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SUPPLEMENTARY INFORMATION: Investigating the Quasi-Liquid Layer on Ice Surfaces: a Comparison of Order Parameters

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Ice surfaces are characterized by pre-melted quasi-liquid layers (QLLs), which mediate both crystal growth processes and interactions with external agents. Understanding QLLs at the molecular level is necessary to unravel the mechanisms of ice crystal formation. Computational studies of the QLLs heavily rely on the accuracy of the methods employed for identifying the local molecular environment and arrangements, discriminating between solid-like and liquid-like water molecules. Here we compare the results obtained using different order parameters to characterize the QLLs on hexagonal ice (Ih) and cubic ice (Ic) model surfaces investigated with molecular dynamics (MD) simulations in a range of temperatures. For the classification task, in addition to the traditional Steinhardt order parameters in different flavours, we select an entropy fingerprint and a deep learning neural network approach (DeepIce), which are conceptually different methodologies. We find that all the analysis methods give qualitatively similar trends for the behaviours of the QLLs on ice surfaces with temperature, with some subtle differences in the classification sensitivity limited to the solid–liquid interface. The thickness of QLLs on the ice surface increases gradually as the temperature increases. The trends of the QLL size and of the values of the order parameters as a function of temperature for the different facets may be linked to surface growth rates which, in turn, affect crystal morphologies at lower vapour pressure. The choice of the order parameter can be therefore informed by computational convenience except in cases where a very accurate determination of the liquid–solid interface is important.

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Table 1 The thickness in Å of the QLLs on the ice Ic (001) surface estimated with different classification methods in the 200-270 K temperature range.

Ice plane	200 K	210 K	220 K	230 K	240 K	250 K	260 K	270 K	Recipe
Ic (001)	2.95 ± 0.12	3.23 ± 0.15	3.46 ± 0.17	3.68 ± 0.22	4.41 ± 0.29	5.53 ± 0.35	6.96 ± 0.46	8.82 ± 0.81	laq_3
	3.06 ± 0.11	3.32 ± 0.14	3.49 ± 0.15	3.71 ± 0.22	4.41 ± 0.27	5.55 ± 0.36	7.02 ± 0.43	10.64 ± 0.82	lq_3
	3.14 ± 0.11	3.40 ± 0.12	3.67 ± 0.14	3.98 ± 0.17	4.65 ± 0.23	5.69 ± 0.29	7.13 ± 0.42	10.64 ± 0.79	le
	2.59 ± 0.11	2.83 ± 0.13	3.04 ± 0.14	3.23 ± 0.18	3.81 ± 0.24	4.72 ± 0.29	5.92 ± 0.41	7.99 ± 0.77	$le+laq_3$
	2.74 ± 0.13	2.93 ± 0.15	3.09 ± 0.16	3.27 ± 0.23	3.80 ± 0.28	4.68 ± 0.36	5.90 ± 0.44	9.15 ± 0.82	$le+lq_3$
	2.66 ± 0.14	2.66 ± 0.14	2.82 ± 0.16	3.03 ± 0.20	3.73 ± 0.29	4.90 ± 0.37	6.39 ± 0.49	10.04 ± 0.83	$nn10$
	1.68 ± 0.10	1.68 ± 0.11	1.79 ± 0.13	1.96 ± 0.17	2.60 ± 0.28	3.69 ± 0.36	5.13 ± 0.49	8.69 ± 0.83	$nn16$

Table 2 The thickness in Å of the QLLs on ice Ih basal, prism1 and prism2 surfaces estimated with different classification methods in the 240-270 K temperature range.

