Selected Research Achievements

Five Research Achievements Enabled by Diverse Expertise

In the Quantum Solid State Materials Group, synthesis, measurement, and theory are not treated as separate, fixed domains. Instead, we begin with a newly created material and combine the research approaches best suited to the questions it raises.

Materials obtained through high-pressure synthesis are examined using neutron and synchrotron radiation techniques, and information derived from structural analysis is incorporated into theoretical models. Key interactions and control parameters identified through calculations are then tested by elemental substitution, crystal-structure control, mixed-anion chemistry, and related approaches.

Our expertise in high-temperature and high-pressure synthesis, perovskite crystal chemistry, mixed-anion chemistry, single-crystal growth, neutron scattering, quantum magnetism, statistical mechanics, and atomistic simulations complements one another and converges on the study of individual materials.

Our work does not end with the synthesis of a material. We determine its structure and physical properties, and then feed the resulting understanding back into the design of the next material. This cycle drives the research of the Quantum Solid State Materials Group forward.

The following five recent achievements illustrate both our research approach and the breadth of expertise within the group.

1. Discovery of Giant Irreversible Expansion through the Release of Structural Frustration

Kazunari YAMAURA

In high-pressure-synthesized barium ruthenate Ba4Ru3O12, we discovered a giant irreversible expansion: the volume increases by approximately 4.4% upon heating and does not return to its original value after cooling. This volume change occurs without altering the crystal symmetry or oxygen content. It is exceptionally large compared with representative materials and constitutes an unusual lattice response for an oxide.

Synchrotron powder X-ray diffraction revealed that, between 450 and 650 K, Ru atoms within the RuO6 trimers undergo a cooperative rearrangement that directly corresponds to the increase in lattice volume.

By combining thermal analysis, electrical-resistivity and magnetization measurements, and first-principles calculations, we further confirmed that the expansion is not caused by decomposition, a redox reaction, an electronic phase transition, or a magnetic phase transition. We proposed a new

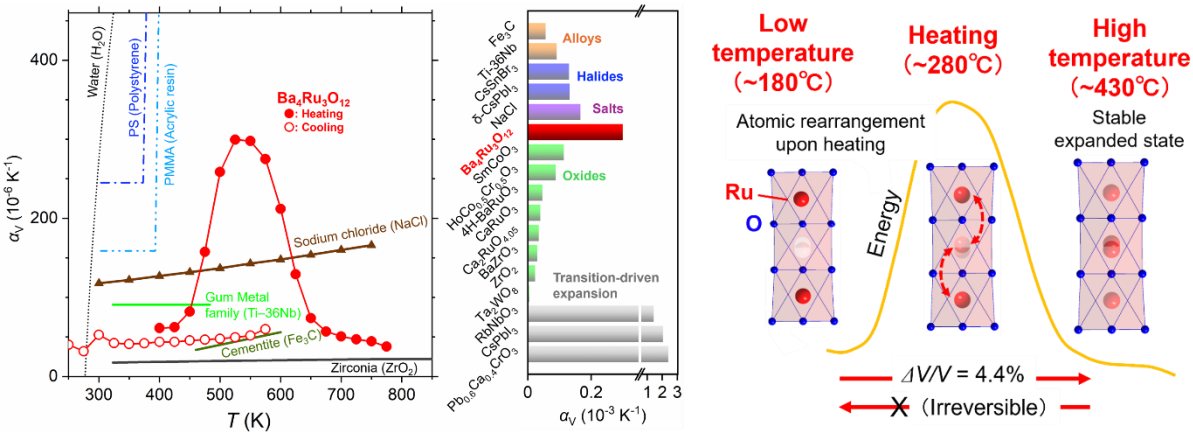
thermal-response mechanism in which heating allows the metastable arrangement of Ru atoms trapped by high-pressure synthesis to transform into a more stable configuration, releasing structural frustration and producing the giant lattice expansion.

Strength Demonstrated by This Achievement

This work creates metastable atomic arrangements through high-pressure synthesis and combines synchrotron structural analysis, physical-property measurements, and first-principles calculations to reveal a new lattice-response mechanism originating from the release of structural frustration.

Representative Publication

Z. Li, H. Yuan, A. A. Belik, T. Tadano, Y. Tsujimoto and K. Yamaura,
“Giant Lattice Expansion through Structural Frustration Release in a Dense Oxide,”
Journal of the American Chemical Society (2026).
DOI: 10.1021/jacs.6c07579



2. Elucidation of a Four-Spin Quantum Singlet and Spin Gap

Masashi HASE

In recent years, f-electron systems, particularly materials containing Yb3+, have attracted considerable attention because spin-orbit coupling and crystal-field effects can produce effective spin-1/2 quantum spin systems.

