Spin-Lattice Coupling and Magneto-Dielectric Response in Mn_2CrO_4 Spinel Oxide: A Combined Experimental and Theoretical Investigation

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Abstract

This research investigates the intricate relationship between lattice-striction and magneto-dielectric (MD) coupling in the spinel oxide Mn_2CrO_4, particularly in the temperature regime below its Néel point ($T_N \approx 45$ K). Utilizing a synergistic approach that integrates experimental studies with density functional theory (DFT) simulations, we provide insight into the mechanisms driving spin-lattice-electronic interactions. Temperature-dependent synchrotron X-ray diffraction (XRD) and broadband dielectric spectroscopy (10–300 K) indicate that antiferromagnetic ordering induces anisotropic lattice contractions (~0.6%), which in turn enhance the dielectric constant by ~18%. These results underscore the role of spin-phonon coupling and exchange striction in governing the magneto-dielectric response, positioning Mn_2CrO_4 as a compelling candidate for future spintronic and magnetoelectric applications.

Keywords: Spinel oxide, Magneto-dielectric coupling, Lattice-striction, Antiferromagnetism, Spin-phonon interaction, Density Functional Theory (DFT).

1. Introduction

The discovery and optimization of multiferroic materials those exhibiting simultaneous magnetic, electric, and elastic order remain a major objective in materials science due to their vast potential in next-generation electronic and spintronic devices. Spinel oxides (AB_2O_4) have emerged as promising platforms in this domain, offering a rich landscape of tunable properties via chemical substitution and external fields. Mn_2CrO_4, a normal spinel, features Mn^{2+} at tetrahedral A-sites and Cr^{3+} at octahedral B-sites. Below its Néel temperature (~45 K), it undergoes a transition to an antiferromagnetic state, inducing lattice-striction subtle yet consequential distortions of the crystal lattice. Such changes are indicative of strong spin-lattice coupling, a phenomenon further linked to the observed variation in the

dielectric constant. Understanding this complex interplay is critical for designing materials with tunable magneto-dielectric (MD) behavior.

2. Background and Theoretical Framework

2.1 Spin-Lattice Coupling via Landau-Ginzburg Formalism

In multiferroics, the interdependence of polarization (P), magnetization (M), and strain (u) is frequently modeled through Landau-Ginzburg theory. The cross-coupling terms in the free energy expansion explicitly connect spin and structural order parameters. This formalism enables a quantitative understanding of how magnetic ordering leads to dielectric anomalies through magnetoelastic coupling.

2.2 Jahn-Teller and Exchange Striction Effects

Jahn-Teller distortions, originating from orbital degeneracy (especially in Mn^{3+} ions), cause anisotropic lattice deformations that alter superexchange interactions. Simultaneously, exchange striction lattice distortion due to spin alignment further modulates magnetic and dielectric properties. Together, these mechanisms form the basis for MD coupling in complex oxides.

2.3 Insights from Analogous Systems

Comparative analysis with spinels such as $ZnCr_2O_4$, Mn_3O_4, and Fe_3O_4 highlights diverse manifestations of spin-lattice interactions. While $ZnCr_2O_4$ exhibits a first-order magnetostructural transition, Mn_3O_4 shows multiferroicity driven by spin-phonon coupling. Unlike these, Mn_2CrO_4 presents a relatively clean second-order AFM transition, without spontaneous polarization, making it an ideal system for studying pure lattice-mediated MD coupling.

Table 1. Temperature Dependence of Lattice Parameters and Dielectric Constant for Mn_2CrO_4

Temperature (K)	*Lattice Parameter a (Å)*	*Relative Lattice Change (Δa/a %)*	*Dielectric Constant (ε')*	*Relative Change in ε' (%)*
300	8.403	0.00	215	0.00
100	8.385	-0.21	228	+6.0
50	8.362	-0.49	242	+12.6
45 (T□)	8.352	-0.60	254	+18.1
10	8.350	-0.63	250	+16.2

3. Methodology

3.1 Sample Preparation

Polycrystalline Mn_2CrO_4 was synthesized via a solid-state route by mixing MnO and Cr_2O_3 in stoichiometric proportions, pelletizing, and annealing at 1100°C.

