

Manipulating the magnetism of NiPS₃ via organic ion intercalation

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Intercalation of layered materials is a powerful fabrication and processing tool to modify the optical, electronic and magnetic properties of the host lattice [1-3]. In this study, we performed an electrochemical intercalation of different alkylammonium cations into NiPS₃ bulk crystals (Fig. 1a). Moreover, we explored an interlamellar alkylammonium-Co(Cp)₂⁺ exchange, which is thermodynamically favored for NiPS₃. X-ray diffraction spectra show the appearance of new (00l) peaks' families at lower 2θ angles, attributed to an increase of host lattice's interlayer distance (Fig. 1b). Raman spectra reveals clear fingerprints of the intercalation. Finally, magnetization measurements demonstrate a dramatic change in the NiPS₃ magnetic properties, with the suppression of the pristine antiferromagnetism and the emergence of ferrimagnetism or paramagnetism (Fig. 1c), depending on the guest specie. Our results indicate that molecular intercalation is a promising route to tailor the magnetic properties of layered materials.

References

- [1] J. Zhou *et al.* Adv. Mater. 33, 2004557 (2021);
- [2] M. Rajapakse, *et al.* 2D Mater. Appl. 5, 30 (2021);
- [3] L. K. Ma *et al.* Phys. Rev. Materials, 4, 124803 (2020)

Figures

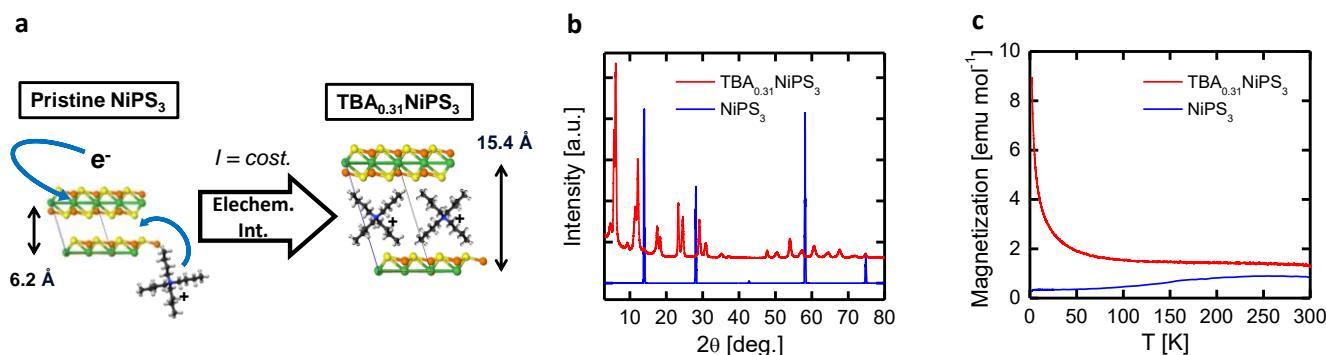


Figure 1: (a) Schematic of the electrochemical intercalation. Upon reduction of NiPS₃ host lattice, TBA⁺ cations are readily intercalated in between the Van der Waals gaps. (b) XRD patterns of pristine NiPS₃ and TBA⁺ int. NiPS₃. (c) Magnetization vs. temperature of pristine NiPS₃ and TBA⁺ int. NiPS₃ at 500 Oe. Pristine-antiferromagnetism is replaced by paramagnetism.