Extended Abstract

Emergent Quasiparticle Dynamics in Correlated Quantum Materials Investigated through Neutron Scattering and Theoretical Framework

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Abstract

Strongly correlated quantum materials exhibit rich magnetic and lattice phenomena arising from the interplay of spin, charge, orbital, and lattice degrees of freedom, which govern the emergence and diverse quasiparticle excitations. In this thesis, neutron scattering (elastic and inelastic) is employed as a microscopic probe to investigate spin-structure, spin excitations and dynamics, crystal-field effects (CEF), spin-orbit coupling (SOC) and lattice-mediated phenomena (phonon and polaron) in transition-metal oxides. Four benchmark compounds are studied, revealing diverse manifestations of correlated quantum behavior. In the single-ion magnet $Sr_2Ca_2Mn_2CoO_9$, neutron diffraction and inelastic neutron scattering reveal the coexistence of single-ion quantum bi-stability and collective magnetic excitations, with magnon excitations persisting to room temperature. This study provides a demonstration of single-ion magnetism (SIM) in a long-range order system and mechanism of relaxation phenomena of SIM. Such a SIM behavior in a long-range order system is quite unusual. Our results yield a route in designing oxide SIM towards application of quantum qubit. The change of concentration of Sr/Ca ion ratio drastically modifies the magnetic behavior. The compound $Sr_4Mn_2CoO_9$ provides a microscopic view of spin dynamics in a low-dimensional single-chain magnet, where crystal-field anisotropy, exchange interactions, and spin–phonon coupling collectively govern the observed excitations. The third system, I studied, is 4d–4f double perovskite, Ba_2DyRuO_6, where neutron diffraction and inelastic neutron scattering uncovers an Ising-like antiferromagnetic ground state and well-defined magnon and crystal-field excitations arising from strong Ru–Dy exchange and magnetic anisotropy. Finally, in a rutile niobate system has been studied, where we document fascinating dispersive room-temperature phonons, which drive Ni^{1+}–Ni^{2+} polaron hopping through coupled spin–charge–phonon interactions. Such an experimental demonstration of dispersive phonon driven polaron is rarely reported. All these experimental results are further complemented with detailed theoretical calculations by solving quantum-mechanical Hamiltonian, spinW modelling of magnon dispersion, modelling of CEF and SOC, and machine-learning-based lattice-dynamics calculations, for all four systems. Together, the results provide a comprehensive microscopic understanding of spin, charge, orbital, phonon and polaron excitations in strongly correlated quantum materials and explain the mechanism of fascinating physical phenomena.

Chapter 1: Introduction

This chapter introduces the fundamental concepts required to understand the magnetic and dynamical properties of the quantum materials investigated in this thesis. The discussion begins with the basic physics of magnetic ions in a crystal environment, including crystal-field splitting, exchange interactions, magnetic anisotropy, spin–orbit coupling, and the origin of magnetic excitations. The principles of single-ion and single-chain magnetism are then introduced, with emphasis on the microscopic mechanisms governing magnetic stability and relaxation. Further, it discusses the physics of 4d–4f magnetic systems, focusing on the interaction between transition-metal and rare-earth magnetic moments, followed by an introduction to polarons and the coupling between charge and lattice degrees of freedom. The fundamental nature of quasiparticle excitations relevant to these systems, including magnons, crystal-field excitations, phonons, and polaronic excitations, is briefly described. Finally, the basic principles of neutron diffraction and inelastic neutron scattering, together with the quantum-mechanical and theoretical approaches used to describe the observed excitations, are

introduced as the foundation for the investigations presented in the subsequent chapters, which deal with unique quasiparticle behavior.

