3.5 SCCMS Projects

Estimation of Hamiltonians of quantum magnets using Bayesian optimization

Takahiro Misawa
Institute for Solid State Physics, University of Tokyo
Kashiwa-no-ha, Kashiwa, Chiba 277-8581

Determination of spin Hamiltonians in magnetic compounds has been recognized as a crucial step in elucidating their magnetic properties. A commonly adopted approach involves the derivation of a Hubbard-type Hamiltonian via *ab initio* downfolding [1], followed by the application of a strong-coupling expansion to obtain an effective spin model. Although this method has been successfully applied to various materials, the absence of a systematic criterion for determining the required order of perturbative expansion has been noted as a limitation. In addition, the Liechtenstein formula [2] has been widely employed to evaluate exchange interactions between classical spins, where the energy variation caused by infinitesimal rotations of magnetic moments around a given magnetic order is computed. Despite its widespread use, the applicability of classical spin models derived from this approach to quantum magnetic systems remains unclear.

Among quantum magnets, α-RuCl$_3$ has been regarded as a prototypical system in which the determination of the spin Hamiltonian continues to be debated. This compound has been intensively investigated as a promising candidate for realizing the Kitaev quantum spin liquid [3]. In conventional approaches, the estimated parameters of the spin Hamiltonian in α-RuCl$_3$, such as the Kitaev interaction (K), off-diagonal terms (Γ, Γ'), and the Heisenberg interaction (J), have been shown to vary significantly depending on the computational method employed [4].

In this project, an alternative strategy is used for determining the spin Hamiltonian of α-RuCl$_3$ through the formulation of an inverse problem constrained by experimental data [5]. Bayesian optimization is employed to identify the parameter set that best reproduces the observed magnetization process. We focus the model that has five essential parameters: the Kitaev interaction (K), the off-diagonal interactions (Γ and Γ'), the Heisenberg interaction (J), and the g-factor along the c-axis (g_c). In the actual calculations, the magnetization curves are computed using the exact diagonalization method implemented in $\mathcal{H}\Phi$ [6] and the Bayesian optimization is performed using Physbo [7]. As a result, we obtain the following optimal parameter set: $K = -6.0$, $\Gamma = 7.5$, $\Gamma' = -0.3$, $J = -1.75$ (in meV), and $g_c = 2.3$. We confirm that this set is found to well reproduce the experimental magnetization curves as well as othe physical quantities such as the specific heat and the spin correlations.

We also peform a similar Bayesian optimization for the spin Hamiltonian of azurite, which is a well-studied one-dimensional quantum magnet. As a result, we also find that the parameters set that well reproduces the experimental magnetization curve. Through these applications, we have demonstrated that the Bayesian optimization provides a systematic and efficient approach for determining spin Hamiltonians in quantum magnets.

References

[1] M. Imada, T. Miyake, J. Phys. Soc. Jpn. **79**, 112001 (2010).

[2] A. I. Liechtenstein, M. Katsnelson, V. Antropov, V. Gubanov, Journal of Magnetism and Magnetic Materials **67**, 65 (1987).

[3] A. Kitaev, Annals of Physics **321**, 2 (2006).

[4] P. A. Maksimov, A. L. Chernyshev, Phys. Rev. Res. **2**, 033011 (2020).

[5] T. Misawa, K. Yoshimi, R. Tamura, Y. Yamaji, in preparation.

[6] K. Ido, M. Kawamura, Y. Motoyama, K. Yoshimi, Y. Yamaji, S. Todo, N. Kawashima, T. Misawa, Comp. Phys. Commun. **298**, 109093 (2024).

[7] Y. Motoyama, R. Tamura, K. Yoshimi, K. Terayama, T. Ueno, K. Tsuda, Computer Physics Communications **278**, 108405 (2022).