Ice plane	240 K	245 K	250 K	255 K	260 K	265 K	270 K	Recipe
Ih basal	4.14 ± 0.22	4.39 ± 0.24	4.73 ± 0.34	5.49 ± 0.45	5.98 ± 0.63	7.09 ± 0.31	7.55 ± 0.35	laq_3
	4.17 ± 0.24	4.43 ± 0.25	4.81 ± 0.29	5.62 ± 0.33	6.13 ± 0.39	7.28 ± 0.56	7.73 ± 0.86	lq_3
	4.85 ± 0.21	5.13 ± 0.22	5.54 ± 0.31	6.34 ± 0.42	7.20 ± 0.43	8.08 ± 0.30	8.62 ± 0.36	le
	3.41 ± 0.19	3.60 ± 0.21	3.89 ± 0.31	4.59 ± 0.42	5.14 ± 0.52	6.02 ± 0.26	6.39 ± 0.31	$le+laq_3$
	3.47 ± 0.21	3.67 ± 0.22	3.97 ± 0.25	4.69 ± 0.27	5.27 ± 0.35	6.13 ± 0.53	6.50 ± 0.84	$le+lq_3$
	3.47 ± 0.24	3.69 ± 0.27	4.08 ± 0.41	4.99 ± 0.57	6.08 ± 0.51	6.93 ± 0.25	7.39 ± 0.34	$nn10$
	4.65 ± 0.27	4.92 ± 0.29	5.35 ± 0.42	6.31 ± 0.56	7.50 ± 0.42	8.24 ± 0.26	8.72 ± 0.36	$nn16$
Ih prism1	4.40 ± 0.25	4.85 ± 0.26	5.20 ± 0.29	5.64 ± 0.30	6.07 ± 0.38	7.05 ± 0.58	10.27 ± 0.88	laq_3
	4.79 ± 0.21	5.25 ± 0.22	5.61 ± 0.31	6.09 ± 0.42	6.52 ± 0.43	7.54 ± 0.30	11.61 ± 0.36	lq_3
	4.71 ± 0.24	5.14 ± 0.25	5.57 ± 0.29	6.12 ± 0.33	6.65 ± 0.39	7.76 ± 0.56	11.87 ± 0.86	le
	3.59 ± 0.21	3.96 ± 0.22	4.25 ± 0.25	4.61 ± 0.27	4.99 ± 0.35	5.85 ± 0.53	8.77 ± 0.84	$le+laq_3$
	3.73 ± 0.19	4.10 ± 0.21	4.40 ± 0.31	4.78 ± 0.42	5.17 ± 0.52	6.04 ± 0.26	9.95 ± 0.31	$le+lq_3$
	3.29 ± 0.30	3.86 ± 0.31	4.21 ± 0.34	4.66 ± 0.34	5.14 ± 0.41	6.12 ± 0.63	10.07 ± 1.24	$nn10$
	4.39 ± 0.34	5.00 ± 0.33	5.42 ± 0.36	5.94 ± 0.36	6.46 ± 0.43	7.51 ± 0.62	11.49 ± 1.24	$nn16$
Ih prism2	4.74 ± 0.31	4.69 ± 0.21	5.12 ± 0.23	5.09 ± 0.26	5.75 ± 0.36	6.43 ± 0.43	11.64 ± 1.34	laq_3
	5.24 ± 0.24	5.17 ± 0.25	5.69 ± 0.29	5.59 ± 0.33	6.26 ± 0.39	6.96 ± 0.56	12.06 ± 0.86	lq_3
	5.02 ± 0.29	5.06 ± 0.22	5.54 ± 0.25	5.75 ± 0.27	6.40 ± 0.35	7.24 ± 0.42	12.13 ± 1.28	le
	3.81 ± 0.23	3.84 ± 0.17	4.08 ± 0.19	4.21 ± 0.22	4.69 ± 0.31	5.28 ± 0.37	10.08 ± 1.27	$le+laq_3$
	4.02 ± 0.21	4.05 ± 0.22	4.31 ± 0.25	4.43 ± 0.27	4.92 ± 0.35	5.52 ± 0.53	10.22 ± 0.84	$le+lq_3$
	3.65 ± 0.31	3.68 ± 0.19	3.96 ± 0.23	4.06 ± 0.27	4.69 ± 0.38	5.32 ± 0.44	10.72 ± 1.35	$nn10$
	4.92 ± 0.35	4.91 ± 0.22	5.32 ± 0.25	5.38 ± 0.28	6.09 ± 0.40	6.81 ± 0.45	12.18 ± 1.35	$nn16$

