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## Exploring Ferrotoroidic Order in $\text{LiNi}_{0.8}\text{Fe}_{0.2}\text{PO}_4$

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Ferrotoroidicity is the fourth ferroic order characterized by the simultaneous breaking of both time and space-reversal symmetries [1]. The intrinsic magnetoelectric effect associated with this order makes it interesting for energy-efficient memory devices. The ability to pole and control ferrotoroidal domains could prove important for non-volatile and high-density data storage applications. [2]

Lithium orthophosphates,  $\text{LiMPO}_4$  ( $M = \text{Mn, Ni, Co, Fe}$ ) are a family of antiferromagnets that crystallize in the olivine crystal structure (orthorhombic space group  $\text{Pnma}$ ) and exhibit a magnetoelectric effect below their ordering temperature [3]. The distinguishing factor among these compounds is the preferred orientation of magnetic moment. For instance, in  $\text{LiNiPO}_4$  and  $\text{LiFePO}_4$  the magnetic moments are aligned along the  $c$  and  $b$  axes respectively [4]. For the stoichiometric compounds, the form of the magnetoelectric tensor ( $\alpha$ ) which connects the electric polarisation to the applied magnetic field is determined by their magnetic point group [5]. Additionally, this family of compounds is notable due to their potential to support ferrotoroidal order. The non-zero off-diagonal elements in  $\alpha$  in  $M = \text{Fe, Co, and Ni}$  suggest the possible existence of ferrotoroidic order [2] which was experimentally confirmed in  $\text{LiCoPO}_4$  [6].

In this study, we focus on the ferrotoroidic order in the mixed compound  $\text{LiNi}_{0.8}\text{Fe}_{0.2}\text{PO}_4$ , where Ni sites are doped with Fe. Previous research on this compound identified a distinct low-temperature magnetic phase due to the combined effect of exchange interaction and mismatched anisotropy. Initially, the magnetic moments align along the  $b$ -axis below 25 K similar to  $\text{LiFePO}_4$ . However, below 21K they rotate towards the  $a$ -axis, a direction different from the easy axes of both parent compounds. In this phase, where we expect four domains, the strength of magnetoelectric coupling is increased by 100-fold [7].

Using spherical neutron polarimetry, we investigated the ferrotoroidic order in  $\text{LiNi}_{0.8}\text{Fe}_{0.2}\text{PO}_4$  giving us direct insight into the magnetic domain distribution and poling behaviour under crossed magnetic and electric fields. Our results suggest mixed-anisotropy ferrotoroidal systems to be a promising route towards next-generation storage devices.

### References

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