Theoretical study on proton transfer with nuclear quantum effect in condensed systems

Tomomi Shimazaki
Graduate School of Nanobioscience,
Yokohama City University, 22-2 Seto, Kanazawa, Yokohama 236-0027

There are two primary mechanisms for proton transport. One is the vehicular mechanism, in which protons move via diffusion, and the other is the Grotthuss mechanism, where protons are transferred through hydrogen-bond network rearrangement and hopping. Since proton hopping is a type of chemical reaction, a quantum chemical approach based on first-principles calculations is required to investigate the Grotthuss mechanism in detail. In this year, we have investigated fundamental behaviors of several condensed systems. We have also studied simulation methodologies to handle such phenomena. [3] However, even with today's advanced computational power, it remains challenging to apply only first-principles or quantum chemical methods to handle entire interesting materials. Therefore, this research project employs a QM/MM approach. In the developed QM/MM program, the MM region utilizes "MDYLAS," enabling highly efficient molecular dynamics (MD) simulations even on the Fugaku supercomputer. For the QM region, the Multi-Component Density Functional Theory (MC-DFT) method is used to consider proton quantum effects. In QM-based MD simulation with the quantum effect are considered, a proton more frequently takes the Zundel structure, where it is located between two oxygen atoms, compared to conventional quantum chemical calculations based on the Born–Oppenheimer approximation. This is likely because nuclear quantum effects lower the reaction barrier for proton transfer, making the Zundel structure more favorable. In the future, we plan to investigate various systems, such as carbon-based fuel cell materials and covalent organic frameworks (COFs), to analyze proton transport behavior. In these materials, the proton transfer plays essential roles.

References

[1] E, Kino and T. Shimazaki, *Phys. Chem. Chem. Phys.*, **27**, 7429 (2025).

[2] T. Wakahara, T. Shimazaki, et al, *Carbon*, 233, 119838 (2025).

[3] T. Shimazaki and M. Tachikawa, *J. Chem. Phys.*, **161**, 014107 (2024).

High-throughput calculations for magnetic materials with configurational and structural disorders

Tetsuya FUKUSHIMA
CD-FMat, National Institute of Advanced Industrial Science and Technology (AIST)
1-1-1 Umezono, Tsukuba, Ibaraki 305-8568, Japan

The purpose of Data Creation and Utilization-Type Material Research and Development Project (Digital Transformation Initiative Center for Magnetic Materials) is to find new functional permanent magnets, soft magnets, and spintronics materials by data-driven approaches.

In this year, we proceed with two research topics using first-principles calculations: (1) data creation of the magnetic anisotropic energies of $Nd_2Fe_{14}B$ and (2) high-throughput calculations of the magnetic damping constants for FeCoNi alloys.

In the first research topic, we systematically investigate the magnetocrystalline anisotropy of $Nd_2Fe_{14}B$ under lattice distortions. The calculations are performed using the all-electron full-potential Korringa–Kohn–Rostoker (KKR) Green's function method. Spin-orbit coupling is included as the $l_z s_z$ term. The neodymium $4f$ orbitals are treated within the open-core approximation, where the $4f$ electrons are considered as core-like states, and their occupations are not explicitly included in the self-consistent calculations. The distortions are introduced by altering the lattice constant a of the tetragonal crystal structures.

Figure 1 shows the calculated values of the magnetocrystalline anisotropy constant K_u and magnetization J_S for $Nd_2Fe_{14}B$ as a function of the distortion δa in the ab-plane. When there is no distortion ($\delta a = 0$), the calculated value of K_u is 4.2 MJ/m^3, which is close to the experimental value of approximately 5 MJ/m^3 reported in previous studies. Additionally, J_S = 1.89 T is in good agreement with the experimental value of 1.85 T at 4.2 K.