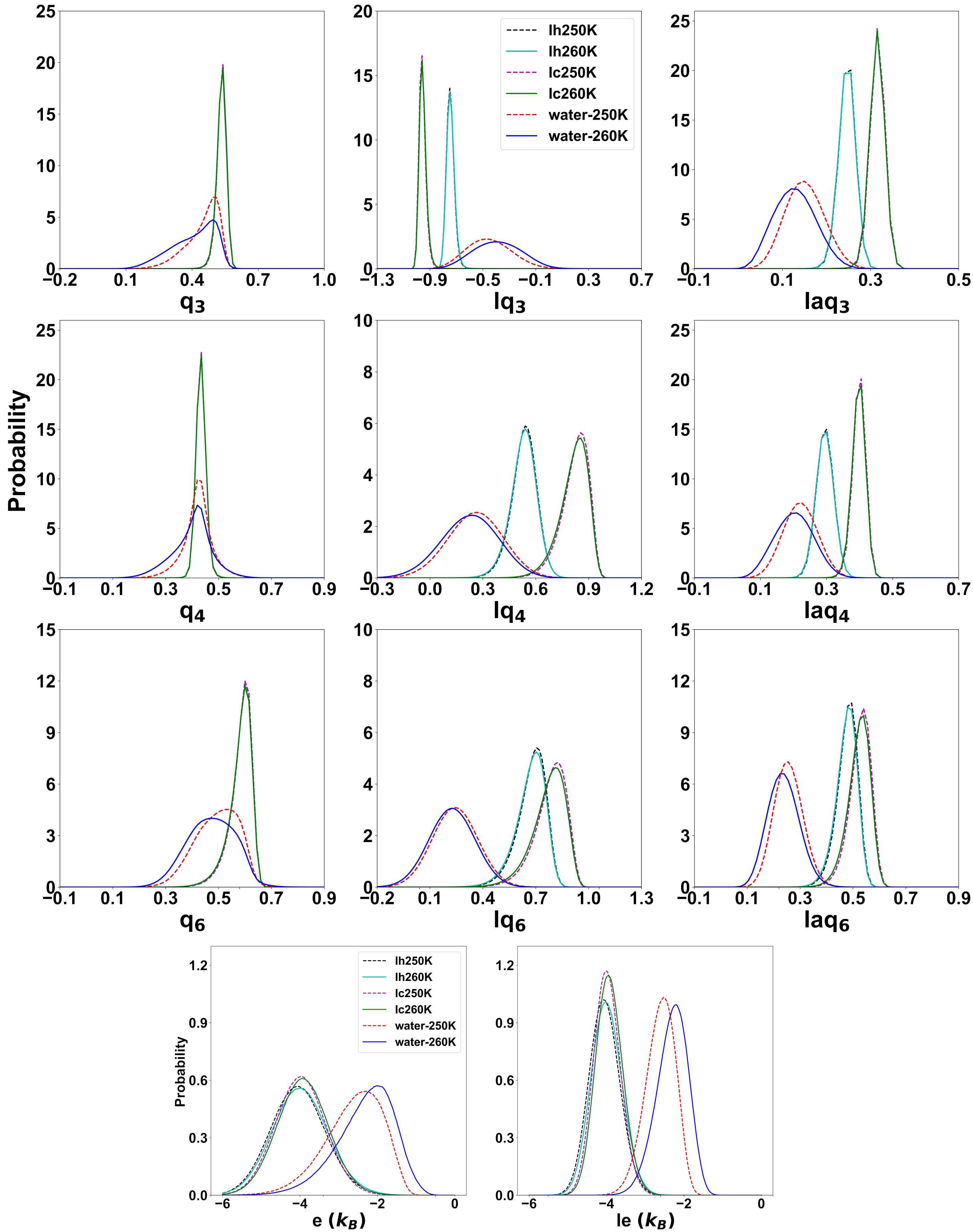

Fig. 1 The probability distribution of the Steinhardt parameters with $l = 3, 4, 6$ and the entropy fingerprint of e and le for bulk water and ice.

