Ordering in magnetic quasicrystals and approximants

T.J. Sato

Institute of Multidisciplinary Research for Advanced Materials, Tohoku University
2-1-1 Katahira, Aoba, Sendai 980-8577, Japan

e-mail: taku@tohoku.ac.jp

Magnetic properties of quasicrystals have been continuously studied from the beginning, as even the first quasicrystal found in the Al-Mn rapidly quenched alloy [1] includes Mn, which is often magnetic. Until recently, however, spin-glass-like random freezing of spins has been commonly observed both in the metastable (quenched) and thermodynamically-stable quasicrystals. This led to the controversy if the quasiperiodic magnetic crystal can exhibit long-range magnetic order.

Recently, several progresses have been made in our understanding of magnetic order in quasicrystals and related rational approximants. First, quite a few magnetic approximants were found to exhibit magnetic long-range order. Examples of such are Cd_6*RE* [2], Au-Si-Tb [3] and Au-Al-Tb [4], just to note a few. The magnetic structures of them were investigated using either x-ray or neutron diffraction techniques, and intriguing non-collinear non-coplanar magnetic structure has been reported in some of the compounds [3,4].

Based on the accumulated knowledge on the magnetic approximants, Tamura et al. recently conjectured that the magnetic order in the approximants is indeed controlled by the electron density [5]. Encouraged by this inspiring conjecture, a new magnetic quasicrystal in the Au-Ga-*RE* (*RE* = Gd and Tb) system has been designed, in which the first ferromagnetic-like order was observed. We have performed neutron diffraction experiment on the magnetic quasicrystals and found clear evidence of long-range magnetic order [6].

In this talk, we will report the observation of long-range magnetic order in approximants as well as quasicrystals. We will also report present status of the magnetic structure analysis in the quasicrystal by applying magnetic representation theory to high-dimensional crystallography.

Acknowledgements: This work has been performed under the collaboration with many researchers including A. Ishikawa, S. Suzuki, T. Kotajima, Y. Tanaka, T. Seki, N. Shibata, T. Yamada, T. Fujii, C. -W. Wang, M. Avdeev, K. Nawa, D. Okuyama, T. Hiroto, H. Cao, T. Hawai, T. Yokoo, S. Itoh, K. Takagi and Y. G. So.. The author thanks late professor A. P. Tsai for his continuous support during this study. This work is partly supported by JSPS KAKENHI (JP22H00101, JP19KK0069, JP19K21839, and 19H05824).

[1] D. Shechtman, I. Blech, D. Gratias, J. W. Cahn, Phys. Rev. Lett. **53** (1984) 1951.
[2] R. Tamura, Y. Muro, T. Hiroto, K. Nishimoto, and T. Takabatake, Phys. Rev. B **82** (2010) 220201R.
[3] T. J. Sato, A. Ishikawa, A. Sakurai, M. Hattori, M. Avdeev and R. Tamura, Phys. Rev. B **100** (2019) 054417.
[4] T. Hiroto, T. J. Sato, H. Cao, T. Hawai, T. Yokoo, S. Itoh, and R. Tamura, J. Phys.: Condens. Matter **32** (2020) 415802.
[5] A. Ishikawa, T. Hiroto, K. Tokiwa, T. Fujii, and R. Tamura, Phys. Rev. B **93** (2016) 024416.
[6] R. Tamura, A. Ishikawa, S. Suzuki, T. Kotajima, Y. Tanaka, T. Seki, N. Shibata, T. Yamada, T. Fujii, C.-W. Wang, M. Avdeev, K. Nawa, D. Okuyama, T. J. Sato, J. Am. Chem. Soc. **143** (2021) 19938.