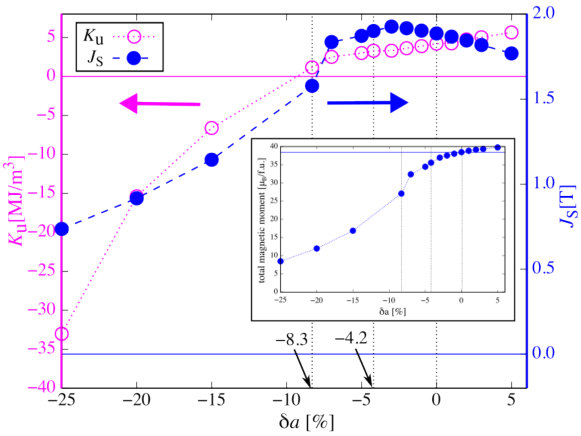

Figure 1: Calculated values of K_u and J_S for $Nd_2Fe_{14}B$ with δa. $\delta a = 0$ corresponds to the experimental lattice constant. The inset figure shows the calculated total magnetic moment as a function of δa.

K_u decreases monotonically with in-plane compression. The $5d$ electron cloud of Nd extends and hybridizes with Fe/B ions within the plane. In-plane compression increases the degree of hybridization, causing the $5d$ orbitals to expand further in the plane. To avoid this $5d$ electron cloud, the $4f$ electrons of Nd ions, which have an oblate shape, spread in the out-of-plane direction, resulting in a reduction of the anisotropy. Furthermore, K_u changes from positive to negative around $\delta a = -8.3$ %. With further compression, the absolute value of K_u

becomes larger and negative. Such extreme compression is observed in strain at interfaces, suggesting that K_{u} becomes negative in these regions.

For J_{S}, no monotonic decrease with respect to δa is observed. However, by examining the total magnetic moment shown in the inset figure of Fig.1, a monotonic decrease is confirmed. Therefore, the reason for the maximum of J_{S} around $\delta a = -4$ % is that the decrease in the crystal volume compensates for the reduction in magnetization. Similarly to K_{u}, J_{S} decreases as δa decreases. Thus, in regions with significant in-plane distortion (such as at interfaces), both K_{u} and J_{S} decrease simultaneously, suggesting that the impact on coercivity is minimal.

The second research topic is for the high-throughput calculations for magnetic damping constants (α), which characterize magnetization dynamics. α strongly affects the operating current and power consumption of spintronics devices; thus, the theoretical approaches to design α are highly desired. However, α exhibits non-monotonic temperature dependence. The conventional frameworks that ignore the temperature effects are insufficient for the practical evaluations. To address this problem, we propose an evaluation framework[1] that incorporates temperature effects into electronic structures via the disordered local moment (DLM) state within first-principles calculations. In this study, we apply this framework to FeCoNi alloys. We systematically calculate α and analyze the magnetic damping mechanisms from an electronic structure perspective.

To evaluate the magnetic damping constant at finite temperatures, the electronic structure calculations are performed using the tight-binding linear muffin-tin orbital method. The local spin density approximation functional is employed, and atomic configuration averaging is incorporated using the coherent potential approximation (CPA). For the evaluation of α, the torque correlation method proposed by Kamberský[2] and Gilmore et al.[3] is adopted. This framework evaluates the intrinsic α due to spin-orbit interaction. The temperature effects are incorporated into the spin fluctuations using the DLM-CPA method, and the effect of these fluctuations is accounted for by evaluating the thermodynamic potential via the electronic structure calculations. The calculations are performed for the FeCoNi alloys with BCC and FCC structures.

Analyzing the calculated data, we confirm that the composition with the minimum α depends on the temperatures. In the BCC structure, the composition with the minimum α is $\mathrm{Fe}_{0.75}\mathrm{Co}_{0.25}$ from 0 K to 500 K, while at temperatures above 1200 K, the minimum *alpha* is achieved in the pure-Fe. These results are consistent with the previous experiments. Furthermore, from the analysis of the electronic structures, a strong correlation is confirmed between α and the density of states at the chemical potential. With increasing temperature, the correlation becomes stronger. These results are quite useful for the design of spintronic materials with small magnetic damping constants.

References

[1] R. Hiramatsu, D. Miura, and A. Sakuma, Appl. Phys. Express **15**, 013003 (2021).