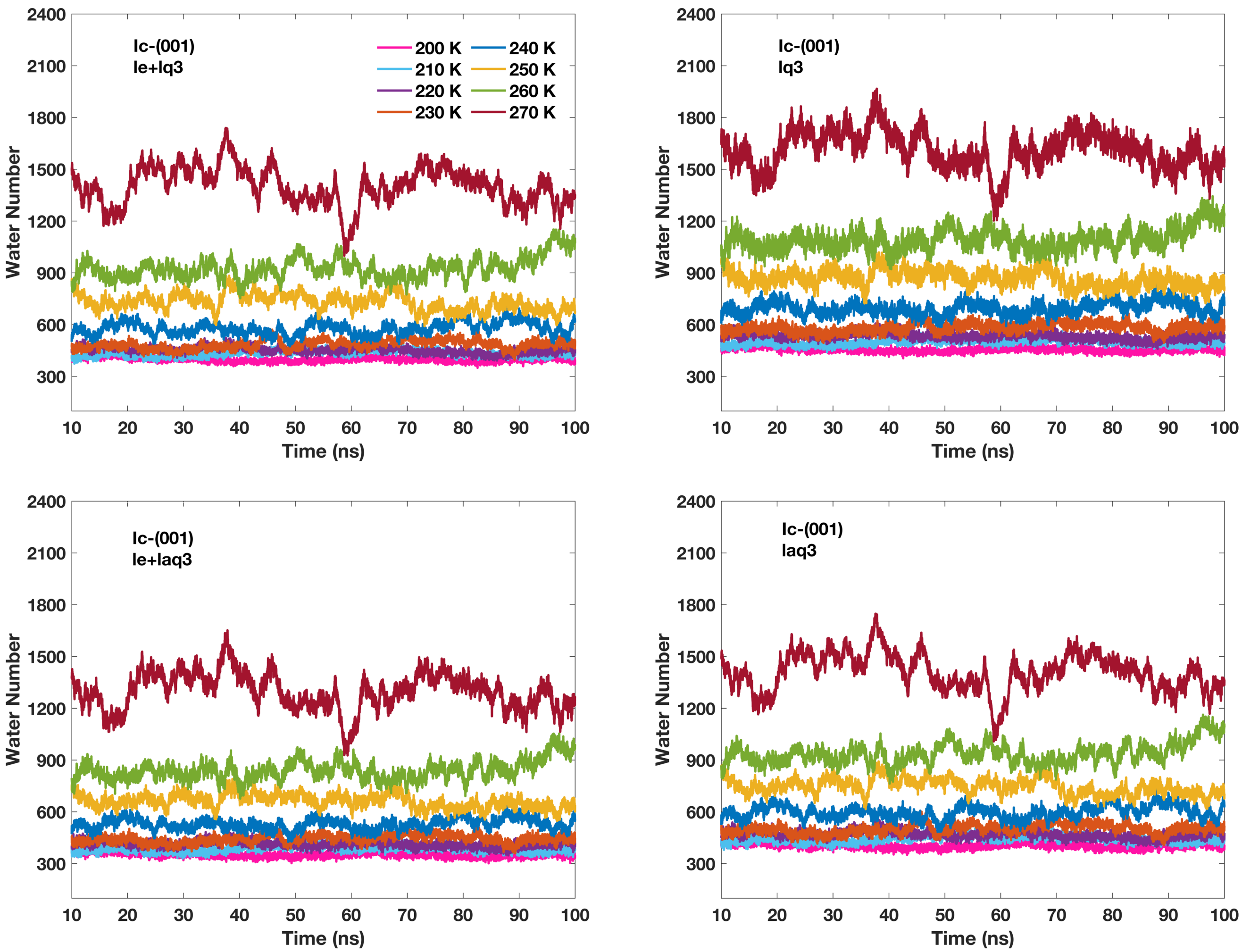

Fig. 2 Time-evolution of the number of water molecules in QLLs on the (001) surface of Ic in the 200-270 K temperature range, using the lq_3, laq_3, and le as order parameters.

