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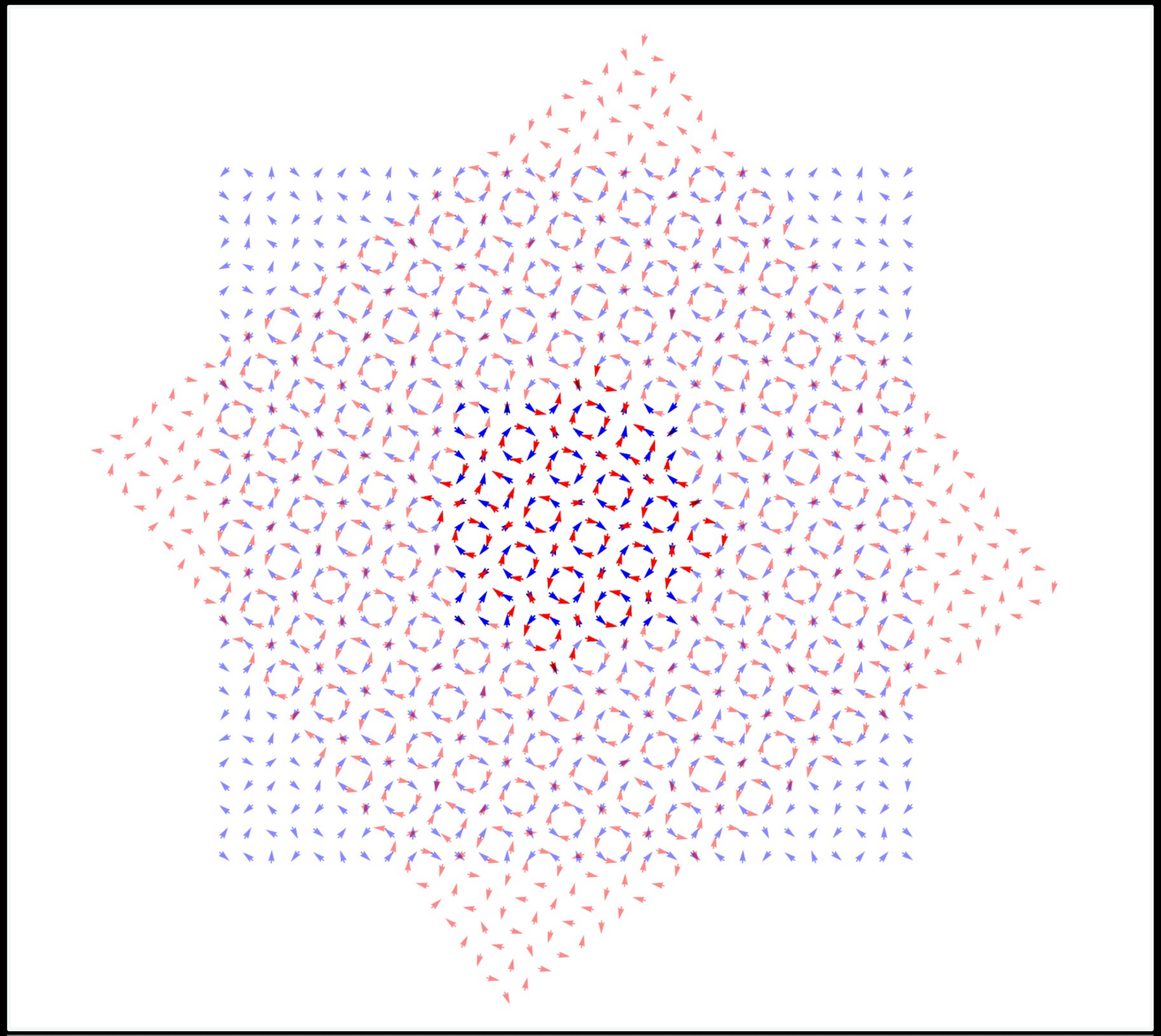
Chiral magnetic phases in Moire bilayers of Magnets



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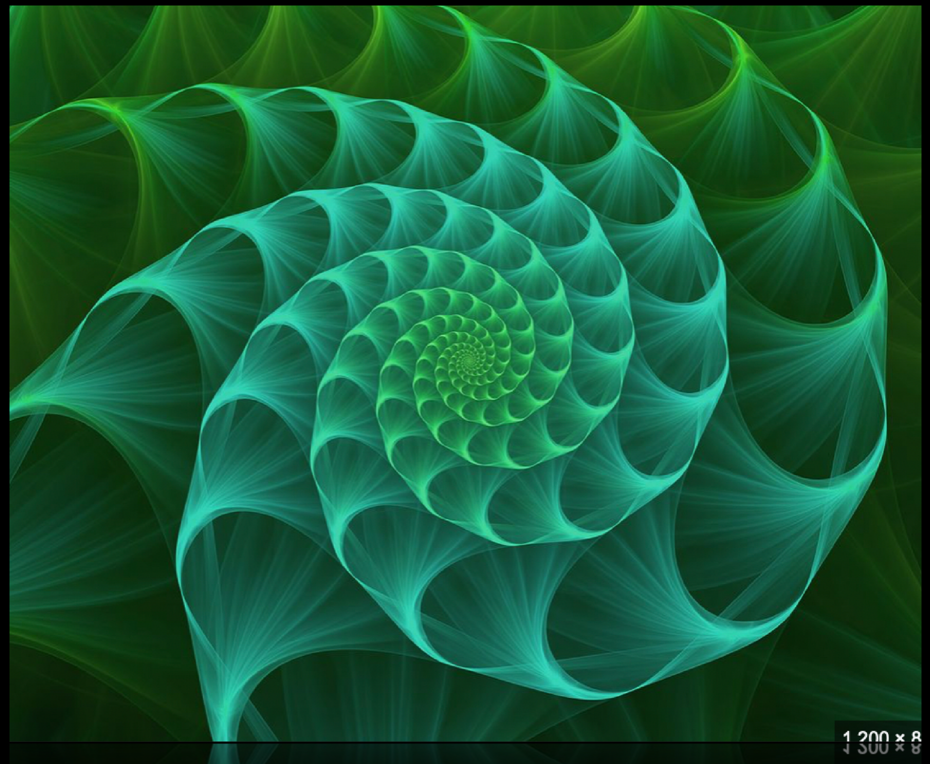
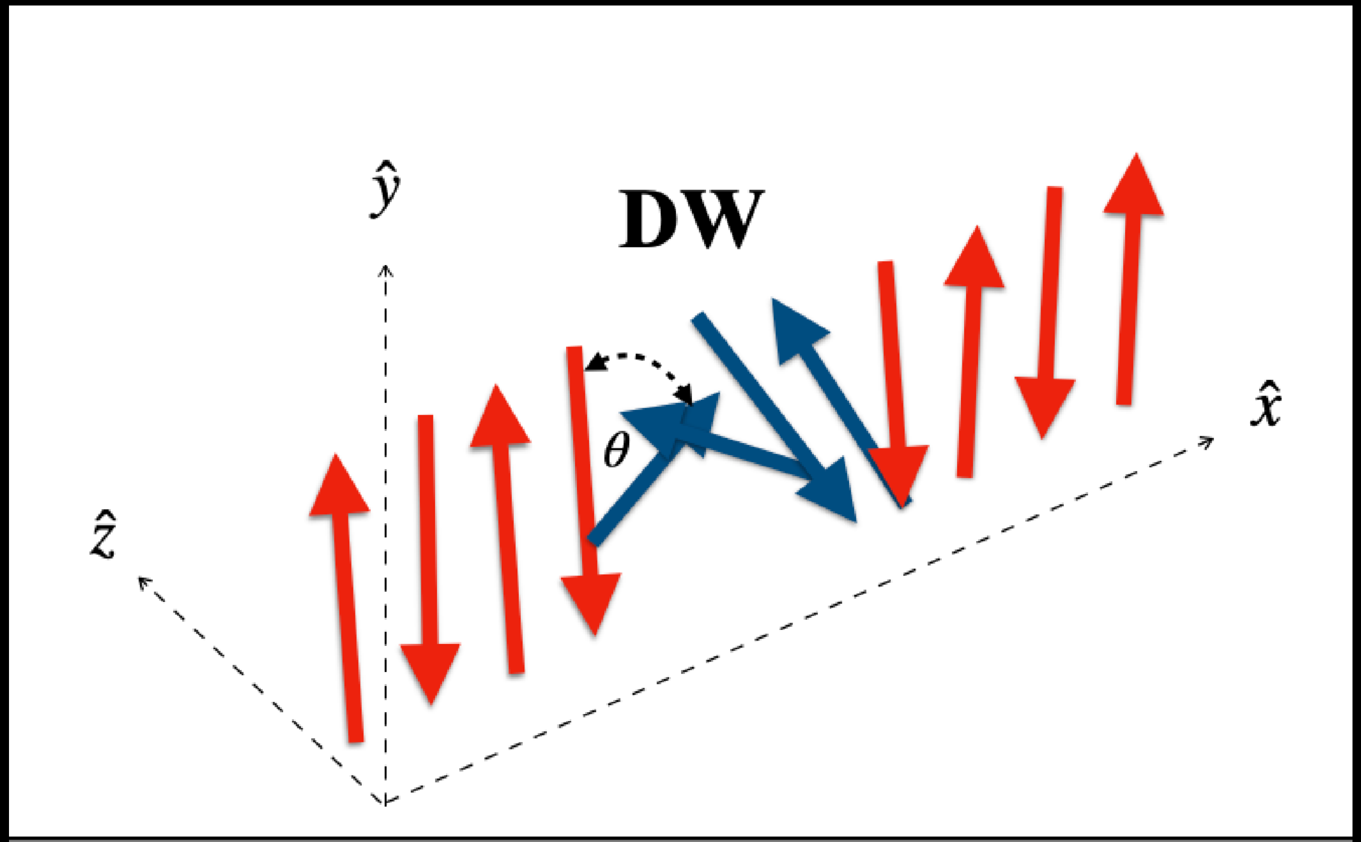
Chiral Fields





$$H_{i,j}^{(\mathrm{DM})} = \mathbf{D}_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j)$$

$$\mathcal{H}_{\mathrm{dip}} = \frac{\gamma}{2} \sum_{i \neq k = 1}^{n} \frac{\hat{\boldsymbol{m}}_i \cdot \hat{\boldsymbol{m}}_k - 3(\hat{\boldsymbol{m}}_i \cdot \hat{\boldsymbol{e}}_{ik})(\hat{\boldsymbol{m}}_k \cdot \hat{\boldsymbol{e}}_{ik})}{|\boldsymbol{r}_i - \boldsymbol{r}_k|^3}$$