Various magnetic phases in Tsai-type 2/1 quasicrystal approximants

T. Sugimoto[1#], S. Suzuki[2], R. Tamura[3], T. Yamada[4] and T. Tohyama[4]

[1]*Center for Quantum Information and Quantum Biology, Osaka University, Toyonaka, Osaka 560-0043, Japan*
[2]*Department of Physical Science, Aoyama Gakuin University, Sagamihara, Kanagawa 252-5258, Japan*
[3]*Department of Material Science and Technology, Tokyo University of Science, Katsushika, Tokyo 125-8585, Japan*
[4]*Department of Applied Physics, Tokyo University of Science, Katsushika, Tokyo 125-8585, Japan*

[#]e-mail: sugimoto.takanori.qiqb@osaka-u.ac.jp

Recent experimental studies on Tsai-type quasicrystal approximants including rare-earth ions have shown various magnetic phases and successive variation in the Curie-Weiss temperature [1]. Particularly, a Tsai-type 1/1 approximant Au-Al-Gd exhibits several phase transitions among ferromagnetism, antiferromagnetism, and spin glass, as the Au concentration changes. In this family, magnetic moments of Gd ions located at vertices of icosahedra in the Tsai clusters, are coupled via the so-called Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction [1,2]. The Gd ions have no orbital angular momentum, and thus the single-ion anisotropy due to the spin-orbit interaction should be strongly suppressed [2]. Therefore, the Friedel oscillation of magnetic exchange energy in the RKKY interaction is supposed to play the key role to exhibit such the interesting phenomenon. Considering the crystal structure of 1/1 approximants, we have succeeded in the numerical reproduction of these various magnetic phases by means of classical Monte-Carlo simulation combined with an appropriate intuitive picture of a cluster model [3,4]. Here we extend the theoretical analysis of magnetic orders to Tsai-type 2/1 quasicrystal approximants. We assume that the primary effect of the Au concentration on the magnetic system is a shift of the Fermi wavenumber. We performed classical Monte-Carlo simulations at zero temperature in the 2/1 approximant structure, resulting in a successive reproduction of several phase transitions among ferromagnetic, antiferromagnetic, incommensurate magnetic-ordered, and other exotic-ordered phases [5]. In real materials, there is always randomness, e.g., chemical disorder and defects in the crystal structure. Thus, because of the randomness, we consider the incommensurate state as the experimentally-observed spin glass order in the real compound. Based on these results, we will discuss what kinds of magnetic orders are realized in the limit of increasing the approximation degree toward quasicrystals.

Acknowledgements: The authors acknowledge financial support from JSPS KAKENHI Grant Numbers JP19H05821.

[1] A. Ishikawa, T. Hiroto, K. Tokiwa, T. Fujii, and R. Tamura, Phys. Rev. B **93**, 024416 (2016); A. Ishikawa, T. Fujii, T. Takeuchi, T. Yamada, Y. Matsushita, and R. Tamura, Phys. Rev. B **98**, 220403 (2018); S. Suzuki, A. Ishikawa, T. Yamada, T. Sugimoto, A. Sakurai, and R. Tamura, Mater. Trans. **62**, 298 (2021).
[2] T. Sugimoto, T. Tohyama, T. Hiroto, and R. Tamura, J. Phys. Soc. Jpn. **85**, 053701 (2016).
[3] H. Miyazaki, T. Sugimoto, K. Morita, and T. Tohyama, Phys. Rev. Mater. **4**, 024417 (2020).
[4] S. Suzuki, R. Tamura, and T. Sugimoto, Mater. Trans. **62**, 367 (2021).
[5] T. Sugimoto, S. Suzuki, R. Tamura, T. Yamada, T. Tohyama, in preparation.

Classical spin models on the 1/1 Tsai-type approximant

Daniel Qvarngård[1] and P. Henelius[2]

[1]*Department of Physics, Royal Institute of Technology, SE-106 91 Stockholm, Sweden*
[2]*Faculty of Science and Engineering, Åbo Akademi University, Åbo, Finland*

[#]e-mail: danielqv@kth.se

In this talk, we present a study of classical spin models on the 1/1 Tsai-type approximant lattice using Monte Carlo and mean-field methods. We assume the presence of the RKKY interaction in order to mimic the experimentally observed dependence of the magnetic properties on the electron concentration. To investigate whether the differences between Gd- and Tb-based approximant phase diagrams can be traced to the crystal-electric field, we make an easy-axis ansatz and model the Tb ions as Ising spins while modeling Gd-ions as classical Heisenberg spins.