[2] V. Kamberský, Phys. Rev. B **76**, 134416 (2007).

[3] K. Gilmore, Y. U. Idzerda, and M. D. Stiles, Phys. Rev. Lett. **99**, 027204 (2007).

[4] M. A. W. Schoen, D. Thonig, M. L. Schneider, T. J. Silva, H. T. Nembach, O. Eriksson, O. Karis, and J. M. Shoaw, Nat. Phys. **12**(9), 839 (2016).

All-atom analysis of the solubilities of polypeptides

Kazuo YAMADA and Nobuyuki MATUBAYASI
Division of Chemical Engineering, Graduate School of Engineering Science,
Osaka University, Toyonaka, Osaka 560-8531

Artificial polypeptides, synthesized from amino acids, have garnered increasing interest as promising candidates for biopharmaceutical applications due to their biodegradability and tunable functionality. They are particularly being investigated as alternatives to PEGylated protein-based drugs, offering intrinsic biodegradability and a lower risk of long-term side effects. Additionally, they are viewed as potential medium-molecular-weight peptide therapeutics. The chemical diversity of artificial polypeptides is remarkably vast. Even with only the 20 canonical amino acids, the number of possible sequences is immense. The incorporation of non-natural amino acids further expands the chemical space, enabling the design of peptides with improved stability, specificity, and bioavailability. This versatility supports the development of peptide-based drugs with reduced off-target effects compared to conventional small-molecule therapeutics.

Among their physicochemical properties, aqueous solubility and partition coefficients are particularly important, as they directly influence pharmacokinetic properties including absorption, distribution, metabolism, and excretion (ADME). Therefore, a systematic understanding of how amino acid sequence affects solubility is essential for rational peptide drug design.

To address this challenge, we developed a computational framework to predict the aqueous solubility of peptide-based drugs. As a key physicochemical descriptor, we use the solvation free energy, $\Delta\mu$, which quantifies the free energy change when transferring a solute from vacuum to a solvent. $\Delta\mu$ reflects the specificities of the solute-solvent interactions sensitively. Solvation free energies were calculated using all-atom molecular dynamics (MD) simulations combined with the energy representation (ER) method, implemented via ERmod software [1]. The ER method estimates $\Delta\mu$ from the distribution of solute–solvent interaction energies, using only two endpoint states: the fully coupled solute–solvent system and a decoupled reference state. This approach circumvents the need for intermediate coupling states required by conventional methods such as thermodynamic integration (TI) or free energy perturbation (FEP), significantly reducing computational cost while maintaining chemical accuracy. We computed $\Delta\mu$ for 15 different homo-tetrapeptides in both single-component pure solids and in aqueous solutions at infinite dilution. The difference between the two solvation free energies, denoted as $\Delta\Delta\mu$ = $\Delta\mu$(water) − $\Delta\mu$(pure solid), directly represents the magnitude for dissolution into water. MD simulations were performed using GROMACS 2024 [2] on GPU nodes for equilibration and sampling, while ERmod calculations were conducted on CPU nodes [1].

To elucidate the molecular factors governing peptide solubility, we decomposed the calculated $\Delta\mu$ values into interaction components. Figure 1 shows that the excluded-volume contribution, $\Delta\Delta\mu^{\mathrm{excl}}$, exhibits the strongest correlation with peptide solubility (correlation coefficient r = 0.86). This component represents the free energy penalty associated with displacing solvent molecules from the volume occupied by the solute.

In contrast, the correlation between peptide solubility and the direct interaction energy between solute and solvent molecules, defined as the energy difference $\Delta\epsilon = \epsilon(\mathrm{peptide} - \mathrm{water}) - \epsilon(\mathrm{peptide} - \mathrm{peptide})$, was weakly negative ($r = -0.13$). These results suggest that excluded-volume effects, which are strongly related to molecular size and steric occupancy, exert a greater influence on aqueous solubility than specific interactions such as hydrogen bonding. In other words, the spatial footprint of a peptide in solution contributes more significantly to its solubility than the strength of its direct interactions with the solvent. Overall, the excluded-volume component represents the free energy cost associated with peptide insertion into the solvent matrix, increasing with molecular size. These findings underscore that, beyond hydrophilic/hydrophobic balance, molecular size and steric effects are key determinants of peptide solubility in aqueous environments.