Non-local interactions, connect the lattice with the spin symmetry anisotropically

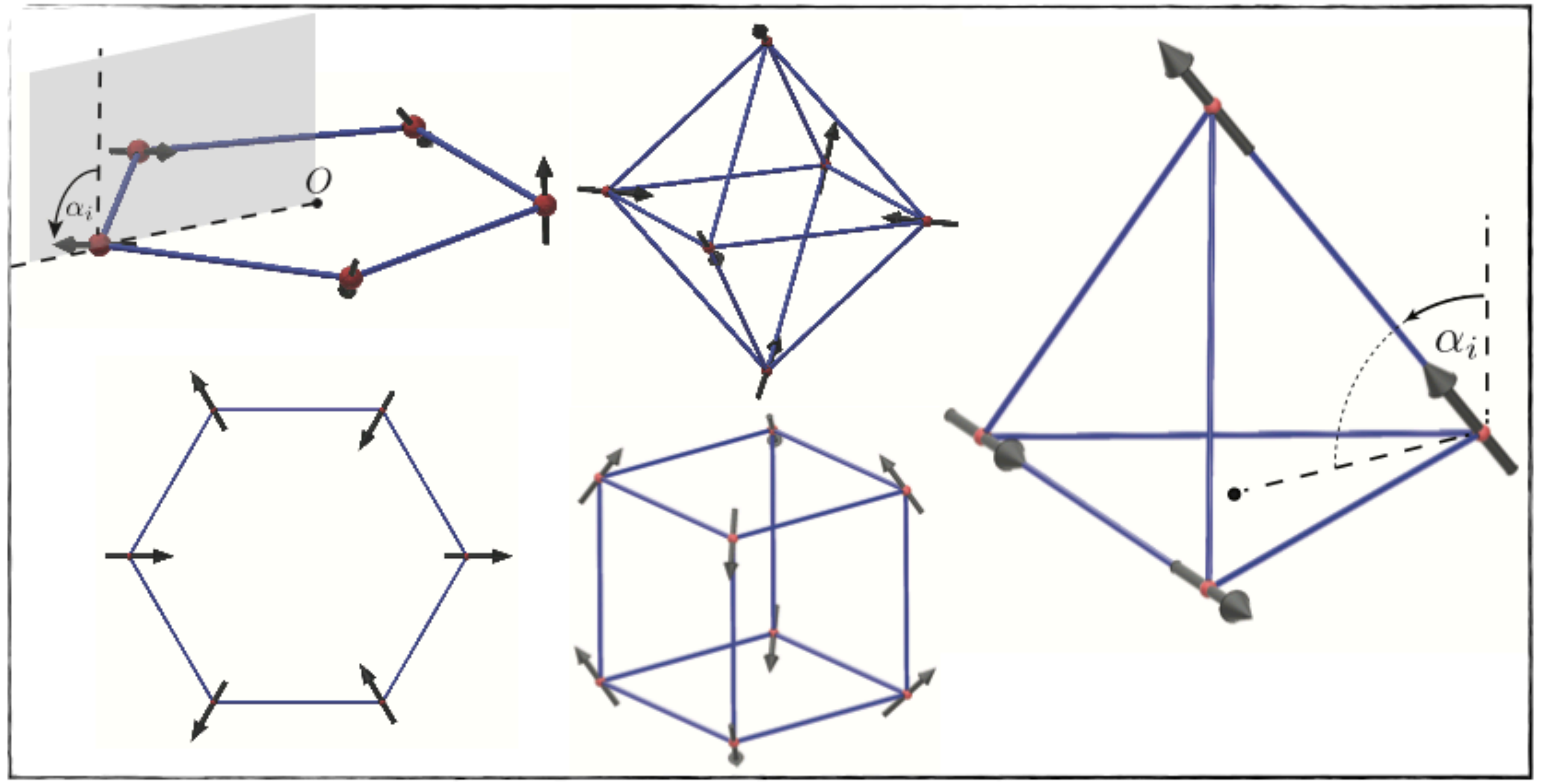
Chiral Phases due to Dipolar Interactions

$$E^{(\mathrm{odd})} = \gamma \sum_{k=1}^{s} \frac{1}{\Delta_k^3} \left[\sum_{i=-s}^{s} \hat{\boldsymbol{m}}_i \cdot \hat{\boldsymbol{m}}_{i+k} + \frac{3}{2} \tan\left(\frac{\pi}{n} k\right) \sum_{i=-s}^{s} (\hat{\boldsymbol{m}}_i \times \hat{\boldsymbol{m}}_{i+k}) \cdot \hat{\boldsymbol{z}} \right],$$

symmetric

Antisymmetric

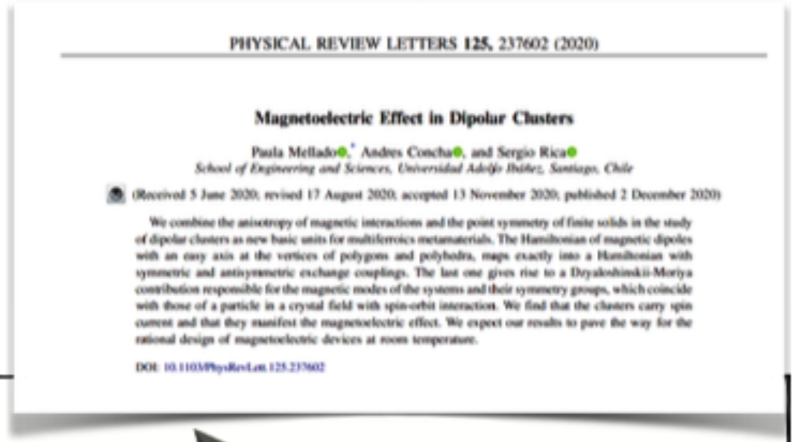
$$E^{(\mathrm{even})} = \gamma \sum_{k=1}^{n/2-1} \left[\frac{1}{\Delta_k^3} \sum_{i=1}^{n} \hat{\boldsymbol{m}}_i \cdot \hat{\boldsymbol{m}}_{i+k} + \frac{3}{2\Delta_k^3} \tan\left(\frac{\pi}{n} k\right) \sum_{i=1}^{n} (\hat{\boldsymbol{m}}_i \times \hat{\boldsymbol{m}}_{i+k}) \cdot \hat{\boldsymbol{z}} \right] + \frac{\gamma}{\Delta_{n/2}^3} \sum_{i=1}^{n/2} [-2\hat{\boldsymbol{m}}_i \cdot \hat{\boldsymbol{m}}_{i+n/2} + 3\hat{m}_i^z \hat{m}_{i+n/2}^z].$$



Chiral Phases due to Dipolar Interactions

Magnetoelectric Effect in Dipolar clusters

$$\mathscr{F}(\mathbf{E},\mathbf{H}) = \mathbf{F_0} - \frac{\epsilon_{\mathbf{ij}}}{\mathbf{8\pi}}\mathbf{E_iE_j} - \frac{\mu_{\mathbf{ij}}}{\mathbf{8\pi}}\mathbf{H_iH_j} - \mathbf{Q_{ij}E_iH_j} + \ldots$$


PHYSICAL REVIEW LETTERS 125, 237602 (2020)

Magnetoelectric Effect in Dipolar Clusters

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We combine the anisotropy of magnetic interactions and the point symmetry of finite solids in the study of dipolar clusters as new basic units for multiferroics metamaterials. The Hamiltonian of magnetic dipoles with an easy axis at the vertices of polygons and polyhedra, maps exactly into a Hamiltonian with symmetric and antisymmetric exchange couplings. The last one gives rise to a Dzyaloshinskii-Moriya contribution responsible for the magnetic modes of the systems and their symmetry groups, which coincide with those of a particle in a crystal field with spin-orbit interaction. We find that the clusters carry spin current and that they manifest the magnetoelectric effect. We expect our results to pave the way for the rational design of magnetoelectric devices at room temperature.

