Topological spin moiré: stability, dynamics, and controllability

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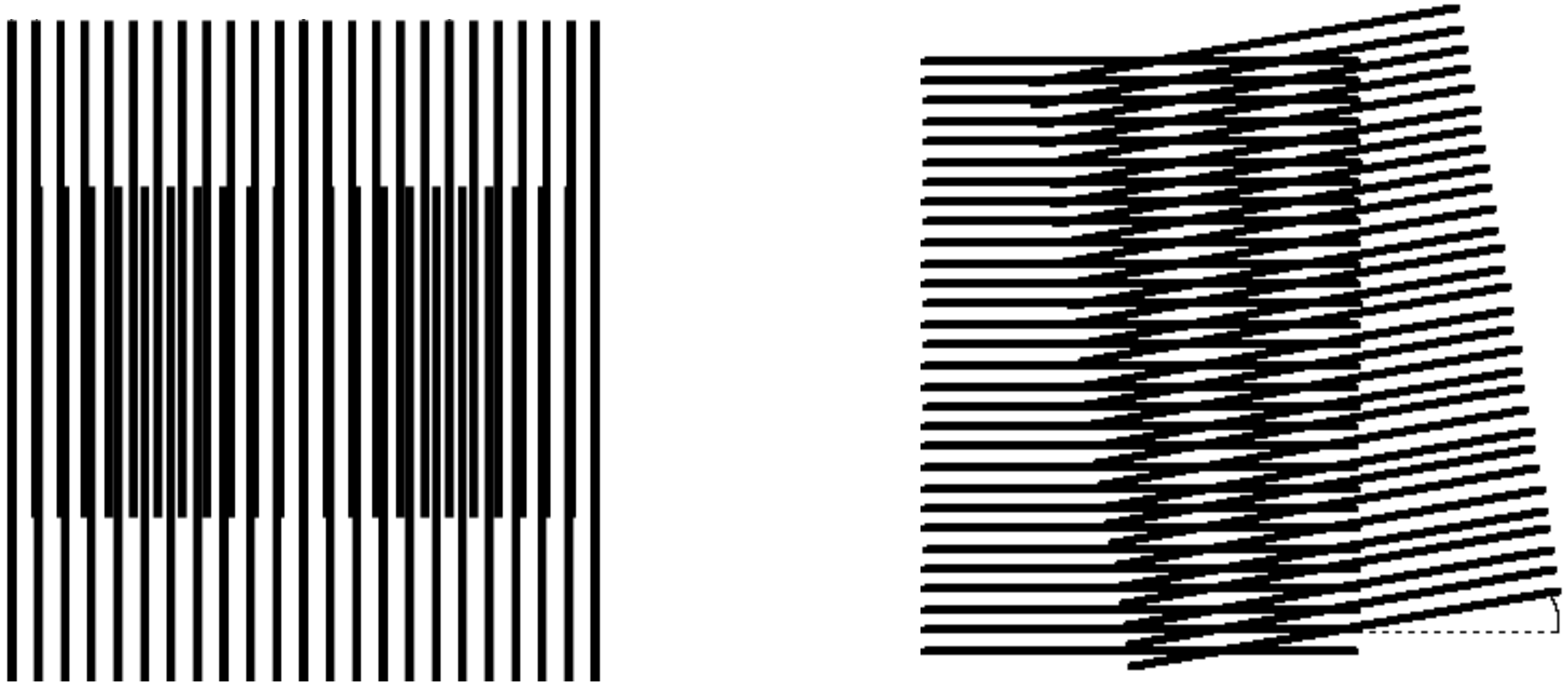
Plan of this talk

- spin moiré
  - superstructure, topology, and emergent electromagnetic field
  - spin moiré engineering: type, number, amplitude of waves, twist angle, phase shift
  - complete topological phase diagram for 2D skyrmions and 3D torons
- itinerant frustration
  - localized spin systems vs itinerant electron systems
  - effective long-range/multiple spin interactions by the Fermi surface effects
  - applications to 2D skyrmion crystals and 3D toron crystals

spin moiré

Moiré

- interference pattern generated by a superposition of multiple waves



figures are taken from Wikipedia

Superstructures are flexibly controlled by many parameters:

number of superposed waves, pitches, amplitudes, relative phases, twist angles, etc.

Moiré in condensed matter physics

- moiré superstructures in twisted 2D van der Waals materials

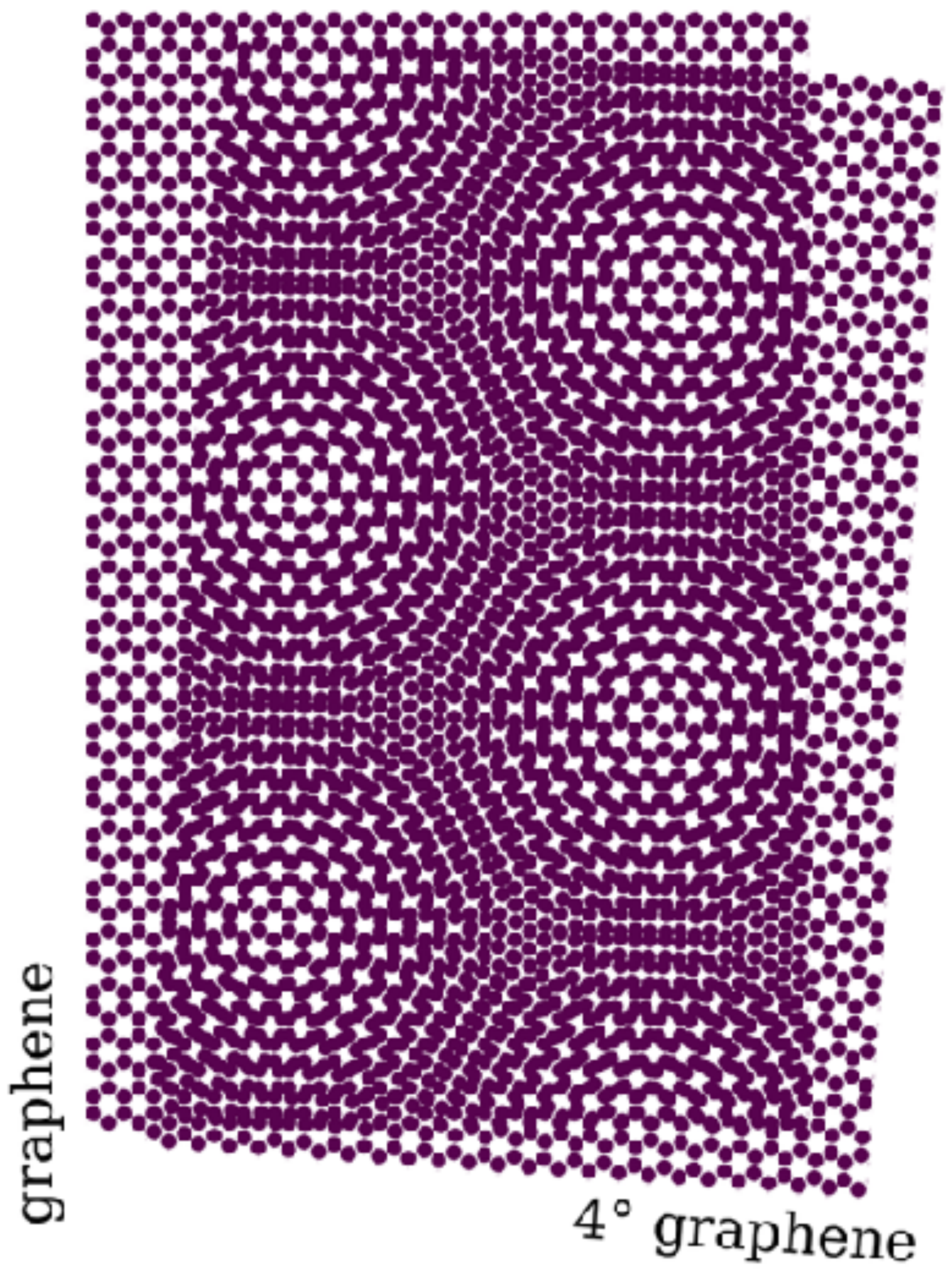
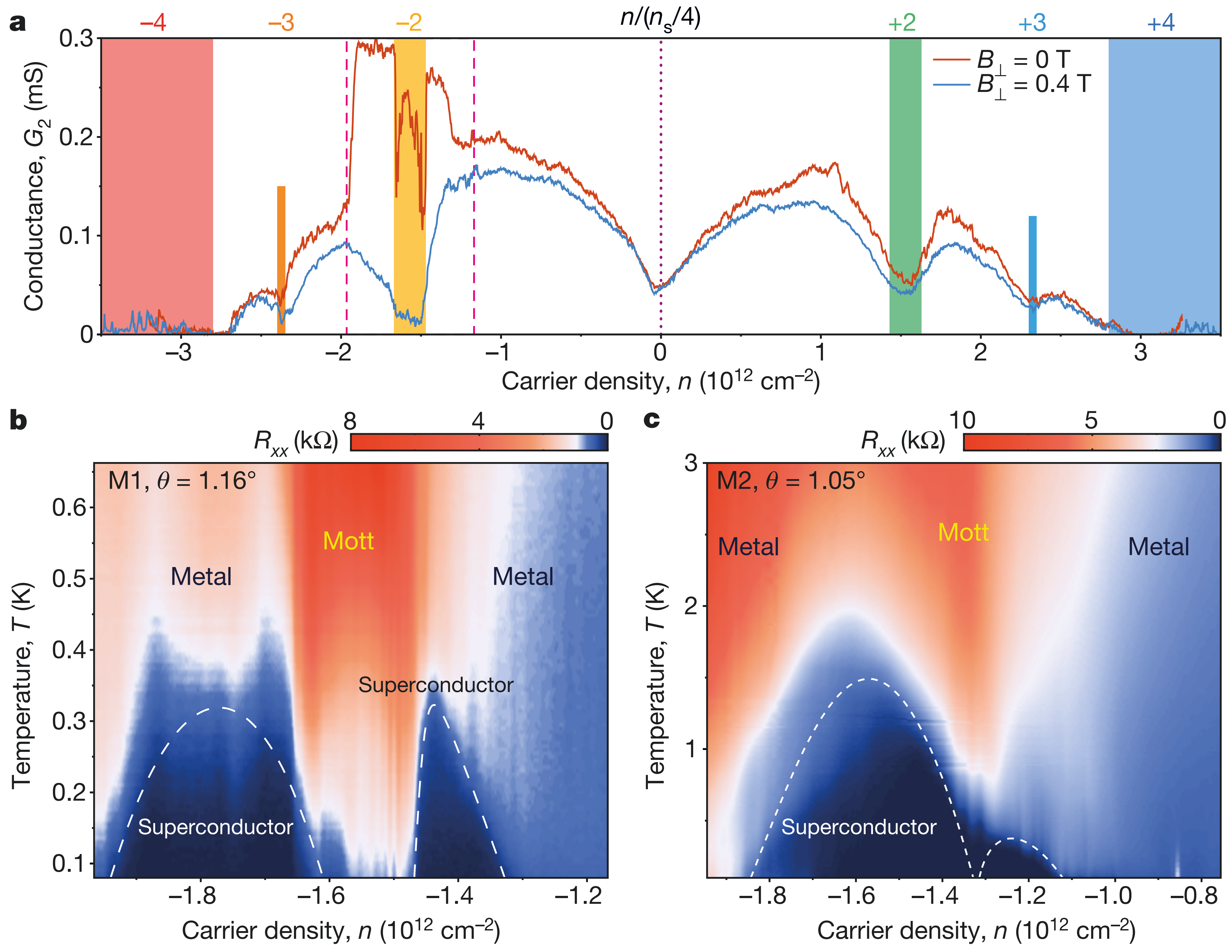


figure is taken from Wikipedia

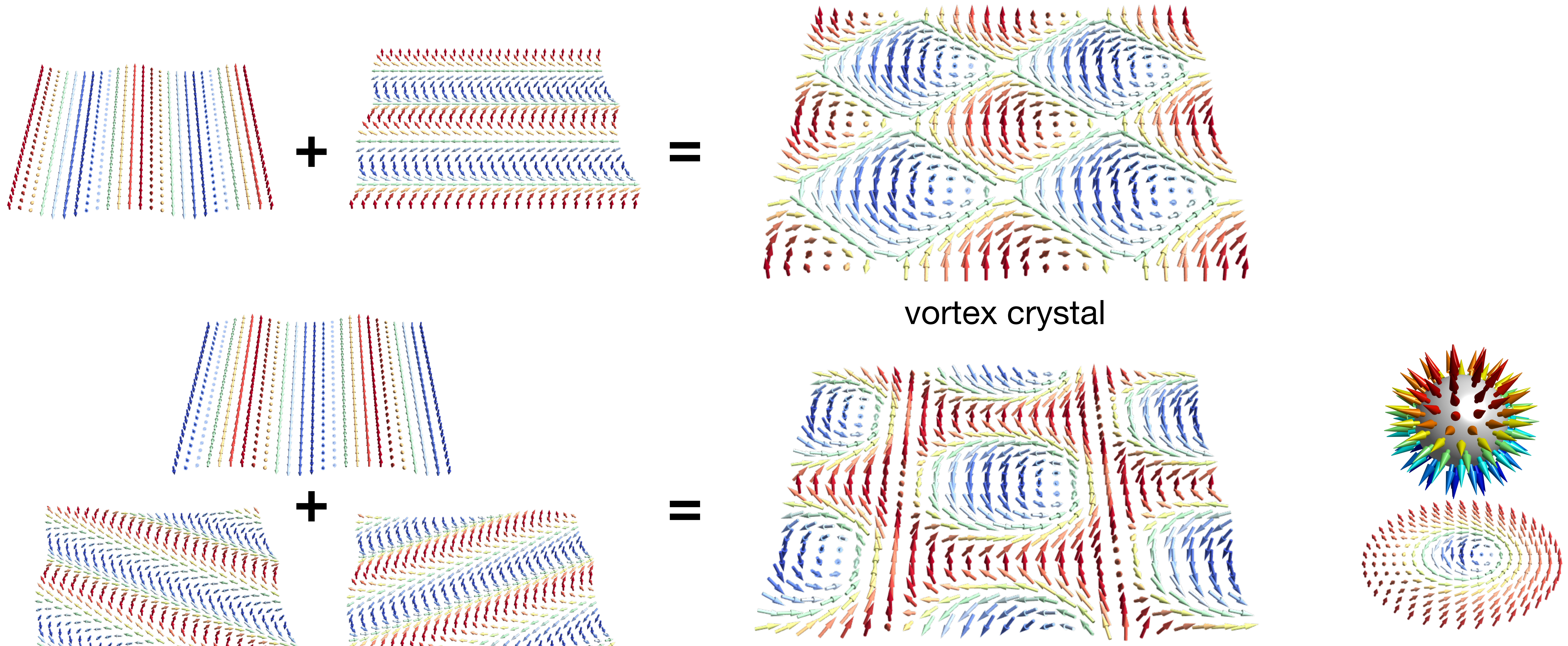


Y. Cao *et al.*, Nature **556**, 43 (2018)

twistronics by modulated flat band and electron correlation in moiré superstructures

Spin moiré

- interference pattern generated by a superposition of spin density waves

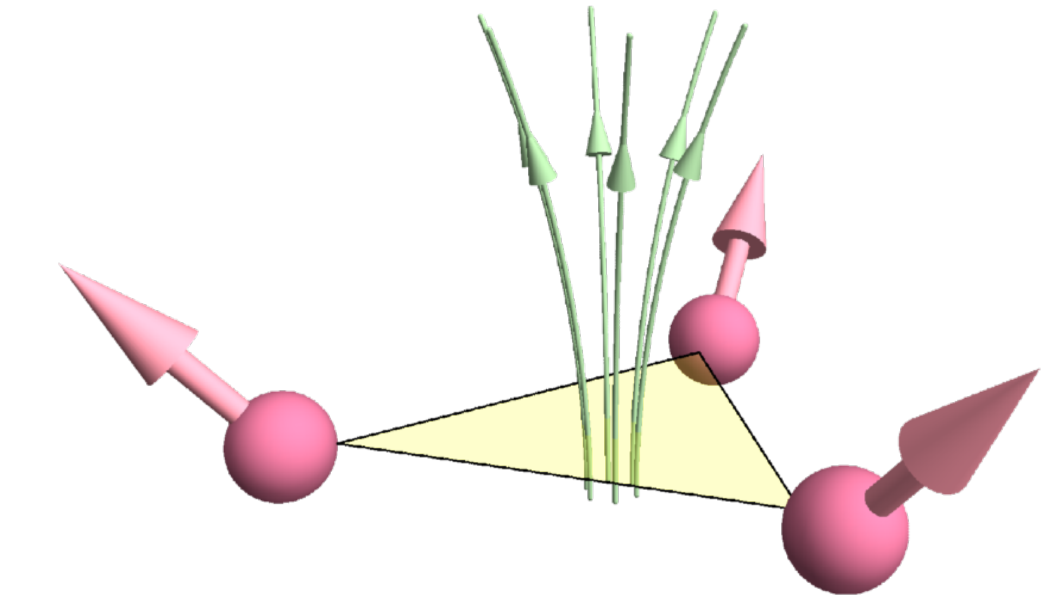


$$N_{\rm sk} = \frac{1}{4\pi}\int d^2\mathbf{r}\, \mathbf{n}\cdot(\partial_x \mathbf{n}\times\partial_y \mathbf{n})$$

- richer variety of textures due to the vector fields
- richer physics due to the topological properties

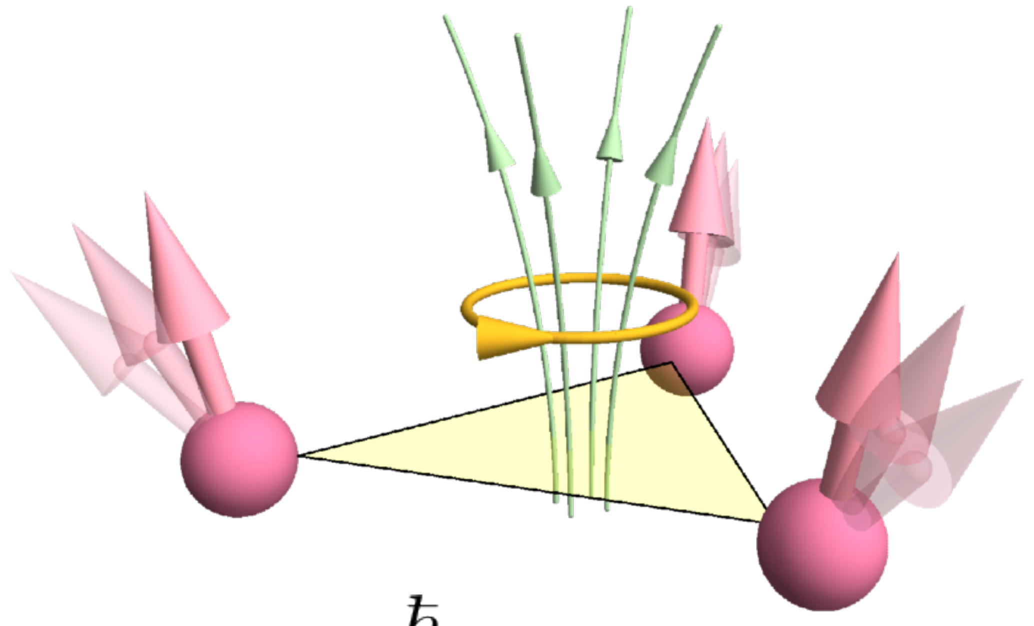
Emergent electromagnetic field

◉ emergent magnetic field

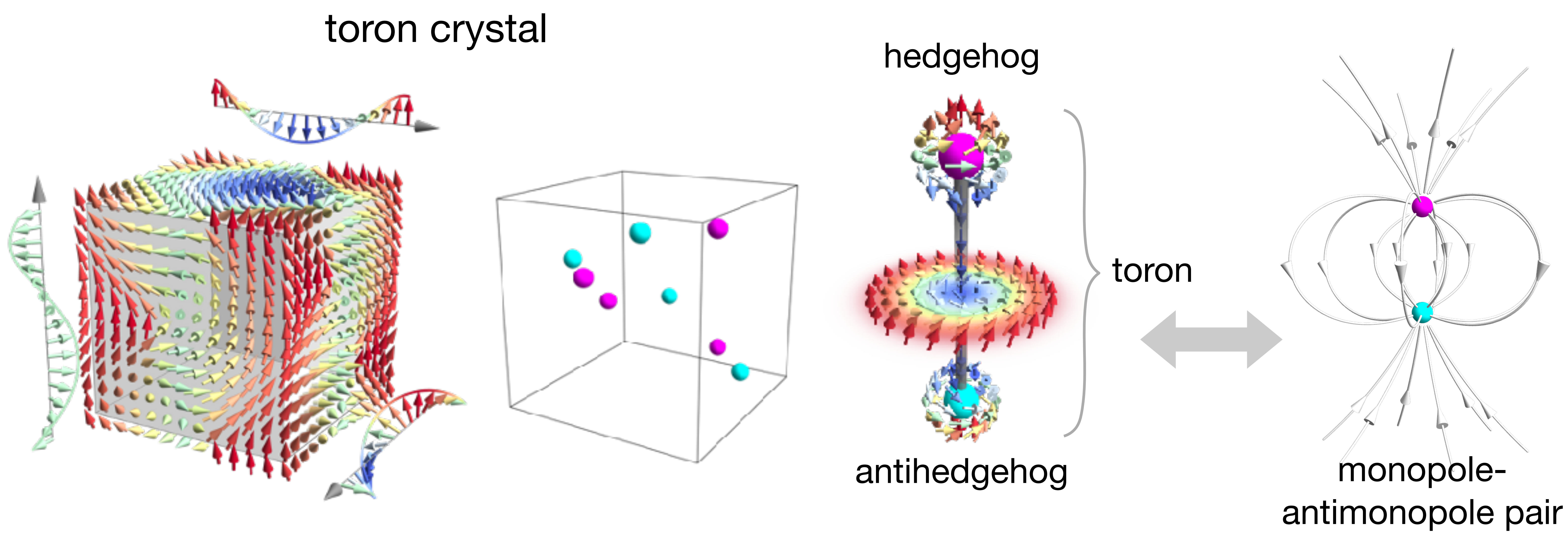


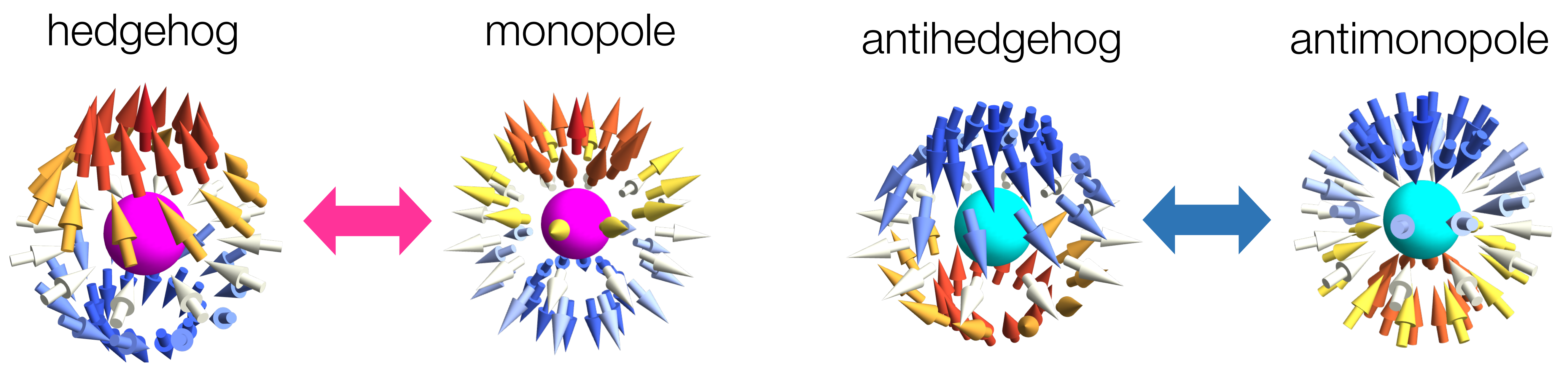
$$B_i^{\mathrm{em}} = \frac{\hbar}{2|e|}\frac{1}{2}\varepsilon^{ijk}\mathbf{n}\cdot(\partial_j\mathbf{n}\times\partial_k\mathbf{n})$$