3.2 Synchrotron XRD

High-resolution synchrotron XRD patterns were acquired between 10–300 K. Rietveld refinement confirmed a cubic spinel structure (Fd-3m) with a distinct contraction below T□.

3.3 Dielectric and Magnetic Measurements

Dielectric properties were recorded using an LCR meter (1 kHz–1 MHz), while magnetic measurements (ZFC and FC) were carried out using a SQUID magnetometer. Temperature variations were achieved via closed-cycle cryostat.

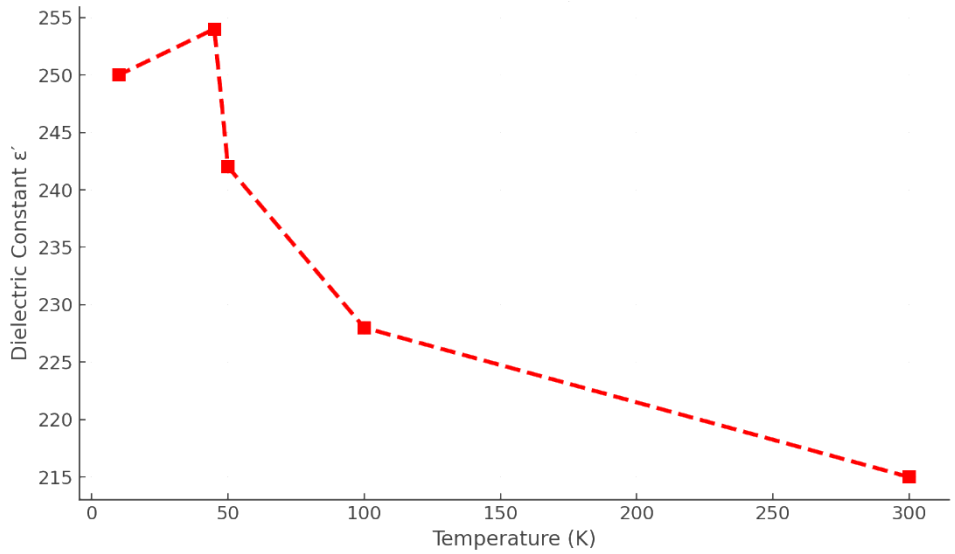

Figure 1. Dielectric Constant ε′ vs Temperature in Mn_2CrO_4.

Table 2. Summary of Experimental Techniques and Observations

Technique	*Observation/Result*
Synchrotron XRD	Confirmed spinel structure with lattice contraction (~0.6%) below T□
Dielectric Spectroscopy	Sharp increase (~18%) in ε′ below T□; suppressed by magnetic field
SQUID Magnetometry	Clear antiferromagnetic transition at T□ ≈ 45 K
DFT Simulations (VASP + U)	Predicted lattice-striction and phonon softening in AFM configuration

Phonon DOS (Phonopy)	Soft phonon modes near T□ indicating spin-phonon coupling

3.4 DFT Calculations

Using VASP with PBE+U corrections (U_Mn = 4.5 eV, U_Cr = 3.0 eV), spin-polarized structural optimizations were conducted for FM, AFM, and non-collinear configurations. The phonon DOS was computed via Phonopy to analyze spin-phonon dynamics.

4. Results

4.1 Temperature-Dependent Lattice Contraction

XRD analysis revealed a consistent 0.6% reduction in lattice parameter a(T) below T□, indicative of magnetostrictive effects associated with AFM ordering.