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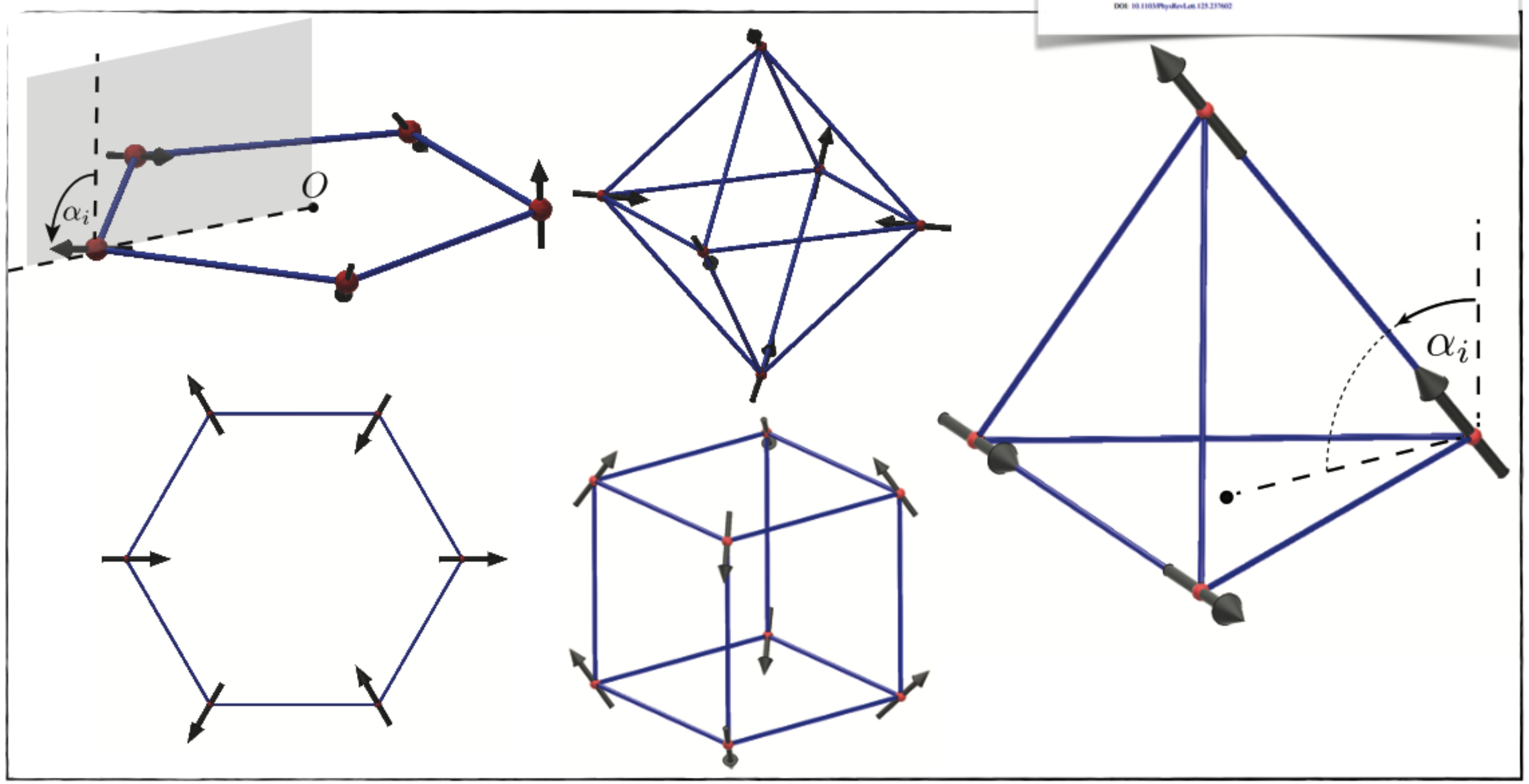
$$\mathbf{P} = \mathbf{QH}$$
$$\mathbf{M} = \underline{\mathbf{Q}}\mathbf{E}$$

$$\frac{\partial \boldsymbol{S}_i}{\partial t} = \frac{1}{i\hbar}[\boldsymbol{S}_i, H] = -\sum_k \boldsymbol{\mathcal{J}}_{ki},$$

$$\frac{\partial \boldsymbol{S}_i}{\partial t} = \boldsymbol{S}_i \times \frac{\partial H}{\partial \boldsymbol{S}_i}, \qquad \boldsymbol{\mathcal{J}}_{ki} = J\boldsymbol{S}_i \times \boldsymbol{S}_k,$$

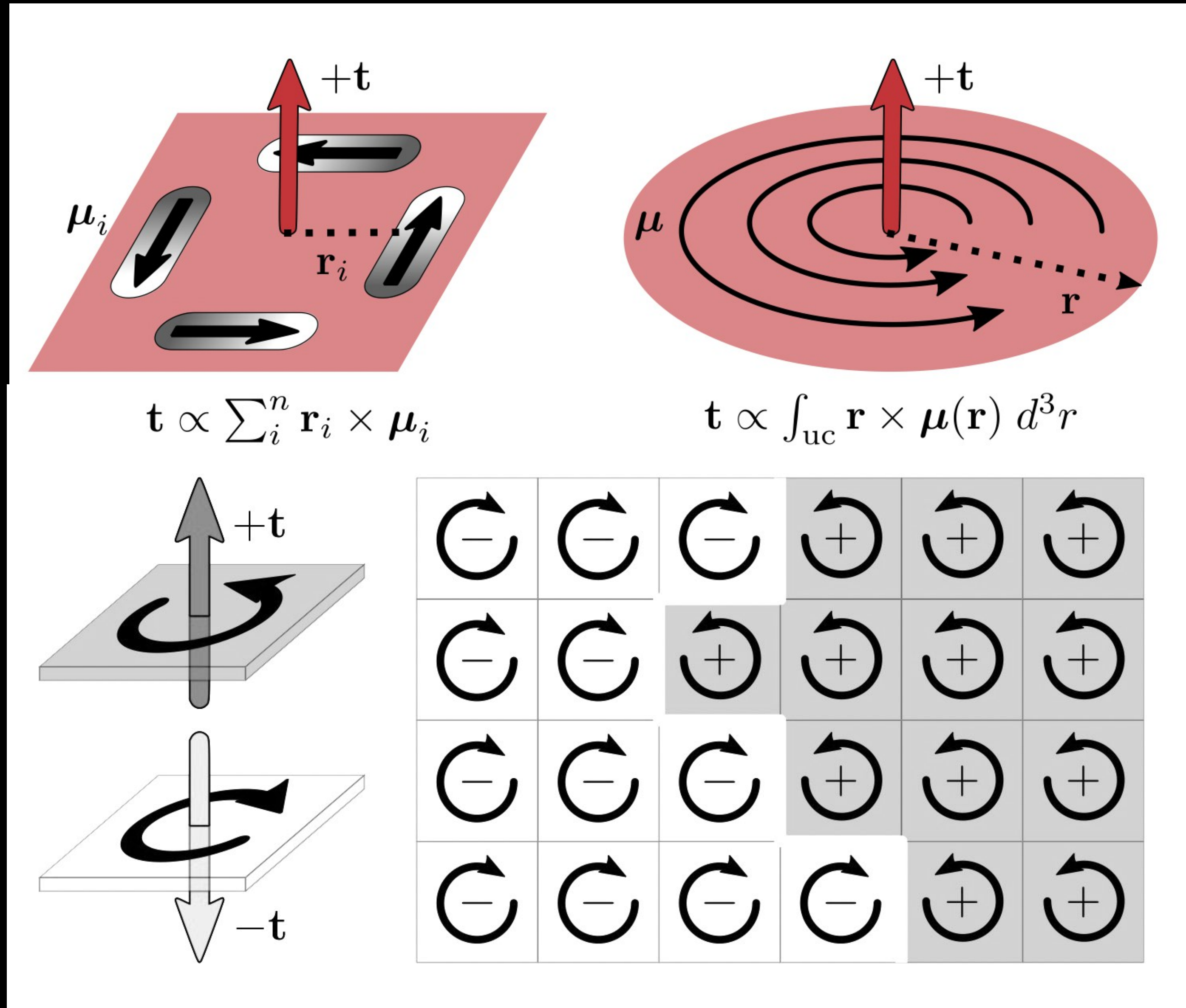
$$\boldsymbol{\mathcal{P}} = \sum_{k=1}^{\frac{n}{2}-1} \hat{\boldsymbol{x}}_{k,k+1} \times \boldsymbol{j}_s^{(k)}$$



Toroidal Orders

Zero magnetization and a spontaneously occurring toroidal moment

$$F(\boldsymbol{r}) = F_0(\boldsymbol{r}) - \boldsymbol{M}\cdot\boldsymbol{B}(\boldsymbol{r}) - \mathcal{Q}_{ij}\partial_i B_j(\boldsymbol{r}) + \cdots$$



Toroidization T is the antisymmetric part of magnetic quadrupolar moment density Qij

$$\mathcal{T}_k = \frac{1}{2}\epsilon_{ijk}\mathcal{Q}_{ij}$$

$$\mathcal{T}(\boldsymbol{r}) = -\lim_{\boldsymbol{B}(\boldsymbol{r})\to 0}\left.\frac{\partial F(\boldsymbol{r})}{\partial(\nabla\times\boldsymbol{B})}\right|_{\boldsymbol{B}(\boldsymbol{r})}$$

It can arise from the orbital and spin moments

Moire Bilayers of Dipoles

Two square lattices of magnetic dipoles stacked along z

$$U_d = \frac{g}{2}\sum_{\mu,\nu}^{2}\sum_{i,j}^{n}\frac{\hat{m}_i^{\mu}\cdot\hat{m}_j^{\nu} - 3(\hat{m}_i^{\mu}\cdot\hat{r}_{ij}^{\mu,\nu})(\hat{m}_j^{\nu}\cdot\hat{r}_{ij}^{\mu,\nu})}{|\mathbf{r}_i^{\mu}-\mathbf{r}_j^{\nu}|^3}$$

$$\hat{m}_i^{\mu} = (m_{i,x}^{\mu}, m_{i,y}^{\mu}) = (\cos\alpha_i^{\mu}, \sin\alpha_i^{\mu})$$