◉ emergent electric field



$$E_i^{\mathrm{em}} = \frac{\hbar}{2|e|}\mathbf{n}\cdot(\partial_i\mathbf{n}\times\partial_t\mathbf{n})$$



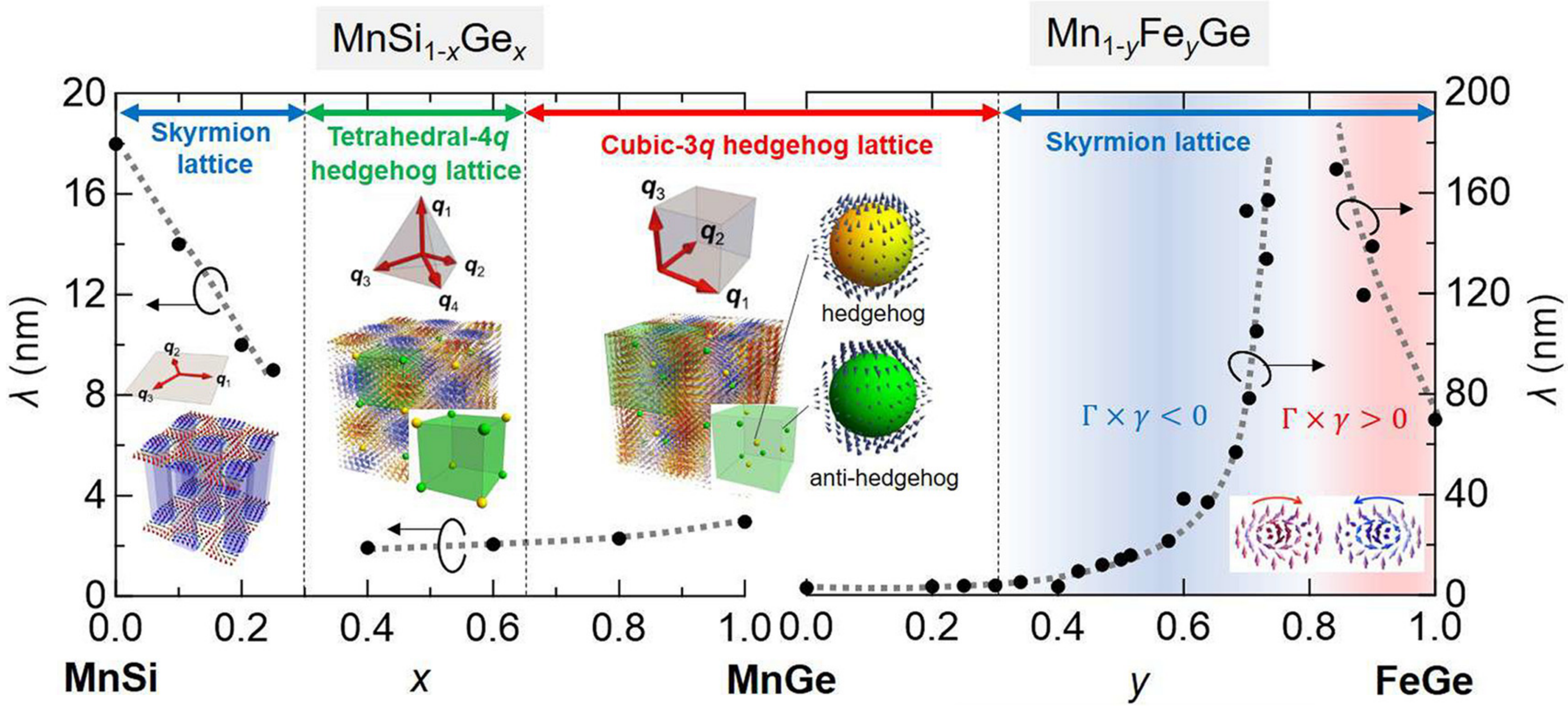


◉ Spin moiré harbors a superstructure of emergent electric and magnetic fields.

➡ interesting magnetic, electric, optical, and transport phenomena

Experimental realization

- Skyrmion and toron crystals have been found in many materials, e.g., in B20 compounds.



Y. Fujishiro, N. Kanazawa, and Y. Tokura, Appl. Phys. Lett. **116**, 090501 (2020)

Motivation-1

- advantages of spin moiré, compare to the structural moiré in twisted 2D materials
  - more variety of moiré patterns owing to vector (spinor) fields
  - nontrivial topology
  - emergent electromagnetic fields, leading to exotic quantum phenomena
  - possible to make more than two-dimensional moiré
  - possible to control by external fields, like magnetic field, pressure, and temperature

Based on the spin moiré picture for multiple spin density waves,

- to investigate the effect of modulations of moiré parameters
- to explore spin textures that have been overlooked in the previous studies
- to systematically elucidate possible phase transitions in both magnetic and topological aspects

Kotaro Shimizu

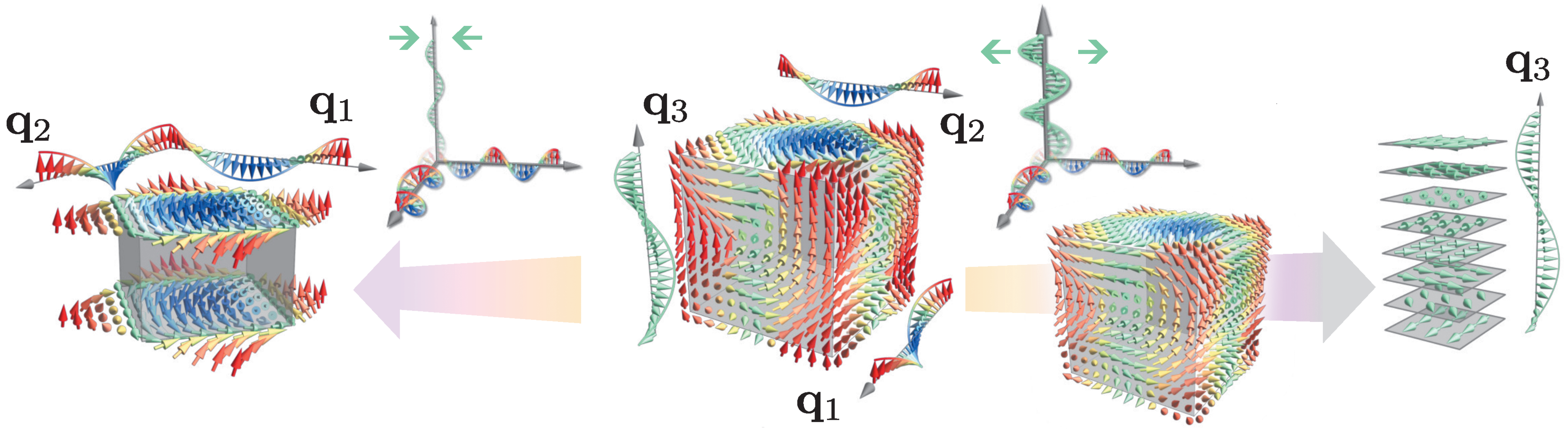
Spin moiré engineering

◉ various ways of moiré modulations

- types of waves (ex. helix or sinusoidal)

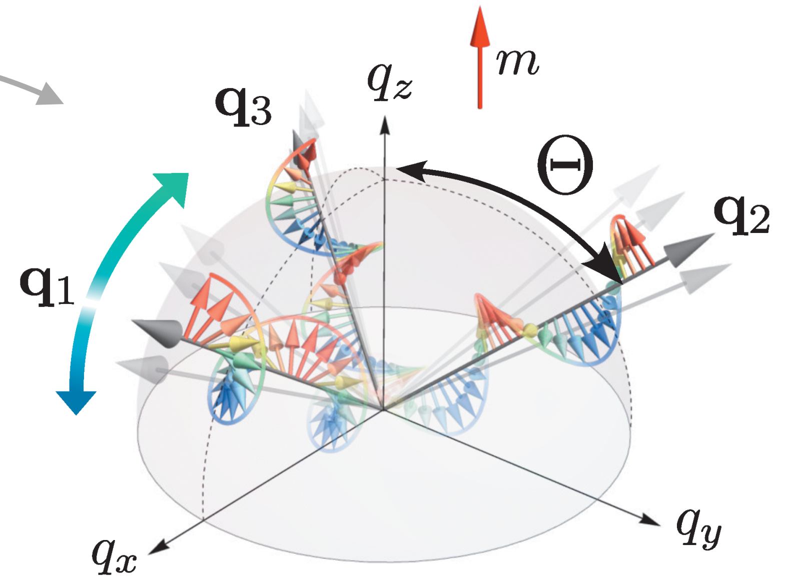
- number of waves
- amplitude of waves

K. Shimizu, S. Okumura, Y. Kato, and Y. Motome, Phys. Rev. B **103**, 054427 (2021)



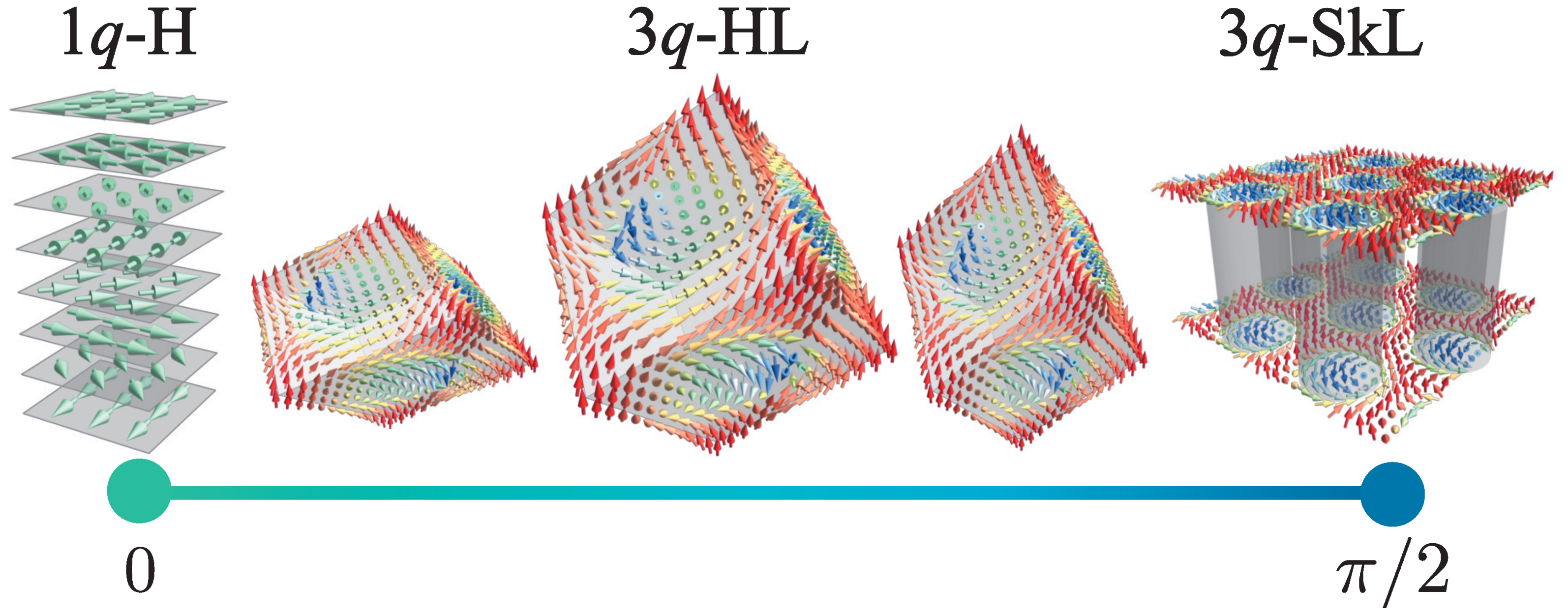
- angle between propagation directions

K. Shimizu, S. Okumura, Y. Kato, and Y. Motome, Phys. Rev. B **103**, 184421 (2021)



- relative phases of waves

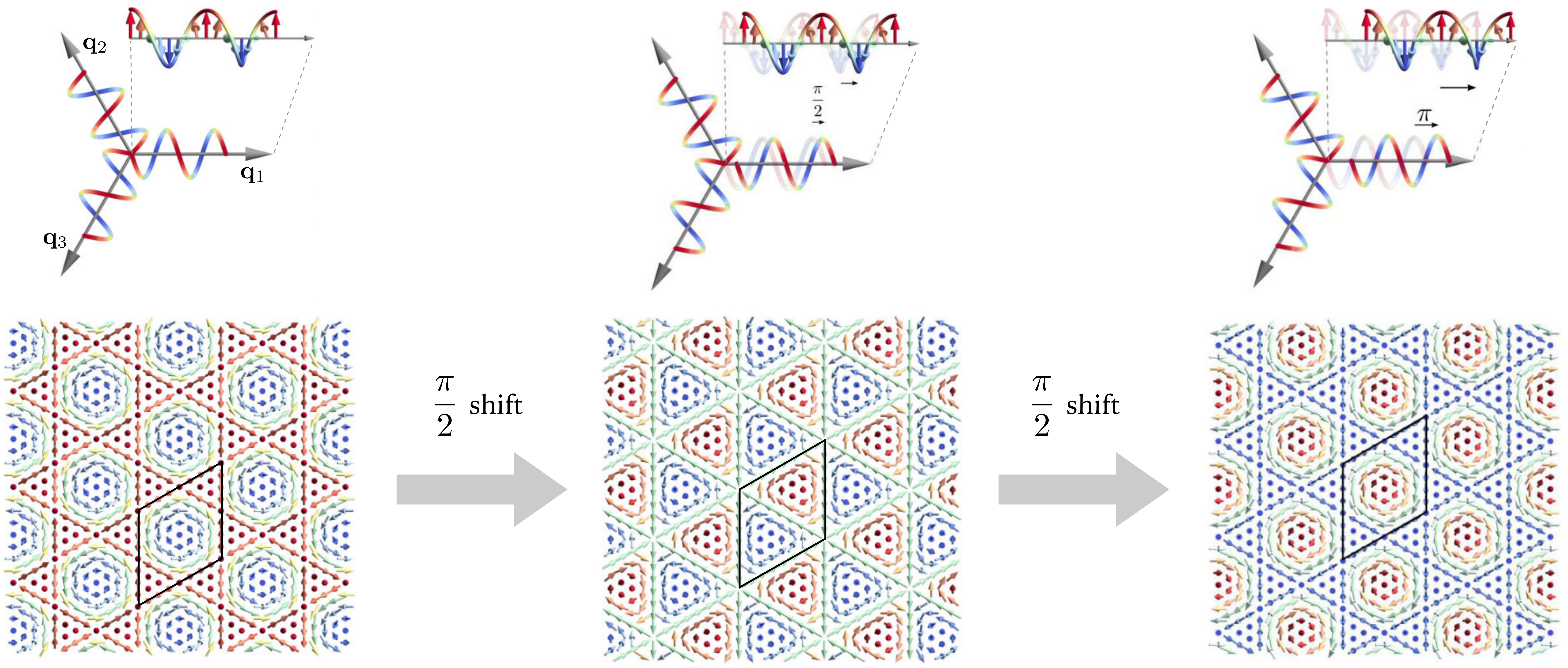
K. Shimizu, S. Okumura, Y. Kato, and Y. Motome, Phys. Rev. B **105**, 224405 (2022)



- etc.

rich variety of magnetic and topological properties, quantum transport, and optical properties

Phase shift in spin moiré



skyrmion crystal with $N_{\rm sk}$=+1 meron-antimeron crystal with $N_{\rm sk}$=0 skyrmion crystal with $N_{\rm sk}$=-1

T. Kurumaji *et al.*, Science **365**, 914 (2019); S. Hayami, T. Okubo, and Y. Motome, Nat. Commun. **12**, 6927 (2021)

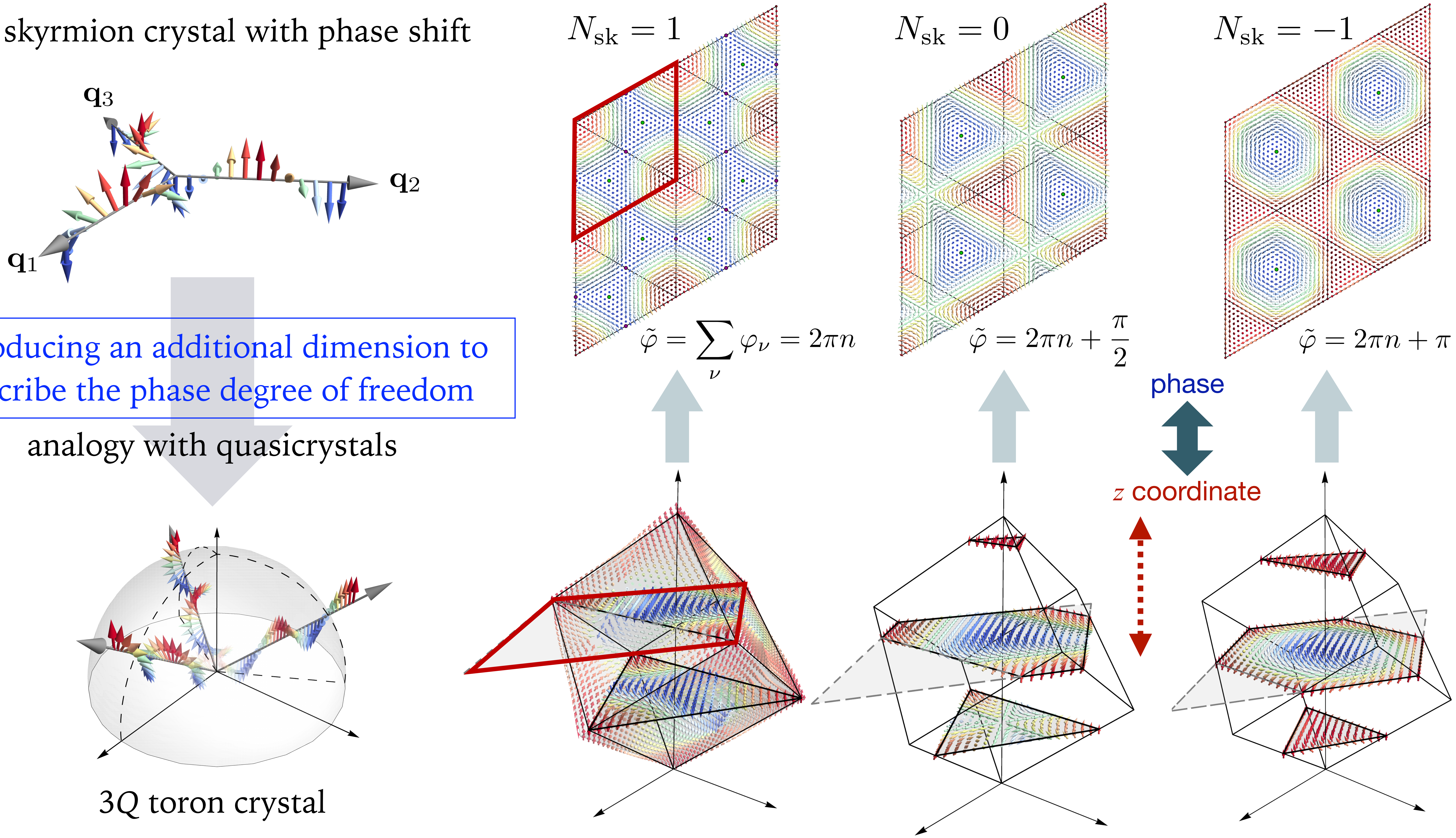
How to investigate the effect of phase shift systematically?

Hyperspace representation

3Q skyrmion crystal with phase shift

introducing an additional dimension to describe the phase degree of freedom

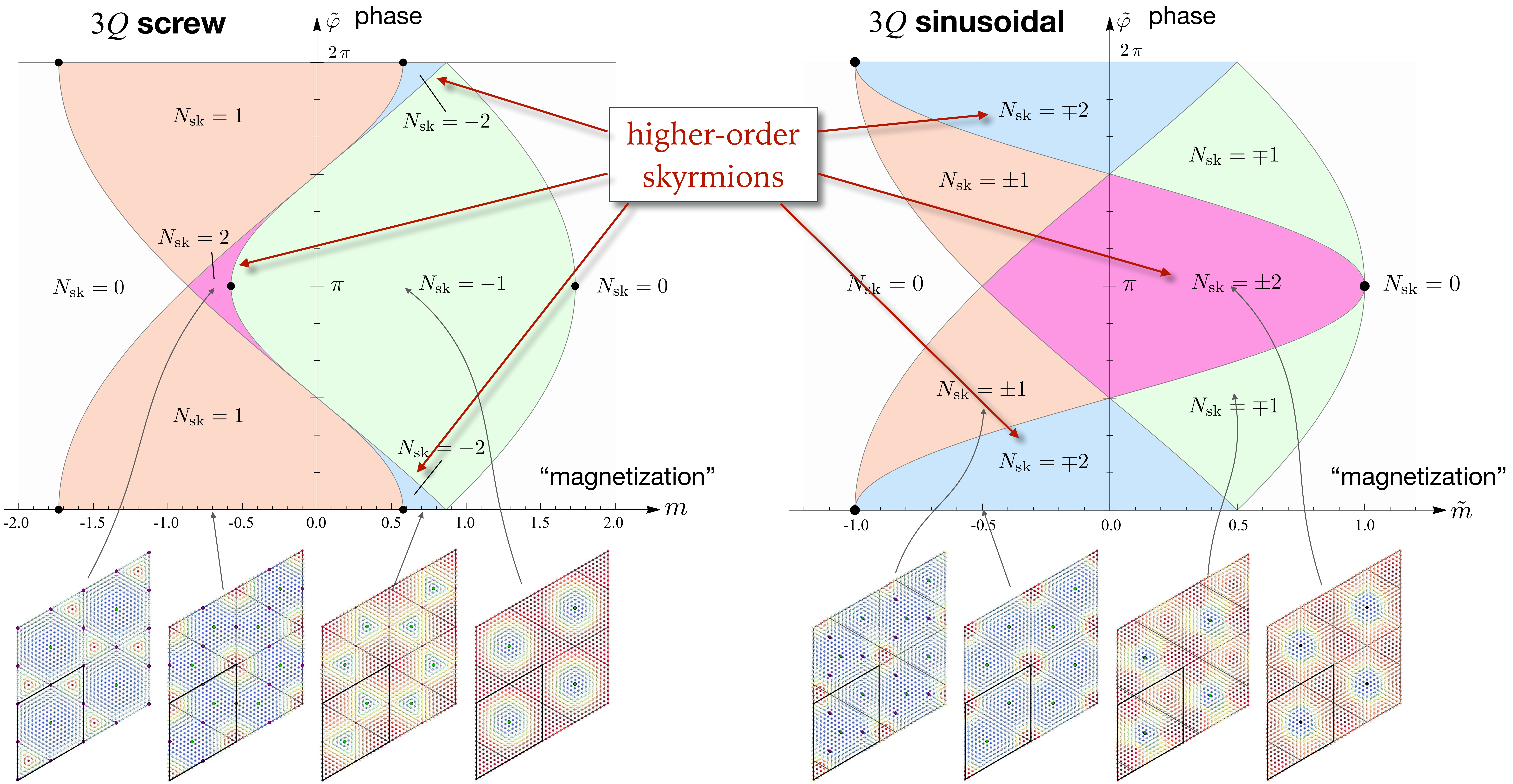
analogy with quasicrystals



3Q toron crystal

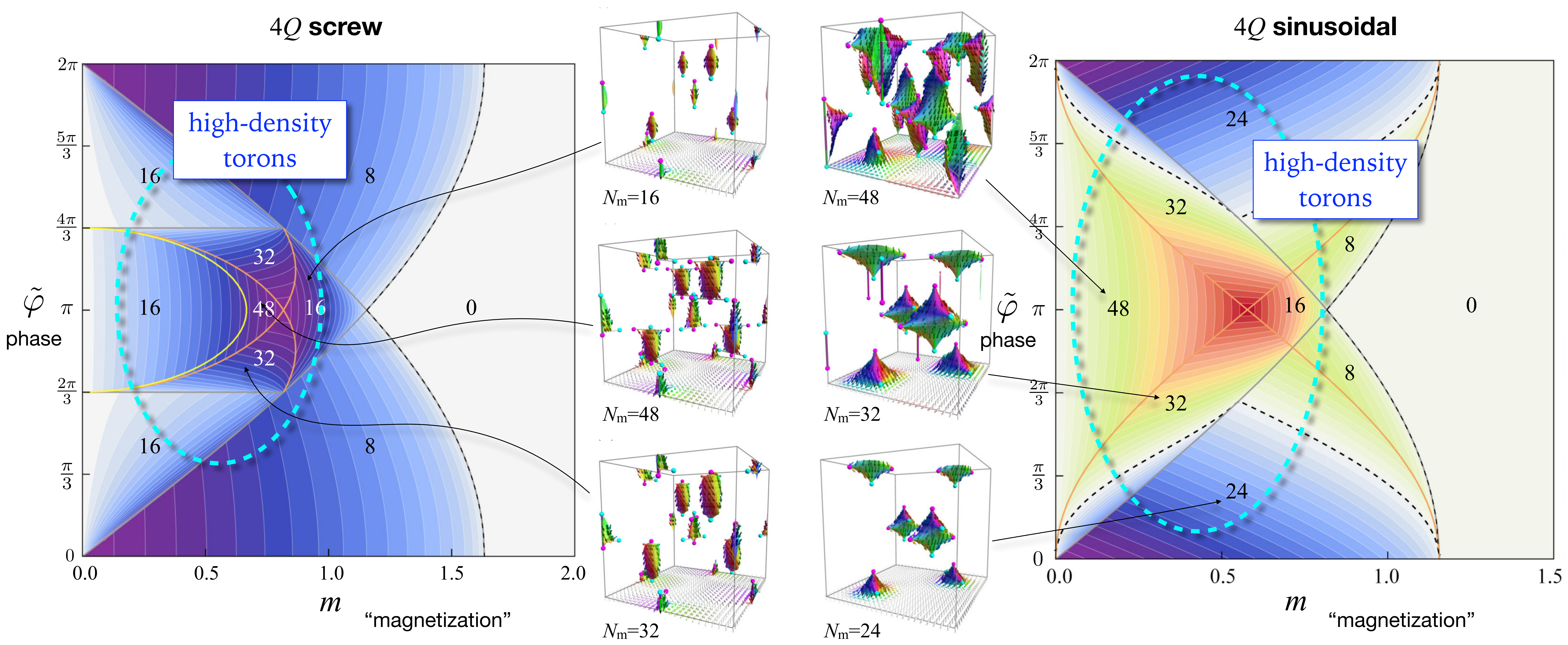
K. Shimizu, S. Okumura, Y. Kato, and Y. Motome, Phys. Rev. B **105**, 224405 (2022)