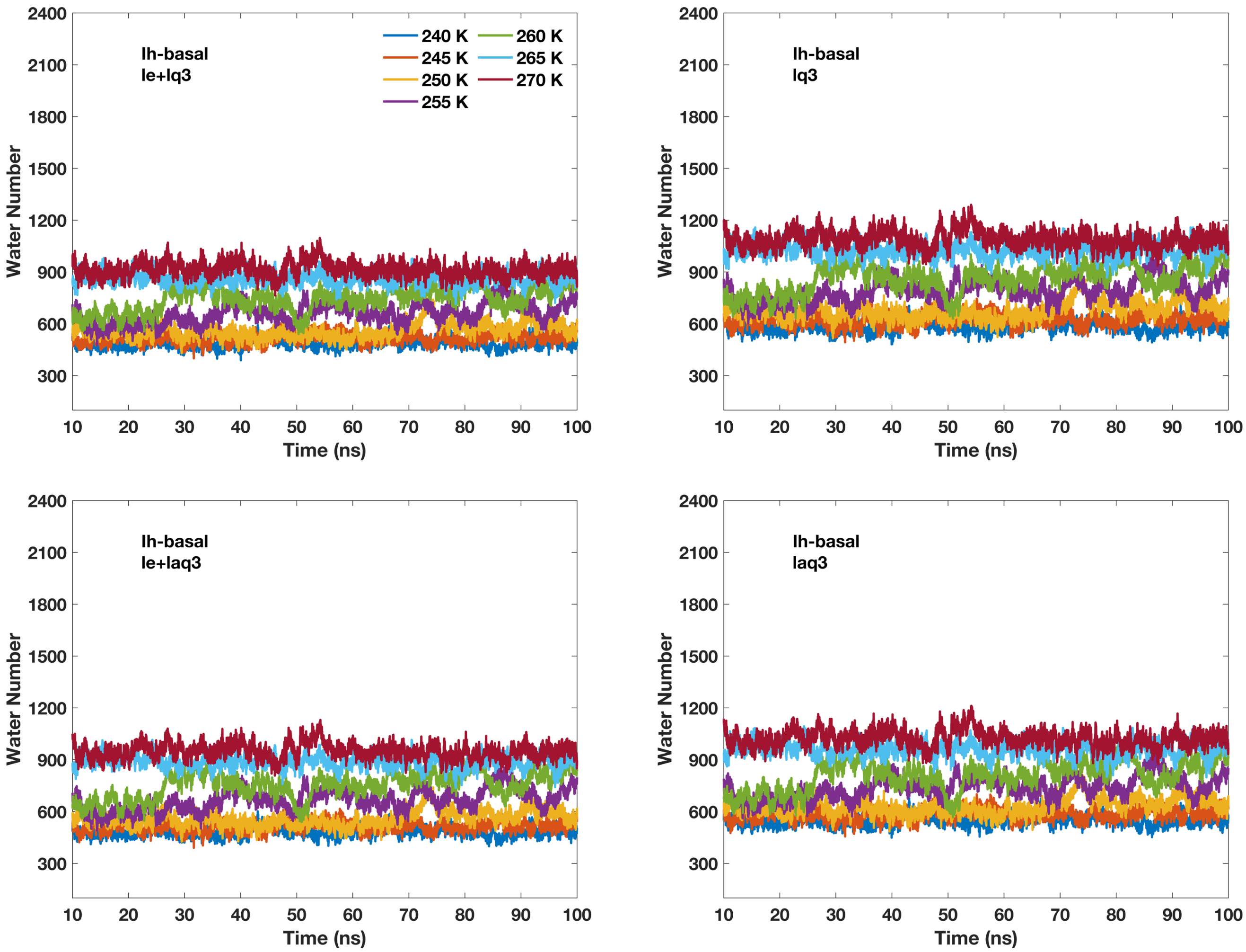

Fig. 3 Time-evolution of the number of water molecules in QLLs on the basal surface of Ih (with the temperature legend for Ih) in the 240-270 K temperature range, using the lq_3, laq_3, and le as order parameters.