In this study, we investigated the rare-earth oxide Yb2SiO5 by combining magnetic-susceptibility, heat-capacity, and inelastic neutron-scattering measurements. The results revealed the formation of a spin-singlet ground state at low temperature and the presence of a spin gap, a finite energy difference between the ground state and magnetic excited states.

Inelastic neutron scattering detected magnetic excitations at 0.7, 1.2, and 1.9 meV. These results can be described by a tetramer model in which four effective spins are antiferromagnetically coupled:

J2(S1･S2 + S3･S4) + J1S2･S3

With J1 = 0.74 meV and J2 = 0.95 meV, the model consistently reproduces the magnetic-excitation energies, magnetic susceptibility, and magnetic heat capacity.

By assigning J1 and J2 to two distinct Yb-Yb exchange interactions, we also accounted for the Q dependence of the neutron-scattering intensity. The combination of bulk-property measurements and neutron scattering thus established that Yb2SiO5 is a quantum-singlet material composed of four antiferromagnetically coupled spins.

Strength Demonstrated by This Achievement

This work combines magnetic-susceptibility and heat-capacity measurements with inelastic neutron scattering to describe the ground state, exchange interactions, and magnetic excitations of a quantum spin system microscopically and quantitatively within a unified spin model.

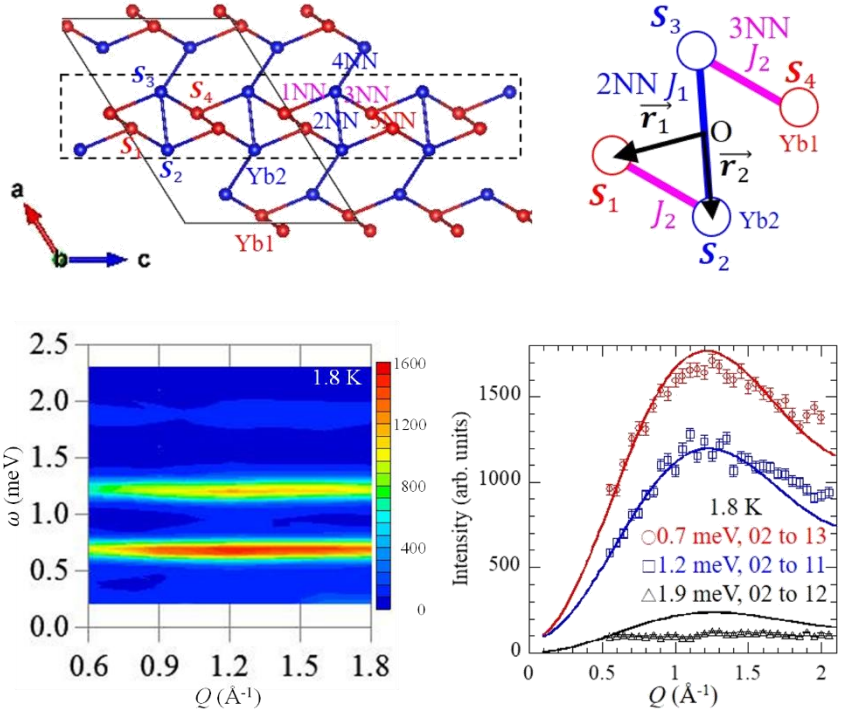
Representative Publication

M. Hase, K. Kaneko, C. Tabata, H. Yamauchi, N. Tsujii and A. Dönni,

“Spin-Singlet Ground State with Spin Gap in Seff = 1/2 Antiferromagnetic Tetramer Compound Yb2SiO5,”

Physical Review B, 111, 094403 (2025).

DOI: 10.1103/PhysRevB.111.094403



3. Cryogenic Magnetic Cooling in a Highly Ordered Perovskite

Alexei A. BELIK

The properties of perovskite-type oxides depend strongly on which elements occupy specific positions within the crystal.

Using high-temperature and high-pressure conditions of 6 GPa, we synthesized the new perovskite Nd2ZnZn(Zn2Sb2)O12, which exhibits distinct cation ordering at both the A and B sites.

In this material, Nd and Zn form columnar order at the A site, while Zn and Sb adopt rock-salt-type order at the B site. High-pressure synthesis enabled this complex crystal structure containing multiple simultaneous cation-ordering patterns.

Magnetization and heat-capacity measurements further revealed a magnetocaloric effect at low temperatures. This result extends the design of complex elemental arrangements toward the exploration of materials for cryogenic magnetic refrigeration.

Strength Demonstrated by This Achievement

This crystal-chemistry research uses high-pressure synthesis to order multiple cations over distinct crystallographic sites and connects the resulting crystal structure with magnetic and thermal functionality.

