Hessisches Kompetenzzentrum
für Hochleistungsrechnen

GPU-accelerated High-Throughput Screening and Multimodal Machine Learning Modeling of Spin Dynamics Derived Spectroscopy Properties of Fe/Mn

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Clusters
Lichtenberg II Cluster Darmstadt

Software
VASP, Quantum ESPRESSO

Additional Software
Castep, QE-gipaw

Institute
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Introduction

Nuclear magnetic resonance and electron paramagnetic resonance spectroscopies are essential experimental techniques for characterizing magnetic materials containing transition metal elements such as iron and manganese. However, interpreting these spectra requires accurate theoretical predictions of NMR and EPR parameters, which are computationally demanding due to the need for precise electronic structure calculations. For transition metal systems, strong electron correlations necessitate advanced methods beyond standard density functional theory, particularly DFT+U corrections. High-performance computing resources are essential for performing systematic high-throughput screening across multiple chemical compositions, structural configurations, and magnetic states. Our project aimed to establish an automated computational framework for predicting spectroscopic properties of Fe/Mn-containing magnetic materials, combining first-principles calculations with machine learning models to accelerate materials discovery. The GPU acceleration was planned for both intensive DFT calculations and training of multimodal machine learning models on large datasets.

Methods

Our initial methodology focused on establishing a robust computational workflow using CASTEP for NMR parameter calculations within the GIPAW (Gauge-Including Projector Augmented Wave) formalism. We successfully implemented high-throughput calculations for non-magnetic oxide systems, validating our approach against established NMR databases. For transition metal systems requiring DFT+U corrections, we discovered fundamental software limitations: CASTEP explicitly does not support DFT+U in combination with NMR calculations. We subsequently investigated Quantum ESPRESSO with GIPAW module as an alternative platform, performing extensive benchmark calculations on simpler systems. We established direct communication with the QE-GIPAW developer to resolve technical compatibility issues between the NMR module and recent Quantum ESPRESSO versions. In parallel, we extended our validated workflow to complex oxide systems, specifically BaTiO3 perovskites. We employed ab initio molecular dynamics simulations at multiple temperatures to generate diverse hypothetical structures, followed by systematic structural relaxation and NMR parameter calculations. Machine learning models were developed using Smooth Overlap of Atomic Positions descriptors to establish structure-property relationships for oxygen NMR parameters across thousands of configurations.

Results

We successfully completed and published our work on non-magnetic oxide systems, creating a comprehensive dataset of over 7,150 compounds with 47,505 oxygen sites, demonstrating the effectiveness of our high-throughput framework. The publication "High Throughput Calculations and Machine Learning Modeling of O-17 NMR in Non-Magnetic Oxides" by Li et al. establishes validated protocols for automated NMR calculations using AiiDA-CASTEP workflow management. We extended this methodology to BaTiO3 perovskite systems, generating perturbed structures based on the prototype material through systematic AIMD sampling at various temperatures. From these simulations, we selected 3,000 representative structures containing over 18,000 oxygen sites for comprehensive NMR calculations. All structures were fully relaxed and complete NMR parameter calculations were performed. Machine learning models were trained on this extensive dataset to predict spectroscopic parameters from structural features, achieving high predictive accuracy for oxygen chemical shifts and electric field gradients. These results were presented at the DPG Spring Meeting 2025 in Regensburg, demonstrating successful application of our methodology to complex perovskite systems. However, the originally proposed Fe/Mn magnetic systems could not be computed due to the discovered incompatibility between DFT+U corrections and NMR calculations in available software packages. The QE-GIPAW alternative showed persistent technical issues that remain unresolved by the development team.

Discussion

This project encountered significant technical challenges that prevented completion of the originally proposed Fe/Mn magnetic

system calculations. The fundamental incompatibility between DFT+U and NMR calculations in CASTEP represents a serious limitation for studying correlated electron systems with magnetic resonance spectroscopy. While Quantum ESPRESSO theoretically offers an alternative, the practical implementation issues we encountered suggest this remains an open technical problem in the computational materials science community. Despite these obstacles, we successfully demonstrated the viability of high-throughput NMR calculations and machine learning modeling for complex oxide systems. The published work on non-magnetic oxides and the extended BaTiO3 study provide valuable contributions to the field. Given the technical impossibility of performing the proposed transition metal calculations with available software, we conserved GPU resources rather than generating scientifically invalid data. Future work will require either fundamental software development to enable DFT+U with NMR calculations, or alternative theoretical approaches such as hybrid functionals or DMFT (Dynamical Mean Field Theory) methods, though these would require substantially different computational strategies. The methodologies established in this project remain applicable to a wide range of materials systems where standard DFT is sufficient.

Publications

Li, Z., Zhao, B., Zhang, H., & Zhang, Y. (2025). High-throughput calculations and machine learning modeling of 17 O NMR in non-magnetic oxides. Faraday Discussions, 255, 72-87.

Reference

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