Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

The below tables present the source data for the plots in Figure-4,5,6,8 and 9. All the plotted data were mean values of 200 samples collected at a frequency of one sample every 1 ps of simulation time, and the standard deviations are shown in parentheses.

Table-1S: Tilt angles for plots in Figure-4.

Temperature (K)	MONO (°)	ORTHA (°)	ORTHB (°)
50	32.71 (0.1)	0 (0)	16.14 (0.1)
100	32.30 (0.1)	0 (0)	15.50 (0.1)
150	32.04 (0.1)	0 (0)	14.52 (0.1)
200	31.75 (0.1)	0 (0)	13.68 (0.1)
250	28.25 (0.1)	0 (0)	0 (0)
300	26.018 (0.1)	0 (0)	0 (0)
320	24.75 (0.1)	0 (0)	0 (0)
330	10.69 (0.2)	6.09 (0.1)	6.09 (0.1)
333	7.05 (0.2)	6.05 (0.1)	6.05 (0.1)

Table-2S: System density for plots in Figure-5.

Temperature (K)	MONO (g/cm^3)	ORTHA (g/cm^3)	ORTHB (g/cm^3)
50	1037.90 (1.2)	1018.32 (0.5)	1007.69 (1.1)
100	1034.90 (1.1)	1004.64 (0.7)	994.81 (1.0)
150	1030.25 (1.1)	990.41 (0.7)	981.54 (1.1)
200	1025.94 (1.2)	975.26 (0.9)	964.84 (1.2)
250	986.98 (1.4)	956.90 (0.8)	957.02 (0.8)
300	942.02 (1.2)	940.54 (0.7)	940.12 (0.7)
320	925.79 (1.3)	922.18 (0.9)	922.39 (0.9)
330	905.05 (1.2)	908.22 (1.0)	908.04 (1.0)
333	902.29 (1.3)	902.36 (1.0)	902.36 (1.0)

Table-3S: Cell dimensions of MONO for plots in Figure-6a.

Temperature (K)	X (Å)	Y (Å)	Z (Å)	A (°)	β (°)	γ (°)
50	96.46 (0)	41.20 (0)	63.56 (0)	0 (0)	15.4 (0.1)	0 (0)
100	96.58 (0)	41.28 (0)	63.53 (0)	0 (0)	15.2 (0.1)	0 (0)
150	96.70 (0.1)	41.45 (0)	63.49 (0.1)	0 (0)	15.0 (0.1)	0 (0)
200	96.48 (0.1)	42.05 (0)	63.61 (0.1)	0 (0)	14.7 (0.1)	0 (0)
250	94.82 (0.1)	44.31 (0)	63.86 (0.1)	0 (0)	13.9 (0.1)	0 (0)
300	93.66 (0.1)	46.24 (0.1)	64.64 (0.1)	0 (0)	13.1 (0.1)	0 (0)
320	93.03 (0.2)	46.16 (0.1)	64.92 (0.1)	0 (0)	12.7 (0.1)	0 (0)
330	85.21 (0.2)	46.24 (0.1)	72.41 (0.1)	0 (0)	10.7 (0.1)	0 (0)
333	81.09 (0.2)	46.30 (0.2)	76.14 (0.1)	0 (0)	7.1 (0.1)	0 (0)

Table-4S: Cell dimensions of ORTHA for plots in Figure-6b.

Temperature (K)	X (Å)	Y (Å)	Z (Å)	α (°)	β (°)	γ (°)
50	73.29 (0)	46.75 (0)	74.17 (0)	0 (0)	0 (0)	0 (0)
100	74.00 (0)	46.75 (0)	74.17 (0)	0 (0)	0 (0)	0 (0)
150	74.61 (0)	46.77 (0)	74.28 (0)	0 (0)	0 (0)	0 (0)
200	75.49 (0)	46.71 (0)	74.86 (0)	0 (0)	0 (0)	0 (0)
250	76.31 (0.1)	46.73 (0)	75.47 (0.1)	0 (0)	0 (0)	0 (0)
300	77.22 (0.1)	46.79 (0.1)	76.54 (0.1)	0 (0)	0 (0)	0 (0)
320	78.37 (0.1)	46.94 (0.1)	76.79 (0.1)	0 (0)	0 (0)	0 (0)
330	80.45 (0.1)	47.11 (0.1)	76.53 (0.1)	0 (0)	0 (0)	0 (0)
333	80.77 (0.1)	47.33 (0.1)	76.17 (0.1)	0 (0)	0 (0)	0 (0)

Table-5S: Cell dimensions of ORTHB for plots in Figure-6c.