Twisted bilayer forms a crystal for a discrete set of commensurate rotation angles.

$$\theta_{p,q} = 2\arctan(p/q)$$

Moire pattern induced by twist has periodicity

$$\mathbf{v}_i = \hat{z}\times\mathbf{a}_i/(\sin\frac{\theta_{p,q}}{2})$$

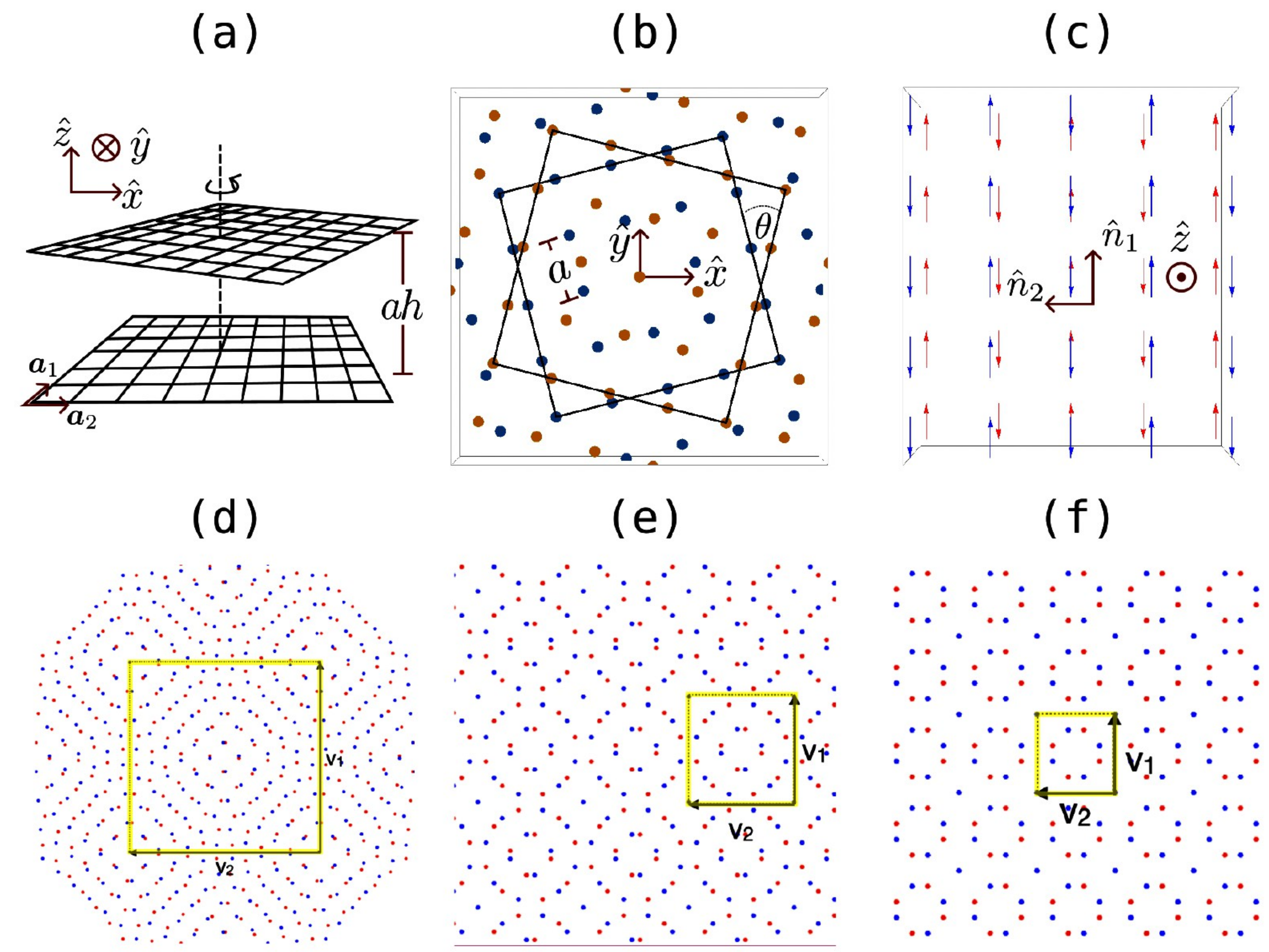


Figure 1: (a) Sketch of the twisted bilayer system. Each square layer is rotated at an angle $\theta/2$ around the z-axis represented by a dashed line. Layers are set apart by a distance ah. The lattice vectors for the bottom layer are denoted as $\mathbf{a}_1$ and $\mathbf{a}_2$. (b) Top view of the sites on both lattices. Blue dots represent sites in the bottom layer, while the yellow dots correspond to the sites in the top layer. The distance between adjacent sites on the same lattice is a. (c) Zig zag phase for the bilayer system with $\theta = 0$. Blue and red arrows show the magnetic moments for the top and bottom layers, respectively. The sites of each layer are directly on top of the sites of the other. The magnetic moments are shown slightly displaced for clarity. (d-f) Moire patterns obtained for twist angles (d) $\theta_{6,7} = 8.78°$, (e) $\theta_{2,3} = 22.62°$ and (f) $\theta_{1,2} = 36.87°$. The yellow square indicates the moire unit cell, with lattice vectors $\mathbf{v}_1$ and $\mathbf{v}_2$.

Magnetic orders in Moire Dipolar bilayers

Molecular Dynamics

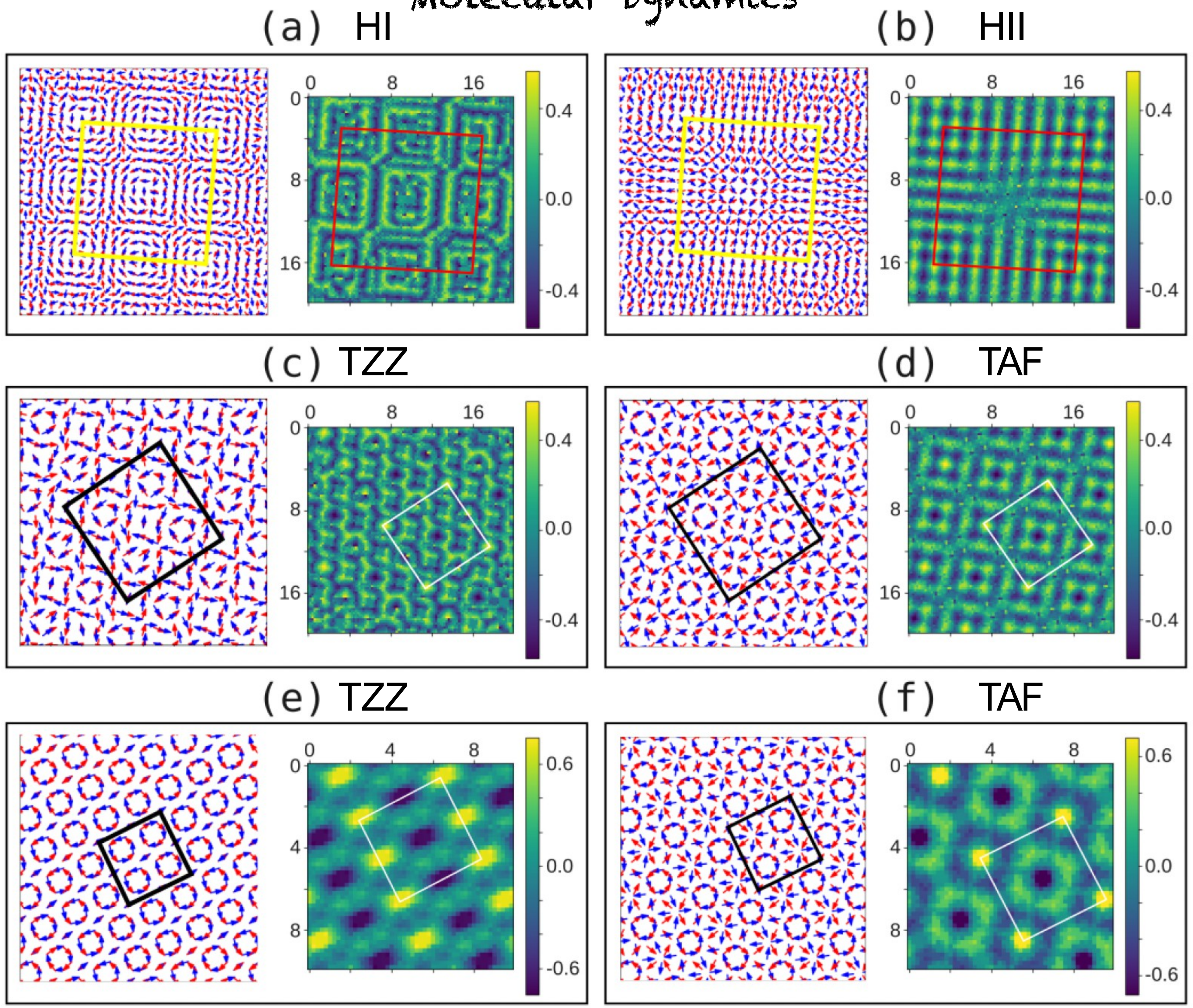


Figure 2: Magnetic phases were obtained at different values of h and $\theta_{p,q}$ for the bilayer system. At each subfigure, the left panel shows the magnetic moments in real space for the bottom (blue) and top (red) layers, and the right panel shows a density plot of the z component of the toroidal moment. The featured phases are (a) HI: $\theta_{6,7} = 8.78°$, $h = 0.1$, (b) HII: $\theta_{6,7} = 8.78°$, $h = 1.4$, (c) TZZ: $\theta_{2,3} = 22.62°$, $h = 0.1$, (d) TAF: $\theta_{2,3} = 22.62°$, $h = 1.4$, (e) TZZ: $\theta_{1,2} = 36.87°$, $h = 0.1$, (f) TAF: $\theta_{1,2} = 36.87°$, $h = 0.9$. Black and white squares indicate the magnetic unit cell, while yellow and red squares indicate the moire unit cell.

