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## **Supporting information**

## Massive hydration-driven swelling of layered perovskite niobate crystals in aqueous solutions of organoammonium bases

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S1. Powder XRD data of KCa<sub>2</sub>Nb<sub>3</sub>O<sub>10</sub> before and after acid treatment.



**Fig. S1** XRD patterns of (a) KCa<sub>2</sub>Nb<sub>3</sub>O<sub>10</sub> and (b) HCa<sub>2</sub>Nb<sub>3</sub>O<sub>10</sub>  $\cdot$  1.5H<sub>2</sub>O obtained by the acid treatment of KCa<sub>2</sub>Nb<sub>3</sub>O<sub>10</sub>. All the peaks can be indexed in terms of orthorhombic [*a* = 0.3875(1) nm, *b* = 0.7709(3) nm, *c* = 2.9486(8) nm] and tetragonal [*a* = 0.3855(1) nm, *c* = 1.6225(2) nm] unit cell dimensions for KCa<sub>2</sub>Nb<sub>3</sub>O<sub>10</sub> and HCa<sub>2</sub>Nb<sub>3</sub>O<sub>10</sub>  $\cdot$  1.5H<sub>2</sub>O, respectively, which are compatible with previous reports.<sup>1,2</sup>

S2. Polarized optical microscope image containing non-swollen component of the crystals.



Fig. S2 Polarized optical microscope image for the sample in the TBAOH solution at TBA $^+/H^+$ 

= 0.1, highlighting the non-swollen component of the crystals.





Fig. S3 XRD data of the swollen samples in the TMAOH (upper panel) and DMAE (lower panel) solutions at indicated  $N^+/H^+$  molar ratios. The numerals on the peak correspond to the interlayer distances.

## Reference

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2. M. Dion, M. Ganne and M. Tournoux, *Mater. Res. Bull.*, 1981, 16, 1429–1435.