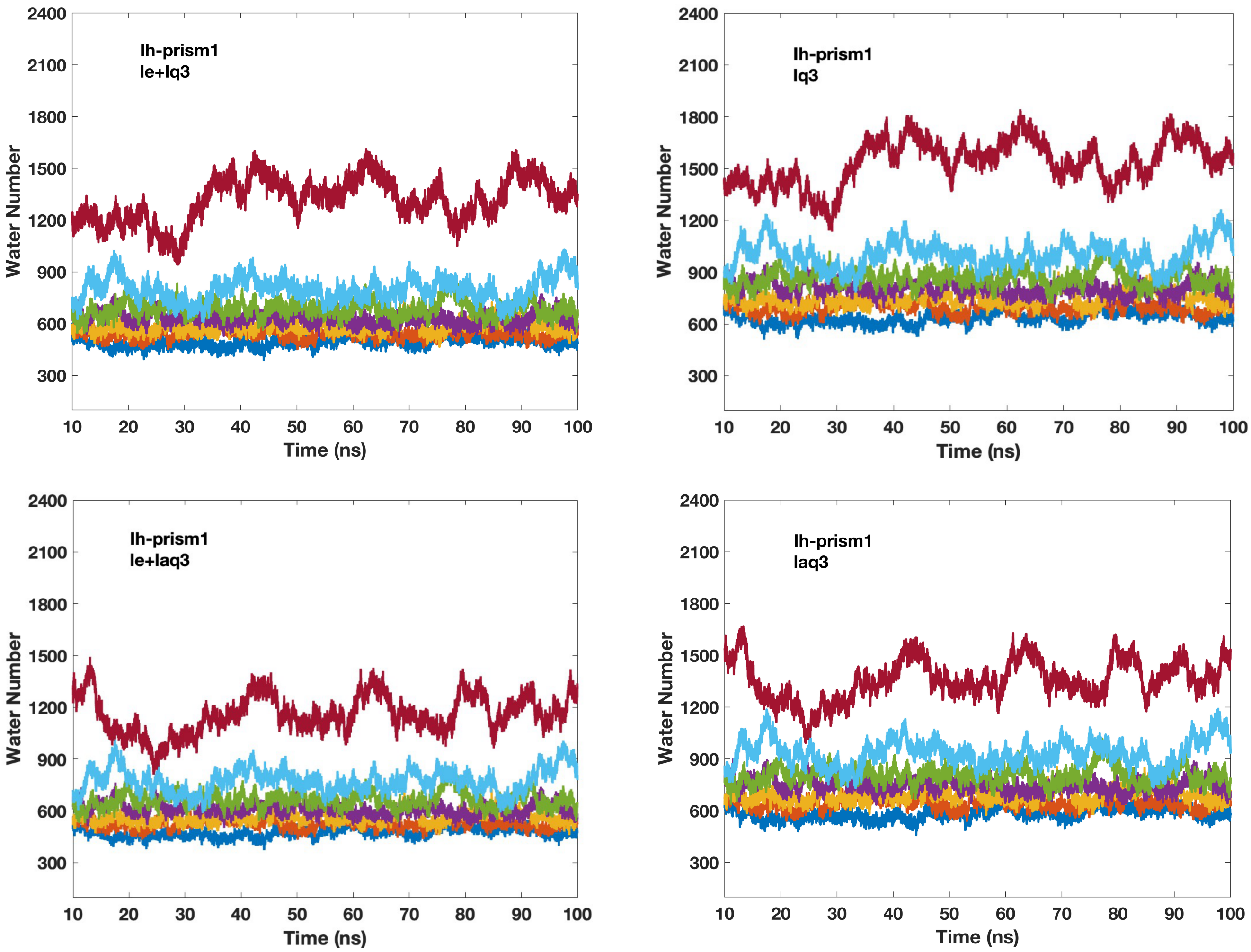

Fig. 4 Time-evolution of the number of water molecules in QLLs on the prism1 surface of Ih (with the same legend as in Fig. 3 for Ih) in the 240-270 K temperature range, using the lq_3, laq_3, and le as order parameters.