Temperature (K)	X (Å)	Y (Å)	Z (Å)	α (°)	β (°)	γ (°)
50	84.93 (0)	41.79 (0)	72.86 (0)	0 (0)	0 (0)	0 (0)
100	83.54 (0)	42.85 (0)	73.04 (0)	0 (0)	0 (0)	0 (0)
150	82.83 (0)	44.05 (0)	73.54 (0)	0 (0)	0 (0)	0 (0)
200	82.17 (0.1)	44.77 (0)	74.00 (0.1)	0 (0)	0 (0)	0 (0)
250	78.44 (0.1)	46.08 (0.1)	78.02 (0.1)	0 (0)	0 (0)	0 (0)
300	78.54 (0.1)	46.27 (0.1)	77.20 (0.1)	0 (0)	0 (0)	0 (0)
320	78.37 (0.1)	46.94 (0.1)	76.79 (0.1)	0 (0)	0 (0)	0 (0)
330	80.45 (0.1)	47.11 (0.1)	76.53 (0.1)	0 (0)	0 (0)	0 (0)
333	80.77 (0.1)	47.33 (0.1)	76.17 (0.1)	0 (0)	0 (0)	0 (0)

Table-6S: Enthalpy and system density of MONO for plots in Figure-8a.

Temperature (K)	Enthalpy (kJ/mol)	Density (g/cm^3)
300	4274.8 (62)	932.0 (1.2)
315	5502.4 (56)	919.3 (1.2)
330	6908.9 (77)	905.0 (1.2)
333	7222.6 (75)	902.2 (1.3)
334	8732. 2 (60)	887. 9 (1.1)
335	10178.4 (63)	861. 5 (1.3)
336	12520.3 (74)	846. 4 (1.3)
340	13270.7 (62)	841. 4 (1.2)
350	14434.6 (90)	826.3 (1.1)
400	18762.3 (93)	742. 1 (1.2)

Table-7S: Enthalpy and system density of ORTH for plots in Figure-8b.

Temperature (K)	Enthalpy (kJ/mol)	Density (g/cm^3)
300	3210.1 (87)	928.1 (0.7)
315	4502.4 (93)	919.3 (0.8)
330	5808.9 (66)	901.0 (1.0)
333	6522.7 (69)	897.3 (1.0)
334	9332.4 (81)	875.6 (0.9)
335	11078.5 (78)	851.4 (0.8)
336	13120.2 (95)	830.3 (0.9)
340	13470.6 (63)	826.7 (1.0)
350	14604.9 (88)	814.6 (1.2)
400	19017.0 (97)	742.4 (1.3)

Table-8S: Liquid density for plots in Figure-9.

Temperature (K)	Experiment (g/cm^3)	TraPPE-UA (This work) (g/cm^3)	TraPPE-UA (Papavasileiou[1]) (g/cm^3)
398.15	738.5	745.7 (0.3)	751.8 (0.2)
423.15	721.3	729.3 (0.3)	735.5 (0.4)
448.15	704	712.4 (0.3)	719.1 (0.4)
473.15	686.7	693.1 (0.3)	702.6 (0.5)
498.15	669.6	678.5 (0.4)	686.1 (0.4)
523.15	653	660.8 (0.4)	668.2 (0.6)
548.15	637	644.4 (0.4)	651.3 (0.6)
573.15	622	625.0 (0.5)	633.2 (1.1)

1. Papavasileiou KD, Peristeras LD, Bick A, Economou IG. Molecular Dynamics Simulation of Pure n-Alkanes and Their Mixtures at Elevated Temperatures Using Atomistic and Coarse-Grained Force Fields. *J Phys Chem B* 2019; **123**: 6229-43.