Order parameters

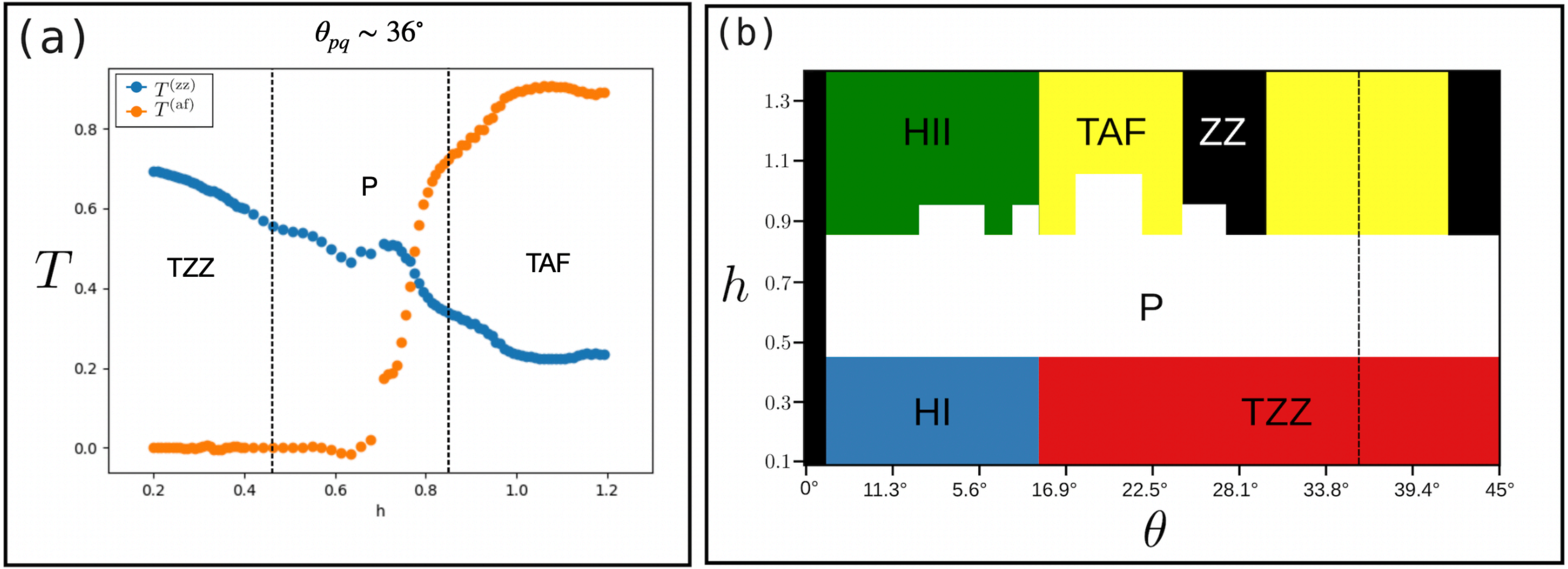
$$\mathrm{T}^{(\mathrm{af})} = \frac{1}{\mathrm{n_c}} \sum_{\mathrm{j}} (\mathbf{t}_{\mathrm{j,s_1}} - \mathbf{t}_{\mathrm{j,s_2}})$$

$$\mathbf{t} = \sum_i r_i \times m_i$$

$$\mathrm{T}^{(\mathrm{zz})} = \frac{1}{\mathrm{n_r}} \sum_{\mathrm{k}} (\mathrm{T}_{2\mathrm{k}+1} - \mathrm{T}_{2\mathrm{k}})$$

$$\mathrm{T_k} = \frac{\mathrm{n_r}}{\mathrm{n_c}} \sum_{\mathrm{j}} \mathbf{t}_{\mathrm{j}}^{\mathrm{k}}$$

Phase Diagram in terms of h and θ

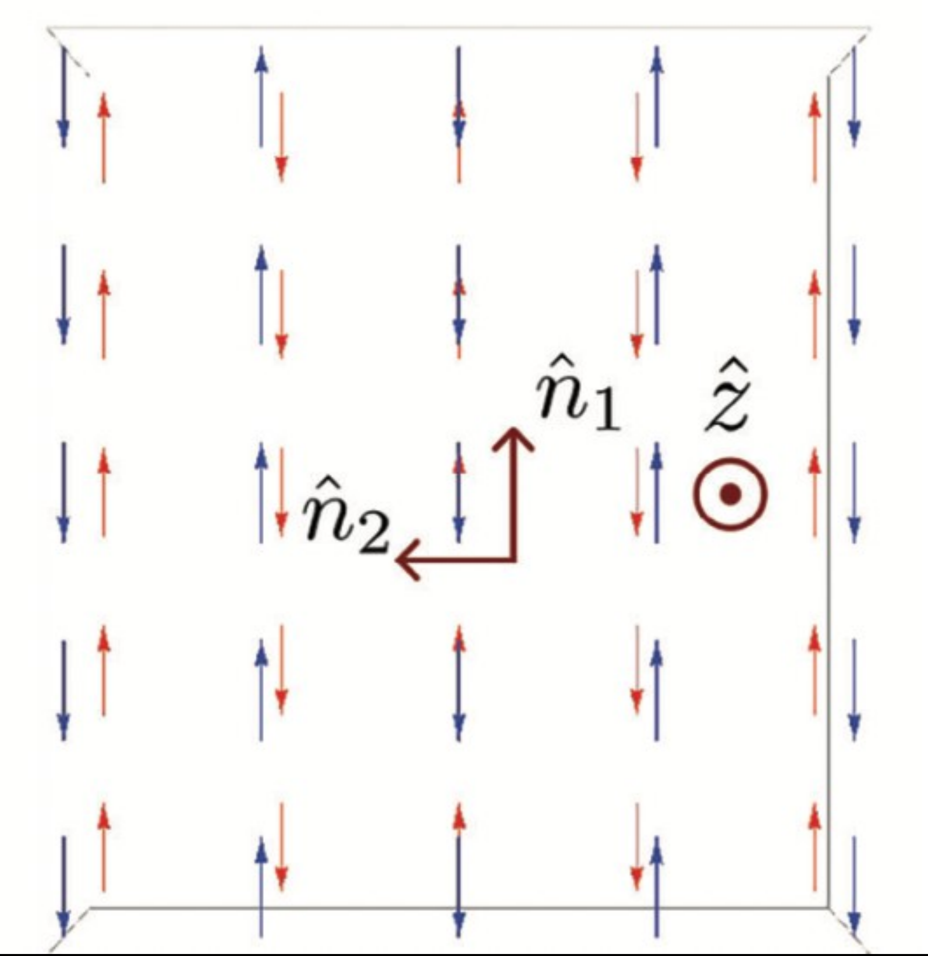


Interlayer Magnetic field at zero twist

ZZ phase

$$\mathbf{B}_i = \sum_m \left[\sum_{n \in s_1} \frac{2m^2 - n^2 - h^2}{(n^2 + m^2 + h^2)^{\frac{5}{2}}} - \sum_{n' \in s_2} \frac{2m^2 - n'^2 - h^2}{(n'^2 + m^2 + h^2)^{\frac{5}{2}}} \right] \hat{y}$$

$\theta_{pq} = 0$



Bi: Magnetic field due to dipoles in layer μ on sites of layer ν. $\hat{x}$ and $\hat{z}$ components of Bi cancel out

$$B_{\theta=0}(x, y) \sim \frac{x^2}{(x^2 + y^2 + h^2)^{5/2}}$$

$$\mathcal{T}(\theta = 0) = \sum_{i,j} \mathbf{m}_j^\nu \times \mathbf{B}^\mu_{i,(\theta=0)} = 0$$

At zero twist Bi is staggered along ferromagnetic chains

Twist affects the internal Magnetic field

in two ways

1) It is a vector quantity, its components change like those of other vectors

$$\mathbf{B}' = \mathbf{B} + \boldsymbol{\theta} \times \mathbf{B}$$

2) B(r) depends on coordinates r, which in terms of rotated ones r′ changes according to

$$\mathbf{B}'(\mathbf{r}') = \mathbf{B}(\mathbf{r}) = \mathbf{B}(\mathbf{r}' - \boldsymbol{\theta} \times \mathbf{r}')$$

$$= \mathbf{B}(\mathbf{r}') - [(\boldsymbol{\theta} \times \mathbf{r}') \cdot \nabla']\mathbf{B}(\mathbf{r}')$$