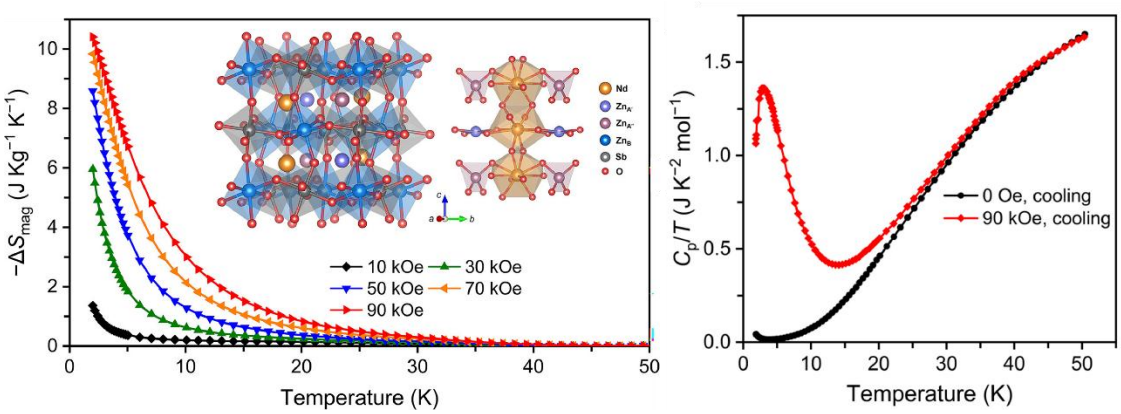
Representative Publication

X. Liang, K. Yamaura and A. A. Belik,

“Cryogenic Magnetocaloric Properties of Zinc-Based Perovskite Nd2ZnZn(Zn2Sb2)O12 with A-Site Columnar-Type and B-Site Rock-Salt-Type Orders,”

Acta Materialia, 302, 121626 (2026).

DOI: 10.1016/j.actamat.2025.121626



4. Atomistic Elucidation of Dy-Induced Spin Reorientation in Permanent Magnets

Masamichi NISHINO

Nd-Fe-B magnets are widely used as high-performance permanent magnets in motors and energy-

efficient devices. Partial substitution of Nd by Dy can enhance coercivity, but a microscopic understanding is needed of how Dy substitution changes the magnetization direction and the spin-reorientation transition.

In this study, we constructed an atomistic spin model based on first-principles calculations. The model incorporates atomic spins, exchange interactions, magnetocrystalline anisotropy, and thermal fluctuations, and was used to analyze the magnetic properties of (Nd1-xDyx)2Fe14B.

The model reproduced the experimentally observed Dy-concentration dependence of the spin-reorientation transition temperature. It also clarified how the different magnetic anisotropies of Nd and Dy determine the temperature-dependent magnetization direction and showed that a Dy-concentration-dependent gap appears in the temperature derivative of the magnetization near the transition temperature.

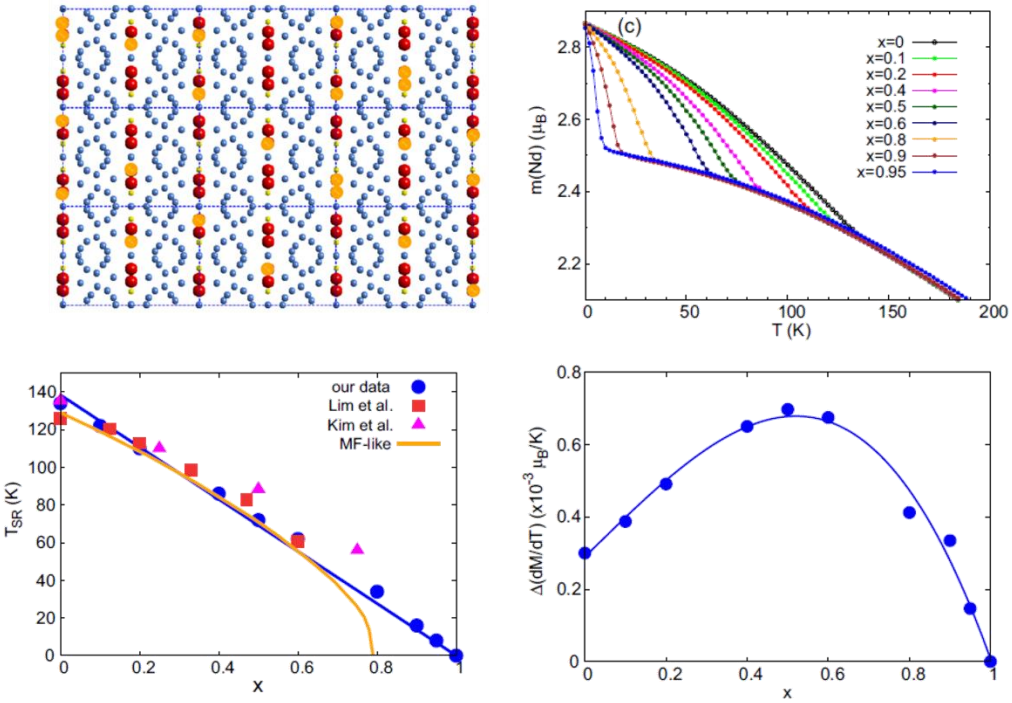
The transition temperature, magnetization direction, and anomalous magnetization response were thereby explained consistently within a single atomistic model, revealing the microscopic origin of the spin-reorientation transition in rare-earth magnets.