Topological phase diagram for 2D skyrmions



K. Shimizu, S. Okumura, Y. Kato, and Y. Motome, Phys. Rev. B **105**, 224405 (2022)

Topological phase diagram for 3D torons



K. Shimizu, S. Okumura, Y. Kato, and Y. Motome, Phys. Rev. B **105**, 224405 (2022)

Short summary

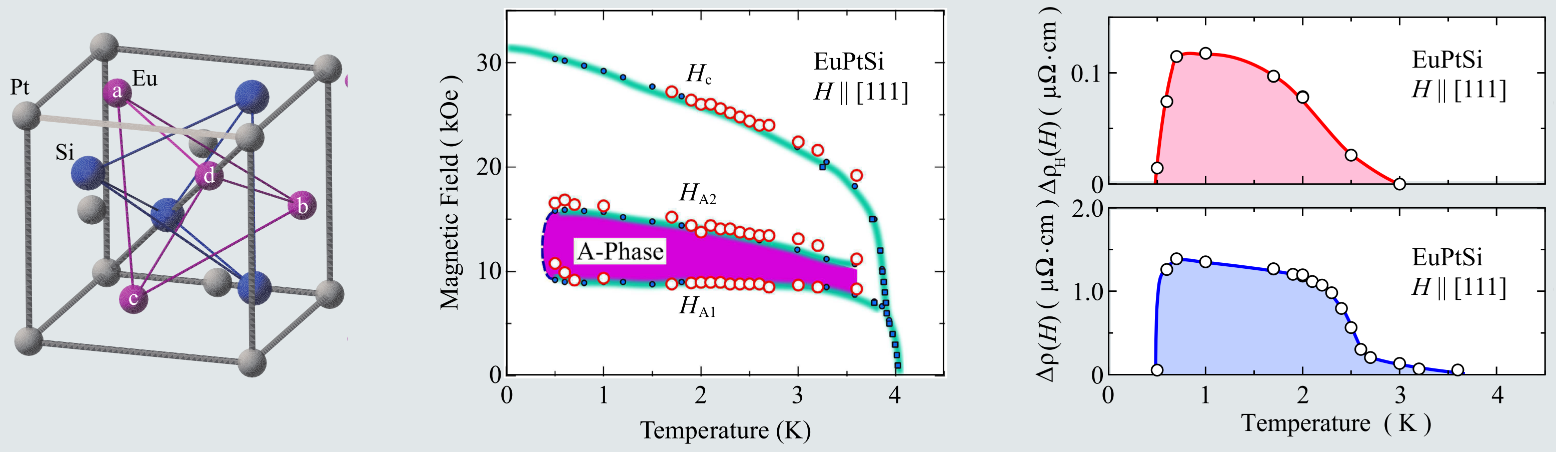
- The spin moiré picture is useful to explore more variety of topological spin crystals.
  - We can exploit the analogy with conventional moiré.
  - There are many advantages of spin moiré, compare to the structural moiré in twisted 2D materials.

- We demonstrated the usefulness of the spin moiré picture for the phase shift in topological spin crystals.
  - complete topological phase diagrams for 2D skyrmions and 3D torons
  - unprecedented topological phases with higher-order skyrmions and high-density torons

itinerant frustration

New generation: nanometer-scale skymions

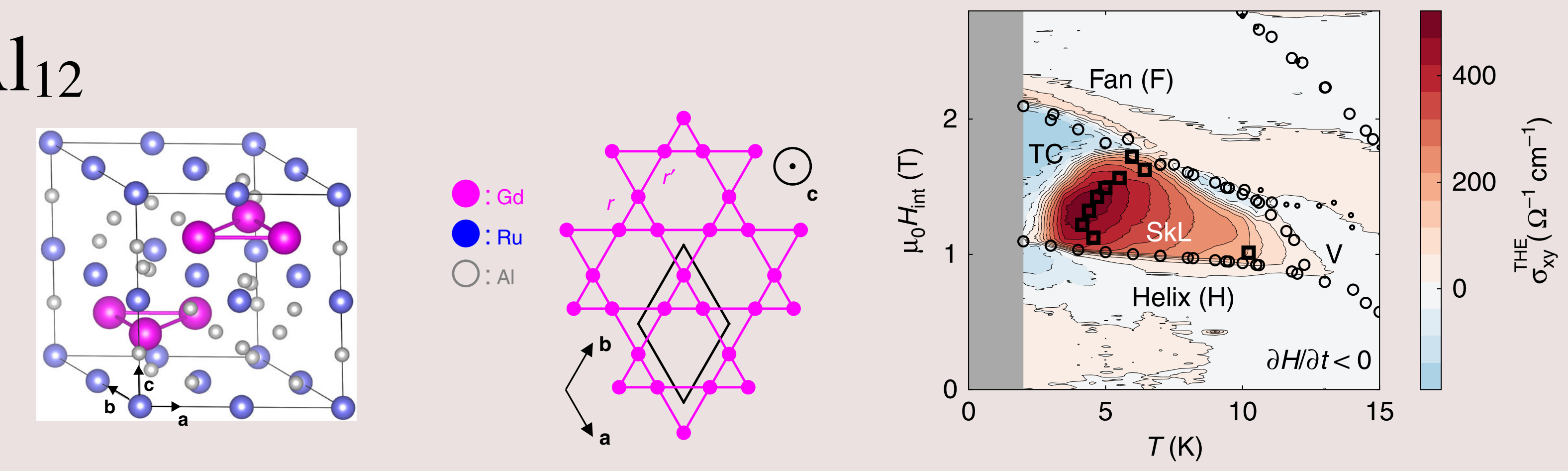
EuPtSi



noncentrosymmetric

M. Kakihana *et al.*, J. Phys. Soc. Jpn. **87**, 023701 (2017)

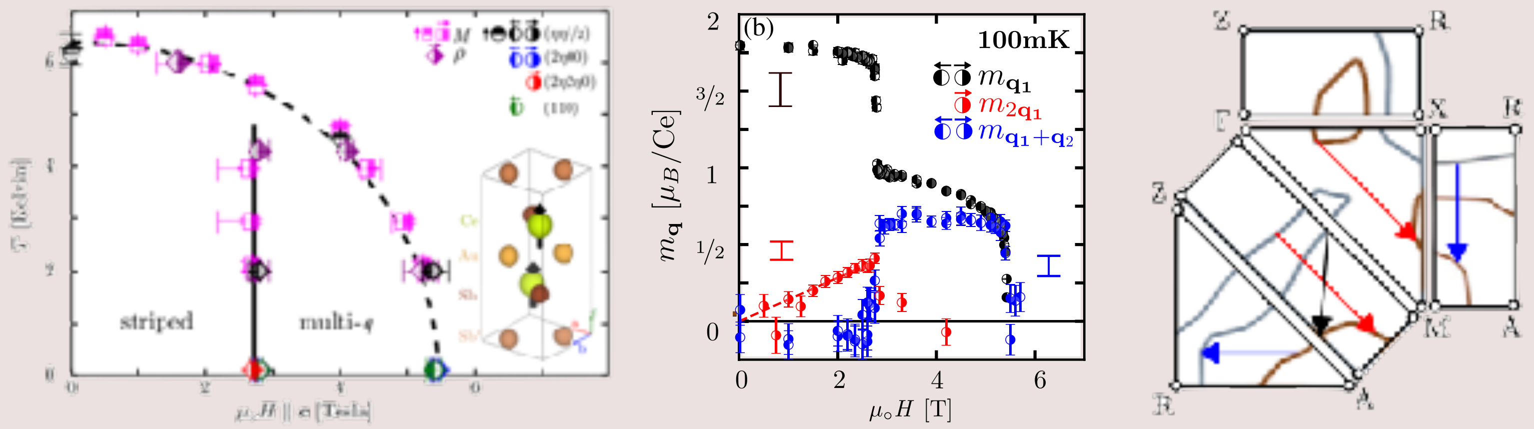
$Gd_3Ru_4Al_{12}$



M. Hirschberger *et al.*, Nat. Commun. **10**, 5831 (2019)

centrosymmetric

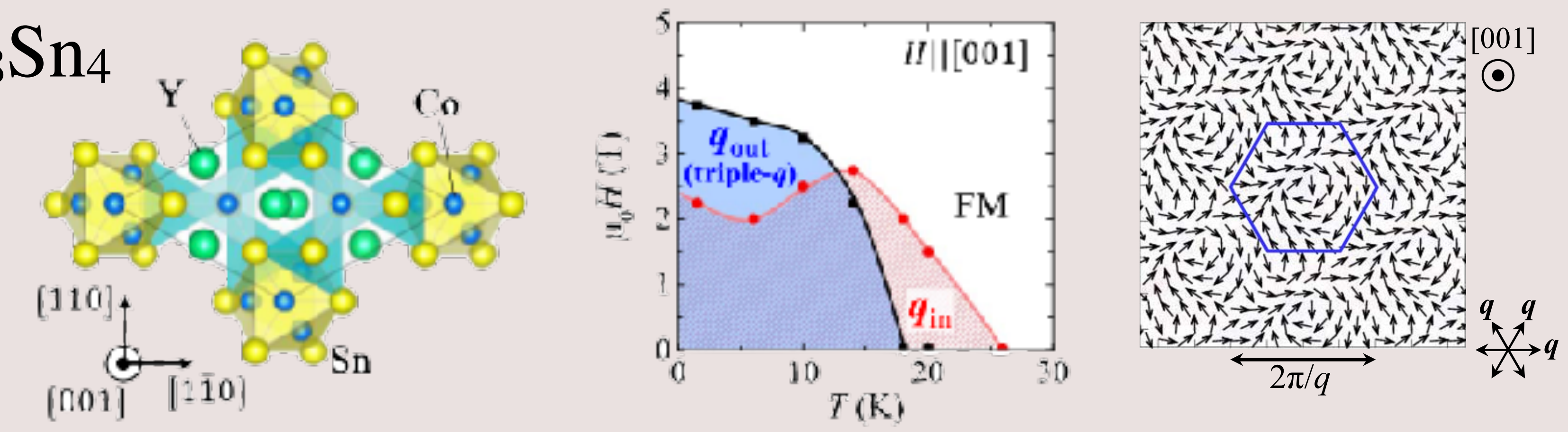
$CeAuSb_2$



centrosymmetric

G. G. Marcus *et al.*, Phys. Rev. Lett. **120**, 097201 (2018)

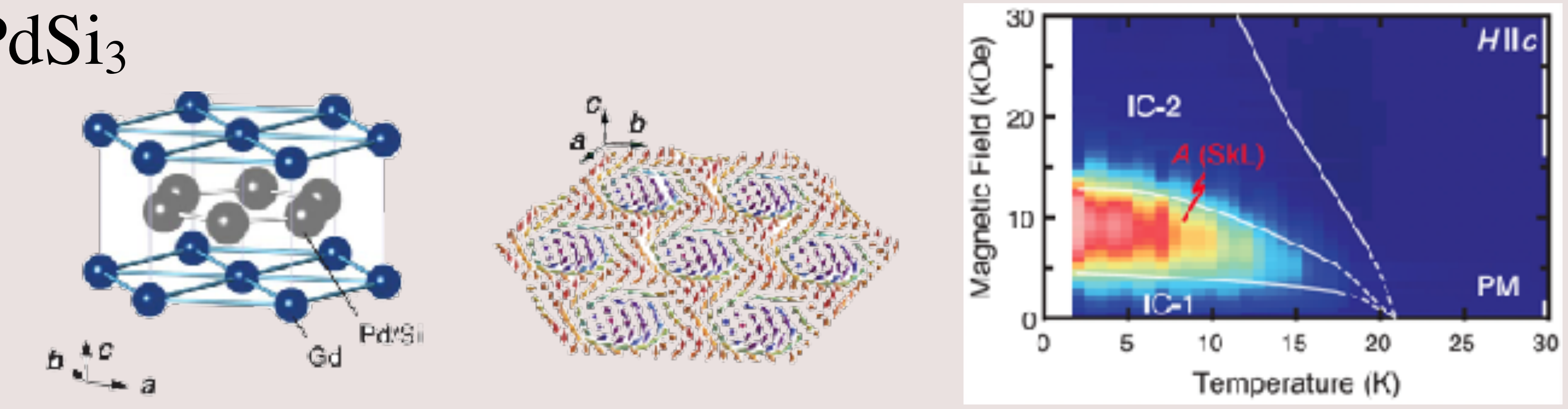
$Y_3Co_8Sn_4$



R. Takagi *et al.*, Sci. Adv. **4**, eaau3402 (2019)

centrosymmetric

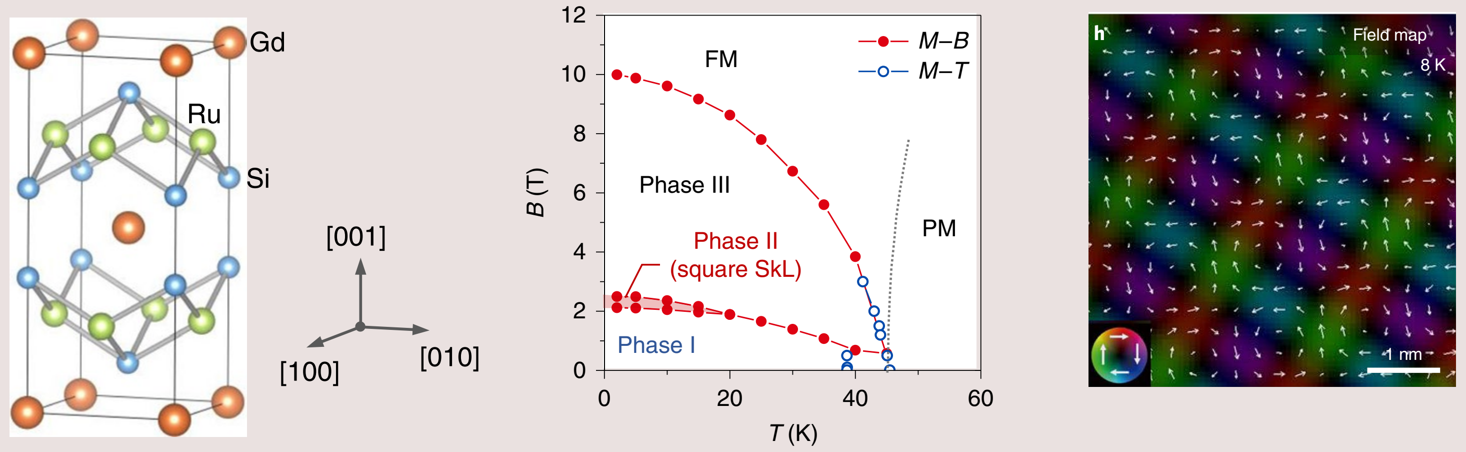
Gd_2PdSi_3



centrosymmetric

T. Kurumaji *et al.*, Science **365**, 914 (2019)

$GdRu_2Si_2$

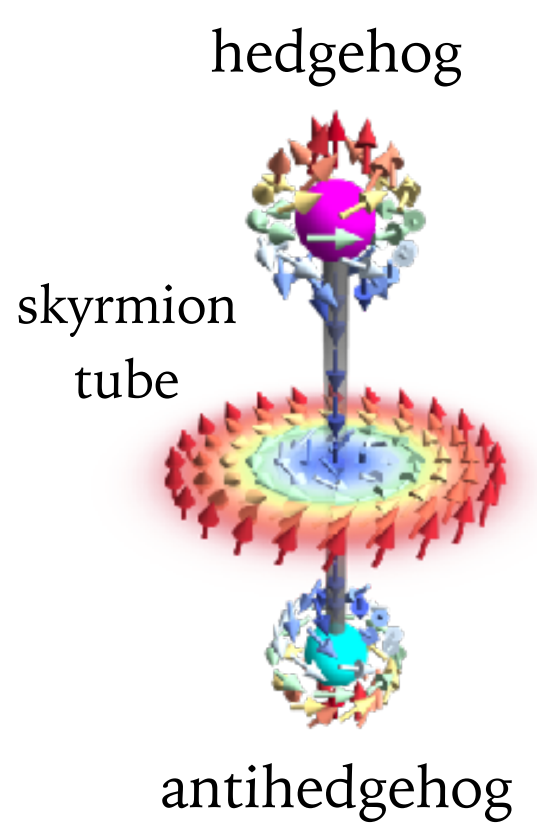


N. D. Khanh *et al.*, Nat. Nanotech. **15**, 444 (2020)

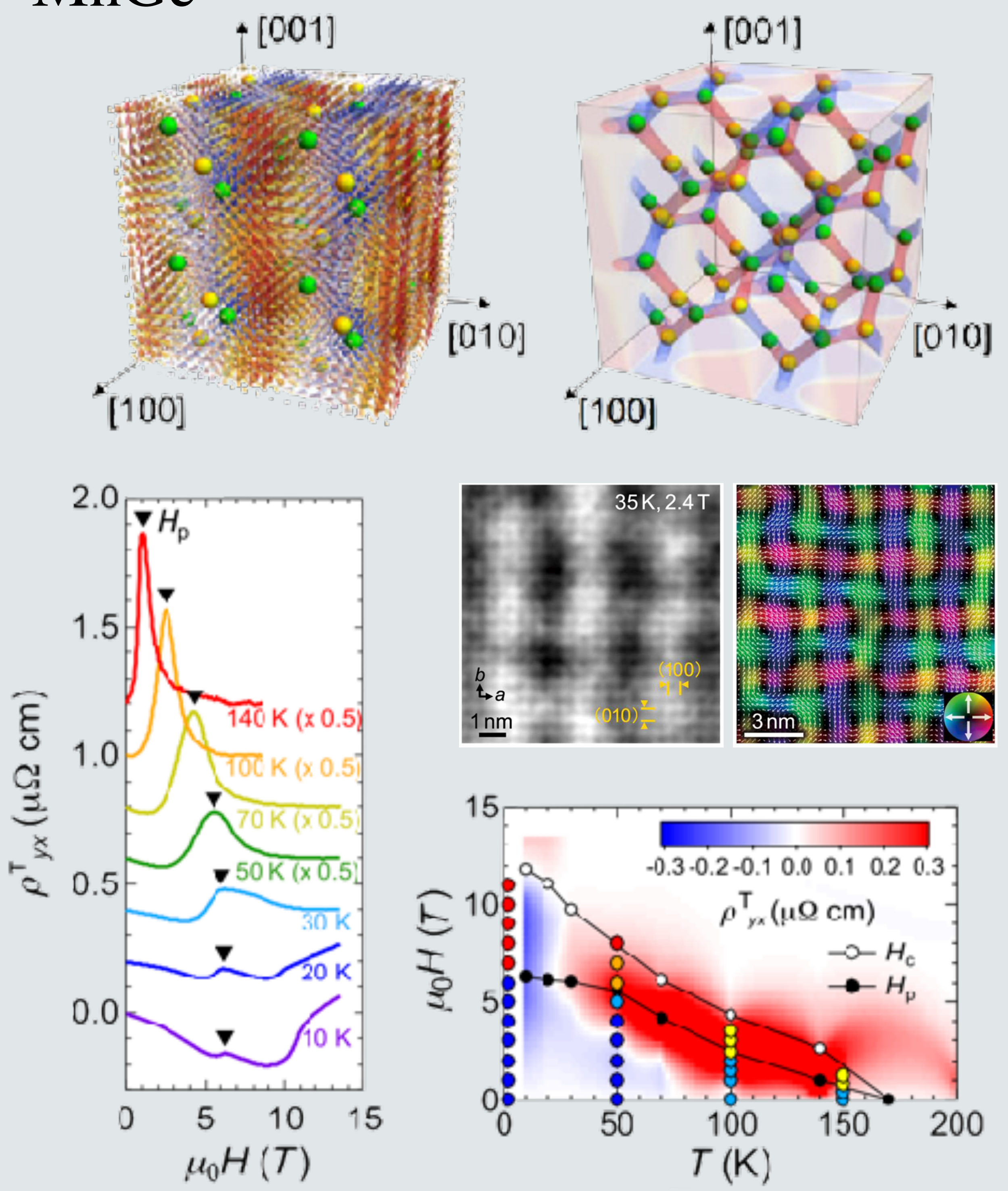
centrosymmetric

extremely short period ~ few nm, in not only noncentrosymmetric but also centrosymmetric systems

