ_4	pcd($\diamond$)
0.0	0.73 ± 0.03	0.85 ± 0.01
9.4	0.72 ± 0.03	0.86 ± 0.01
21.8	0.72 ± 0.03	0.91 ± 0.01
29.4	0.74 ± 0.01	0.89 ± 0.01

Synthetic structure formulation

A square is a special rhombus whose diagonal lengths are equal because the rhombus, in that case, will be constrained to have all its angles as 90°. Hence, we start laying out the problem from a general rhombic structure and control the difference of diagonal lengths to get a square. Fig. 3S shows the unit rhombus of side length l that we consider to find the constrained relation between a diagonal (p) and diagonal length difference with the other diagonal (d). Thus, the other diagonal is $p + d$.

From the geometry of a rhombus and the above definition, we can write,

$$\sqrt{l^2 - \frac{p^2}{4}} - \frac{p}{2} = \frac{d}{2} \tag{1}$$

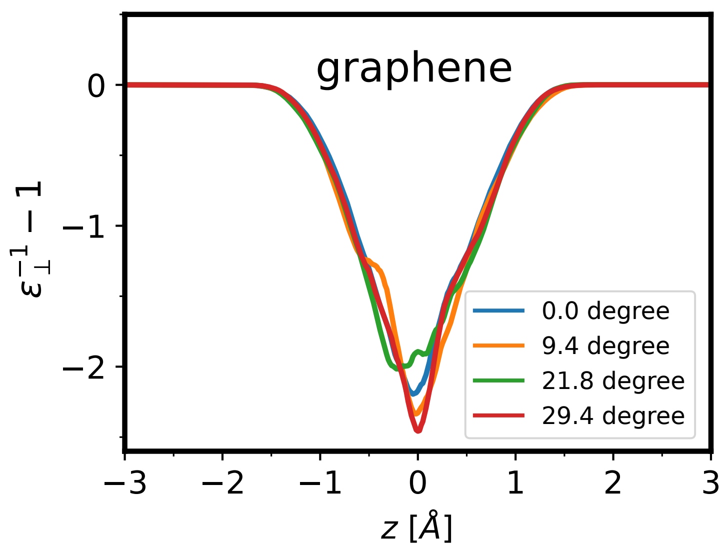

Figure 2S: Confined water dielectric profile inside bilayer graphene for various twist angles.

The solution to the above equation is,

$$\frac{p}{2l} = -\frac{d}{4l} + \frac{1}{4}\sqrt{8 - \frac{d^2}{l^2}} \tag{2}$$

Writing d as a fraction of l, *i.e.* $d = kl$ where $0 \leq k \leq 1$,

$$p = \frac{l}{2}(\sqrt{8 - k^2} - k) \tag{3}$$

We can use this value of p to construct a synthetic lattice that can be tuned between a square and rhombic ordered structure depending on the value of k used. Fig. 4S shows the representation of such a construction based on the lattice constant that can be written as,

$$\begin{aligned} \vec{a} &= \left(\frac{(p + kl)}{2}, \frac{p}{2}\right) \\ \vec{b} &= (p + kl, 0) \end{aligned} \tag{4}$$

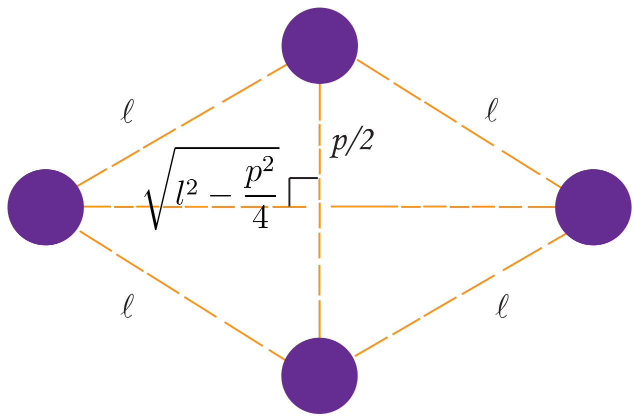

Figure 3S: A unit structure of rhombus. One diagonal of the rhombus is of length p, and all the sides of the rhombus are of length l. When $p = \sqrt{2}l$, both the diagonals will be of equal length, and it will be a perfect square.

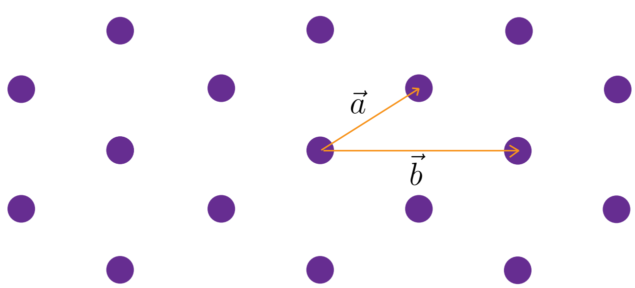

Figure 4S: A rhombic lattice with lattice vectors $\vec{a}$ and $\vec{b}$, constructed from small rhombus units. The structure can be smoothly turned to square and back to any general rhombus by varying k in the evaluation of $\vec{a}$ and $\vec{b}$.