Chapter 2: Experimental Techniques and Methodology

This chapter describes the experimental methods and analysis techniques which are employed throughout the thesis. The details of sample preparation and their structural characterisation through powder X-ray diffraction using the Rietveld refinement method have been discussed. The chapter provides a detailed explanation of neutron scattering techniques, highlighting their importance in probing both crystal and magnetic structures as well as spin dynamics. Powder neutron diffraction is discussed as a key tool for determining magnetic ordering, while inelastic neutron scattering (INS) is introduced as a powerful technique to study magnetic excitations and spin-wave spectra. The principles of data analysis, including Rietveld refinement, representation analysis for magnetic structures, and spin-wave modelling using SpinW software and Point charge CEF calculation for rare-earth, McPhase CEF framework, are explained. The machine-learning framework used to produce the phonon spectra has been discussed. The formulation and solution of relevant quantum-mechanical Hamiltonians are presented to describe the microscopic energy levels, magnetic states, and quasiparticle excitations of the investigated systems. This chapter therefore provides the experimental and theoretical methodology underlying the results presented in the subsequent chapters and establishes the basis for their systematic and reproducible analysis.

Chapter 3: Quantum Bi-stability and Robust Room-Temperature Spin Excitation in a Single-Ion Magnet

The search for single-ion magnetism (SIM) has been largely focused on hybrid metal–organic systems. Here, the spin-relaxation mechanism of SIM is demonstrated in a pure inorganic transition-metal oxide, $Sr_2Ca_2Mn_2CoO_9$, an Ising-chain magnet, employing neutron diffraction and inelastic neutron scattering (INS), complemented by SpinW simulation and a machine-learning framework. Interestingly, the SIM mechanism persists even in the presence of long-range magnetic ordering, a phenomenon rarely observed. This coexistence indicates a novel magnetic domain in which local single-ion quantum bi-stability and collective spin excitations coexist on the same magnetic sublattice, potentially leading to a hybrid quantum-magnonic system in chemically adjustable oxides. Such coexistence establishes a previously unexplored magnetic regime, bridging single-ion quantum bi-stability and collective magnetic order beyond the limits of conventional magnets. This pioneering investigation shows that bi-stability between the two quantum states $|Ms| = \pm 3/2$ is maintained at zero magnetic field, driven by a dominant Orbach spin-relaxation mechanism with an effective energy barrier $U_{eff} \approx 4.4$ meV. This behavior arises from strong spin–phonon coupling in the presence of negative axial anisotropy ($D \approx -2.2$ meV) and the high-spin ($S = 3/2$) Co (II) ion. Our results provide a path in designing oxide SIM towards quantum qubit application. Furthermore, it is observed that magnon excitations persist up to room temperature, reflecting low-dimensional magnetic interactions and extended magnetic correlations within the oxide lattice. The interplay between single-ion magnetism and room-temperature spin excitations underscores the tunability of magnetic anisotropy in oxide lattices. This work presents a new magnetic domain where anisotropy-protected single-ion memory coexists with room-temperature magnons, allowing materials to combine information storage with collective spin transport.

Chapter 4: Microscopic investigation of spin dynamics in the single-chain magnet $Sr_4Mn_2CoO_9$

One-dimensional single-chain magnets offer a unique platform for studying the interplay of crystal-field effects, exchange interactions, and lattice dynamics. Here, we investigate spin excitations in Sr4Mn2CoO9 using inelastic neutron scattering (INS) and theoretical modelling. INS reveals two low-energy magnetic excitations at 4 and 7 meV from Mn–Co–Mn spin chains, alongside higher-energy crystal-electric-field (CEF) excitations from two crystallographically inequivalent Co2+ sites. Interestingly, these spin excitations persist at room temperature, demonstrating dynamic magnetic correlations in the absence of long-range order. Furthermore, the crystal-field modelling, based on Stevens operator formalism, reproduces well the CEF spectra, establishing Ising-like Kramers ground-state doublets with strong uniaxial magnetic anisotropy for both Co2+ ions. In addition, the spin wave simulation using SpinW reproduces the spin excitation spectrum and reveals microscopic exchange interactions in two non-interacting Mn–Co–Mn spin chains. Finally, machine-learning lattice-dynamics calculations confirm the phonon spectrum and spin–phonon coupling. By projecting the exchange Hamiltonian onto CEF ground-state doublets, we estimate exchange-induced splitting, matching the observed excitations. Thus, our results elucidate low-energy spin dynamics arising from combined crystal-field anisotropy and exchange interactions, with the persistent low-energy excitation providing a microscopic pathway for thermally activated spin relaxation. Furthermore, this work delivers a unified microscopic understanding of the interplay between crystal-field effects, magnetic exchange, and lattice dynamics in Sr4Mn2CoO9, advancing insights into spin dynamics in low-dimensional transition-metal oxides.