New generation: nanometer-scale torons

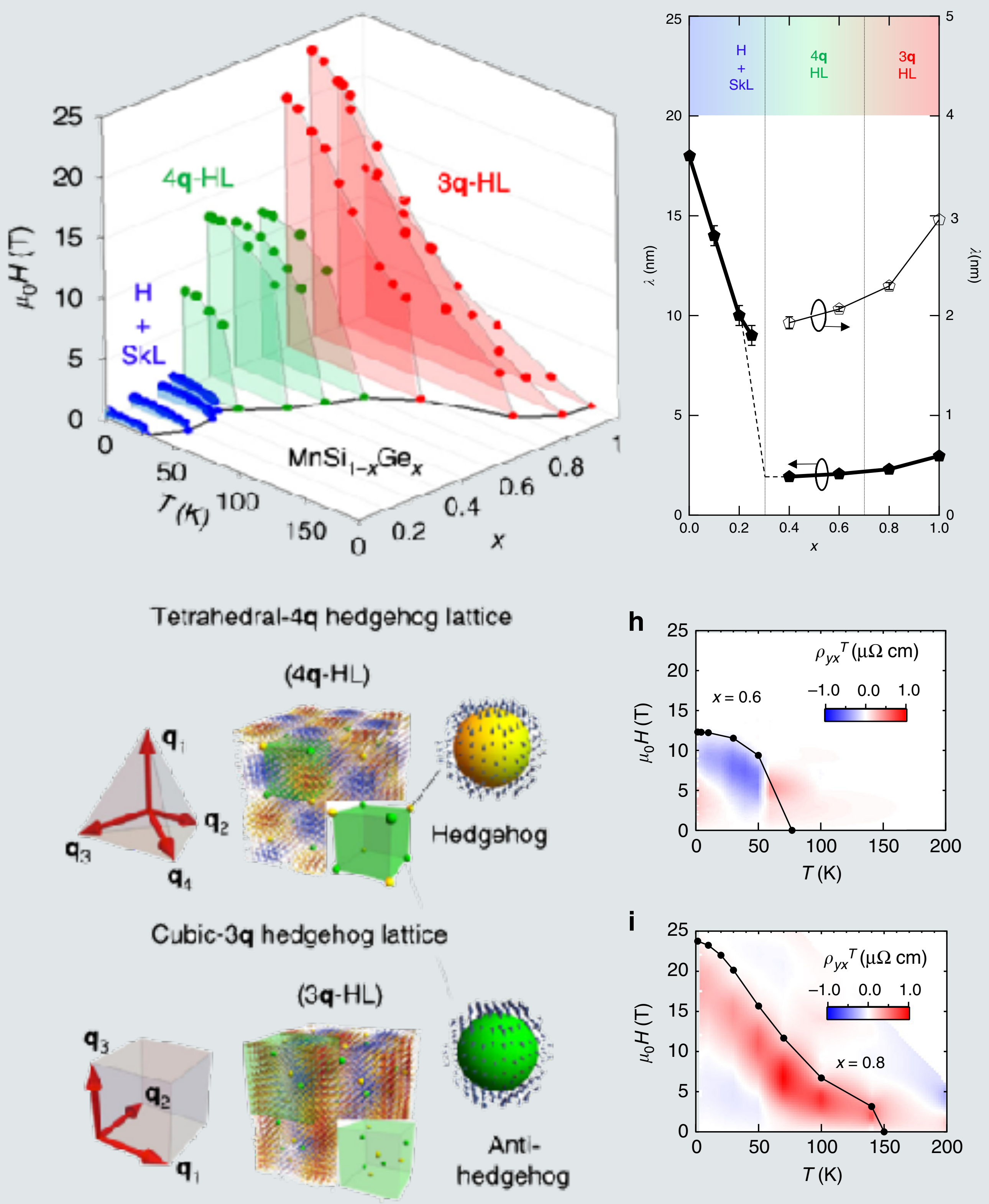


MnGe — noncentrosymmetric



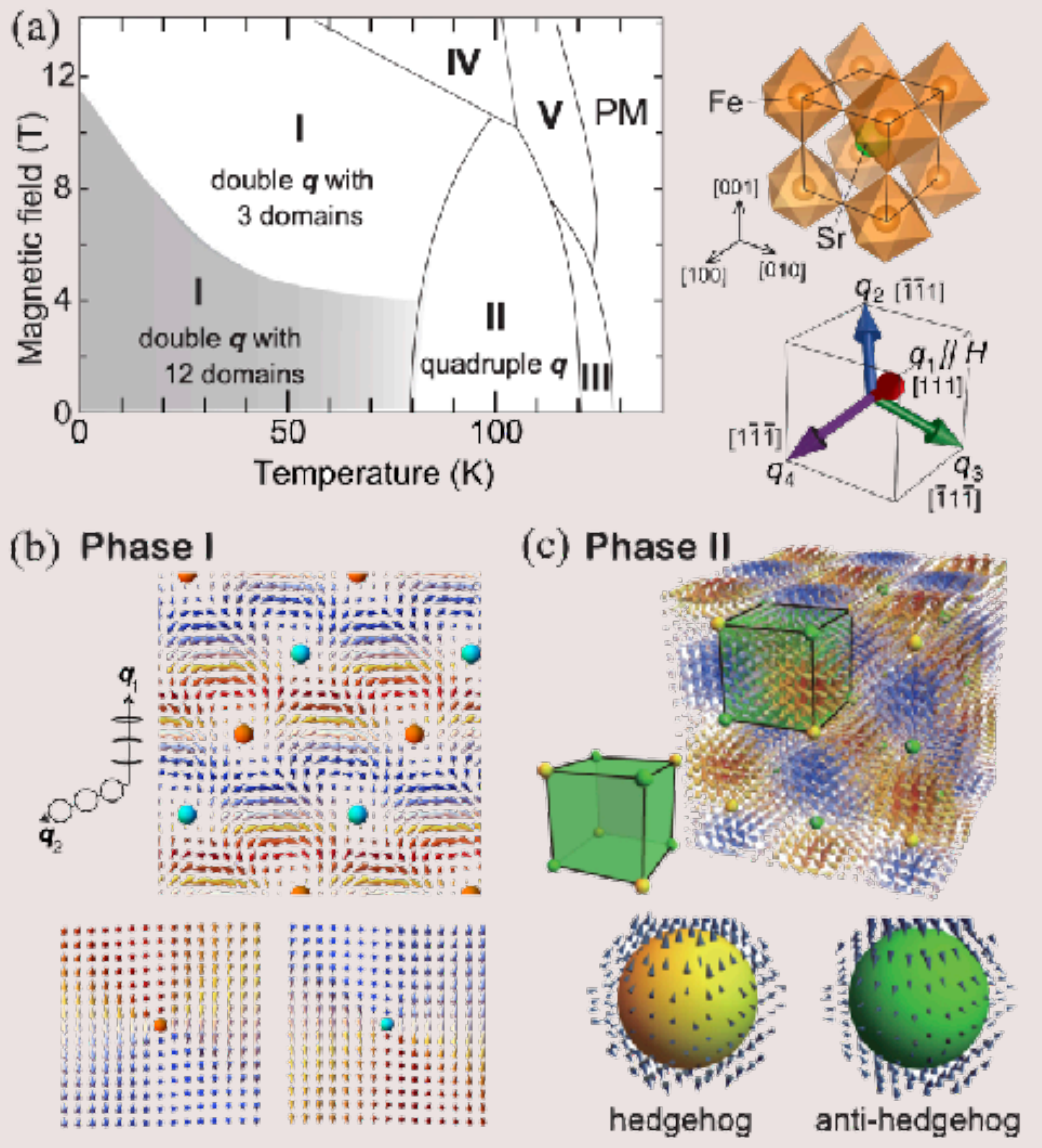
N. Kanazawa *et al.*, Phys. Rev. B **86**, 134425 (2012)
T. Tanigaki *et al.*, Nano Lett. **15**, 5438 (2015)
N. Kanazawa *et al.*, Phys. Rev. Lett. **125**, 137202 (2020), …

$MnSi_{1-x}Ge_x$ — noncentrosymmetric



Y. Fujishiro *et al.*, Nat. Commun. **10**, 1059 (2019)

$SrFeO_3$ — centrosymmetric



S. Ishiwata *et al.*, Phys. Rev B **84**, 054427 (2011)
S. Ishiwata *et al.*, Phys. Rev B **101**, 134406 (2020)

extremely short period ~ few nm, in not only noncentrosymmetric but also centrosymmetric systems

Motivation-2

- ◉ What is the stabilization mechanism for the new-generation topological spin crystals?
  - Conventional mechanism based on the Dzyaloshinskii-Moriya interaction does not explain the extremely short-period textures in centrosymmetric systems.
  - In general, magnetic frustration is active and able to stabilize short-period spin textures even in centrosymmetric systems in the absence of spin-orbit coupling, but it is discussed mostly for insulating systems.

➡ What is a generic mechanism in metallic magnets? What is the role of itinerant electrons?

our proposal: ***itinerant frustration***
as an underlying mechanism to generate frustrated/multiple-spin interactions

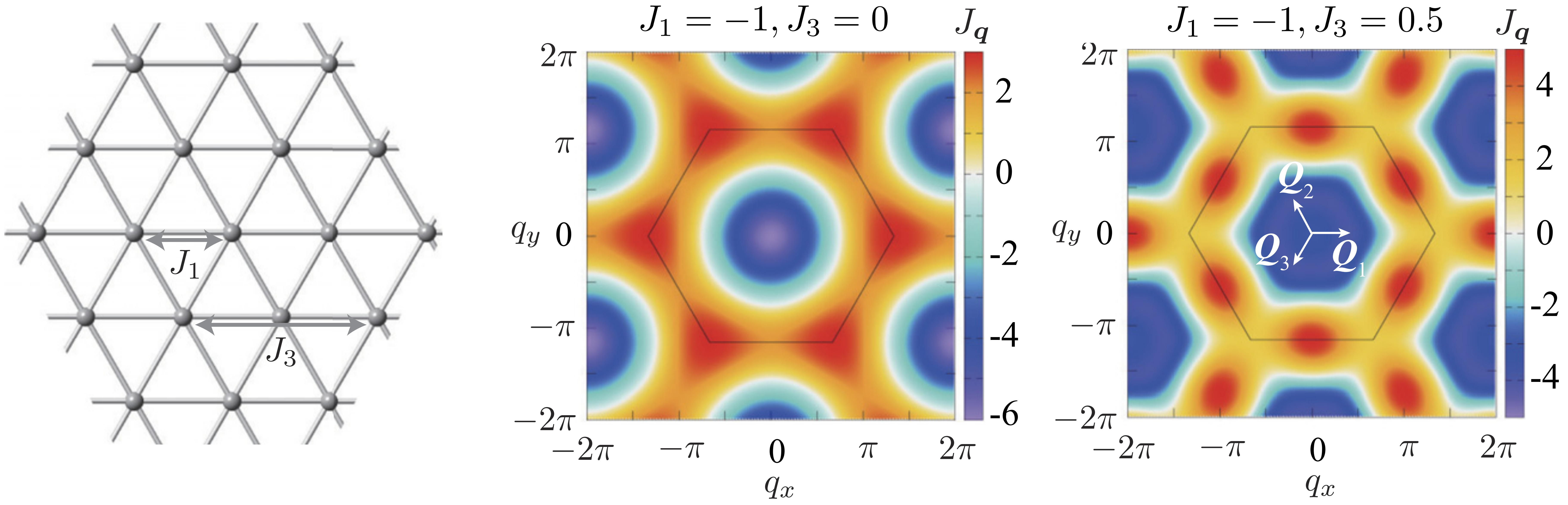
review: S. Hayami and Y. Motome, J. Phys.: Condens. Matter **33**, 443001 (2021)

Frustration in localized spin systems

$$\mathcal{H} = \sum_{ij} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j = \sum_{\mathbf{q}} J_{\mathbf{q}} \mathbf{S}_{\mathbf{q}} \cdot \mathbf{S}_{-\mathbf{q}}$$

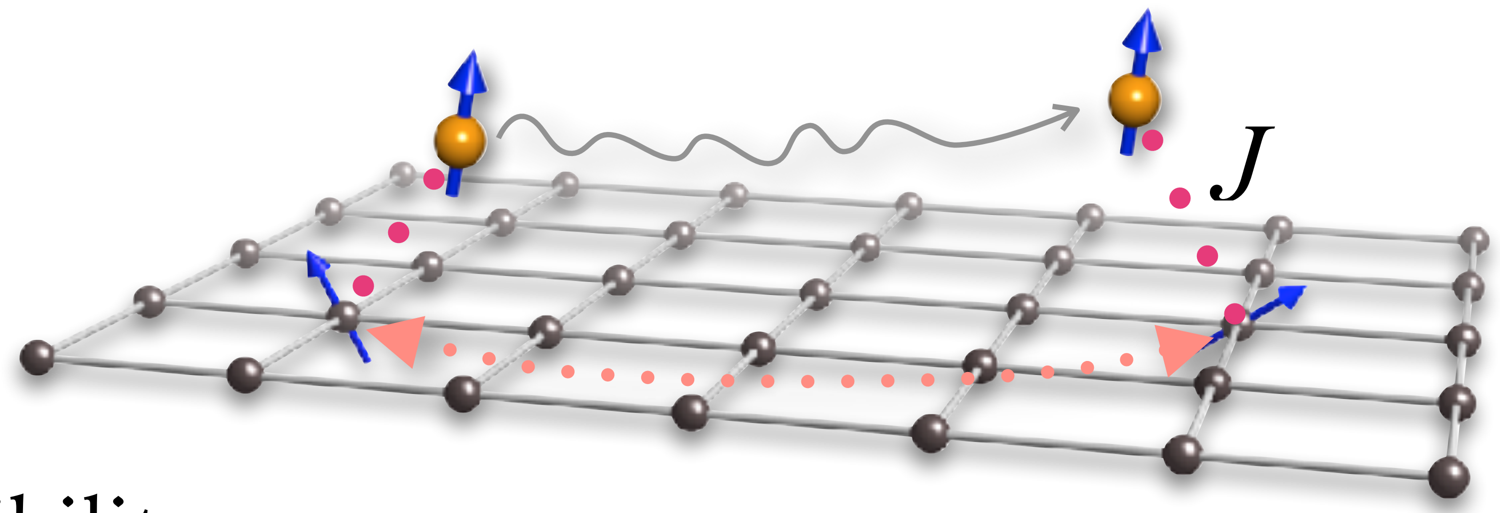
minimization of $J_{\mathbf{q}}$ → stable spin configuration

multiple minima in $J_{\mathbf{q}}$ → frustration



Frustration in itinerant electron systems

effective magnetic interactions mediated by itinerant electrons

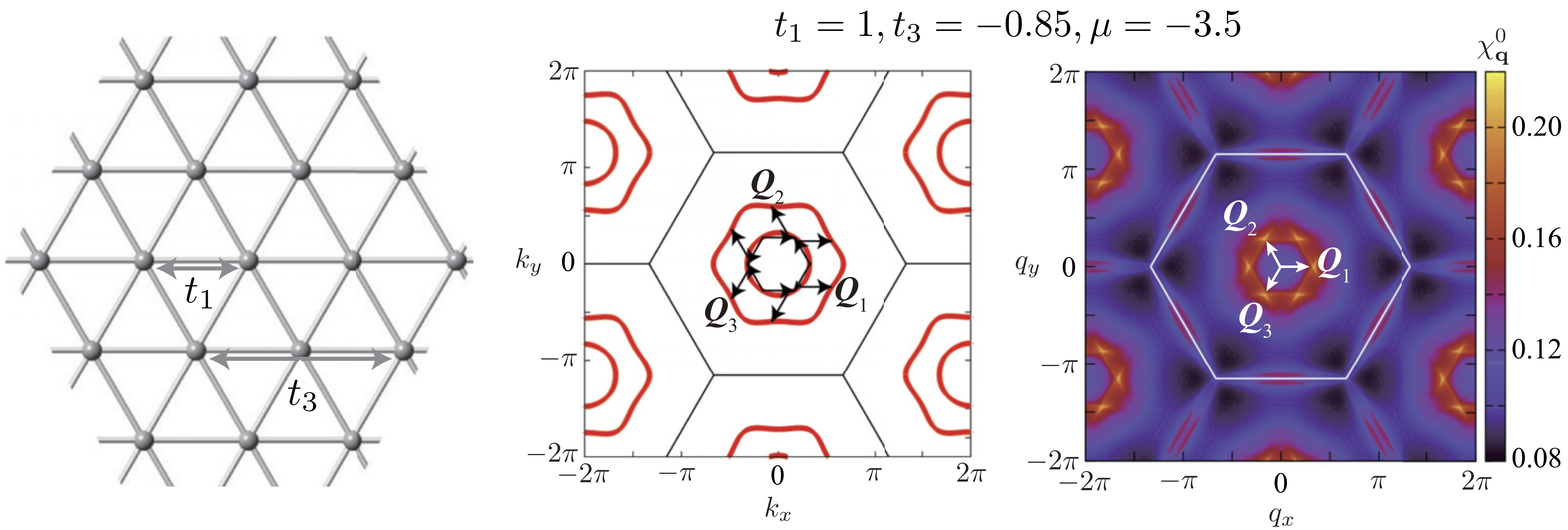


e.g., RKKY interaction

$$\mathcal{H} = \sum_{\mathbf{q}} J^2(-\chi_{\mathbf{q}}^{(0)})\mathbf{S}_{\mathbf{q}} \cdot \mathbf{S}_{-\mathbf{q}}$$

$\chi_{\mathbf{q}}^{(0)}$: bare susceptibility

multiple maxima in $\chi_{\mathbf{q}}^{(0)}$ due to Fermi surface nesting → frustration



Localized vs itinerant frustration

review: S. Hayami and Y. Motome, J. Phys.: Condens. Matter **33**, 443001 (2021)

◉ localized magnets

$$\mathcal{H} = \sum_{\mathbf{q}} J_{\mathbf{q}} \mathbf{S}_{\mathbf{q}} \cdot \mathbf{S}_{-\mathbf{q}}$$

multiple minima in $J_{\mathbf{q}}$ → frustration

degeneracy lifting,

e.g., by multiple-spin interactions arising from higher-order perturbation in t/U

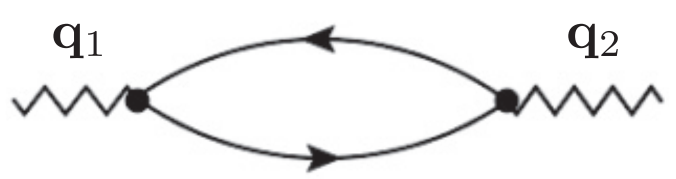
$$\frac{t^{2n}}{U^n}(\mathbf{S}_i \cdot \mathbf{S}_j)(\mathbf{S}_k \cdot \mathbf{S}_l)\cdots$$

exponential decay in real space

◉ itinerant magnets

$$\mathcal{H} = \sum_{\mathbf{q}} (-J^2 \chi^{(0)}_{\mathbf{q}}) \mathbf{S}_{\mathbf{q}} \cdot \mathbf{S}_{-\mathbf{q}}$$

2-spin interaction (RKKY)



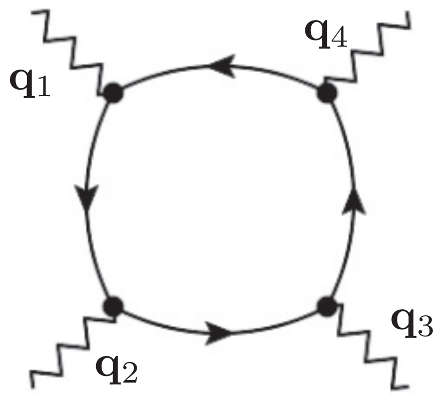
multiple maxima in $\chi^{(0)}_{\mathbf{q}}$ → frustration

degeneracy lifting by …?

➡ multiple-spin interactions arising from higher-order perturbation in J/t

$$J_{\mathbf{q}_1,\mathbf{q}_2,\mathbf{q}_3,\mathbf{q}_4,\cdots}(\mathbf{S}_{\mathbf{q}_1} \cdot \mathbf{S}_{\mathbf{q}_2})(\mathbf{S}_{\mathbf{q}_3} \cdot \mathbf{S}_{\mathbf{q}_4})\cdots$$

4-spin interaction



well described in momentum space: intrinsically long-range

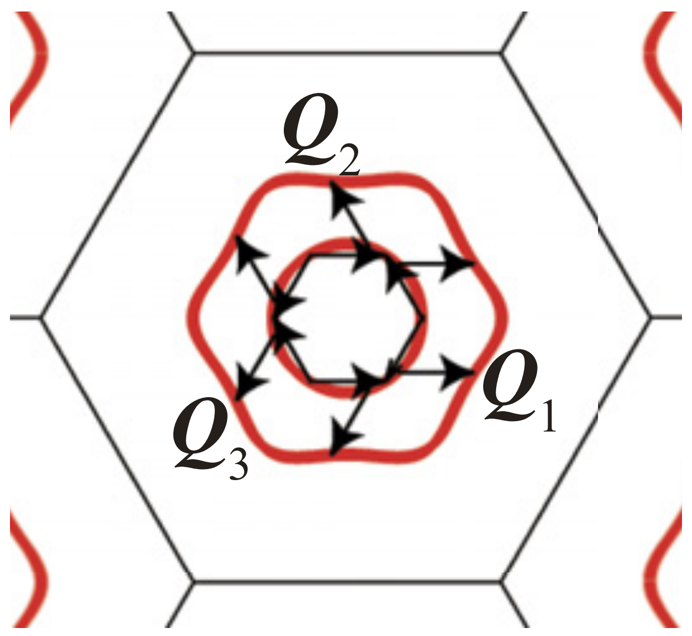
relevant wave vectors are dictated by the Fermi surface nesting

- size of spin textures can be extremely small
- inversion symmetry breaking is not necessary (unlike DMI)

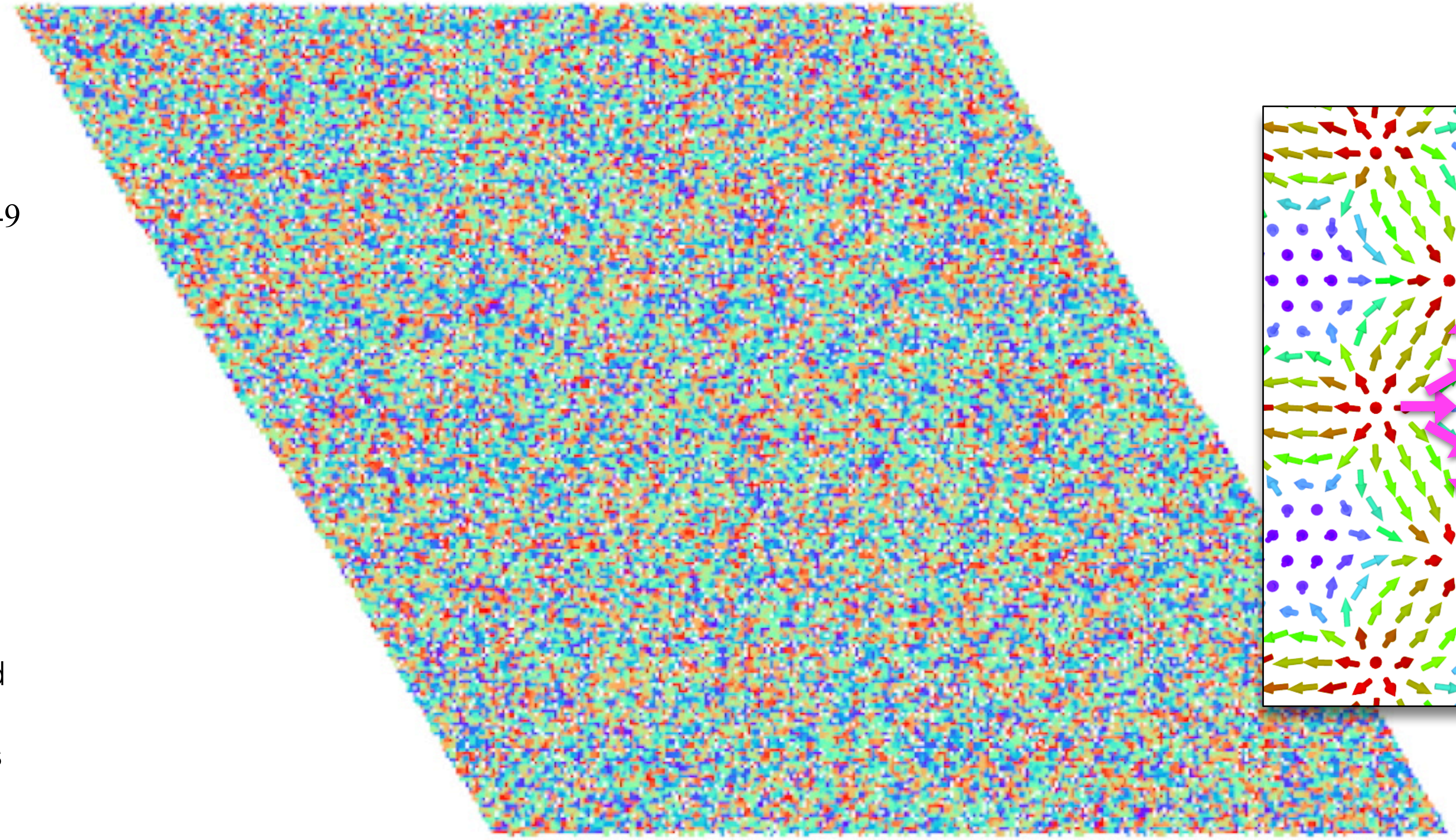
Skyrmion crystal by itinerant frustration