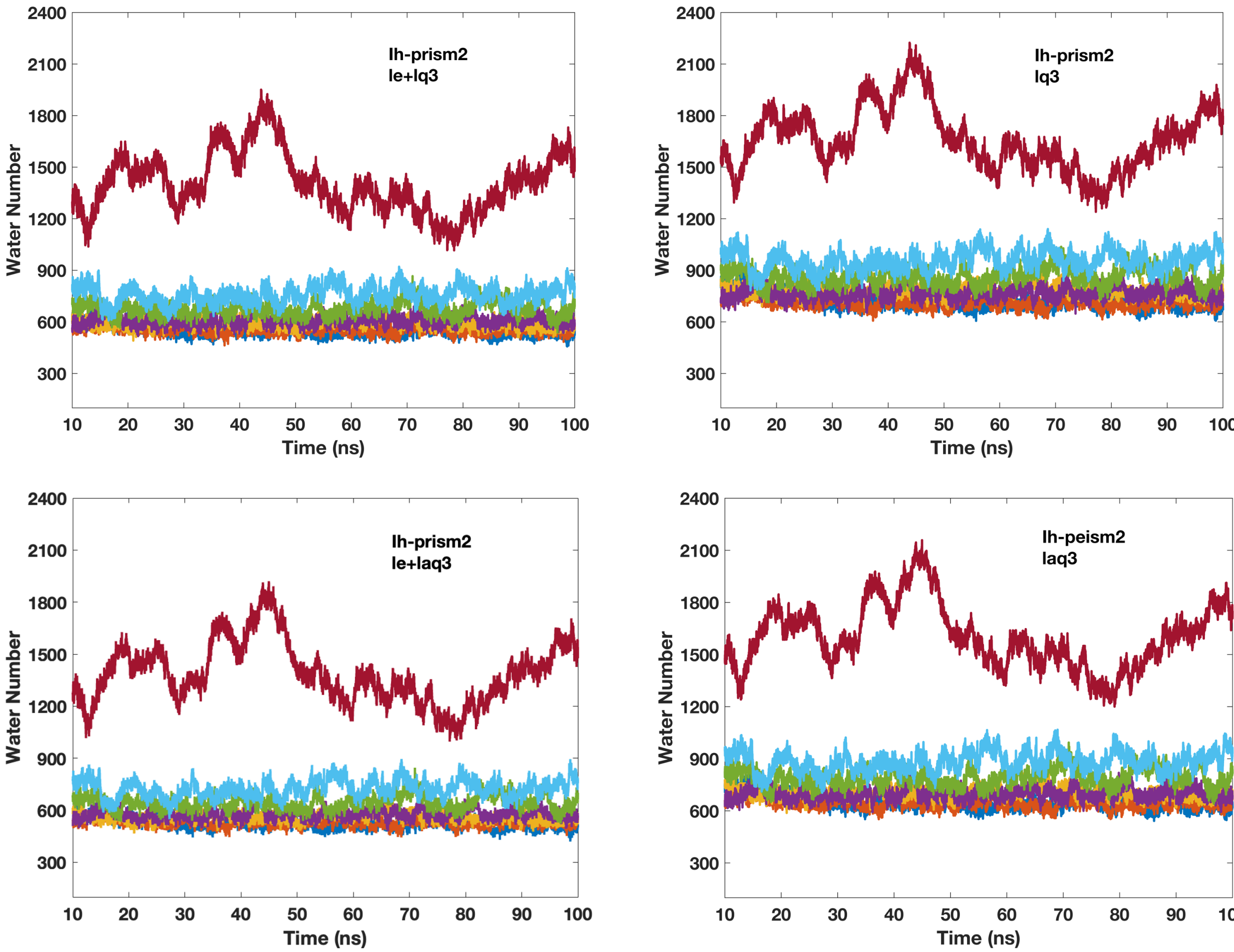

Fig. 5 Time-evolution of the number of water molecules in QLLs on the prism2 surface of Ih (with the same legend as in Fig. 3 for Ih) in the 240-270 K temperature range, using the lq_3, laq_3, and le as order parameters.

