Spin Dynamics and Symmetry Transitions in Cr_2O_3 Probed by NMR

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Abstract:

Chromium(III) oxide (Cr_2O_3) is the prototypical magnetoelectric antiferromagnet, known for its linear magnetoelectric (ME) effect and symmetry-governed spin phenomena. We present a comprehensive ^{53}Cr NMR study of Cr_2O_3, combining field- and temperature-dependent measurements to investigate spin-lattice relaxation, spontaneous sublattice magnetisation, and symmetry evolution across the spin-flop transition.

NMR spectra reveal a first-order spin-flop transition at B_{SF} ≈6.7 T, marked by a collapse of quadrupolar triplets into magnetic doublets. Angular-dependent NMR measurements in the flop phase show sinusoidal frequency shifts and cosine-like FWHM variation, consistent with coherent in-plane spin canting and domain-selective magnetic anisotropy. These features support a symmetry reduction from magnetic point group $\bar{3}'m'$ to $2'/m$.

Spin-lattice relaxation rate ($1/T_1$) measurements across a wide temperature range reveal three distinct regimes: (1) activated BPP behavior below 15 K ($\tau_0 \approx 9$ ns, $E_a \approx 0.56$ meV), (2) a power-law regime ($n \approx 2.3$) dominated by two-magnon Raman scattering, and (3) critical slowing down near $T_N \approx 308$ K with $n \approx 4$. A pronounced relaxation peak near 8 K, invariant in position but suppressed in magnitude under field, suggests low-energy dynamic contributions from domain walls or transverse modes.

Zero-field NMR shifts directly track the internal hyperfine field and show continuous suppression of spontaneous magnetisation approaching T_N. Quadrupolar splitting and line width remain stable, implying minimal structural contributions to the magnetic evolution.

These results establish ^{53}Cr NMR as a sensitive local probe of spin symmetry, magnetic domains, and low-energy excitations in Cr_2O_3, bridging classic magnetoelectric theory with modern spintronic and multipolar perspectives.

Selected References

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