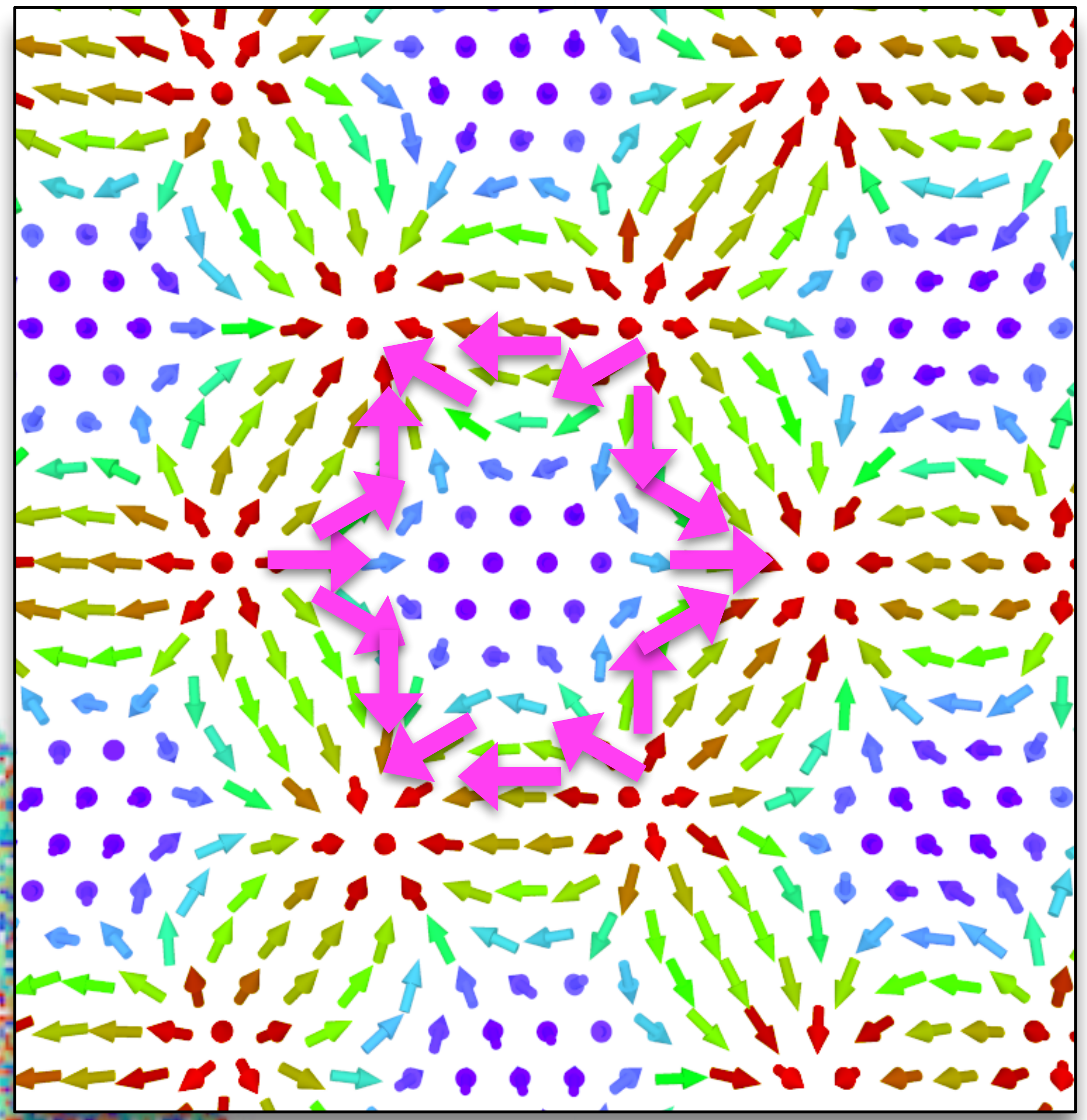
- large-scale numerical simulation of the Kondo lattice model on a triangular lattice

triangular lattice
t_1=1, t_3=-0.85, J=0.3, n~0.149



kernel polynomial method
with Langevin dynamics
192 x 192 ~ 3.7 x 10^4 sites



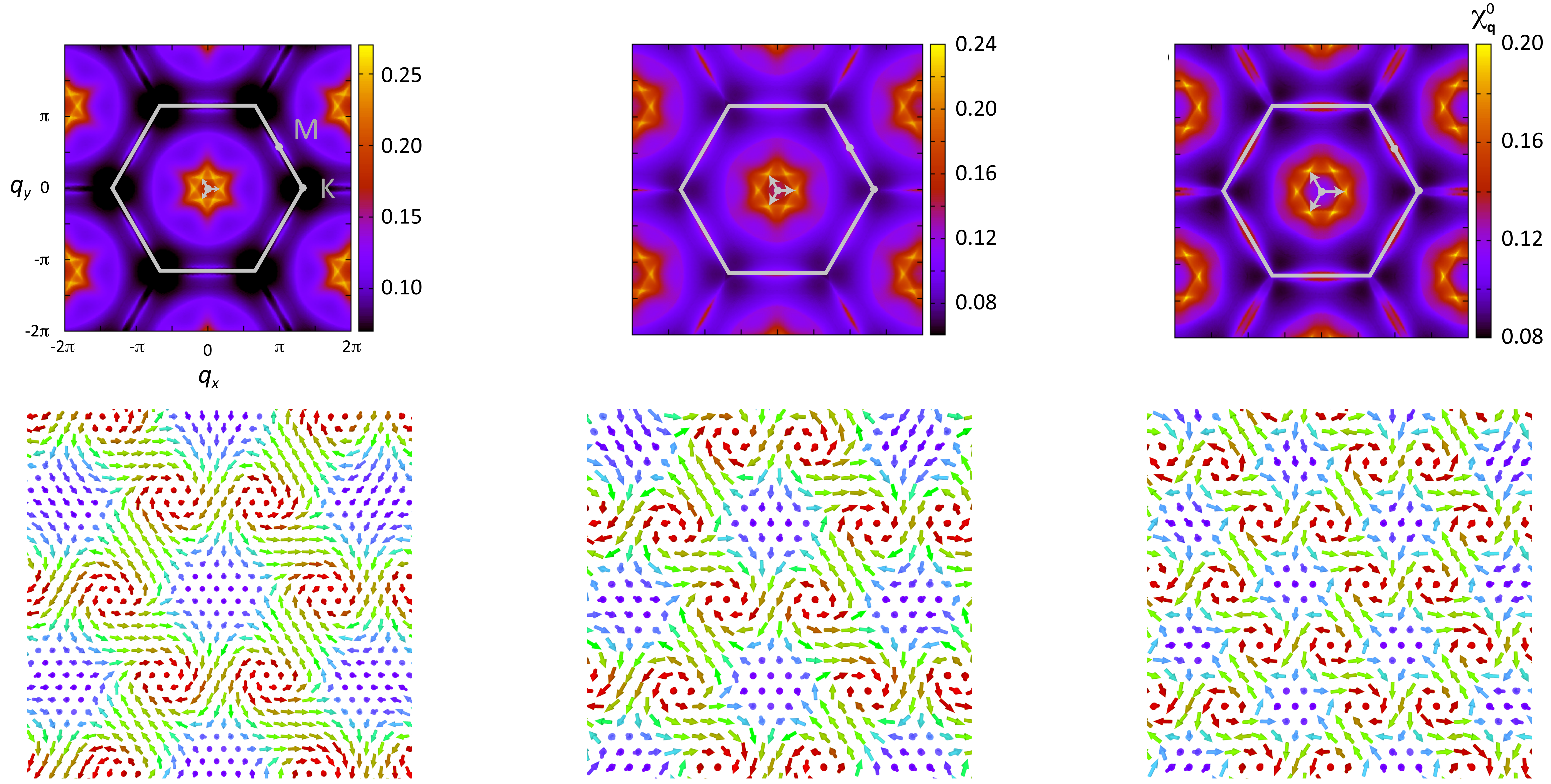


spontaneous formation of 3**q** skyrmion crystal with a high skyrmion number of N_{sk}=2

R. Ozawa, S. Hayami, and Y. Motome, Phys. Rev. Lett. **118**, 147205 (2017)

Control of skyrmion size by Fermi surface

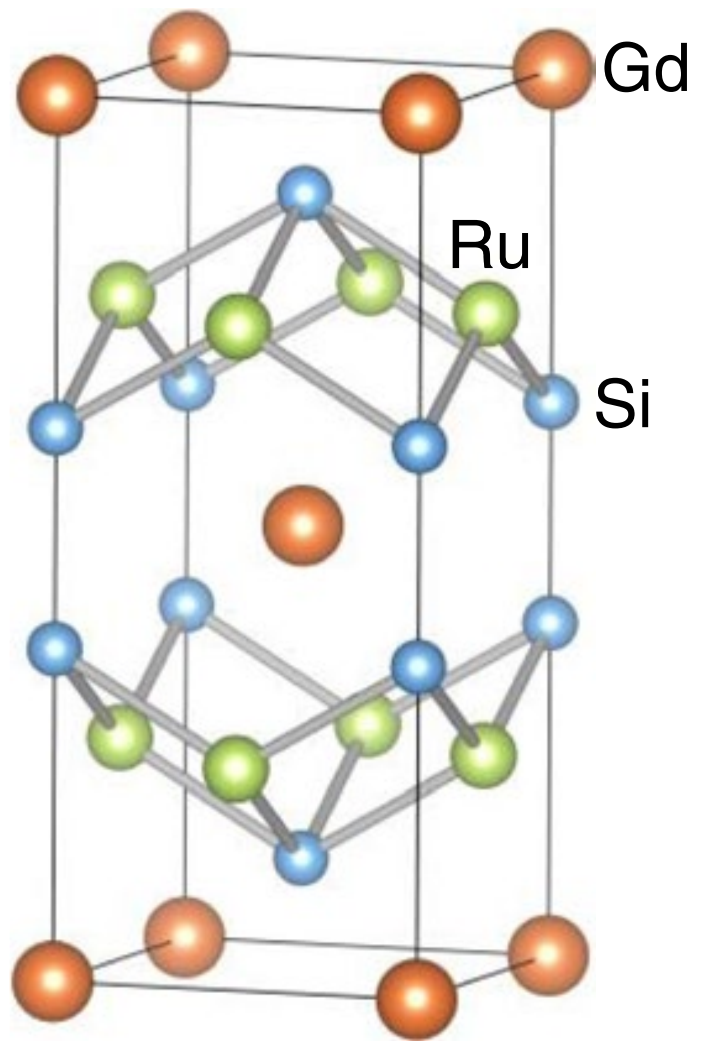
small — nesting vector → large

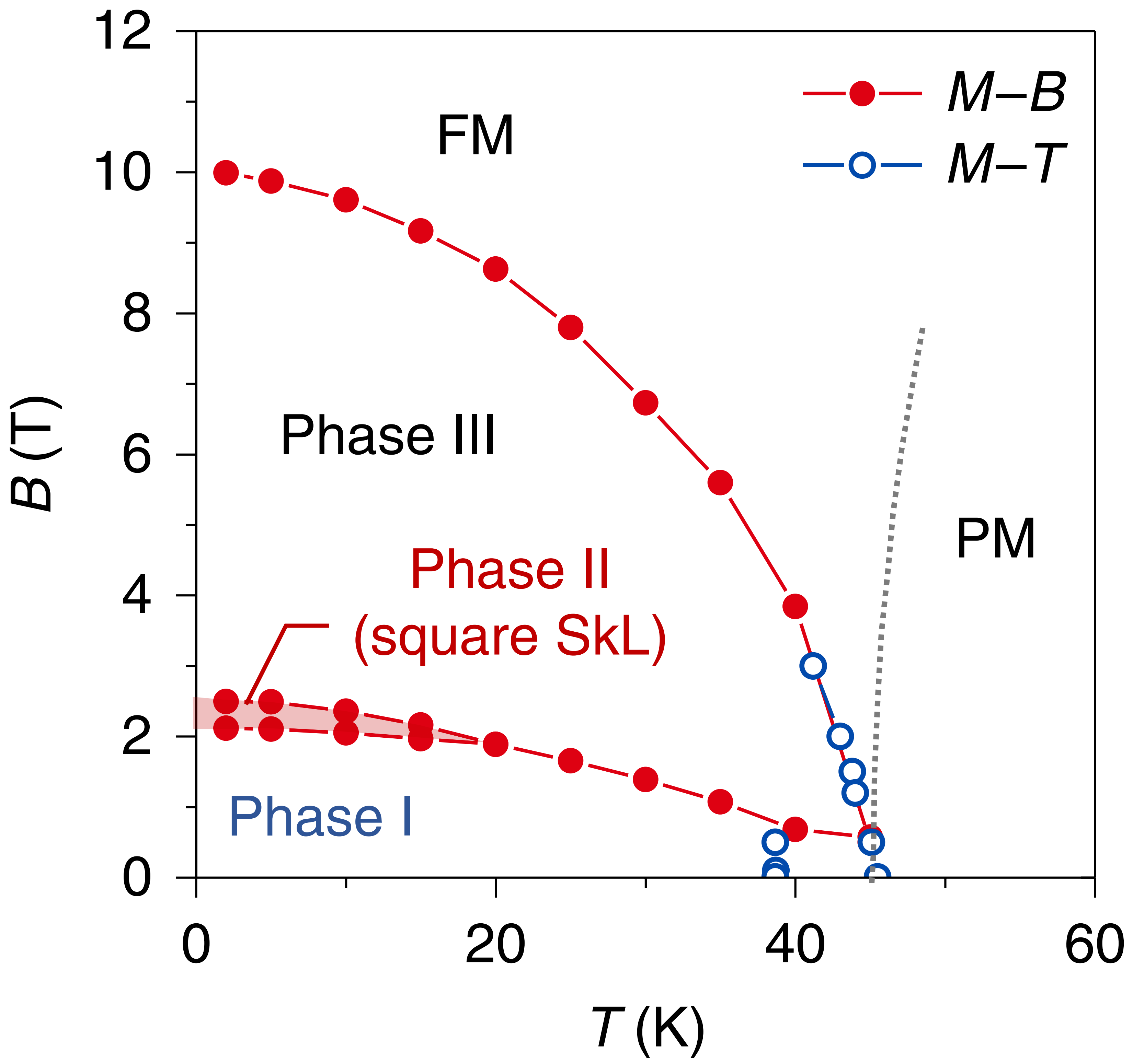


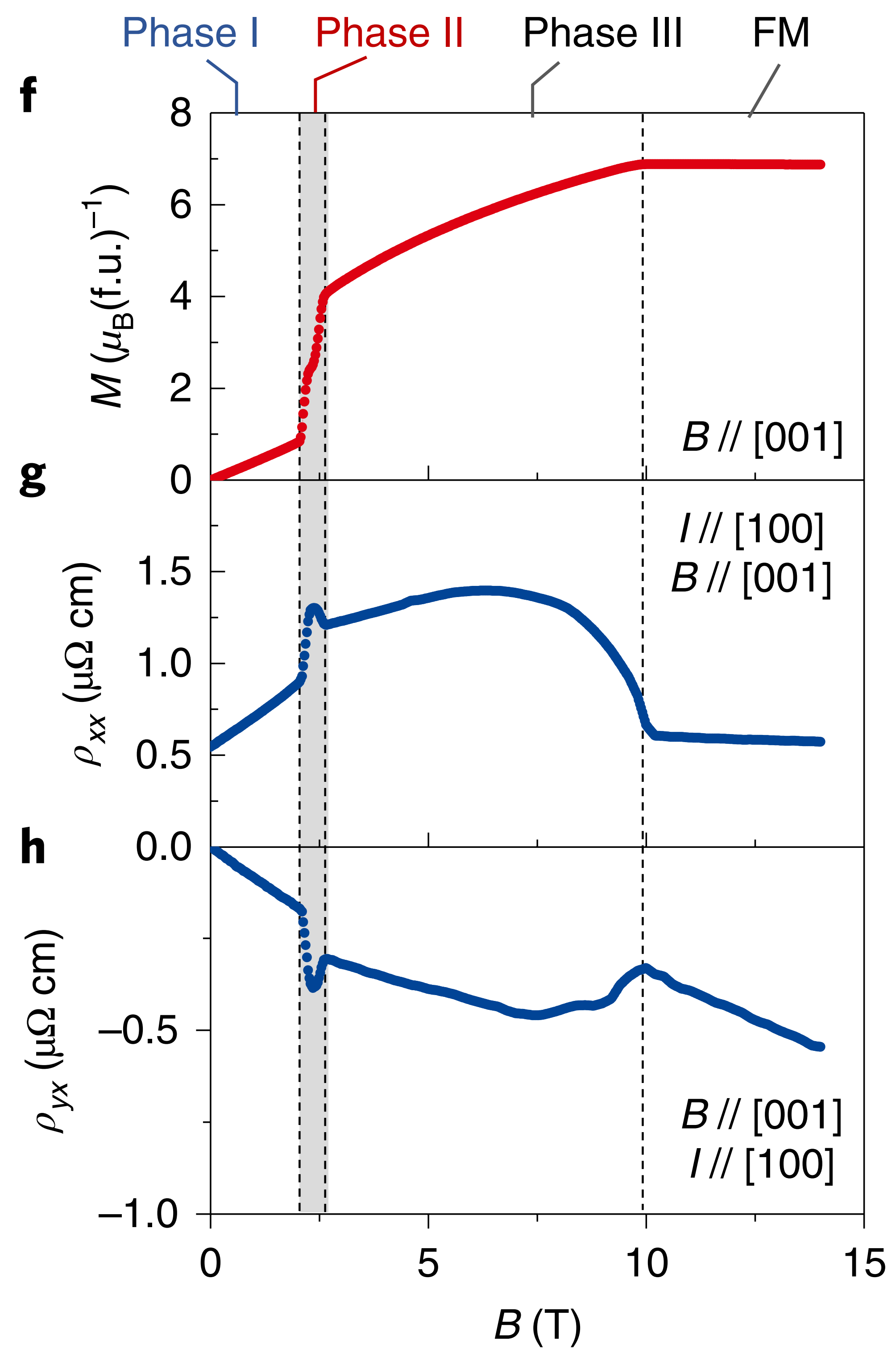
large — skyrmion size → small

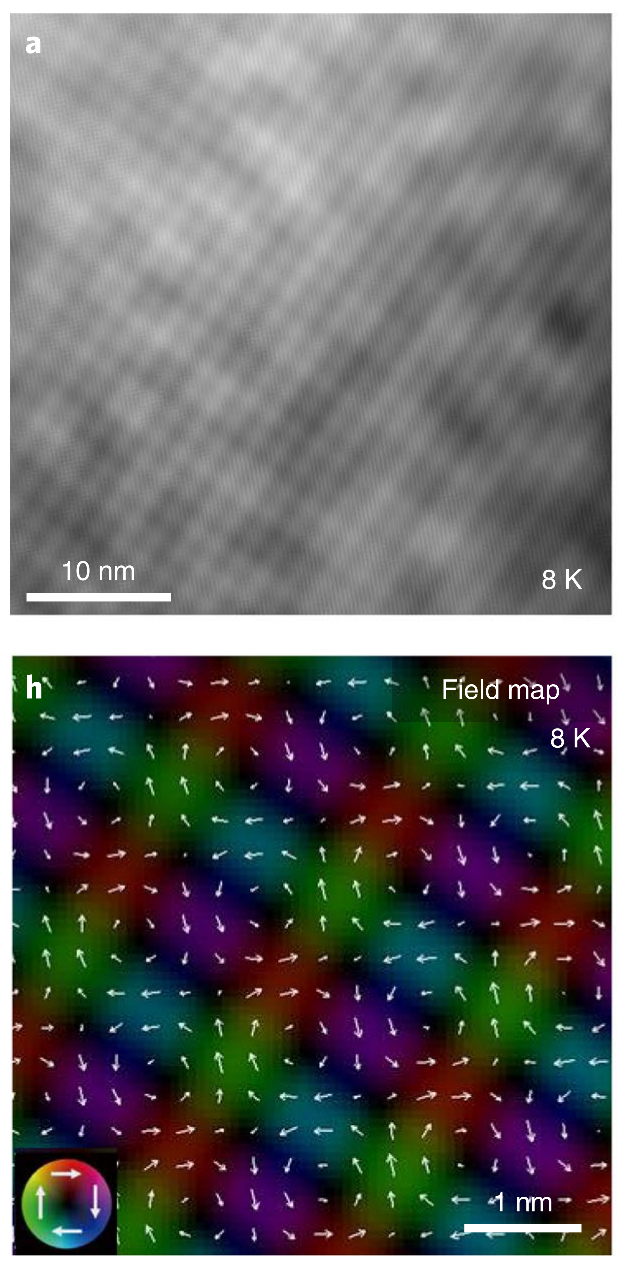
R. Ozawa, S. Hayami, and Y. Motome, Phys. Rev. Lett. **118**, 147205 (2017)
see also R. Ozawa, S. Hayami, K. Barros, G-W. Chern, Y. Motome, C. D. Batista, J. Phys. Soc. Jpn. **85**, 103703 (2016)

Square skyrmion crystal in $GdRu_2Si_2$









centrosymmetric system: Dzyaloshinskii-Moriya interaction is inactive

extremely short period ~1.9 nm: importance of itinerant frustration?