Chapter 5: Quasiparticle dynamics in the 4d-4f Ising-like double perovskite Ba_2DyRuO_6 studied using neutron scattering and machine-learning framework

Double perovskites containing 4d–4f interactions provide a platform to study complex magnetic phenomena in correlated systems. Here, we investigate the magnetic ground state and quasiparticle excitations of the fascinating double perovskite system, Ba_2DyRuO_6, through time-of-flight (TOF) neutron diffraction, inelastic neutron scattering (INS), and theoretical modelling. The compound Ba_2DyRuO_6 is reported to exhibit a single magnetic transition, in sharp contrast to most of the other rare-earth (R) members in this family, A_2RRuO_6 (A = Ca/Sr/Ba), which typically show magnetic ordering of the Ru ions, followed by R-ion ordering. Our neutron diffraction results confirm that long-range antiferromagnetic order emerges at TN ≈ 47 K, primarily driven by 4d–4f Ru^{5+}–Dy^{3+} exchange interactions, where both Dy and Ru moments contribute to the ordered state. The ordered ground state is a collinear antiferromagnet with Ising character, carrying ordered moments of μ_{Ru} = 1.6(1) μ_B and μ_{Dy} = 5.1(1) μ_B at 1.5 K. Low-temperature INS reveals well-defined magnon excitations below 10 meV. SpinW modelling of the INS spectra evidences complex exchange interactions and the presence of magnetic anisotropy, which governs the Ising ground state and accounts for the observed magnon spectrum. Combined INS and Raman spectroscopy reveal crystal-electric-field (CEF) excitations of Dy^{3+} at 46.5 and 71.8 meV in the paramagnetic region. The observed CEF levels are modelled through point-charge CEF calculations, consistent with the Oh site symmetry of Dy^{3+}. A complementary machine-learning approach is used to calculate and analyse the phonon spectrum, enabling a direct comparison with the INS data. These combined experimental and theoretical results provide a

comprehensive understanding of the origin of phonon and magnon quasiparticle excitations and their influence on the crystal structure and ground-state magnetism of Ba_2DyRuO_6.

Chapter 6: Dispersive-phonon-driven room-temperature Ni^{1+}-Ni^{2+} polaron hopping in spin-charge coupled rutile niobate

Understanding how lattice dynamics mediate polaron hopping is essential for designing multifunctional correlated oxides. Here, we demonstrate room-temperature dispersive phonon excitations and elucidate the Ni^{1+}–Ni^{2+} polaron-hopping mechanism and the presence of rare spin-charge-phonon coupling even in a magnetically short-range-ordered state in rutile niobate, a rare room-temperature magnetodielectric system. We reveal room-temperature dispersive phonon excitations using inelastic neutron scattering (INS), complemented by machine-learning-based phonon calculations, to establish the microscopic origin of the polaron-hopping mechanism. Experimental evidence of dispersive phonon-driven polaron hopping is scarce. INS measurements show significant dispersive phonon excitations at 21, 33, and 47 meV, implying collective lattice dynamics that enable delocalized polaron propagation via coupled charge-spin-phonon interactions. Dispersive phonons couple to charge carriers and promote correlated NiO6 lattice distortions, facilitating delocalized polaron hopping. Low-energy magnetic excitations at 4 and 8 meV indicate the presence of local short-range magnetic correlations or spin-orbit coupling-induced anisotropy in deformed NiO_6 octahedra, which are discussed in detail. Machine-learning phonon calculations replicate experimentally observed phonon excitations and demonstrate lattice instability, consistent with dynamic local distortions induced by polaron production. These findings provide microscopic evidence for a coupled charge-spin-phonon mechanism that mediates polaron hopping in rutile oxide systems.