We find that the ground state magnetic ordering vectors under the RKKY interaction are unaffected by the anisotropy – even when including a dipolar perturbation. However, the dipolar perturbation induces ground state magnetic ordering in the Ising system reminiscent of the recently resolved ferromagnetic structure in Au-Si-Tb approximants discussed by Hiroto et al[1]. In the Heisenberg system, the dipolar interaction breaks the rotational symmetry of the ground state, and we predict that Gd-based approximants have magnetic ordering along the 111-axis.

Acknowledgements: We thank Cesar Pay Gómez and Johan Hellsvik for insightful discussions, and Simon "Raba" Ranefjärd for valuable input on figure design. This work has been financially supported by the Knut and Alice Wallenberg foundation (grant number KAW 2018.0019). The simulations were performed on resources provided by the Swedish National Infrastructure for Computing (SNIC) at the Center for High Performance Computing (PDC) at the Royal Institute of Technology (KTH).

[1] T. Hiroto et al, J. Phys.: Condens. Matter **32** (2020) 415802

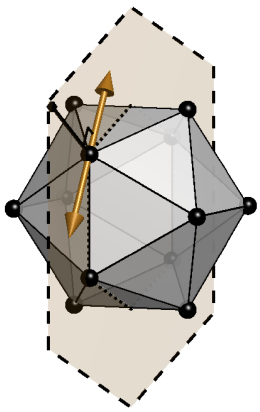


Fig.1: Easy axis ansatz, Tb magnetic moments assumed normal to the Tsai-type rare-earth cluster

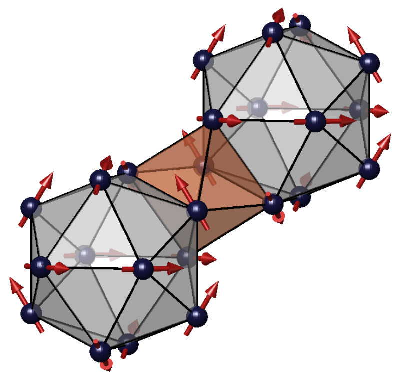


Fig.2: Ising model ground state under RKKY and Dipole interactions

Exact quantum ground state of a two-dimensional quasicrystalline antiferromagnet

P. Ghosh[1#]

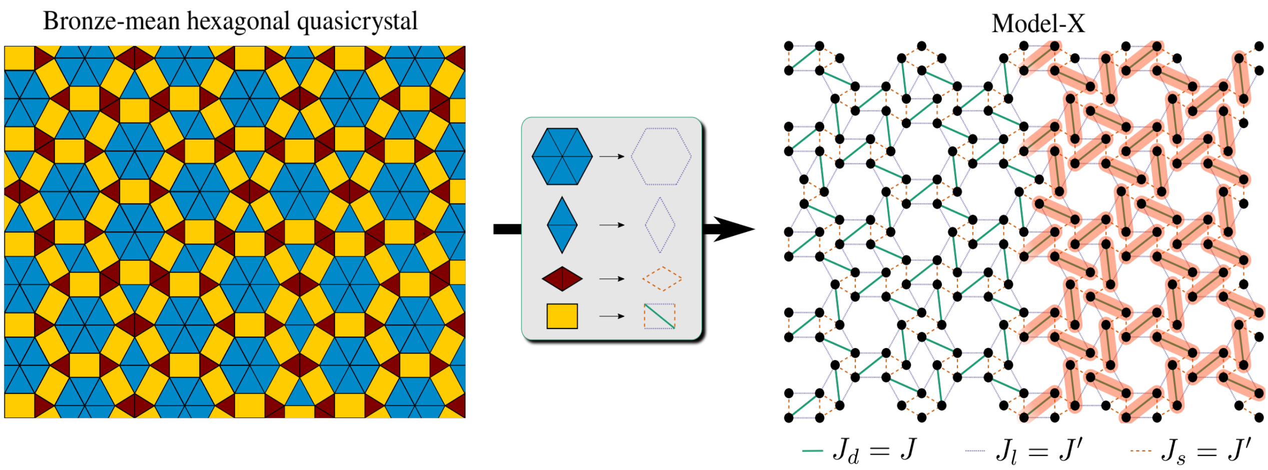
[1]*Institut für Theoretische Physik und Astrophysik and Würzburg-Dresden Cluster of Excellence ct.qmat, Universität Würzburg, Am Hubland Campus Süd, Würzburg 97074, Germany*