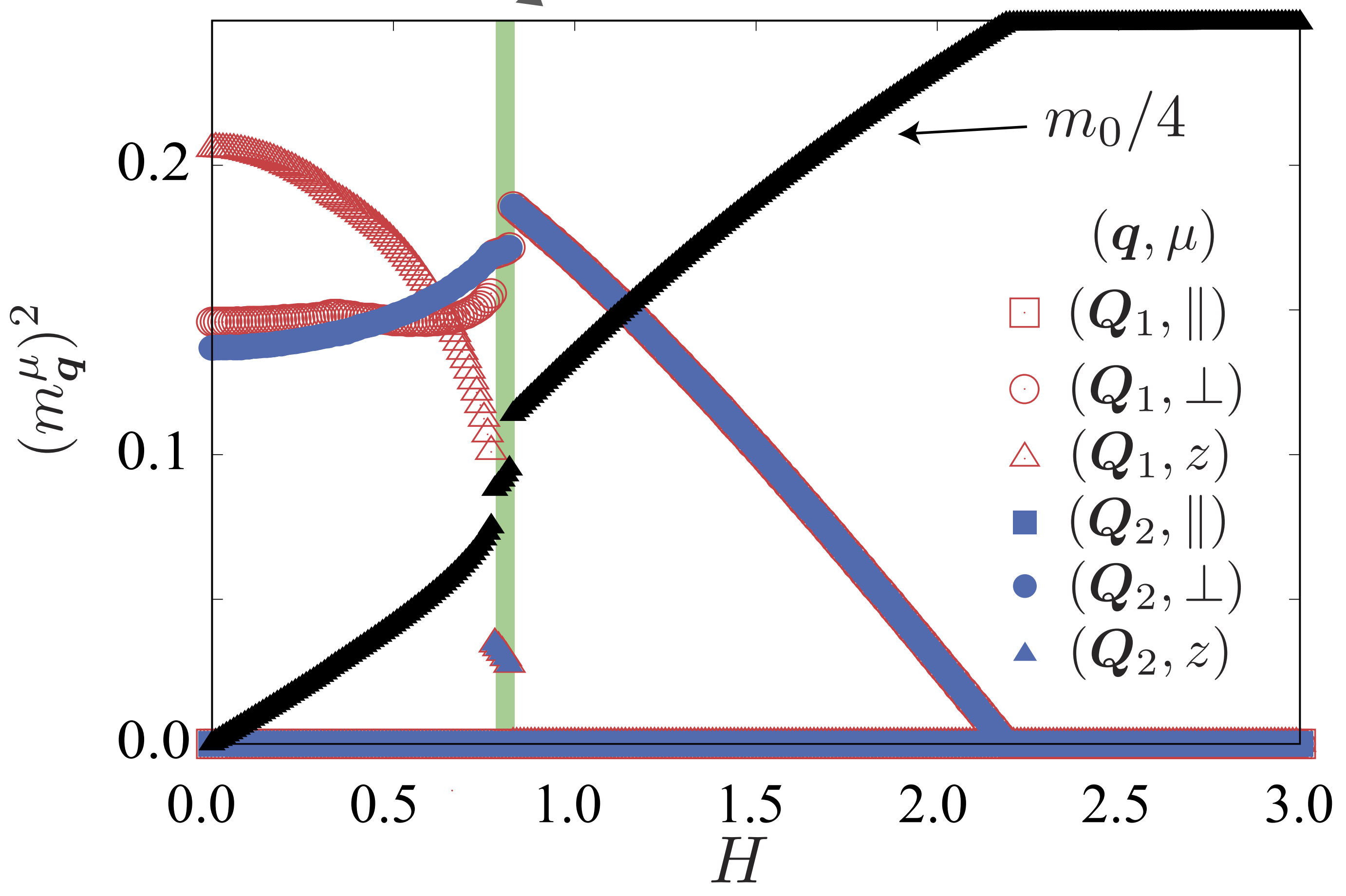
N. D. Khanh et al., Nat. Nanotech. **15**, 444 (2020)

Square skyrmion crystal: theory

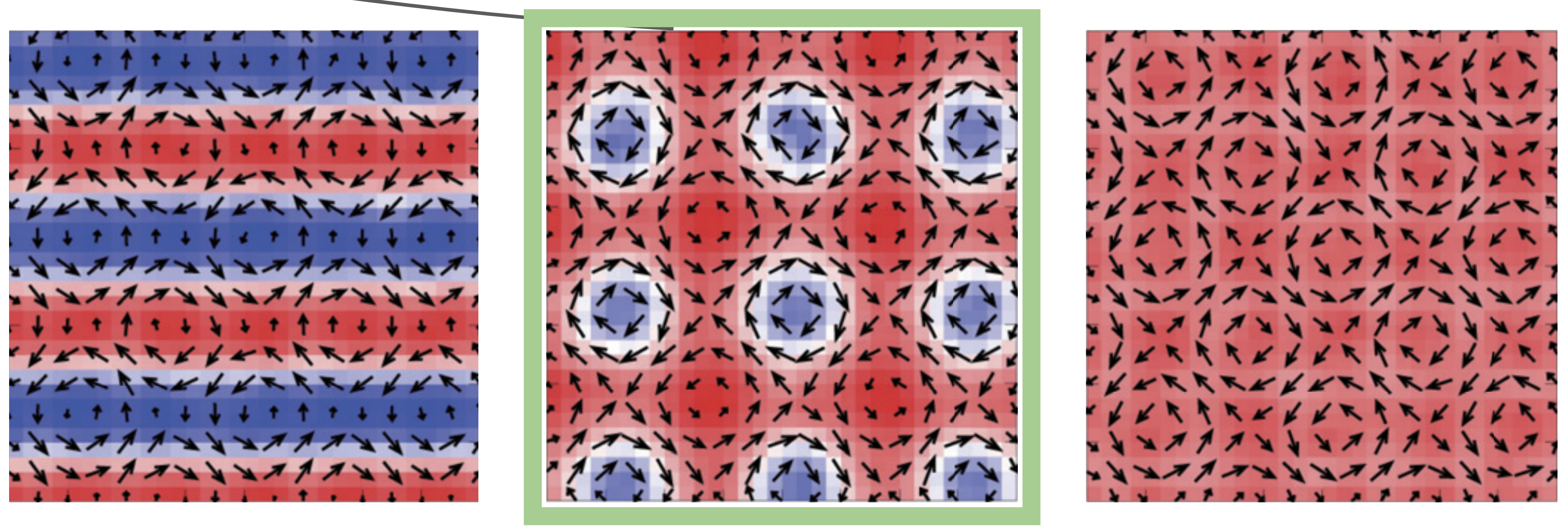
2-spin interaction (RKKY) 4-spin biquadratic interaction

$$\mathcal{H} = 2\sum_{\nu}\Big[-J\sum_{\alpha\beta}\Gamma^{\alpha\beta}_{\mathbf{Q}_\nu}S^{\alpha}_{\mathbf{Q}_\nu}S^{\beta}_{-\mathbf{Q}_\nu} + \frac{K}{N}\Big(\sum_{\alpha\beta}\Gamma^{\alpha\beta}_{\mathbf{Q}_\nu}S^{\alpha}_{\mathbf{Q}_\nu}S^{\beta}_{-\mathbf{Q}_\nu}\Big)^2\Big]$$

bond-dependent anisotropy from spin-orbit coupling



magnetic field // [001]



2q vortex crystal (low field)

2q skyrmion crystal (intermediate field)

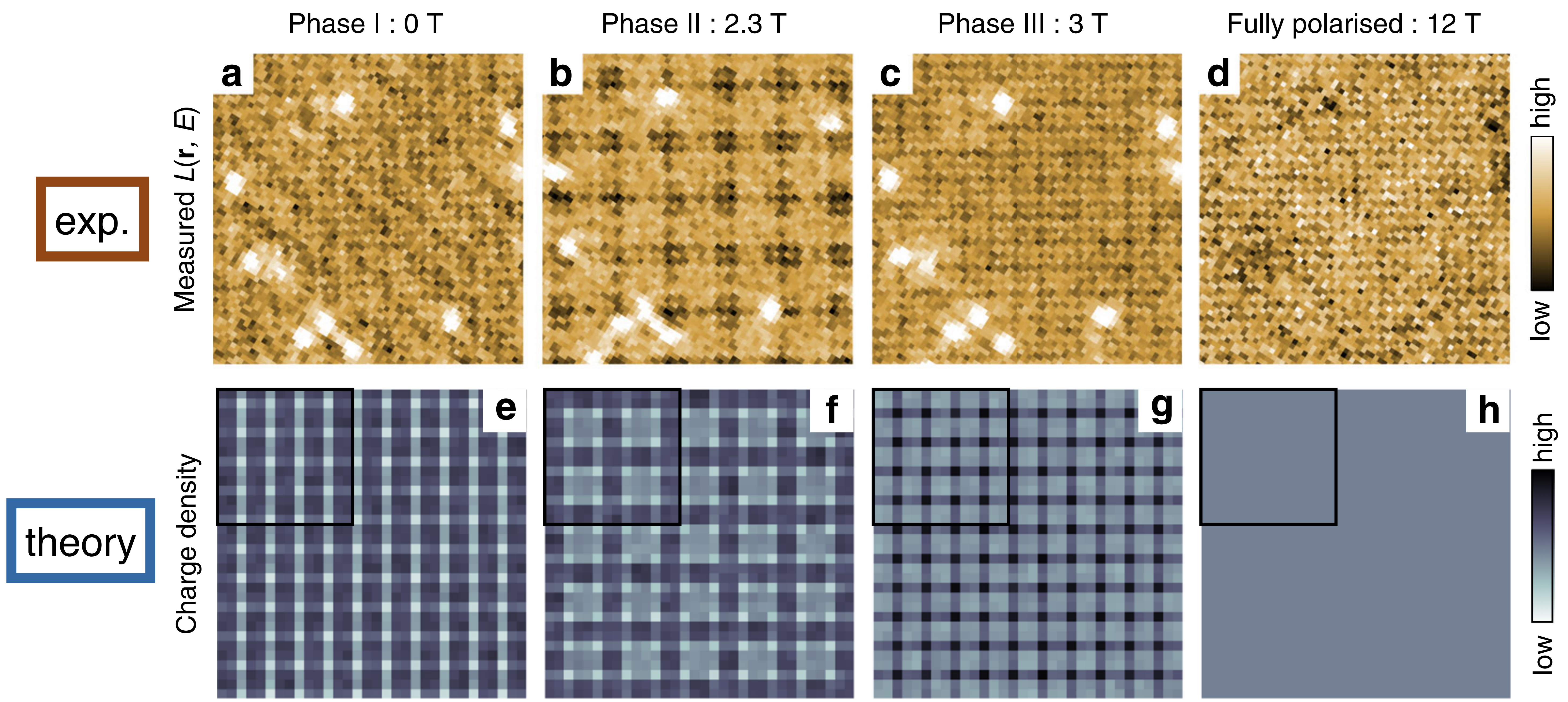
2q vortex crystal (high field)

Square-lattice type skyrmion crystal with short period is realized in a magnetic field.
Phase sequence and regions are qualitatively consistent with the experimental results.

S. Hayami and Y. Motome, Phys. Rev. B **103**, 024439 (2021)

Comparison between experiments and theory

- STM measurement for $GdRu_2Si_2$

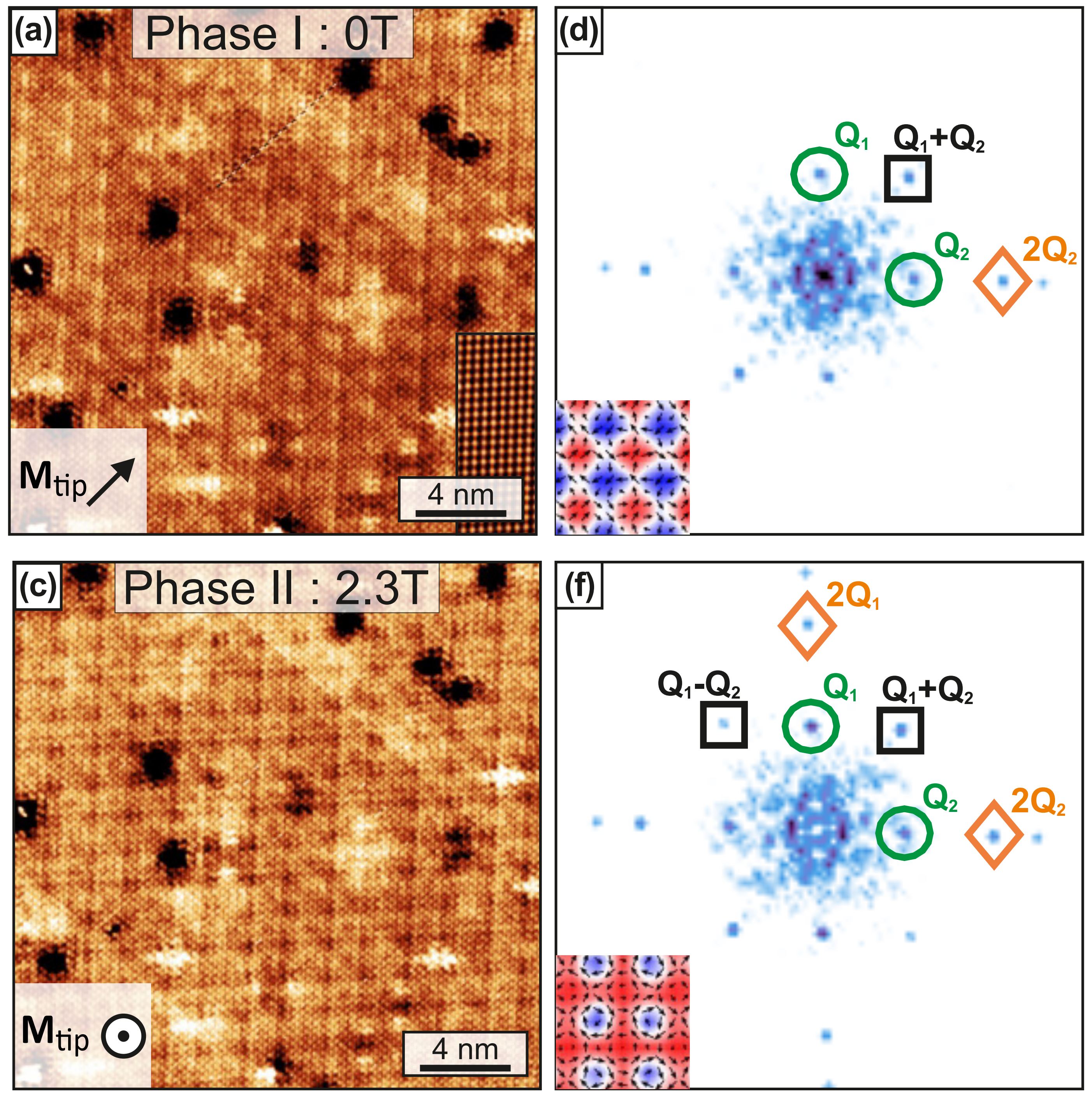


charge modulations concomitant with spin textures:
evidence for spin-charge coupling

Y. Yasui, C. J. Butler, N. D. Khanh, S. Hayami, T. Nomoto, T. Hanaguri, Y. Motome, R. Arita, T. Arima, Y. Tokura, and S. Seki, Nat. Commun. **11**, 5925 (2020)

S. Hayami and Y. Motome, Phys. Rev. B **104**, 144404 (2021)

- spin-polarized STM measurement



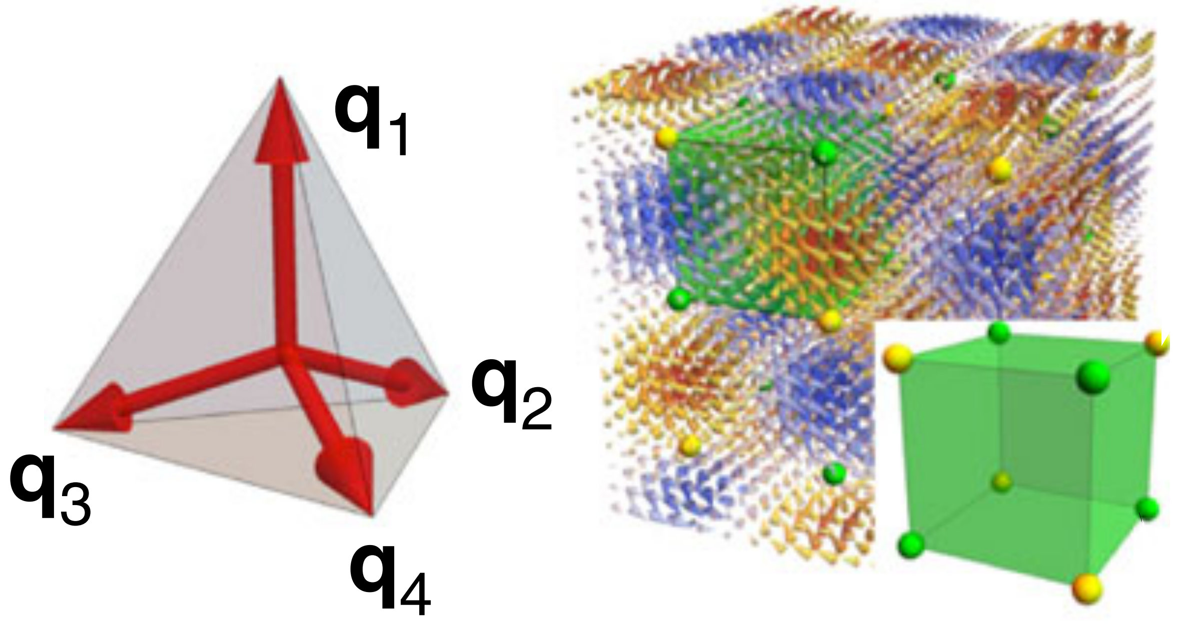
good agreement with theory

J. Spethmann, N. D. Khanh, H. Yoshimochi, R. Takagi, S. Hayami, Y. Motome, R. Wiesendanger, S. Seki, and K. von Bergmann, Phys. Rev. Materials **8**, 064404 (2024)

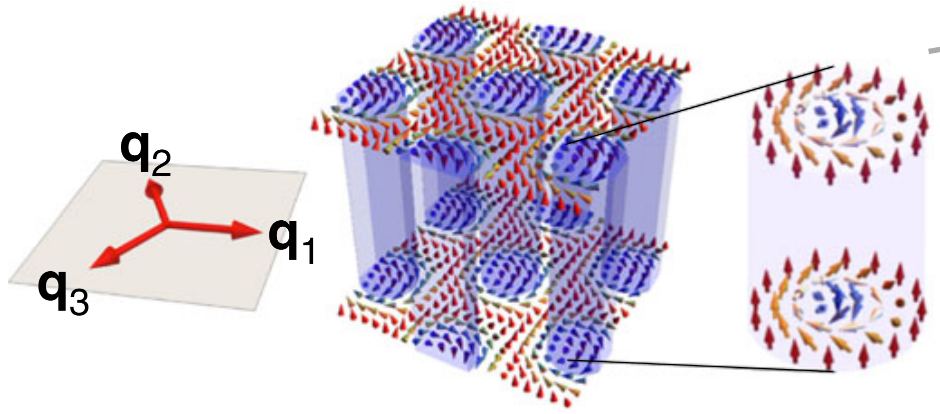
Application to 3D toron crystals

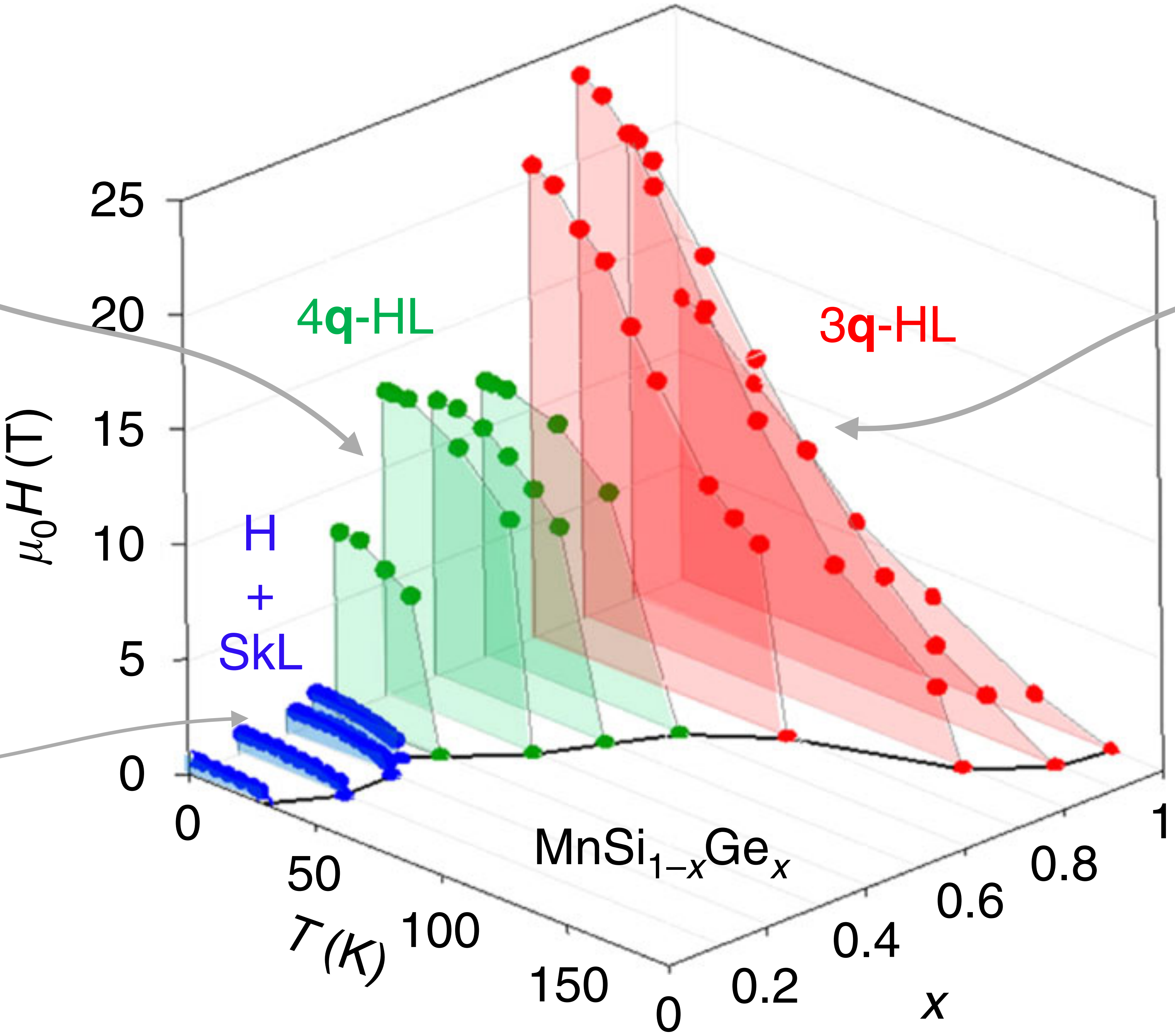
◉ phase diagram for $MnSi_{1-x}Ge_x$

tetrahedral-4**q** toron crystal

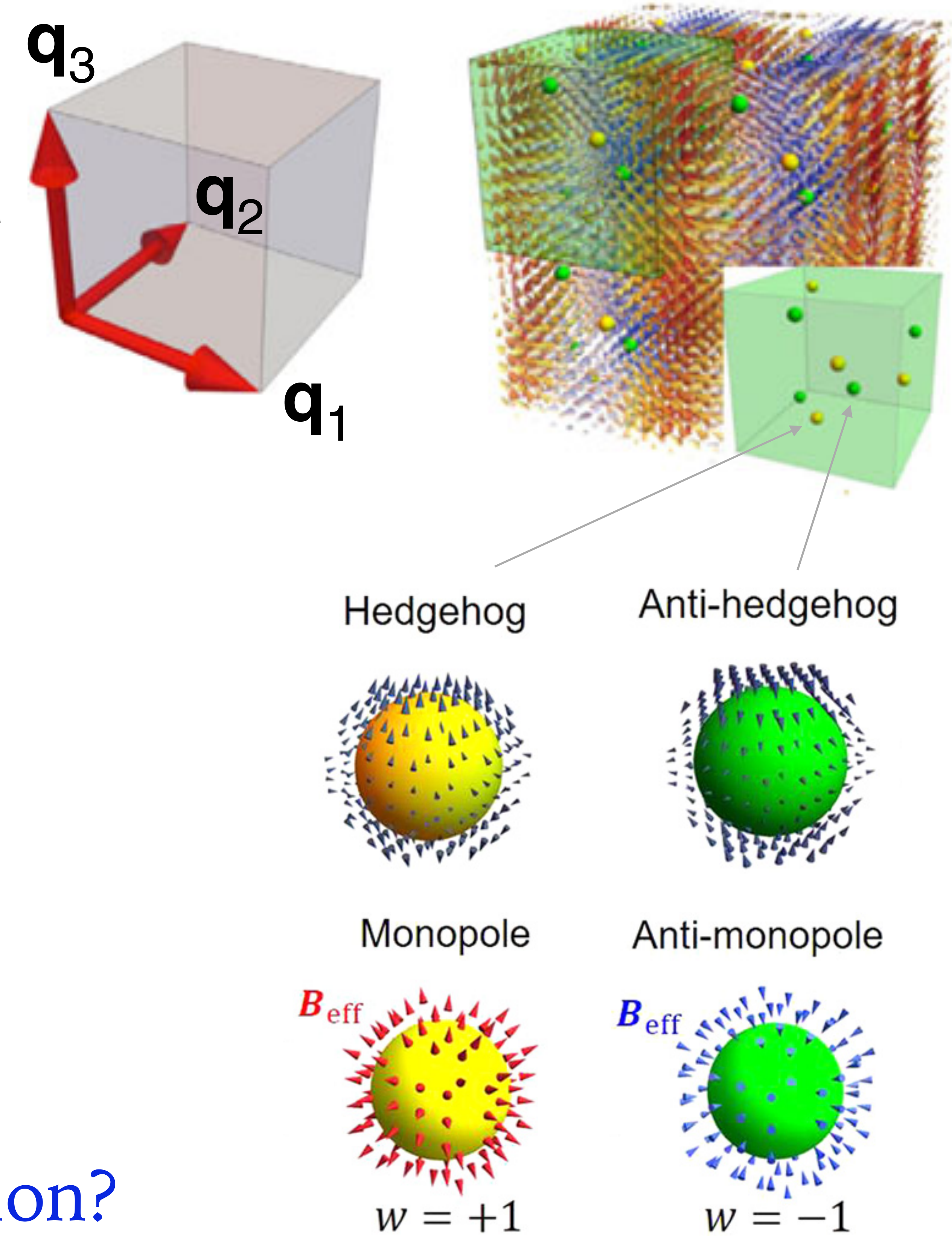


skyrmion crystal





cubic-3**q** toron crystal



period ~2-3 nm: importance of itinerant frustration?