Outcome of the thesis:

1. Demonstration of single-ion quantum bi-stability coexisting with long-range magnetic order and robust room-temperature magnon excitations in an inorganic oxide that is a rare phenomenon and experimentally explored for the first time (see Fig. 1). Our results predict a path in designing SIM in pure oxide environment towards quantum qubit for potential application in long-run.

2. Microscopic elucidation of spin dynamics in the single-chain magnet, revealing the combined roles of crystal-field anisotropy, exchange interactions, and spin–phonon coupling in a low-dimensional system.

3. Establishment of the microscopic origin of quasiparticle excitations in the 4d–4f Ising-like double perovskite 4d-4f strongly correlated system, including magnon, crystal-field, and phonon excitations governed by Ru–Dy exchange and magnetic anisotropy.

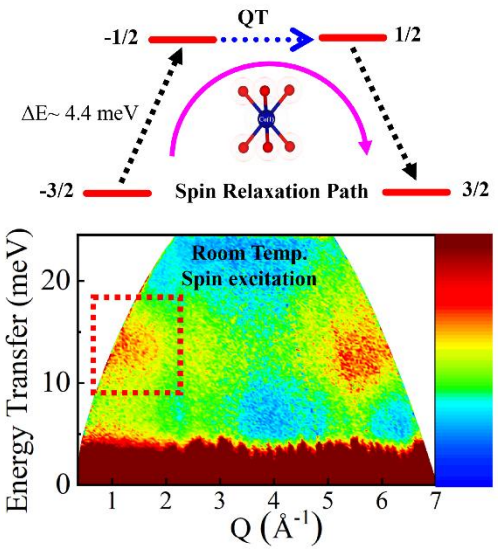

Figure 1: ***Anisotropy-protected quantum bi-stability and robust room-temperature spin excitations emerge in the single-ion magnet***

4. Identification of dispersive phonons as a driving mechanism for room-temperature Ni1+–Ni2+ polaron hopping through coupled spin–charge–phonon dynamics in a rutile niobate.

Overall, the work establishes neutron scattering combined with quantum-mechanical Hamiltonian calculations, theoretical modelling, and machine-learning lattice dynamics as an effective framework for uncovering the microscopic origin and interplay of magnetic, electronic, and lattice quasiparticles in strongly correlated quantum materials.

Publications (related to thesis):

1. **Roy, G.***, Kushwaha, E., Kumar, M., Ghosh, S., Orlandi, F., Le, M.D., Stone, M.B., Sannigrahi, J., Adroja, D.T. and Basu, T.*, 2026. Quasiparticle dynamics in the 4d-4f Ising-like double perovskite Ba2DyRuO6 studied using neutron scattering and machine-learning framework. ***Physical Review B***, *113*(17), p.174416.

2. **Roy, G.,** Kumar, M., Sannigrahi, J., Caignaert, V., Adroja, D.T., Kushwaha, E., Ghosh, S., Prellier, W., Hardy, V., Basu, T.*, et al., 2026. Quantum bi-stability and robust room-temperature spin excitation in a single-ion magnet. *Research Square Preprint* (2026). DOI: 10.21203/rs.3.rs-8591958/v1 (Accepted in ***Advanced Functional Materials, 2026***)

3. **Roy, G.*** and Basu, T.*, 2026. From molecules to qubits: evolution of single-molecule magnets. ***Journal of Physics: Condensed Matter***.

4. **Roy, G**.*, *Kumar, M., Ghosh, S., Kushwaha, E., Le, M.D., Stone, M.B., Adroja, D.T. and Basu, T.**, 2026. Dispersive-phonon-driven room-temperature Ni^{1+}–Ni^{2+} polaron hopping. (**Under review**).