The overall infinitesimal change in B(r)

$$\delta \mathbf{B}^{\text{twist}} = \boldsymbol{\theta} \times \mathbf{B} - \boldsymbol{\theta} \cdot (\mathbf{r} \times \nabla)\mathbf{B}$$

Toroidal Conjugate Field

Twist arises a new $\hat{x}$ component

$$\delta \mathbf{B}_i^{\text{twist}} = \theta \left[B_{i,(\theta=0)}, \left(y \frac{\partial B_{i,(\theta=0)}}{\partial x} - x \frac{\partial B_{i,(\theta=0)}}{\partial y} \right), 0 \right],$$

Component orthogonal to chains gives rise to torque

$$\mathcal{T}^{\text{twist}}(h, \theta) = m_0 \theta B_{(\theta=0)} \hat{\mathbf{z}}$$

Gradient expansion F(r) of layer μ in the inhomogeneous magnetic field created by dipoles in layer ν (δB^{twist} small and varies slowly in space.)

$$T_k = \frac{1}{2} \epsilon_{ijk} Q_{ij}$$

$$F(r) = \mathbf{F}_0(r) - N \cdot \delta \mathbf{B}^{\text{twist}}(r) - Q_{ij} \partial_i (\delta \mathbf{B}_j^{\text{twist}}(r)) + \ldots$$

Toroidal Conjugate Field

linear-response

$$\mathbf{T}(r) = -\lim_{\delta\mathbf{B}^{\text{twist}}\to 0} \frac{\partial F(r)}{\partial(\nabla \times \delta\mathbf{B}^{\text{twist}})}\bigg|_{\delta\mathbf{B}^{\text{twist}}}$$

$$\mathbf{G} = \nabla \times \delta\mathbf{B}^{\text{twist}}$$

$\hat{z}$ component couples to the toroidal moment

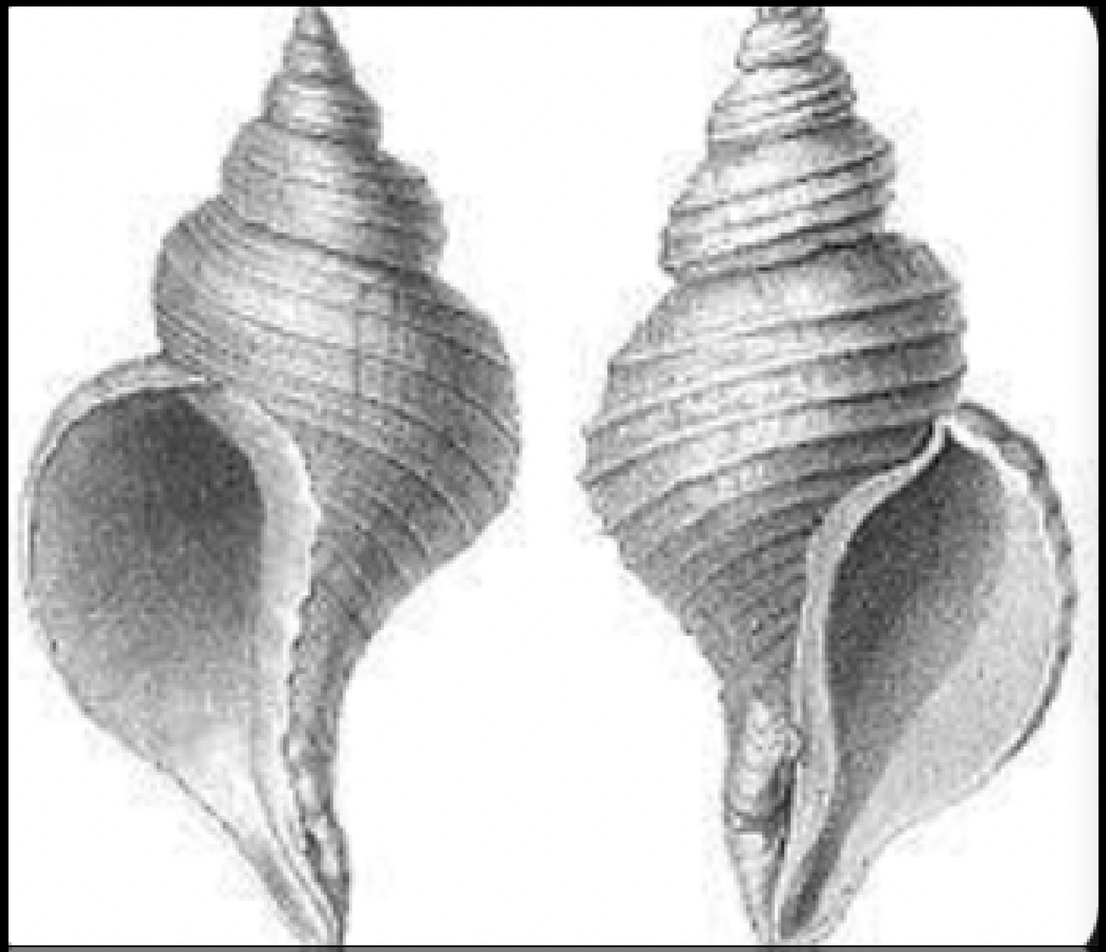
$$G_z = (\partial_x \delta\mathbf{B}_y^{\text{twist}} - \partial_y \delta\mathbf{B}_x^{\text{twist}}) \sim \theta\partial_y B_{\theta=0}$$

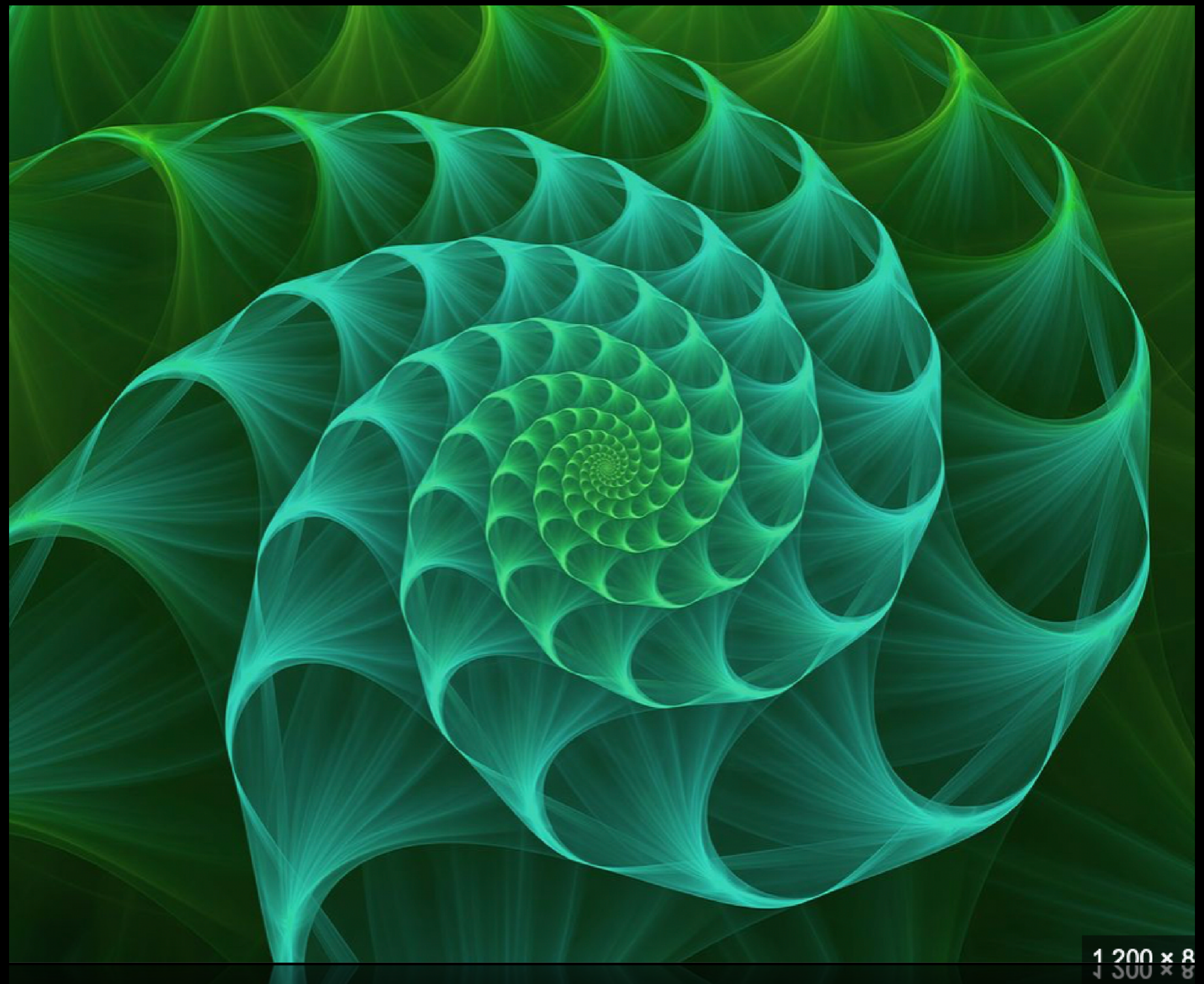
Conclusions

- Toroidization can be induced by a twist, it can be tuned by h and θ
- Twist between layers triggers a net magnetic torque orthogonal to the bilayer system. It leads to new magnetic orders.
- New magnetic phases are magnetization-free, break time, and inversion symmetry and depict chiral motifs that realize a toroidal moment orthogonal to the bilayer.
- Conjugate field to the toroidization is proportional to the twist.