[#]e-mail: pratyay.ghosh@physik.uni-wuerzburg.de

We present the exact dimer ground state of a quantum antiferromagnet defined on a quasicrystal constructed from the Bronze-mean hexagonal quasicrystal [1, 2]. A coupling isotropy on the first and second-neighbor bonds is sufficient to stabilize a product state of singlets on the third-neighbor bonds. The ground state here is similar to the exact dimer ground state of celebrated Shastry-Sutherland [3] and Maple leaf model [4]. We also provide a systematic approach for constructing additional crystals, quasicrystals, and amorphous structures that can sustain an exact dimer ground state.

Acknowledgements: The work is supported by the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation) through Project-ID 258499086-SFB 1170 and the WürzburgDresden Cluster of Excellence on Complexity and Topology in Quantum Matter – ct.qmat Project-ID 390858490-EXC 2147.

[1] P. Ghosh, arXiv:2301.11331 (2023).
[2] T. Dotera, S. Bekku, and P. Ziherl, Nature Materials **16**, 987 (2017).
[3] B. S. Shastry and B. Sutherland, Physica B+C **108**, 1069 (1981)
[4] P. Ghosh, T. Müller, and R. Thomale, Phys. Rev. B **105**, L180412 (2022)



Search for long-range magnetic order in icosahedral quasicrystals

R. Tamura

[1]*Dept. of Mater. Sci. & Tech., Tokyo University of Science, Tokyo 125-8585, Japan*

[#]e-mail: tamura@rs.tus.ac.jp

Since the discovery of QCs in 1982 [1], one of the fundamental issues in the QC community is to realize long-range magnetic order (LRMO) in QCs. In this respect, the discoveries of rare-earth (*R*) containing QCs, such as Zn-Mg-*R* [2], Cd-Mg-*R* [3] and Cd-*R* [4] i QCs, opened up a new field to investigate the magnetism of localized moments embedded in a quasiperiodic crystal. However, all the *R* containing QCs commonly exhibited a spin-glass behavior and no long-range magnetic order was observed. On the contrary, Tsai-type 1/1 periodic approximants (PAs) were found to exhibit rich magnetic orders including ferromagnetic, antiferromagnetic and spin-glass states [5].

As a result of the extensive investigation on Tsai-type PAs for the last decade, it has been found that the magnetism of PAs is well controllable by the electron-per-atom (*e/a*) ratio [5]: The physical quantities such as the Weiss temperature (Θ_p) and Curie temperature (T_c) are well described by the *e/a* ratio. In addition, the study of the magnetism in the composition-tunable Tsai-type 1/1 PAs has enabled us to understand and also predict the magnetism of Tsai-type QCs to considerable extent. The absence of the LRMO in the previously studied QCs is now understood by the fact that their *e/a* ratio happens to be located in the strongly frustrating region. Recently, new Au-Ga-*R* QCs were synthesized in the ferromagnetic region by the melt-spinning method and they were found to exhibit ferromagnetic transitions as expected [6], which is the first LRMO observed in *i*-QCs. In this presentation, we will also show our recent work [7] on the synthesis of a high-phase purity ferromagnetic Au-Ga-*R* QC and discuss its bulk magnetic properties in detail.

Acknowledgements: This work was supported by Japan Society for the Promotion of Science through Grants-in-Aid for Scientific Research Grants No. JP19H05817, JP19H05818, JP21H01044 and JST, CREST Grant No. JPMJCR22O3, Japan.

[1] D. Shechtman, *et al*., Phys. Rev. Lett. **53** (1984) 1951.
[2] Z. Luo, *et al*., Scr. Metall. Mater. **28** (1993) 1513.
[3] J. Guo *et al*., Jpn. J. Appl. Phys. **39** (2000) L770.
[4] A. I. Goldman, *et al*., Nat. Mater. **12** (2013) 714.
[5] S. Suzuki *et al*., Mater. Trans. **62** (2021) 298.
[6] R. Tamura *et al*., J. Am. Chem. Soc. **143** (2021) 19938.
[7] R. Takeuchi, *et al*., Phys. Rev. Lett. in press.