Supporting Information

Structural features that modulate the sharpness of the spin crossover transition in [Fe\textsuperscript{III}(5-X-qsal)\textsubscript{2}]\textsuperscript{+} based salts

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Fig. S1: Chain Layers supramolecular structure in 4.PF\textsubscript{6}.1.5H\textsubscript{2}O. Cations belonging to the same layer share the same color: top layer orange cations, middle layer light blue cations and bottom layer green cations.

Fig. S2: Overlap of the Qn fragments in the interchain connectivity between cations of 4.CF\textsubscript{3}SO\textsubscript{3}.iPrOH (top), 4.CF\textsubscript{3}SO\textsubscript{3}.nPrOH (center) and 4.CF\textsubscript{3}SO\textsubscript{3}.MeOH (bottom). The short contacts colour code corresponds to the difference, \(\Delta\) in Å, between the distance of the contact and the sum of the van der Waals radii of the involved atoms: violet \(\Delta < -0.1\); light blue \(\Delta < 0.0\).

Fig. S3: Interlayer DD and DAD connectivity of cations regarding 2.PF\textsubscript{6}.MeCN (a and d), 3.PF\textsubscript{6}.H\textsubscript{2}O (b and e) and 5.PF\textsubscript{6}.1.5H\textsubscript{2}O (c and f). The short contacts colour code corresponds to the difference, \(\Delta\) in Å, between the distance of the contact and the sum of the van der Waals radii of the involved atoms: violet \(\Delta < -0.1\); light blue \(\Delta < 0.0\); orange \(\Delta < 0.1\).

Fig. S4: Interlayer DD and DAD contacts in 1.I\textsubscript{3}. Cations in the same layer have the same color (top layer orange and bottom layer light blue). The short contacts colour code corresponds to the difference, \(\Delta\) in Å, between the distance of the contact and the sum of the van der Waals radii of the involved atoms: violet \(\Delta < -0.1\); light blue \(\Delta < 0.0\). (adapted from Figure S18 of reference [7]).
**Fig. S1:** Chain Layers supramolecular structure in $4\text{PF}_6\cdot1.5\text{H}_2\text{O}$. Cations belonging to the same layer share the same color: top layer orange cations, middle layer light blue cations and bottom layer green cations.
Fig. S2: Overlap of the Qn fragments in the interchain connectivity between cations of 4.CF₃SO₃-iPrOH (top), 4.CF₃SO₃-nPrOH (center) and 4.CF₃SO₃-MeOH (bottom). The short contacts colour code corresponds to the difference, \( \Delta \) in Å, between the distance of the contact and the sum of the van der Waals radii of the involved atoms: violet \( \Delta < -0.1 \); light blue \( \Delta < 0.0 \).
Fig. S3: Interlayer DD and DAD connectivity of cations regarding 2.PF₆·MeCN (a and d), 3.PF₆·H₂O (b and e) and 5.PF₆·1.5H₂O (c and f). The short contacts colour code corresponds to the difference, \( \Delta \), in Å, between the distance of the contact and the sum of the van der Waals radii of the involved atoms: violet \( \Delta < -0.1 \); light blue \( \Delta < 0.0 \); orange \( \Delta < 0.1 \). (adapted from Figure S18 of reference [7]).
Fig. S4: Interlayer DD and DAD contacts in 1.Ia. Cations in the same layer have the same color (top layer orange and bottom layer light blue). The short contacts colour code corresponds to the difference, $\Delta$ in Å, between the distance of the contact and the sum of the van der Waals radii of the involved atoms: violet $\Delta < -0.1$; light blue $\Delta < 0.0$. 