Magnetic Chirality

Chirality: all mirror symmetries are broken (or absent).

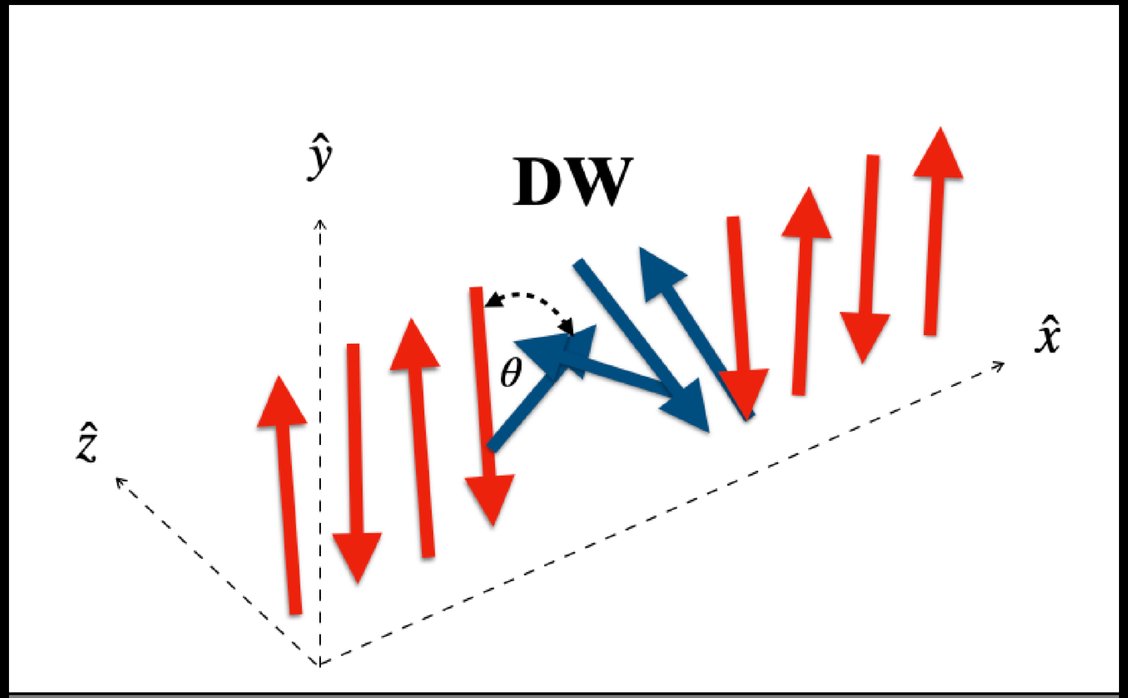




Dzyaloshinskii_Moriya interaction

(DMI) is a chiral field: it twists the

magnetization and leads to winding textures,



Passing through the chiral spin textures, electrons undergo

precession and exchange angular momentum with it.

Semiclassical theory of spin toroidization

PHYSICAL REVIEW B **97**, 134423 (2018)

Microscopic theory of spin toroidization in periodic crystals

Yang Gao,[1] David Vanderbilt,[2] and Di Xiao[1]
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Spatial inhomogeneity of B(r)

$$\hat{H}_{\mathrm{F}} = \hat{H}(i\hbar\partial_{\boldsymbol{r}},\boldsymbol{r}) - \frac{g\mu_B}{\hbar}\boldsymbol{B}(\boldsymbol{r})\cdot\hat{\boldsymbol{s}}.$$

translational invariant

Bloch electrons subject to perturbations varying slowly in space

Local approximation: the system can be described by a set of local Hamiltonians Hc that respect the lattice translational symmetry and with eigenstates that are Bloch states.

1) The phase space density of states D is modified through the Berry curvature between the real and momentum space.
2) The band energy is affected by the spatial inhomogeneity

$$\varepsilon_0' = \tilde{\varepsilon}_0 + \mathrm{Im}\langle\partial_{k_{ci}}\tilde{u}_0|(\tilde{\varepsilon}_0 - \hat{H}_c)|\partial_{r_{ci}}\tilde{u}_0\rangle.$$

Correction to free energy density at first order

$$\delta F = -\int^{\mu}\frac{d\boldsymbol{k}_c}{(2\pi)^3}\,\mathrm{Im}\langle\partial_{k_{ci}}\tilde{u}_0|(\tilde{\varepsilon}_0 + \hat{H}_c - 2\mu)|\partial_{r_{ci}}\tilde{u}_0\rangle.$$

$$\mathcal{T}(\boldsymbol{r}) = -\lim_{\boldsymbol{B}(\boldsymbol{r})\to 0}\left.\frac{\partial F(\boldsymbol{r})}{\partial(\nabla\times\boldsymbol{B})}\right|_{\boldsymbol{B}(\boldsymbol{r})}.$$

$$\mathcal{T} = \frac{1}{2}\int^{\mu}\frac{d\boldsymbol{k}}{(2\pi)^3}\,\mathrm{Im}\langle\partial_{\boldsymbol{k}}\tilde{u}_0|\times(\tilde{\varepsilon}_0 + \hat{H}_c - 2\mu)|\partial_{\boldsymbol{B}}\tilde{u}_0\rangle\big|_{\boldsymbol{B}\to 0}.$$

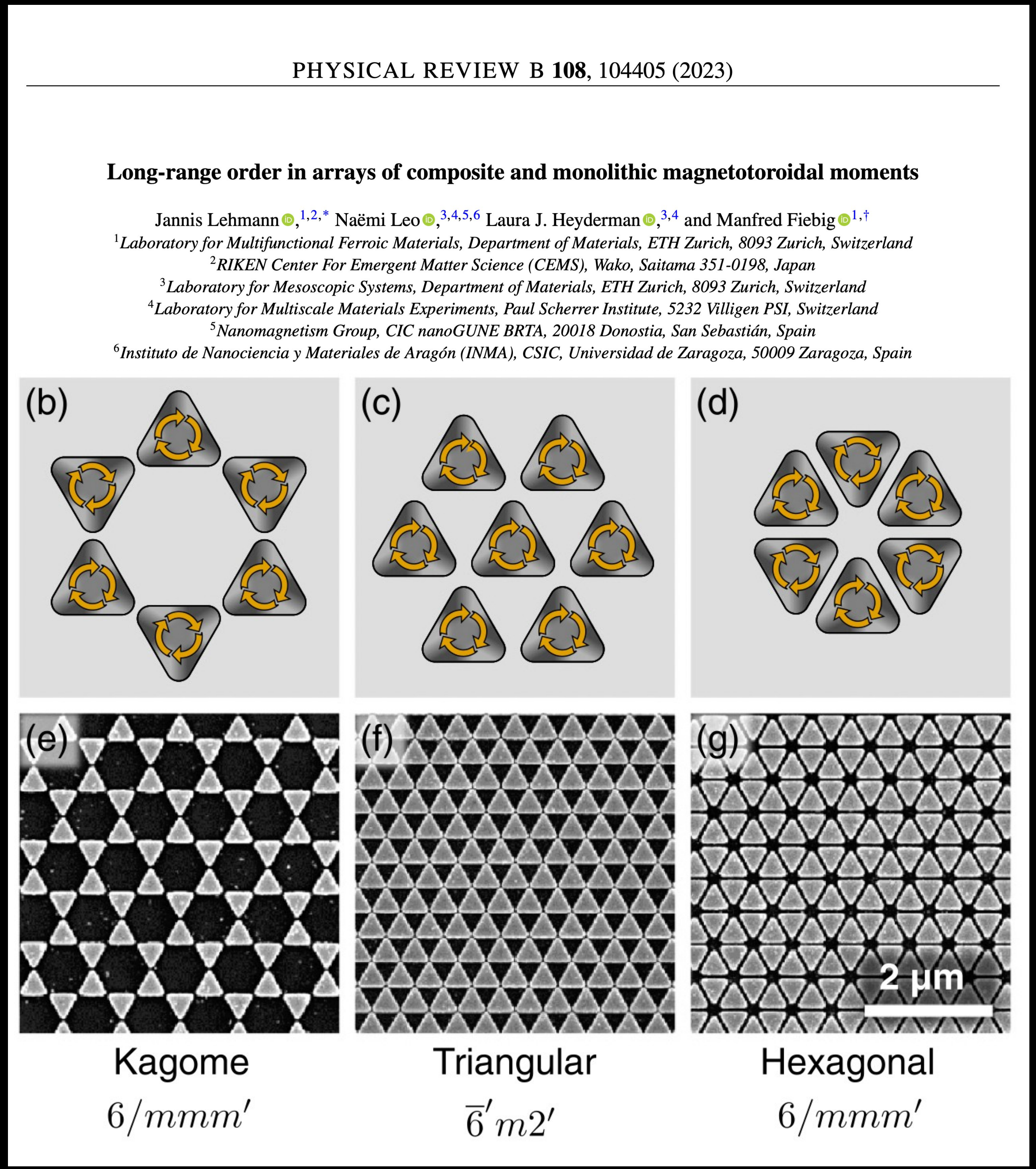
Variation of the j-th component of B(r) induces a change of the polarization: magnetoelectric polarizibility

$$\Delta P_i = e\int\frac{d\boldsymbol{k}dB_j}{(2\pi)^3}\,\mathrm{Im}\langle\partial_{k_i}\tilde{u}_0|\partial_{B_j}\tilde{u}_0\rangle.$$

$$\alpha_{ij} = \left.\frac{\partial P_i}{\partial B_j}\right|_{\boldsymbol{B}\to 0} = e\int\frac{d\boldsymbol{k}}{(2\pi)^3}\,\mathrm{Im}\langle\partial_{k_i}\tilde{u}_0|\partial_{B_j}\tilde{u}_0\rangle\big|_{\boldsymbol{B}\to 0}.$$

$$e\frac{\partial\mathcal{T}_k}{\partial\mu} = -\frac{1}{2}\epsilon_{ijk}\alpha_{ij},$$

