O05

Unraveling the Strongly Entangled Spin-Dipole-Lattice Behavior: Phenomena Driven by Helical Magnetic Order

Arkadeb Pal[a,b], T. W. Yen[b], V. G. Sathe[c], Y. C. Chuang[d], and H. D. Yang[b]

[a]*ZIAM, University of Groningen, 9700 AB Groningen, The Netherlands*
[b]*Department of Physics, National Sun Yat-sen University, Kaohsiung 804, Taiwan, R. O. C.*
[c]*UGC-DAE Consortium for Scientific Research, Khandwa Road, Indore-452001, India*
[d]*National Synchrotron Radiation Research Center, Hsinchu, 30076, Taiwan, R. O. C.*

The growing demand for advanced spintronic devices has created a strong impetus for research into multifunctional materials capable of responding dynamically to a variety of external stimuli. These innovative materials are distinguished by the intricate coupling of microscopic order parameters, including spin, orbital, lattice, dipole, and phonon, which result in a diverse array of interesting properties. Researchers have focused on the discovery and development of new multiferroic and magnetodielectric materials that have the potential for numerous real-world applications across technology sectors. However, these materials are relatively scarce; their scarcity can be attributed to the antagonistic nature of their magnetic and electric properties, which are often limited by low critical temperatures, typically below 40 K. The challenge of engineering materials to achieve higher critical temperatures remains a significant obstacle, with only a handful of compounds, such as CuO (T_N ~ 230 K) and certain hexaferrites, demonstrating coupled properties at significantly elevated temperatures.

An especially fascinating phenomenon within this context is Spin-Phonon Coupling (SPC), which occurs in strongly correlated systems. SPC is characterized by magnetic and structural phase transformations that are reflected in the phonon spectra of these materials. This coupling serves as a valuable gauge for monitoring a variety of exotic properties, which include magnetoelectric coupling, the spin Seebeck effect, magnetostriction, the phonon Hall effect, the spin-Peierls transition, and the thermal Hall effect, all of which are pivotal in the study of multiferroics.

In this report, we demonstrate a spectrum of simultaneously occurring and highly entangled phenomena induced by helical spin ordering within the polar and spin-frustrated magnetic system $Fe_3(PO_4)O_3$. Key phenomena observed in this unique system include magnetodielectric coupling in conjunction with weak ferroelectric ordering. Additionally, we note a distinct magnetostriction effect, which is evidenced by a dramatic reduction in the thermal variation of the lattice parameters. The system also exhibits pronounced spin-phonon coupling, characterized by unique and anomalous hardening and softening of various phonon modes at temperatures reaching T_N = 163 K.

To investigate these effects, we utilized high-resolution synchrotron X-ray diffraction (SXRD) to probe the structural changes that occur across the magnetic transition temperature. If the spontaneous lattice distortion resulting from magnetic ordering is sufficiently pronounced, the resultant magnetostriction effect can be quantitatively assessed using SXRD data. This approach offers notable advantages over more conventional techniques such as capacitive and strain-gauge measurements. Our findings revealed a clear downturn in the lattice parameters, including the unit-cell volume, indicating a significant volume magnetostriction effect. This effect plays a crucial role in contributing to the various phenomena we observed, including the magnetodielectric, multiferroic, and SPC properties.

O05

Moreover, the dielectric peak we observed appears to be closely associated with the structural distortions driven by the strong magnetostriction effect. It is particularly noteworthy that the magnetostriction effect, which is often regarded as negligible in other SPC systems, plays a vital role in modulating phonons in this specific system, leading to an indirect form of spin-phonon coupling. Nevertheless, this system exemplifies a highly entangled interaction among spin, lattice phonons, and dipoles, which activates a diverse range of intriguing phenomena. As a result, this places it among the rare and exceptional materials in the field.

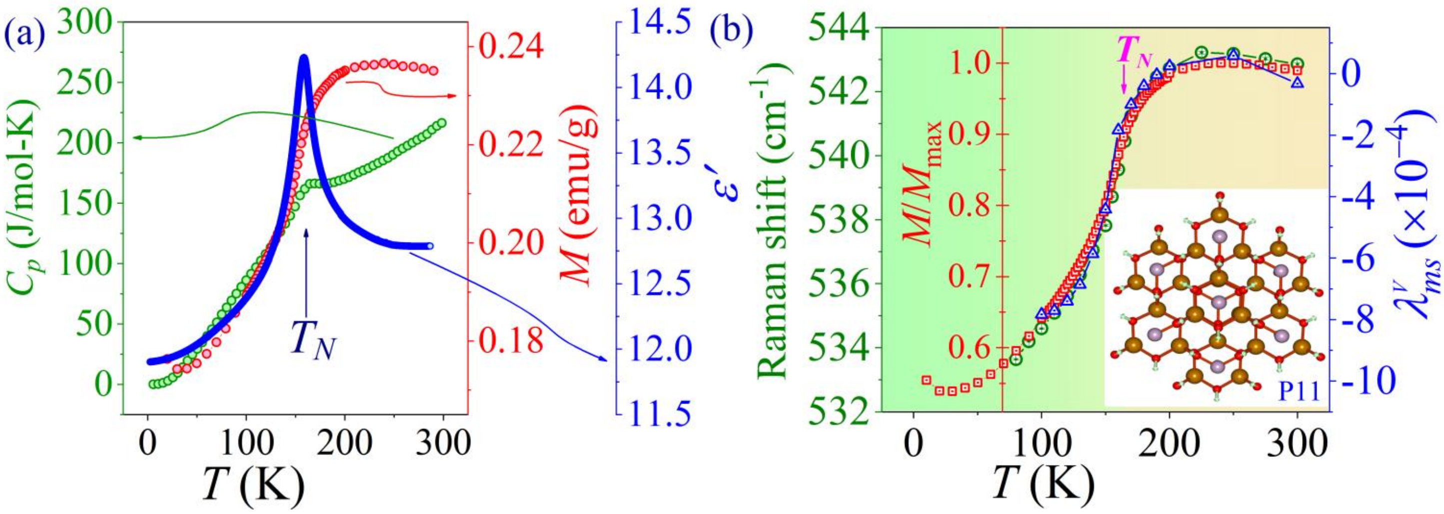

FIGURE 1. : (a): Specific heat (C_p), Magnetization (M), and Dielectric constant ($\mathcal{E}'$), variations as a function of temperature. (b): Raman shift, normalized magnetization, and volume magnetostriction coefficient variations as a function of temperature. All these curves unambiguously suggest the highly entangled behavior of spin, lattice, phonon, and dipolar degrees of freedom in this system.

REFERENCES

[1] Pal et. al., Phys. Rev. B **106**, 094404 (2022)