Huge Spin-driven Polarization and Magnetic Frustration in Multiferroics from Neutron Scattering

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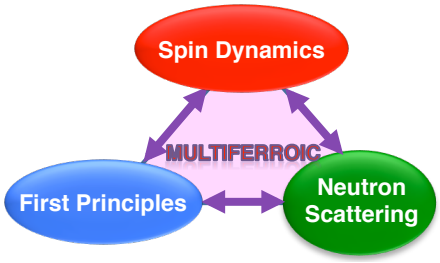
The strong interplay of order parameters including spin, electric polarization, and orbital ordering are responsible for various intriguing phenomena such as multiferroicity and magnetic frustration. While cooperative coupling in multiferroics can induce simultaneous order or more than one degree of freedom, frustrated geometries such as the pyrochlore lattice hamper such ordering and can produce more complex phases such as spin ice or spin liquid.



Neutron scattering measurements are a powerful tool to uncover the hidden couplings between order parameters. In particular, neutron scattering combined with first-principles calculations and spin models provide synergistic methods to understand the microscopic origins of the strong couplings and related phenomena in multiferroic oxides. We discuss two case studies.

As a representative case fr revealing hidden multiferrocities with neutron scattering, we show huge spin-driven ferroelectric polarizations arise in a type-*I* multiferroic $BiFeO_3$. Although $BiFeO_3$ is one of the most thoroughly investigated multiferroics, its magnetoelectric couplings are barely understood on an atomic level. By combining a first-principles approach with a spin-cycloid model, we report hidden but huge spin-driven polarizations at room temperature in bulk $BiFeO_3$. One of the polarizations reaches ~3.0 μC/cm², which is larger than any other spin-driven polarization in a bulk material by one order of magnitude [1] as shown the comparison between our results (lines) and elastic neutron scattering data (dots) in Fig. 1. We disentangle all the spin-driven polarizations due to exchange-striction, spin-current, and single-ion-anisotropy. The broken inversion symmetries of *R*3c $BiFeO_3$ induce the strong response of the magnetic interactions to an electric field and are responsible for the associated huge spin-driven polarizations. This neutron scattering study may motivate future investigation of the hidden spin-driven ferroelectric polarizations in various type-*I* multiferroics including $YMnO_3$ and $BiMnO_3$, as well as $BiFeO_3$, whose macroscopic magneto-capacitance measurements are critically limited.

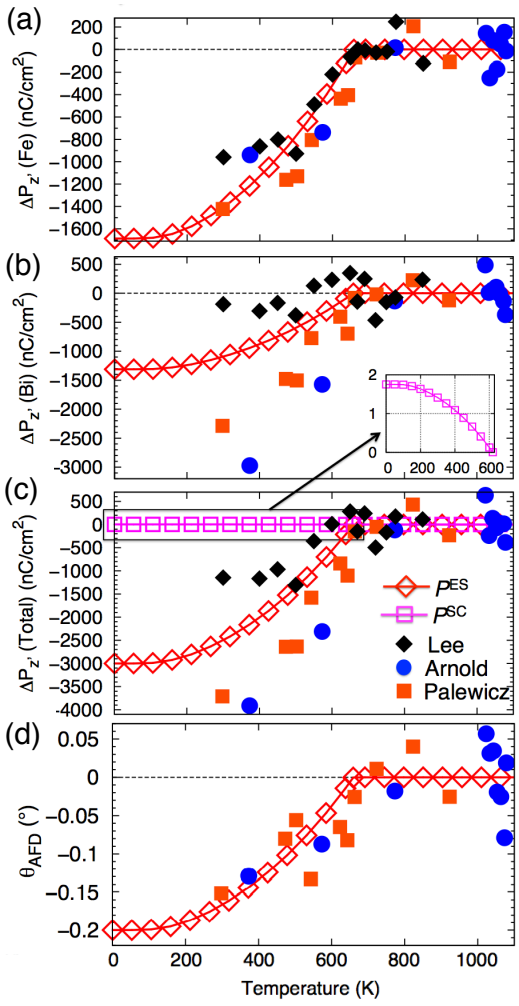


Figure 1. *Spin-driven polarizations by* ***(a)*** *Fe* ***(b)*** *Bi* ***(c)*** *Fe+Bi* ***(d)*** *oxygen in $BiFeO_3$ from elastic neutron scattering.*