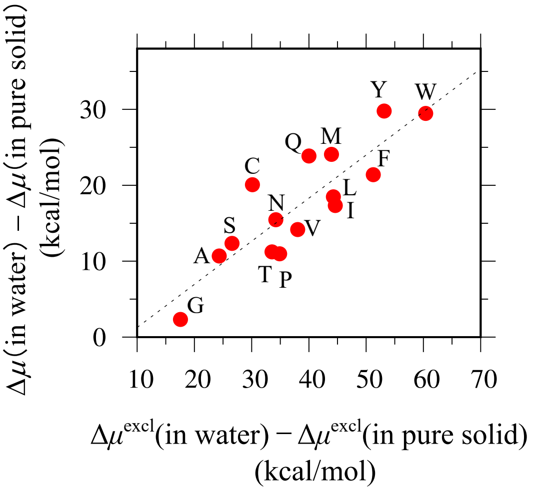

Figure 1: Correlation between $\Delta\Delta\mu$ and $\Delta\Delta\mu^{\mathrm{excl}}$ for 15 homo-tetrapeptides, where each is defined as the difference between values in the water solvent and in the pure solid. A strong correlation is observed with a correlation coefficient r = 0.86.

References

[1] S. Sakuraba and N. Matubayasi: J. Comput. Chem. **35** (2014) 1592-1608

[2] Abraham, M. et. al., “GROMACS 2024 Manual”, Zenodo., (2024)

High-throughput first-principles exploration of stable Heusler alloys

Terumasa TADANO
National Institute for Materials Science
Sengen 1-2-1, Tsukuba, Ibaraki 305-0047

Heusler alloys have attracted significant attention due to their exceptional magnetic and multifunctional properties such as high saturation magnetization, large magnetocrystalline anisotropy, high T_c, notable magnetocaloric effects, and promising thermoelectric performance, which have prompted high-throughput (HTP) computational studies. However, previous HTP screenings have focused mostly on thermodynamic stability and omitted dynamical stability. Since unstable phonon modes can lead to structural phase transitions that may affect performance, our study incorporates phonon stability as an additional crucial metric of stable Heusler alloys.

Our HTP screening encompasses 27,864 Heusler compositions, including regular and inverse Heusler compounds with the X_2YZ composition, half-Heusler compounds with the XYZ composition, and X_3Z compounds (where $X = Y$). Initial structures are generated using primitive cells with lattice parameters taken from the Open Quantum Materials Database (OQMD) when available, or estimated from averages of related compounds. These structures are optimized using density functional theory (DFT) with the Vienna *ab initio* Simulation Package (VASP) under the generalized gradient approximation (GGA). Multiple magnetic configurations, including high-spin and low-spin states as well as various alignment patterns, are tested to determine the magnetic ground state.

Thermodynamic stability is determined by the following two criteria: The formation energy (ΔE) must be negative relative to the elemental reference states, and the energy above the convex hull (ΔH) must be less than 0.3 eV/atom. Only compounds meeting these criteria are then assessed for dynamical stability via phonon calculations, carried out within the harmonic approximation using the ALAMODE package. A compound is considered dynamically stable if all phonon frequencies at commensurate q points are nonnegative. The magnetic transition temperature T_c is calculated within the mean-field approximation using exchange constants from the spin-polarized relativistic Korringa-Kohn-Rostoker (SPRKKR) code.

Out of 106,235 computed structures (including both ground states and metastable states), our workflow identifies 8,191 compounds that satisfy the thermodynamic stability criteria. Phonon calculations were successful for 8,180 compounds, of which 4,011 are dynamically stable (Fig. 1). This indicates the effectiveness of considering the phonon stability for a robust prediction of stable Heusler compounds.