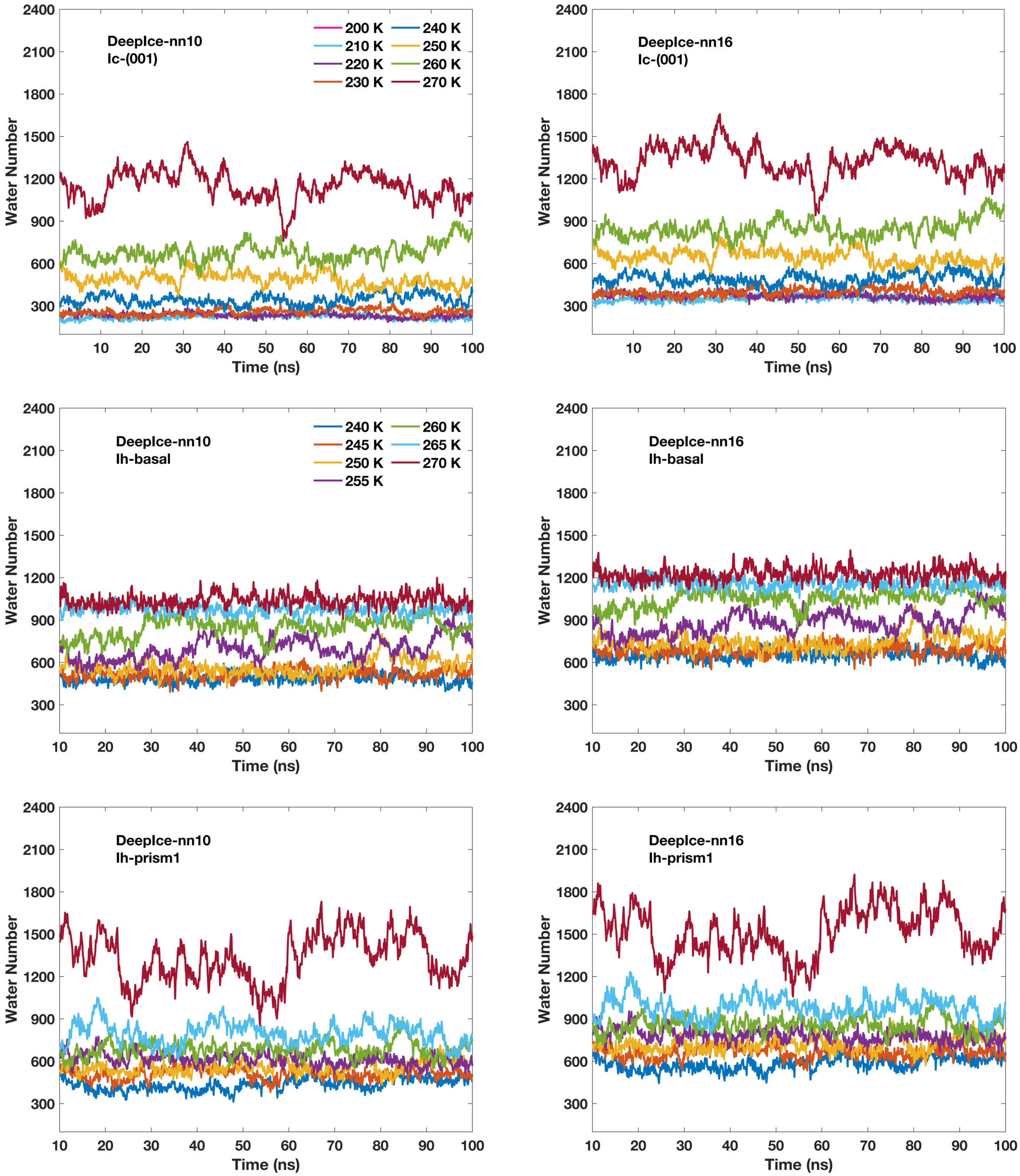

Fig. 6 Time-evolution of the number of water molecules in QLLs on the (001) surface of Ic (top panels, with temperature legend for Ic) in the 200-270 K temperature range and on the basal (middle panels, with temperature legend for Ih), prism1 (bottom panels) surfaces of Ih in the 240-270 K temperature range, using DeepIce-nn10 and DeepIce-nn16.