Strength Demonstrated by This Achievement

This theoretical work uses a first-principles-based atomistic spin model to reproduce experimentally observed transition temperatures and magnetization responses, providing a unified explanation of the spin-reorientation mechanism produced by magnetic anisotropy and thermal fluctuations.

Representative Publication

M. Nishino, R. Lamouri and H. Akai,
“Atomistic Model Analysis of the Spin Reorientation Transition in (Nd1−xDyx)2Fe14B Systems,”
Physical Review B, 113, 064427 (2026).
DOI: 10.1103/kdjg-bhy8



5. Two-Dimensional Lithium-Ion Conduction in New Layered Oxysulfides

Yoshihiro TSUJIMOTO

Replacing some of the oxygen in oxides with sulfur, nitrogen, halogens, or other anions enables the design of materials with crystal structures, electronic states, and ion-transport properties not found in conventional oxides. This mixed-anion approach has attracted attention as a powerful route to new functional solid-state materials.

In this study, we successfully synthesized the Ruddlesden-Popper-type layered oxysulfides LiLnTiO3S (Ln = La-Gd) by single-crystal growth using a halide flux. These materials adopt tetragonal structures containing two-dimensional LiO planes and were identified as globally rare lithium-ion conductors among oxysulfides.

In the Nd-containing system, partial substitution of Ti by Nb introduced Li vacancies, improving the lithium-ion conductivity beyond that of known related oxides. Bond-valence analysis further demonstrated the formation of two-dimensional lithium-ion conduction pathways within the LiO planes.

This achievement introduces oxysulfides as a new materials platform for lithium-ion conductors, a field previously centered mainly on oxides, and points to a new direction for developing solid electrolytes through mixed-anion chemistry.

Strength Demonstrated by This Achievement

This work integrates the design of anion species and arrangements, layered crystal structures, and elemental substitution, combining single-crystal growth, structural analysis, and ion-conduction

measurements to develop a new class of mixed-anion ion conductors.

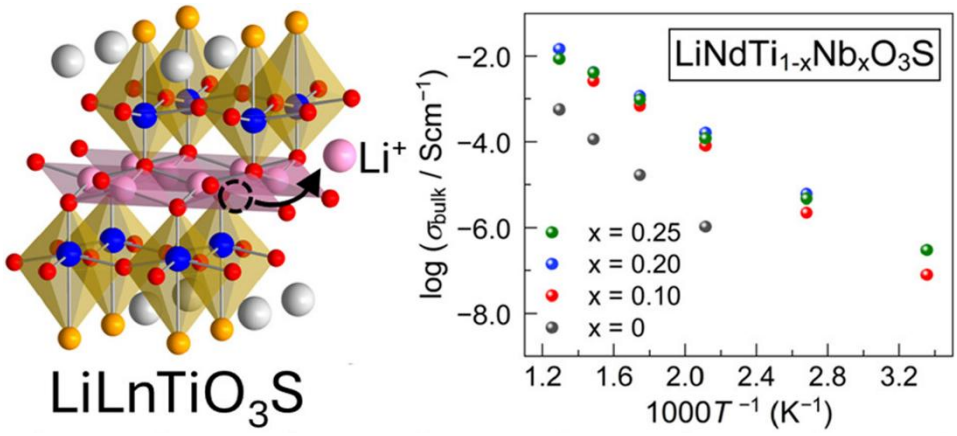
Representative Publication

H. Yuan, K. Sugii, G. Hasegawa, Y. Meng, Y. Matsushita, H. Akamatsu, N. Kuwata, K. Yamaura and Y. Tsujimoto,

“Flux Crystal Growth of Ruddlesden–Popper Titanium Oxysulfides LiLnTiO3S (Ln = La–Gd): Lithium-Ion Mobility in Li1−xNd(Ti1−xNbx)O3S (x = 0–0.25),”

Chemistry of Materials, 38, 1457–1468 (2026).

DOI: 10.1021/acs.chemmater.5c02924



A defining feature of our research is the way we connect individual areas of expertise and cycle continuously from materials creation to analysis and understanding, and then back to the design of the next material.