chem2Dmat
European Conference on Chemistry of Two-Dimensional Materials

# Tuning the magnetism of $NiPS_3$ and $MnPS_3$ through organic ion intercalation

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Organic ion intercalation is a powerful tool to tailor the physical properties of layered materials [1-3]. Here, we report on the tuning of the magnetic properties of antiferromagnetic $NiPS_3$ ($T_N$ = 155 K) through electrochemical intercalation of tetrabutylammonium ions ($TBA^+$), followed by cobaltocenium ($Co(Cp)_2^+$) – $TBA^+$ exchange in solution (Figure 1a)[4]. On the other hand, $MnPS_3$ was processed in aqueous solutions of four alkylammonium bromide salts ($R_4NBr$, with $R_4$ = tetramethyl, tetraethyl, tetrabutyl, cetyltrimethyl), leading to the corresponding hybrid intercalates via direct cation exchange. X-ray diffraction, Raman, and gravimetric studies were carried out for structural characterizations. Finally, magnetometry measurements revealed the suppression of pristine antiferromagnetism and the emergence of molecular-dependent ferrimagnetism for both intercalated host materials. We found that $TBA_{0.25}NiPS_3$ ($T_C$ = 78 K) and $[Co(Cp)_2]_{0.25}NiPS_3$ ($T_C$ = 98 K) show doping-induced magnetizations in the order of $10^{-2}$ $\mu_B$/atom (Figure 1b) [4]; conversely, the four $R_4N$-$MnPS_3$ intercalates ($T_C$ ~ 45-55 K) display a guest's size-dependent magnetism with saturation magnetization reaching up almost 1 $\mu_B$/atom. Our results establish organic ion intercalation as an effective tool to control the magnetism of layered magnetic materials.

## References

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## Figures

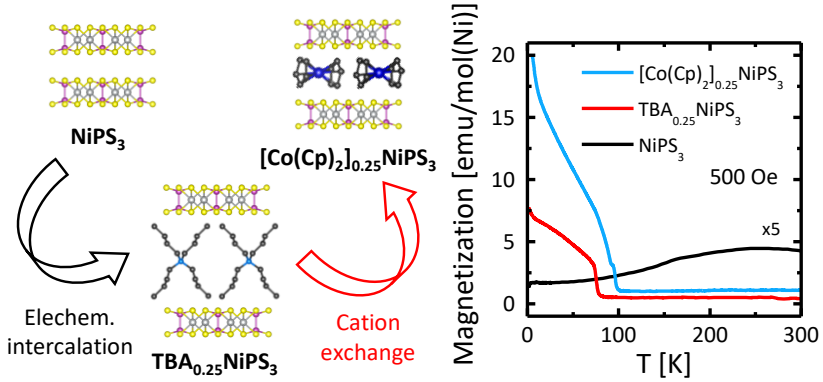


**Figure 1: a)** Schematic of the two-step intercalation process for $NiPS_3$: electrochemical intercalation of $TBA^+$ ions is followed by a $TBA^+/Co(Cp)_2^+$ exchange; **b)** Molar magnetization vs. temperature of pristine $NiPS_3$, $TBA_{0.25}NiPS_3$ and $[Co(Cp)_2]_{0.25}NiPS_3$.