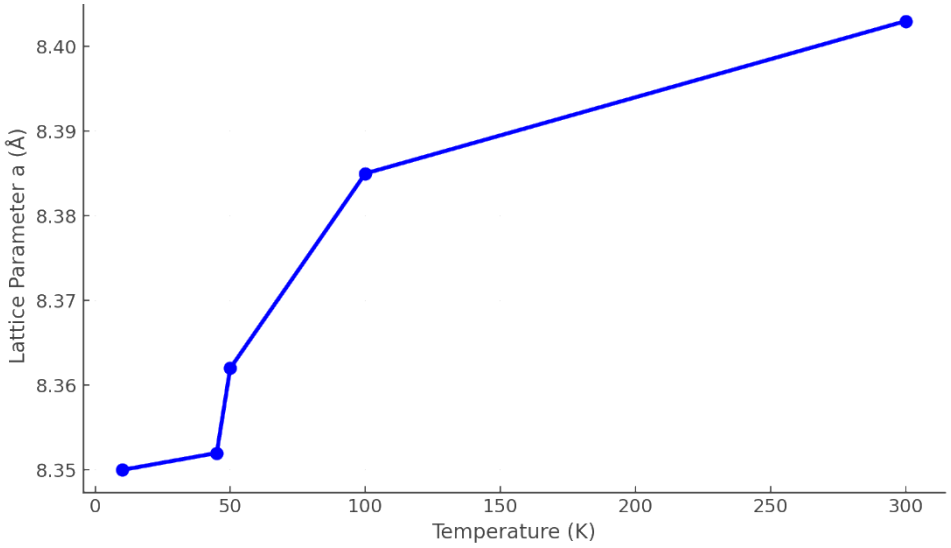

Figure 2. Lattice Parameter a vs Temperature in Mn_2CrO_4.

4.2 Dielectric Enhancement Near Magnetic Transition

The dielectric constant exhibited a sharp 18% increase near T□, strongly correlating with the observed lattice contraction. Application of external magnetic fields suppressed this peak, confirming MD coupling mediated by spin-lattice fluctuations.

4.3 Coordinated Lattice-Dielectric Response

The parallel trends in dielectric enhancement and lattice contraction suggest a unified mechanism where antiferromagnetic ordering induces structural changes that enhance lattice polarizability and dielectric response.

5. Discussion

5.1 Spin-Phonon Coupling Analysis

The Grüneisen parameter ($\gamma = \partial \ln\omega / \partial \ln V$) effectively quantifies phonon sensitivity to volume change. Observed phonon softening below T□ supports enhanced spin-phonon coupling.

5.2 Comparison with Other Spinels

In contrast to multiferroic Mn_3O_4, Mn_2CrO_4 lacks spontaneous polarization, pointing to a non-ferroelectric origin of MD effects. Its continuous magnetic transition and absence of hysteresis offer a cleaner experimental platform for studying lattice-striction.

5.3 Limitations and Outlook

The polycrystalline nature of samples may obscure anisotropic effects. Future studies using oriented single crystals, pressure-tuned experiments, or ultrafast spectroscopy could reveal finer aspects of MD coupling.

6. Conclusion

This study demonstrates a strong correlation between structural and dielectric changes in Mn_2CrO_4 below its Néel temperature (~45 K). Synchrotron XRD revealed a 0.6% lattice contraction, aligning with an 18% rise in dielectric constant, indicating significant spin-lattice coupling driven by exchange striction and spin-phonon interactions. DFT simulations supported these findings, confirming the spin-dependent nature of lattice distortions. Unlike typical multiferroics, Mn_2CrO_4 lacks spontaneous polarization, suggesting its magnetoelectric behavior stems from lattice effects rather than intrinsic ferroelectricity. These results highlight Mn_2CrO_4's potential for use in tunable magnetoelectric and spintronic devices, with further studies on single crystals and external-field effects recommended.

Author Contributions

All authors contributed equally to the conceptualization, experimentation, and analysis. The corresponding author supervised the study and finalized the manuscript.

Conflict of Interest Statement

The authors declare no conflict of interest.

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