- Order of magnetotoroidal moments in arrays of nanoscale building blocks made from a soft-magnetic alloy. Local control of ferrotoroidicity achieved by scanning a magnetic tip


PHYSICAL REVIEW B **108**, 104405 (2023)

Long-range order in arrays of composite and monolithic magnetotoroidal moments

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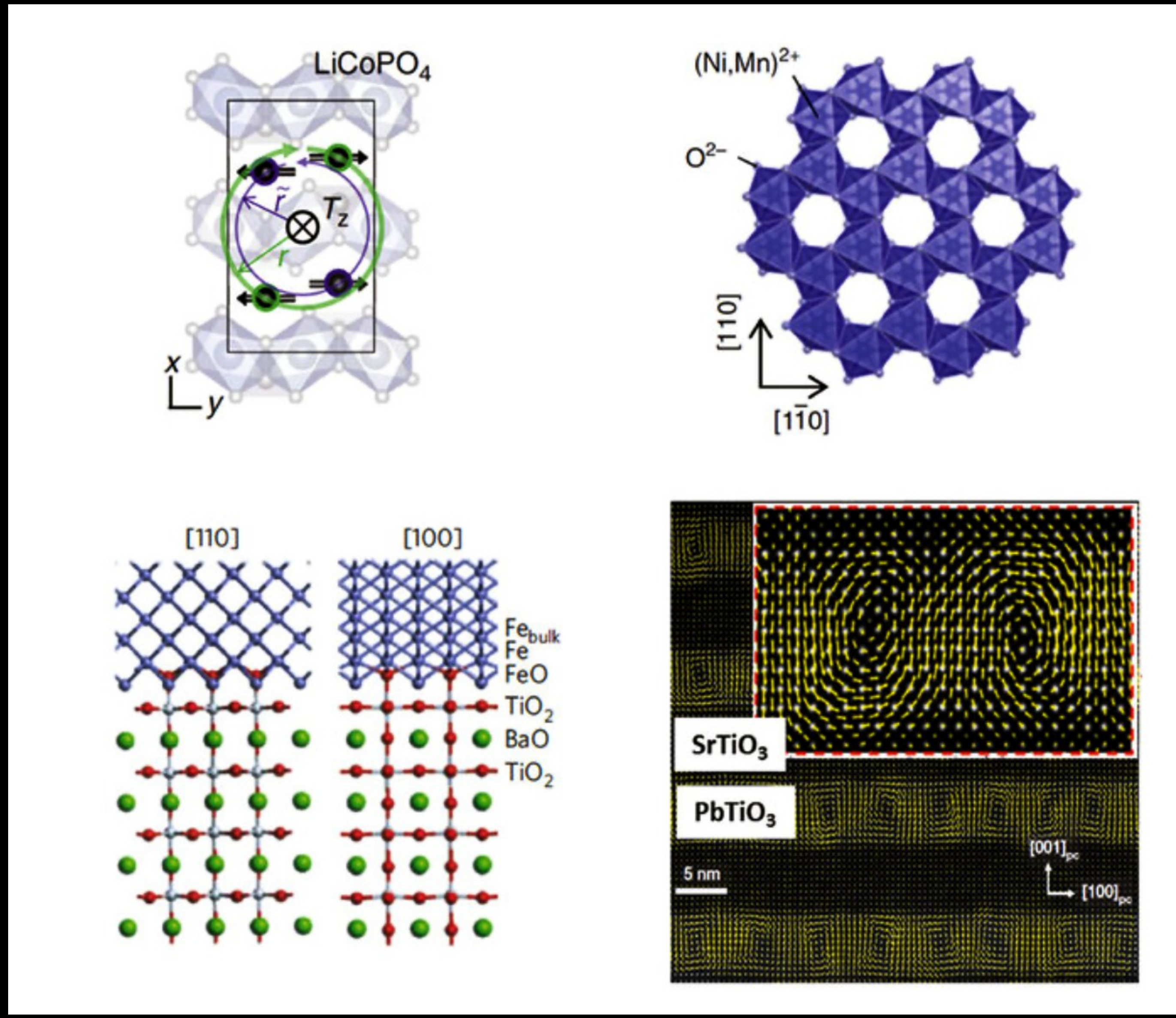
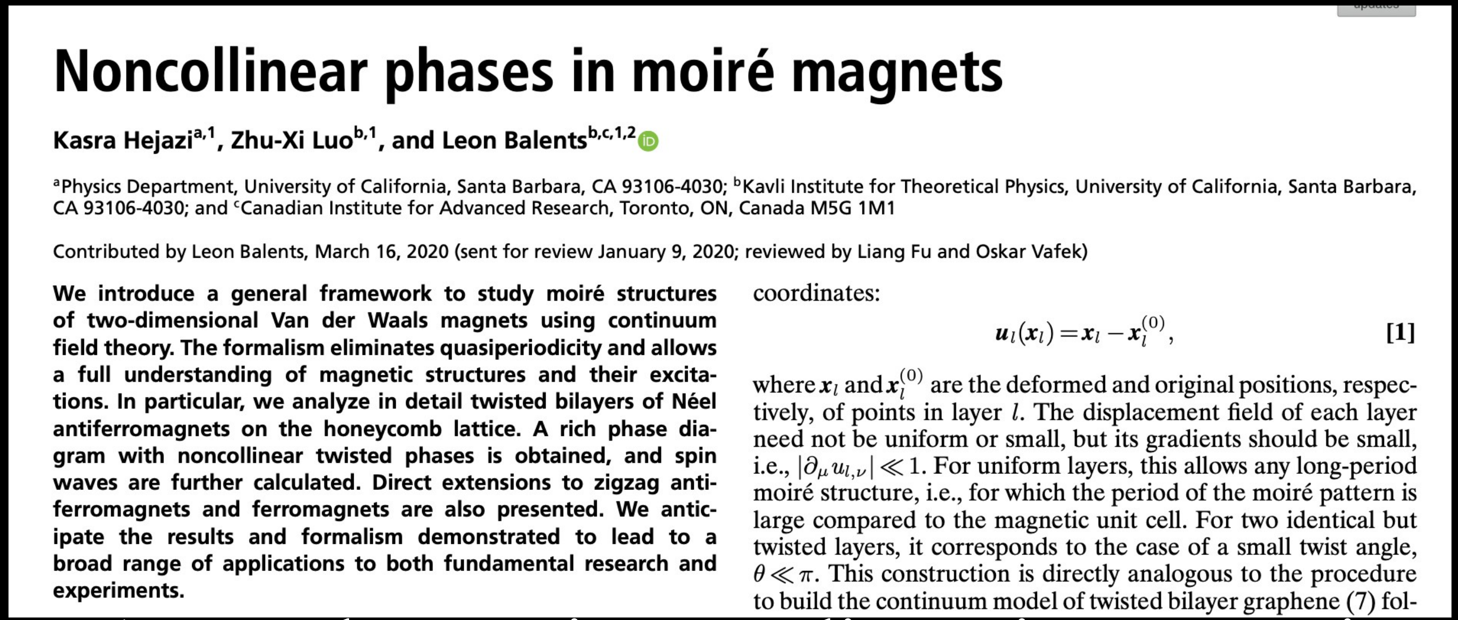
$6/mmm'$ $\quad \bar{6}'m2'$ $\quad 6/mmm'$

- Ferrotoroidal order coexisting with AFM in LiCoPO4

- Linear coupling of polarization with antiferromagnetic order parameter in MnTiO3




Noncollinear phases in moiré magnets

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Contributed by Leon Balents, March 16, 2020 (sent for review January 9, 2020; reviewed by Liang Fu and Oskar Vafek)

We introduce a general framework to study moiré structures of two-dimensional Van der Waals magnets using continuum field theory. The formalism eliminates quasiperiodicity and allows a full understanding of magnetic structures and their excitations. In particular, we analyze in detail twisted bilayers of Néel antiferromagnets on the honeycomb lattice. A rich phase diagram with noncollinear twisted phases is obtained, and spin waves are further calculated. Direct extensions to zigzag antiferromagnets and ferromagnets are also presented. We anticipate the results and formalism demonstrated to lead to a broad range of applications to both fundamental research and experiments.

coordinates:

$$\boldsymbol{u}_l(\boldsymbol{x}_l) = \boldsymbol{x}_l - \boldsymbol{x}_l^{(0)}, \qquad [1]$$

where $\boldsymbol{x}_l$ and $\boldsymbol{x}_l^{(0)}$ are the deformed and original positions, respectively, of points in layer l. The displacement field of each layer need not be uniform or small, but its gradients should be small, i.e., $|\partial_\mu u_{l,\nu}| \ll 1$. For uniform layers, this allows any long-period moiré structure, i.e., for which the period of the moiré pattern is large compared to the magnetic unit cell. For two identical but twisted layers, it corresponds to the case of a small twist angle, $\theta \ll \pi$. This construction is directly analogous to the procedure to build the continuum model of twisted bilayer graphene (7) fol-

- AF and F spin couplings in magnetic insulators can give rise to noncollinear magnetic textures for small twist angles