5. **Roy, G.*,** *Kumar, M., Ghosh, S., Kushwaha, E., Le, M.D., Stone, M.B., Adroja, D.T. and Basu, T.*, 2026. Microscopic understanding of spin excitations in the single-chain magnet $Sr_4Mn_2CoO_9$. (**Under review**)

Publications (not included in my thesis):

1. Kushwaha, E., **Roy, G**., Kumar, M., dos Santos, A.M., Ghosh, S., Adroja, D.T., Caignaert, V., Perez, O., Pautrat, A. and Basu, T.*, 2024. Origin of spin-driven ferroelectricity and effect of external pressure on the complex magnetism of the 6H perovskite $Ba_3HoRu_2O_9$. ***Physical Review B***, 109, p.224418. (**Equal first author contribution**).

2. Kumar, M., **Roy, G**., Ghosh, S., Kushwaha, E., Singh, K. and Basu, T.*, 2026. Spin-chain incipient magnetocaloric effect and rare-earth (R) controlled switching in the Haldane-chain system, R_2BaNiO_5. ***Advanced Quantum Technologies***, 9(6), p.e70356.

3. Ghosh, S., **Roy, G**., Kushwaha, E., Kumar, M. and Basu, T.*, 2026. Spin-correlation driven ferroelectric quantum criticality in a perovskite quantum spin-liquid system, $Ba_3CuSb_2O_9$. ***Journal of Materials Chemistry C***, 14(20), pp.8482–8487.

4. Kushwaha, E., **Roy, G**., dos Santos, A.M., Kumar, M., Ghosh, S., Heitmann, T. and Basu, T.*, 2025. Unconventional S-orbital state of Tb and cooperative Ru (4d)–Tb (4f) spin-ordering in the strongly correlated 4d–4f system $Ba_3TbRu_2O_9$. ***Journal of Materials Chemistry C***, 13(30), pp.15384–15389.

5. Kumar, M., Ghosh, S., **Roy, G**., Kushwaha, E., Caignaert, V., Prellier, W., Majumdar, S., Hardy, V. and Basu, T.*, 2026. Intriguing magnetocaloric effect in multiferroic $Ba_3RRu_2O_9$ (R = Ho, Gd, Tb, Nd) with strong 4d–4f correlations. ***ACS Applied Energy Materials***, 9(8), pp.5141–5147.

6. Kushwaha, E., Ghosh, S., Sannigrahi, J., **Roy, G**., Kumar, M., Cottrell, S., Stone, M.B., Fang, Y., Adroja, D.T., Ke, X. and Basu, T.*, 2025. Interplay between trimer structure and magnetic ground state in $Ba_5Ru_3O_{12}$ probed by neutron and μSR techniques. ***Physical Review B***, 112(9), p.094410.

7. Panda, S., **Roy, G**., Basu, T. and Panda, D., 2024. Facet {100} fosters resonance energy transfer in Ni/Co-doped $CsPbBr_3$ nanocrystals. ***ACS Applied Energy Materials***, 7(21), pp.10179–10188.

8. **Roy, G.,** Basu, T., et al., 2026. Crystal-to-Spin: Machine-Learning Software for Orbital Splitting and Virtual Electron Hopping. (**Under preparation**).

9. **Roy, G**.*, Basu, T., et al., 2026. Quantum-Mechanical Formulation of Virtual Electron Hopping. ***arXiv preprint*** (2026)

Achievements:

1. Awarded the highly prestigious Indo-French **Raman–Charpak Fellowship** (2024) by CEFIPRA for the Indo–French collaborative research proposal entitled “Design and Investigation of Single-Ion Magnetism (SIM) in Inorganic Metal Oxides”.

2. Won the **Best Poster Award** at the 67th DAE-SSPS Conference organized by **BARC, DAE** on December 20-24, 2023.

3. Beamtime accepted in the **WISH** diffractometer as co-PI at ISIS, STFC, UK for Time-of-Flight Neutron Diffraction.

I received **travel funding** awarded by the **DST-RAL India-UK program** to perform experiments in 2023.

4. Beamtime accepted in **MARI and MERLIN, ISIS**, **UK** STFC, several times as co-PI.