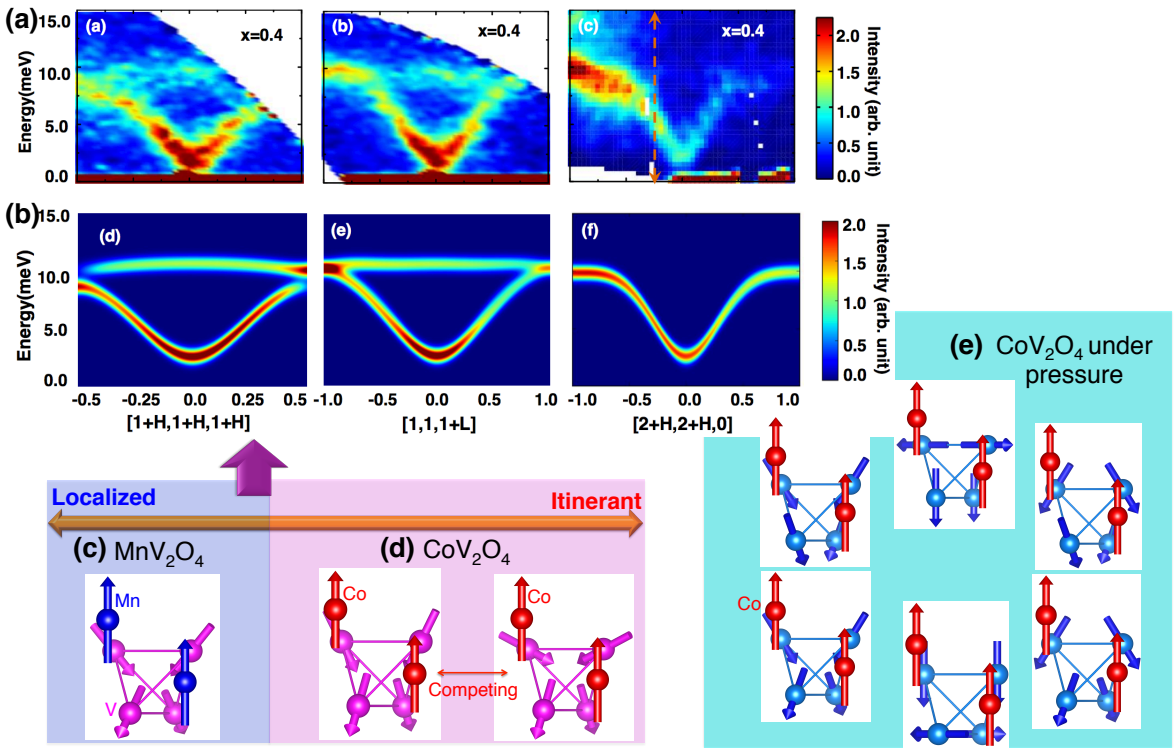


Fig 2. *Inelastic neutron scattering for multiferroic $Mn_{0.4}Co_{0.6}V_2O_4$ from* ***(a)*** *experiment and* ***(b)*** *spin model.* ***(c,d)*** *Iso-symmetric spin states.* ***(e)*** *Pressure-induced spin-glass state in CoV_2O_4.*

As a representative case for how magnetic frustration is revealed by neutron scattering measurements, we study a multiferroic spinel where frustration is driven by electronic itinerancy. By applying the same methodology as for $BiFeO_3$, we uncover novel phases induced by the competition between localized and itinerant electrons in the geometrically-frustrated system $Mn_{1-x}Co_xV_2O_4$ (Fig. 2). At low Co doping, the orbital ordering (OO) of the localized V^{3+} spins suppresses magnetic frustration by triggering a tetragonal distortion. With Co doping, however, electronic itinerancy melts OO and revives the suppressed frustration by weakening the structural and magnetic anisotropies, thereby producing spin-glass and spin-liquid-like degenerate states [2]. The measured non-collinear spin states in the Co-rich region exhibit the strong competition between localized spins and electronic itinerancy and give rise to novel phases in a geometrically-frustrated lattice [3]. This frustration may also be responsible for emergent phenomena including skyrmions in various multiferroic spinel compounds.

References

[1] J. H. Lee and R. Fishman (In review by *Nature Communications*)
[2] J. H. Lee, S. Hahn *et al.* (In review by *Physical Review X*)
[3] J. Ma, J. H. Lee, S. Hahn *et al.* (In review by *Physical Review X*)

* These works have been collaborated with Randy Fishman, Masaaki Matsuda, Steven Hahn, Jie Ma, Huibo Cao, Tao Hong, Satoshi Okamoto (ORNL), and Heidong Zhou (UTK).