N. Kanazawa *et al.*, Phys. Rev. B **86**, 134425 (2012); Y. Fujishiro *et al.*, Nat. Commun. **10**, 1059 (2019)

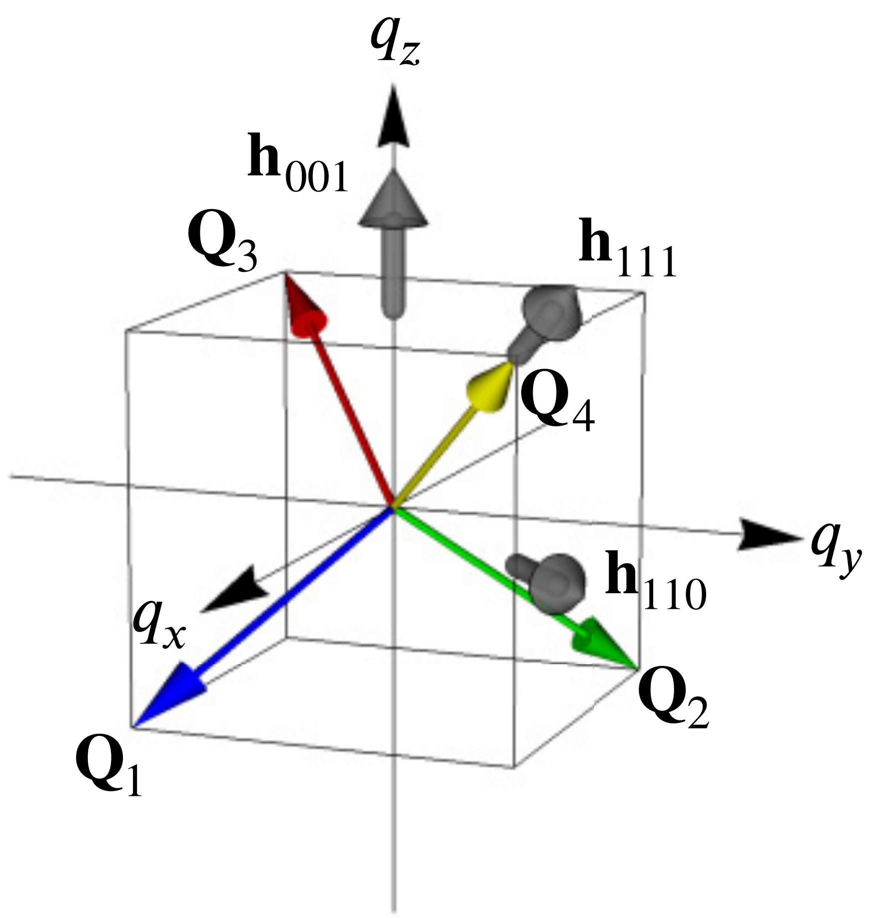
Toron crystals: theory

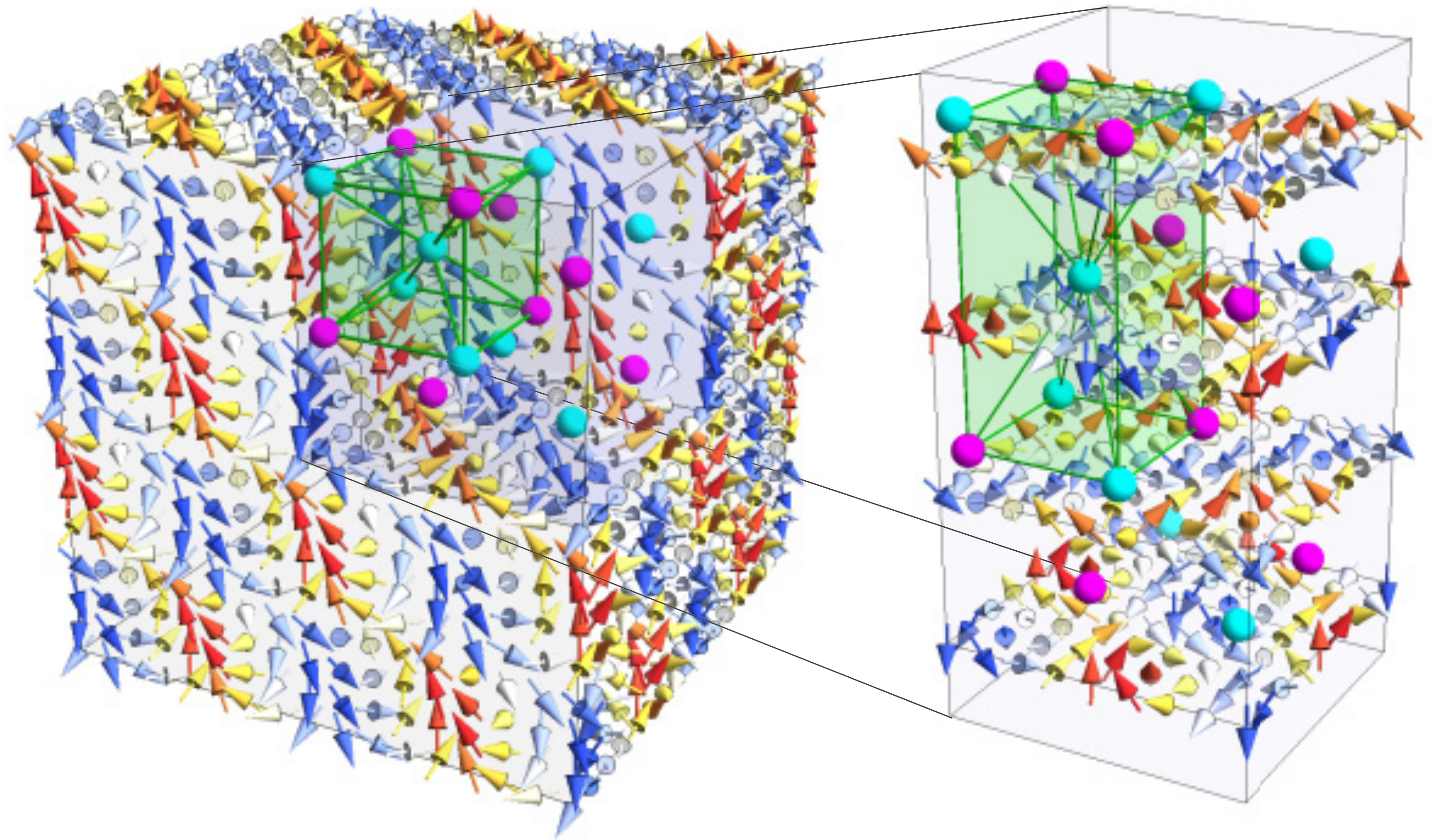
Shun Okumura

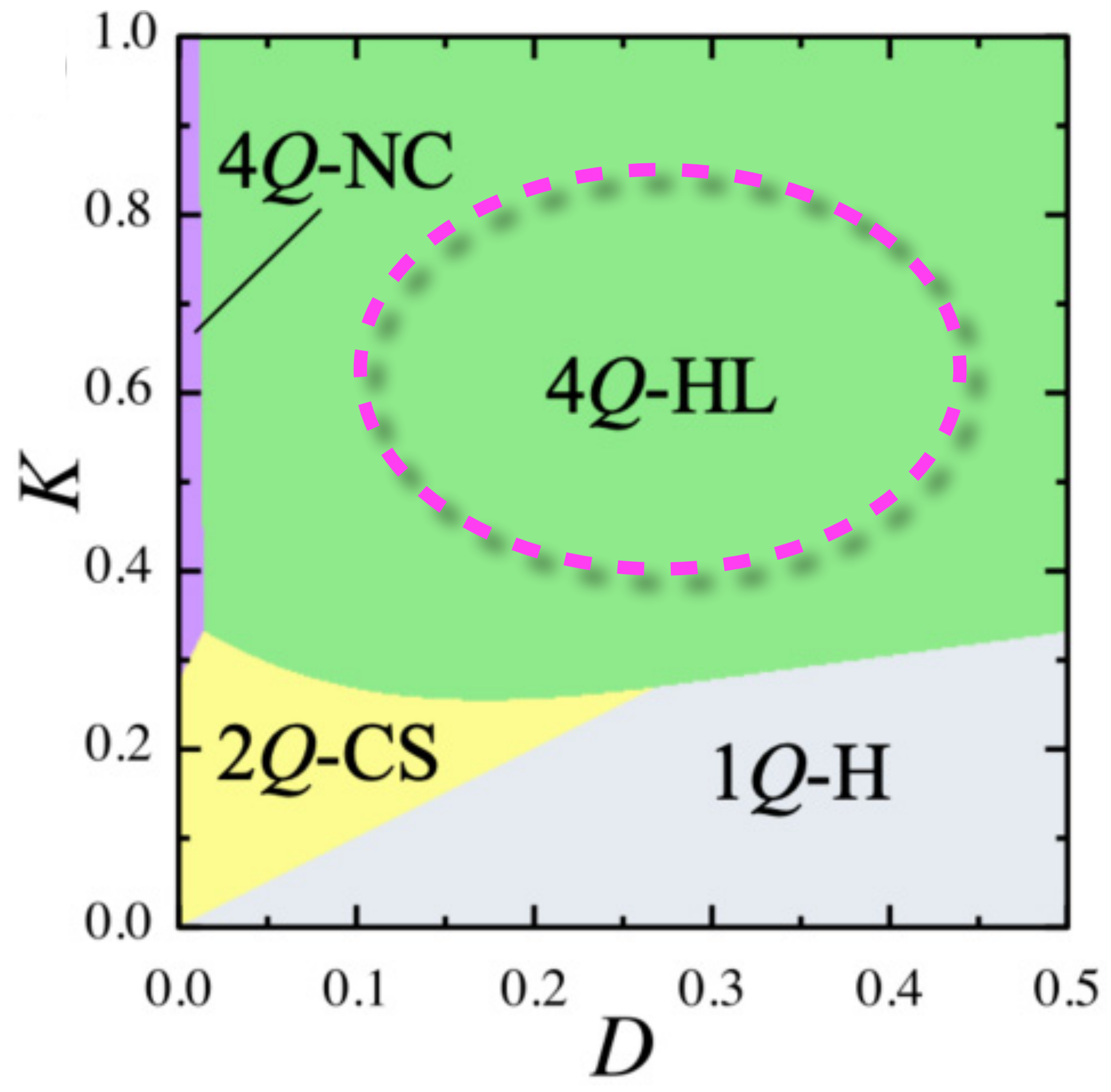
$$\mathcal{H} = 2\sum_{\nu}\Big[-J\sum_{\alpha\beta} S^{\alpha}_{\mathbf{Q}_\nu} S^{\beta}_{-\mathbf{Q}_\nu} + \frac{K}{N}(\mathbf{S}_{\mathbf{Q}_\nu}\cdot\mathbf{S}_{-\mathbf{Q}_\nu})^2 - i\mathbf{D}_\nu\cdot(\mathbf{S}_{\mathbf{Q}_\nu}\times\mathbf{S}_{-\mathbf{Q}_\nu})\Big]$$

RKKY interaction — biquadratic interaction — DM-type interaction

tetrahedral-4**q** model → tetrahedral-4**q** toron crystal







Cooperation between biquadratic interaction and DM-type interaction stabilizes toron crystals.

(similar results for cubic-3**q** toron crystal)

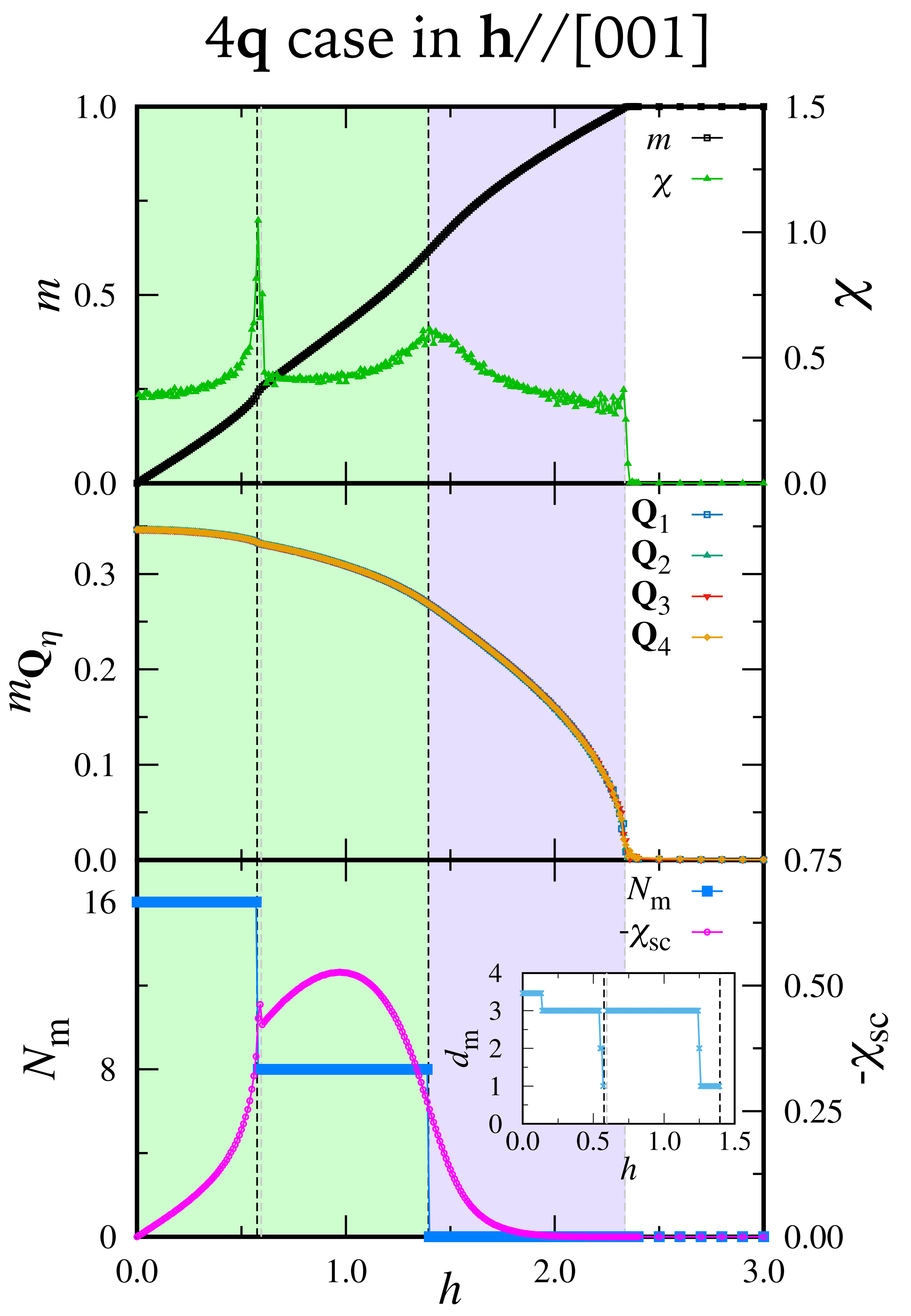
S. Okumura, S. Hayami, Y. Kato, and Y. Motome, Phys. Rev. B **101**, 144416 (2020)

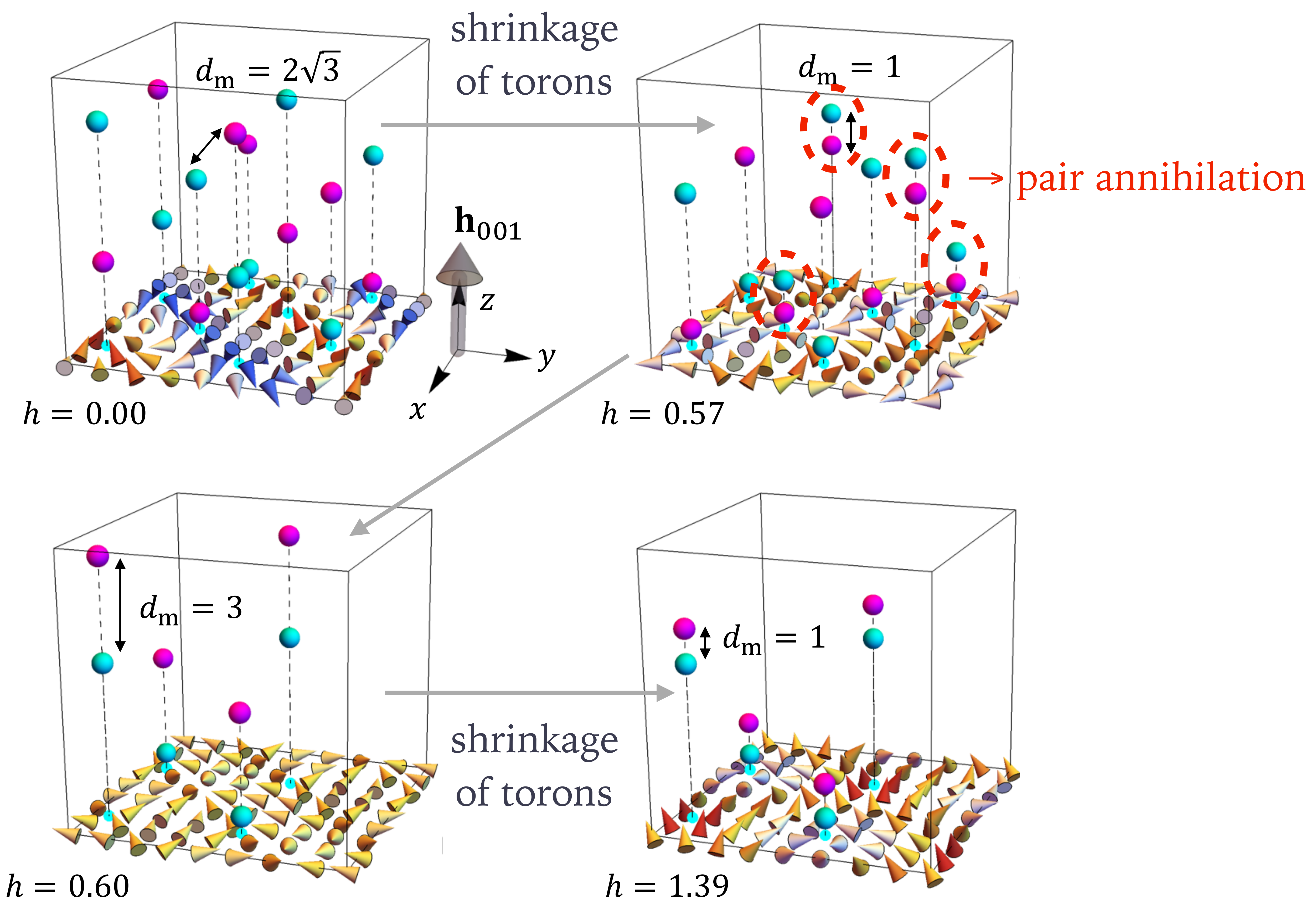
Toron crystals: magnetic field

magnetization and susceptibility

magnetic moment with wave vector $\mathbf{Q}_\eta$

number of hedgehogs and antihedgehogs in a magnetic unit cell



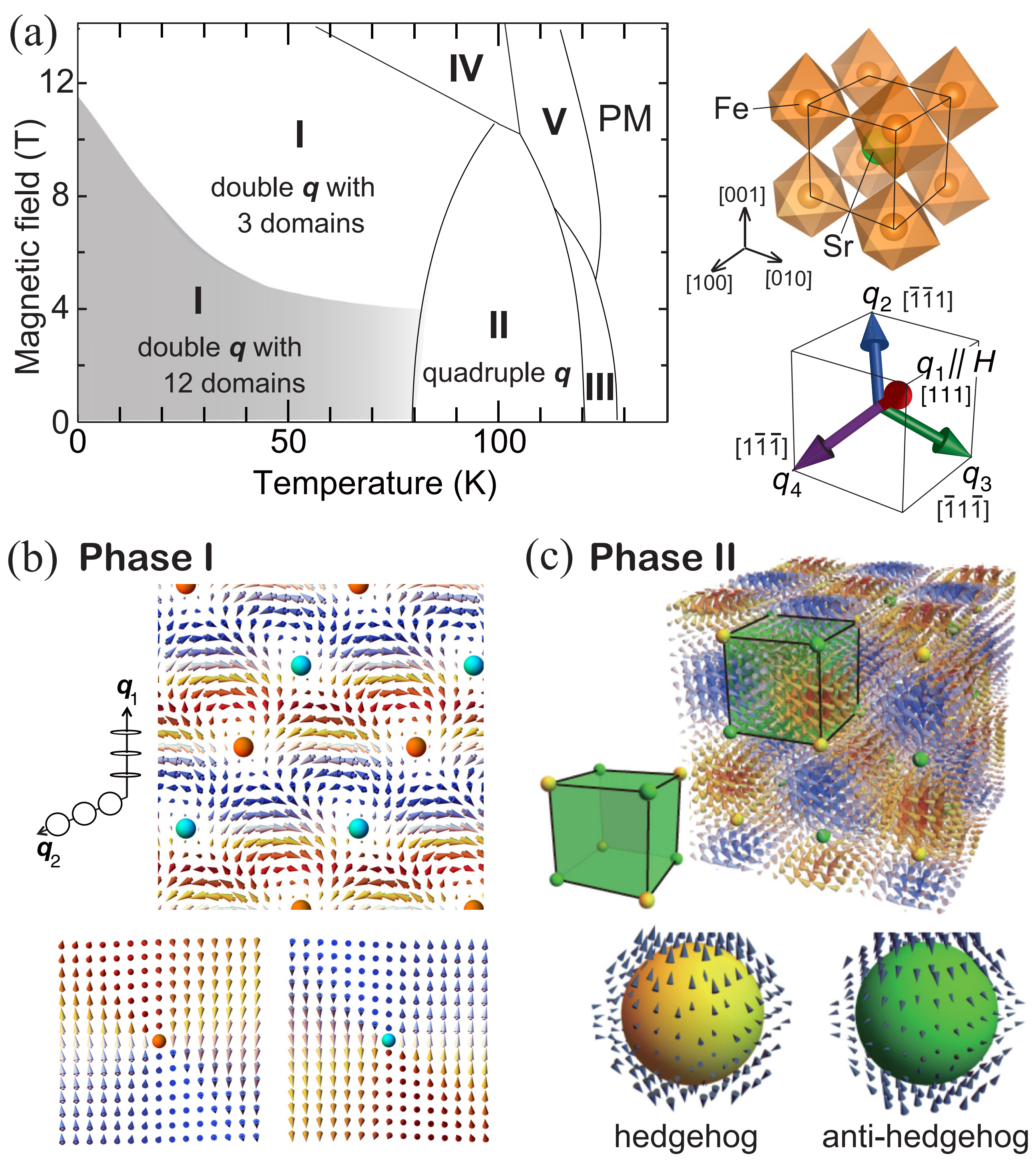


topological transitions by pair annihilation of hedgehogs and antihedgehogs depending on the **h** direction

S. Okumura, S. Hayami, Y. Kato, and Y. Motome, Phys. Rev. B **101**, 144416 (2020); Y. Kato and Y. Motome, Phys. Rev. B **107**, 094437 (2023)

