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Perfect restacking of 2D-materials from their liquid-crystalline nanosheets suspensions

Davidson, Patrick^{1,*}; Cherni, Lina²; El Rifaii, Karin¹; Wensink, Henricus H.¹; Chevrier, Sarah M.²; Goldmann, Claire¹; Michot, Laurent J.³; Gabriel, Jean-Christophe²

*Corresponding author: <u>patrick.davidson@universite-paris-saclay.fr</u>

¹Laboratoire de Physique des Solides, Université Paris-Saclay, CNRS, Orsay, France; ²Université Paris-Saclay, CEA, CNRS, NIMBE-LICSEN, Gif-sur-Yvette, France; ³PHENIX, Sorbonne Université, CNRS, Paris, France.

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Aqueous suspensions of H₃Sb₃P₂O₁₄ mineral nanosheets are one of the rare examples of colloids based on 2D materials exhibiting a lamellar liquid crystalline (LC) phase [1,2]. Because the lamellar period can reach several hundred nanometers, the suspensions show vivid structural colors and because these colors are sensitive to various chemicals, the suspensions can be used as sensors [3]. Such applications require the well-ordered deposition of the nanosheets from suspension to prepare solid-state devices. We report here the crystalline restacking of the layered $H_{3(1-x)}M_{3x}Sb_3P_2O_{14}$, (where M = Li, Na, K, Rb, Cs and $0 \le x \le 1$) dielectric materials from their nanosheets dispersed in colloidal LC suspension, induced by a simple pH change using alkaline (MOH) bases [4]. Using polarized-light microscopy and X-ray scattering, we show that thanks to the lamellar LC order of the colloidal suspension, the restacking is highly accurate and avoids the turbostratic disorder of the nanosheets classically observed with other 2D materials. Strikingly, the restacking process exhibits features highly reminiscent of a firstorder phase transition, with a phase coexistence region where both ~ 1 nm and ~ 120 nm lamellar periods are observed simultaneously, which we have described theoretically. This crystalline restacking of nanosheets from their lamellar LC suspensions opens the way to the production of complex integrated electronic, spintronic or photonic structures.

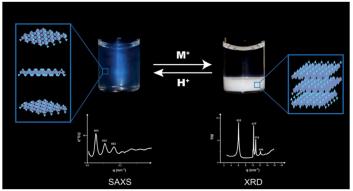


Figure 1: Crystalline restacking (right) of the 2D phosphatoantimonic H_{3(1-x)}M_{3x}Sb₃P₂O₁₄ materials from their nanosheets dispersed as a colloidal lamellar LC suspension (left), induced by a simple pH change using alkaline bases.

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