Among the dynamically stable compounds, 1,356 are magnetic. When filtered by $T_\mathrm{c} > 300$ K, 631 promising candidates are identified, which include 47 low-moment compensated ferrimagnets. The identified list includes not only Mn-based Heuslers but also Cr-, Ti-, and V-based Heuslers, whose anomalous Hall conductivity can be larger than $250\,\mathrm{S\,cm^{-1}}$.

Our analysis also reveals clear correlations

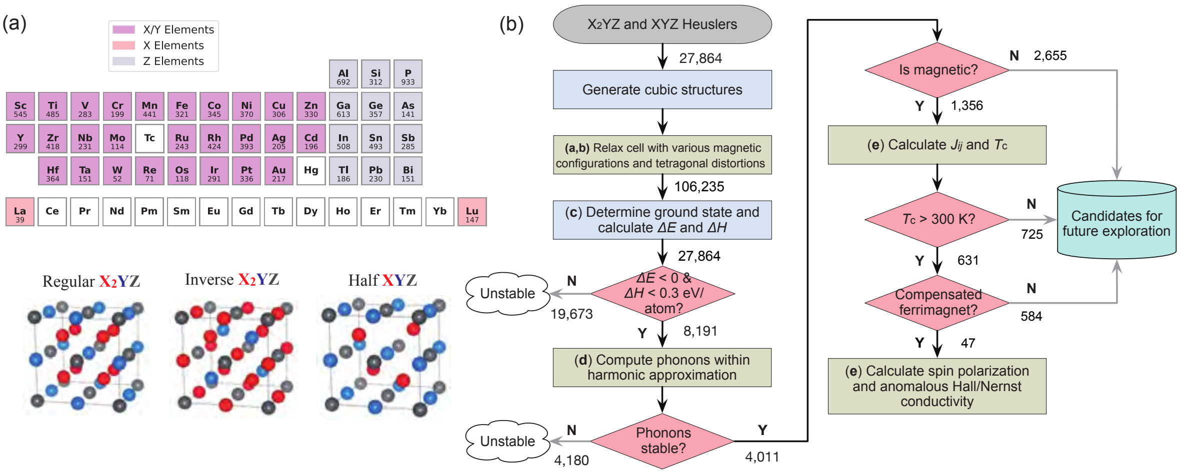

Figure 1: (a) The compositions covered in the high-throughput search and distribution of stable compositions. (b) Workflow of the high-throughput search for stable Heusler compounds. Adapted from Ref. [1].

between stability and atomic properties. In X_2YZ compounds, stability is enhanced when the Z element has a small atomic radius and low ionization energy. For half-Heusler compounds, a necessary condition for stability is that the atomic radius of X is smaller than that of Y. These trends offer useful guidelines for selecting elemental combinations.

In summary, our high-throughput study identifies 631 stable magnetic Heusler compounds with promising functional properties, including 47 candidates of low-moment compensated ferrimagnets. These findings, combined with the comprehensive dataset generated in this work, will contribute significantly to the discovery of next-generation materials for spintronic and energy-harvesting applications and provide a valuable resource for future machine-learning efforts in materials design.

This study was supported by MEXT Program: Data Creation and Utilization-Type Material Research and Development Project (Digital Transformation Initiative Center for Magnetic Materials) Grant Number JPMXP1122715503 and as "Program for Promoting Researches on the Supercomputer Fugaku" (Data-Driven Research Methods Development and Materials Innovation Led by Computational Materials Science, JPMXP1020230327). This study used computational resources of supercomputer Fugaku provided by the RIKEN Center for Computational Science (Project ID: hp240223), the computer resources provided by ISSP, U-Tokyo under the program of SCCMS, and the computer resources at NIMS Numerical Materials Simulator.

References

[1] E. Xiao and T. Tadano, arXiv:2502.17946.