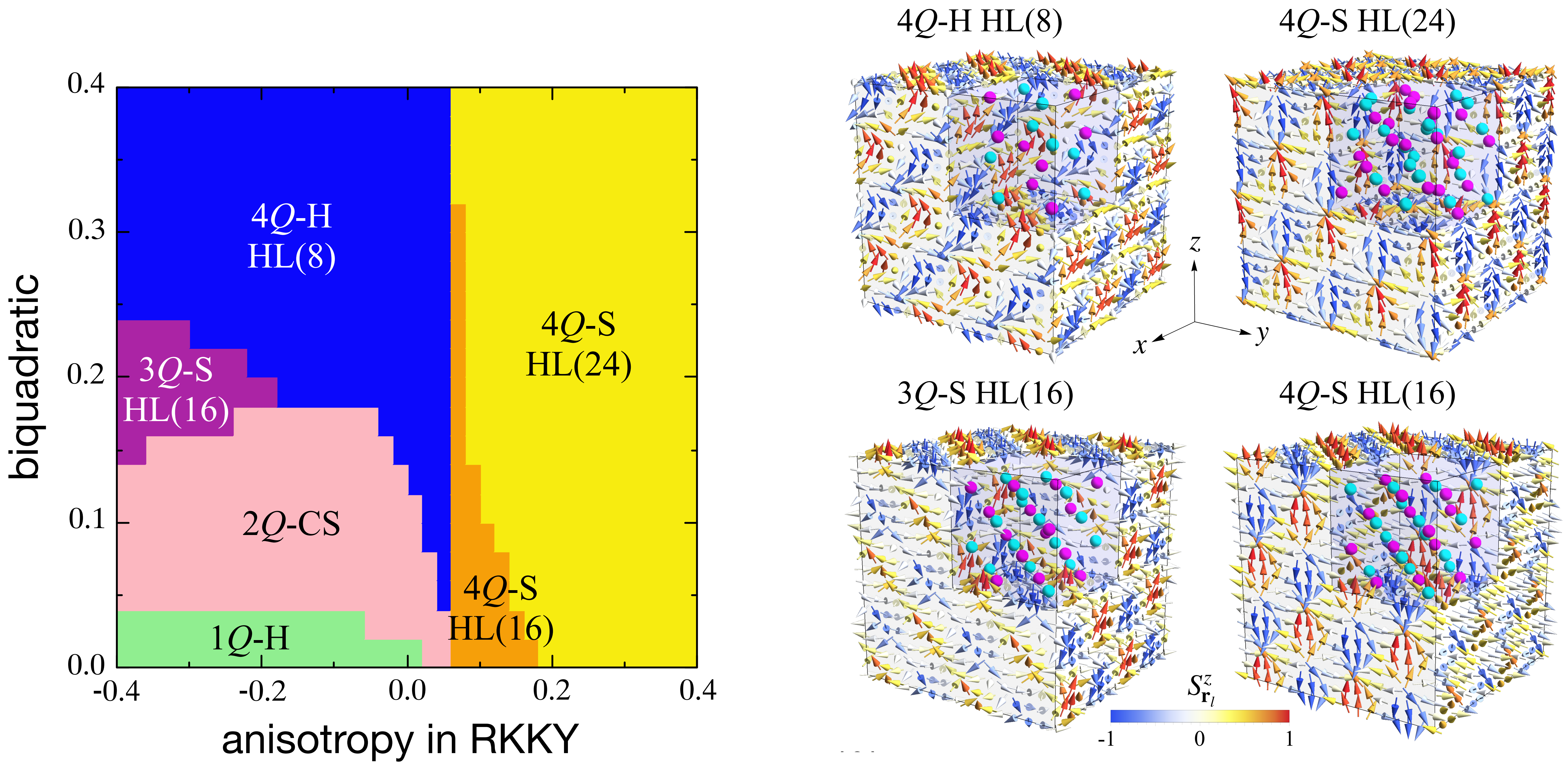
Toron crystals: centrosymmetric case

- experiment in $SrFeO_3$



S. Ishiwata *et al.*, Phys. Rev B **84**, 054427 (2011)
S. Ishiwata *et al.*, Phys. Rev B **101**, 134406 (2020)

- theory: RKKY + biquadratic



a variety of toron crystals even in the centrosymmetric case,
including high-density toron crystals found in the spin moiré analysis

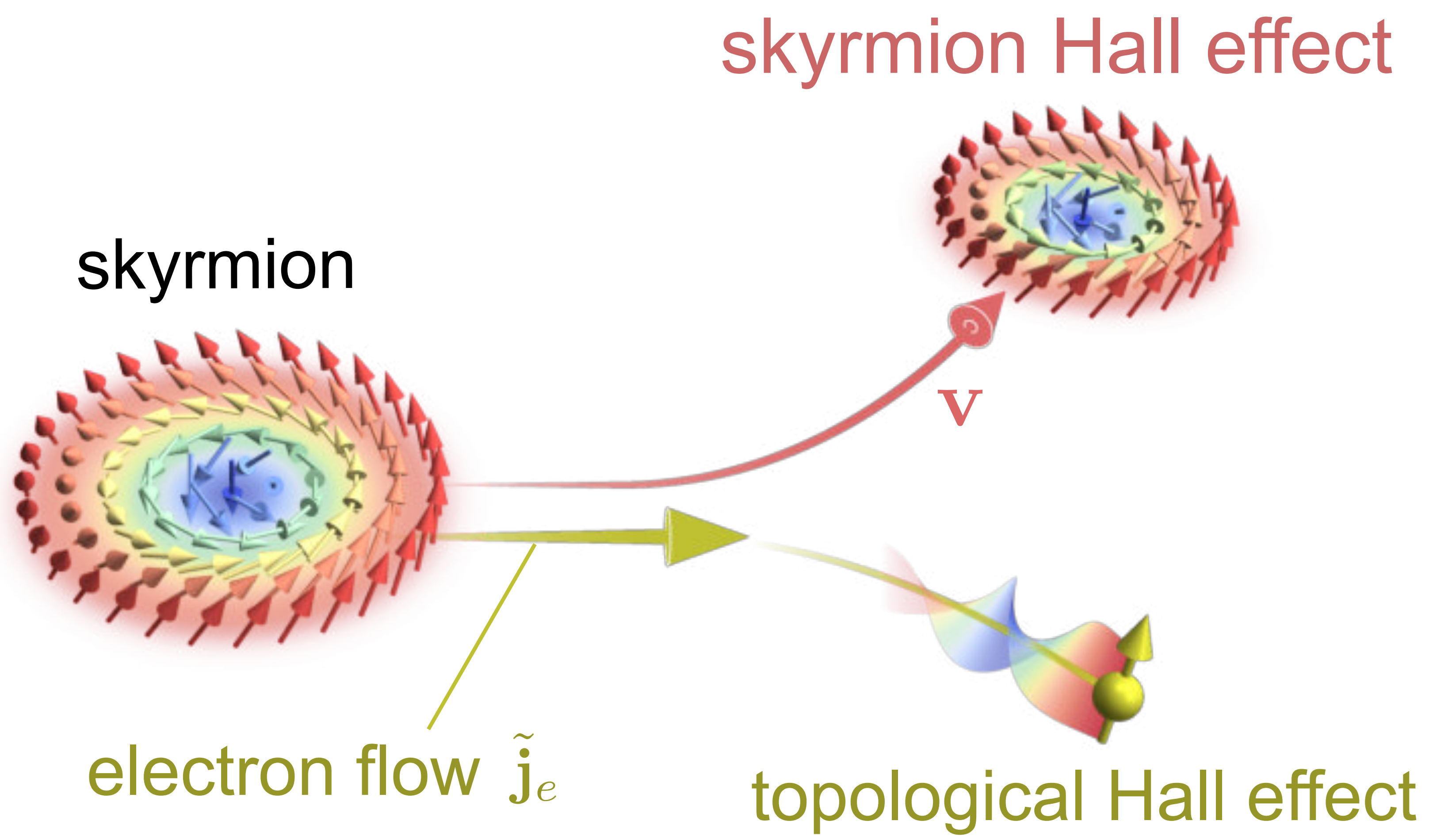
qualitative understanding of the experimental phase diagram of $SrFeO_3$

S. Okumura, S. Hayami, Y. Kato, and Y. Motome, in preparation

Current-induced dynamics of torons

Kotaro Shimizu

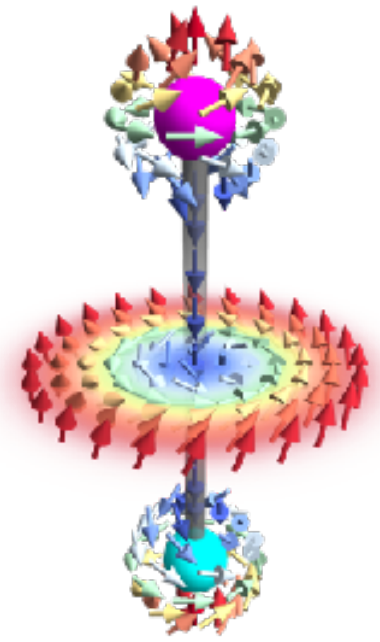
◉ 2D skyrmion



- driven by ultralow current density
- transverse motion of skyrmion by Magnus force: skyrmion Hall effect

➡ hindering application like racetrack memory

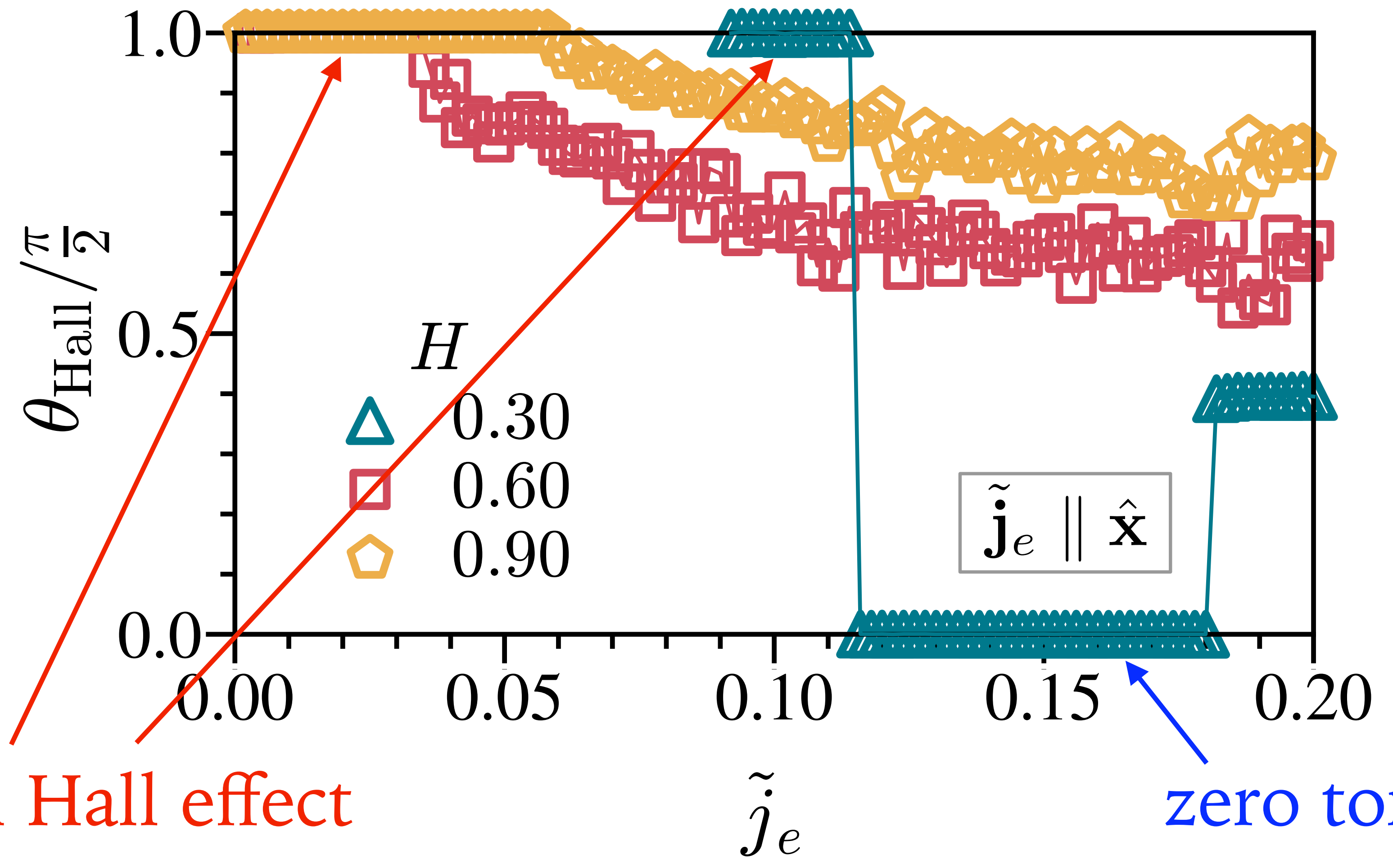
◉ 3D toron



How do the torons respond to an electric current?
especially, short-period torons by itinerant frustration

➡ controllable in a wide range by an electric current density and an external magnetic field

Hall angle of toron motion



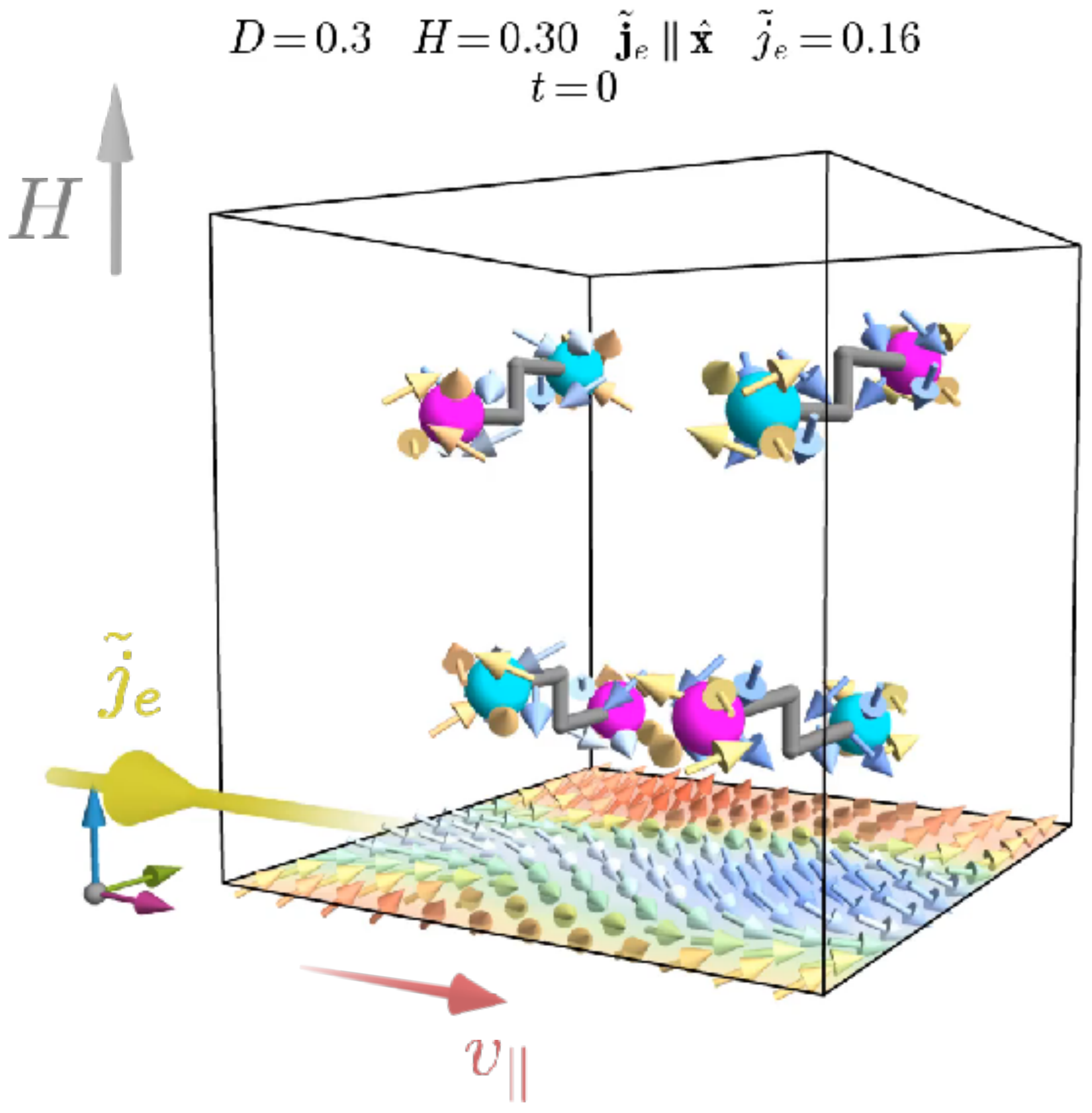
perfect toron Hall effect (only transverse motion)

zero toron Hall effect (only longitudinal motion)

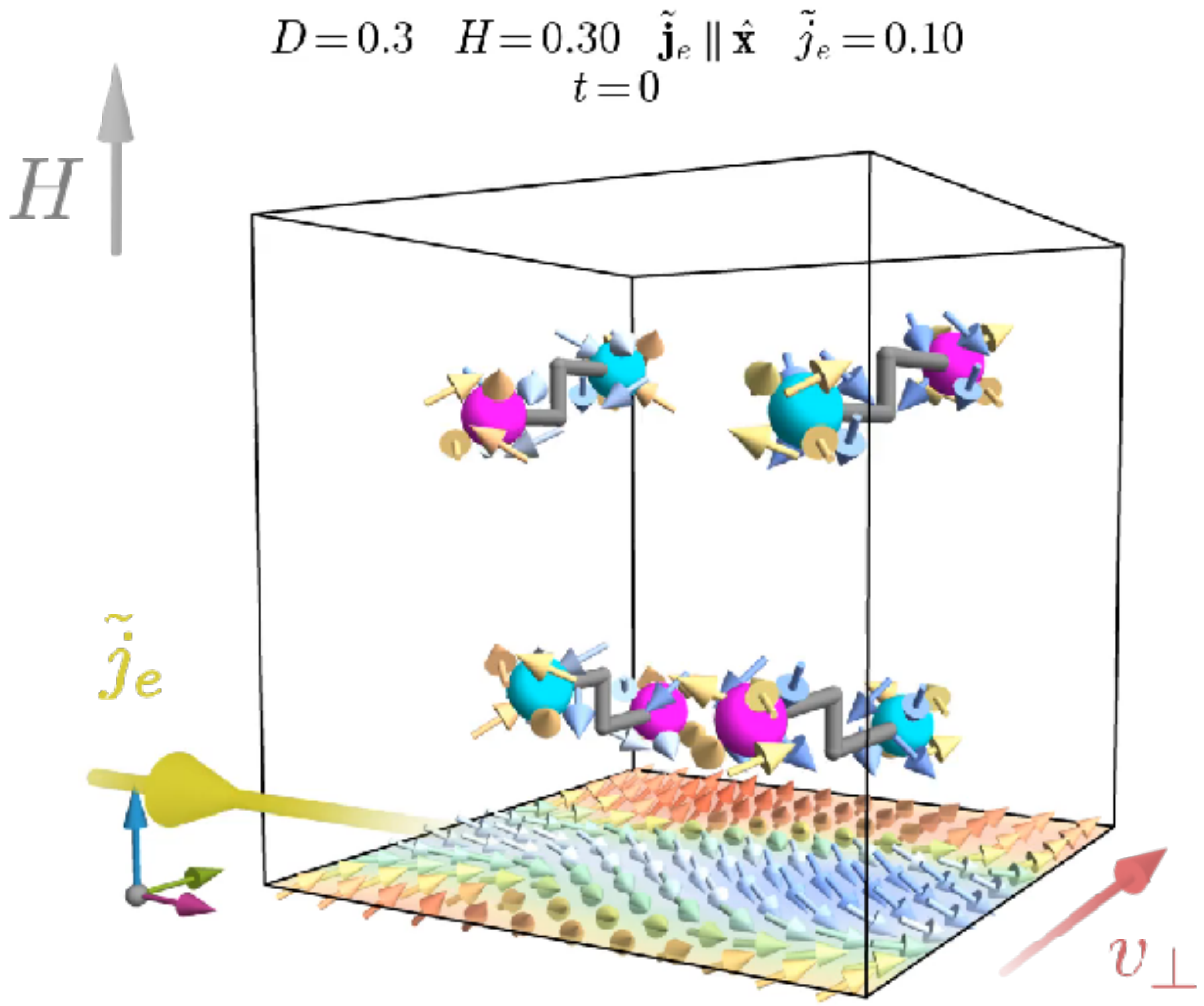
K. Shimizu, S. Okumura, Y. Kato, and Y. Motome, preprint (arXiv:2407.02983)

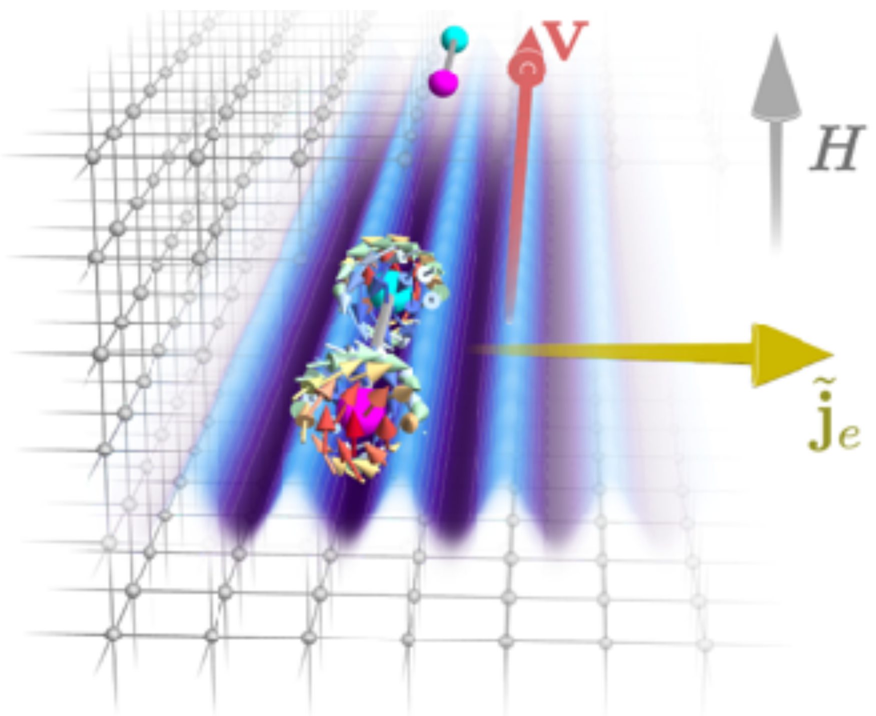
Zero-to-perfect toron Hall effect

zero toron Hall effect (only longitudinal)



perfect toron Hall effect (only transverse)





Lattice discretization significantly affects the motions of short-period torons.

K. Shimizu, S. Okumura, Y. Kato, and Y. Motome, preprint (arXiv:2407.02983)

Short summary

- We have proposed a new mechanism to stabilize topological spin textures, *itinerant frustration*.
  - generic mechanism for generating frustrated/multi-spin interactions in metallic magnets
  - many applications for new-generation topological spin crystals with extremely short period in centrosymmetric systems

- We presented applications to 2D skyrmion crystals and 3D toron crystals.
  - 2D: higher-order skyrmions, flexible change of the magnetic period, application to $GdRu_2Si_2$
  - 3D: application to $MnSi_{1-x}Gd_x$, phase diagrams, effect of magnetic fields, unconventional current-induced motions of torons

Conclusion

- spin moiré picture
  - superstructure, topology, and emergent electromagnetic field
  - spin moiré engineering: type, number, amplitude of waves, twist angle, phase shift
  - complete topological phase diagram for 2D skyrmions and 3D torons
- itinerant frustration
  - localized spin systems vs itinerant electron systems
  - effective long-range/multiple spin interactions by the Fermi surface effects
  - applications to 2D skyrmion crystals and 3D toron crystals

Both concepts will be useful for further exploration of topological spin superstructures and their physical properties.

➡ new multi-**q** states, new topological transitions, topological responses, etc.

Acknowledgement

Collaborators

Present and former members of my group (Tokyo):

Yutaka Akagi (UTokyo), **Satoru Hayami** (Hokkaido), **Yasuyuki Kato** (Fukui), Shoya Kasai, **Shun Okumura**, **Ryo Ozawa**, **Kotaro Shimizu**, Masafumi Udagawa (Gakushuin), …

Others:

Kipton Barros (LANL), Gia-Wei Chern (Virginia), Cristian D. Batista (Tennessee), …

Experimental colleagues:

Nguyen Duy Khanh, Shinichiro Seki, Taka-hisa Arima, Yoshinori Tokura (UTokyo/RIKEN), Yuuki Yasui (UTokyo), Christopher J. Butller, Tetsuro Hanaguri (RIKEN), Kirsten von Bergmann, Roland Wiesendanger (Hamburg), …

Fundings

科研費 KAKENHI

JST CREST