Electronic Orbital Currents and Polarization in Mott Insulators

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L. N. Bulaevskii et al, Phys. Rev. B 78, 024402 (2008).

Motivations

- Complete the picture by showing that spin ordering in Mott insulators can drive **orbital electric currents** in addition to charge-density-waves of purely electronic origin.

- Provide a clear connection between “scalar spin chirality” and orbital currents or magnetic moments.

- Identify the spin textures that are accompanied by ordering of charge or electric currents and understand the crucial role of geometric frustration.

- Find unusual physical responses of the charge degrees of freedom in Mott insulators.

- Understand the interplay between electric field and topological defects (domain walls) of certain magnetic orderings.

Outline

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- Provide a clear connection between “scalar spin chirality” and orbital currents or magnet moments.

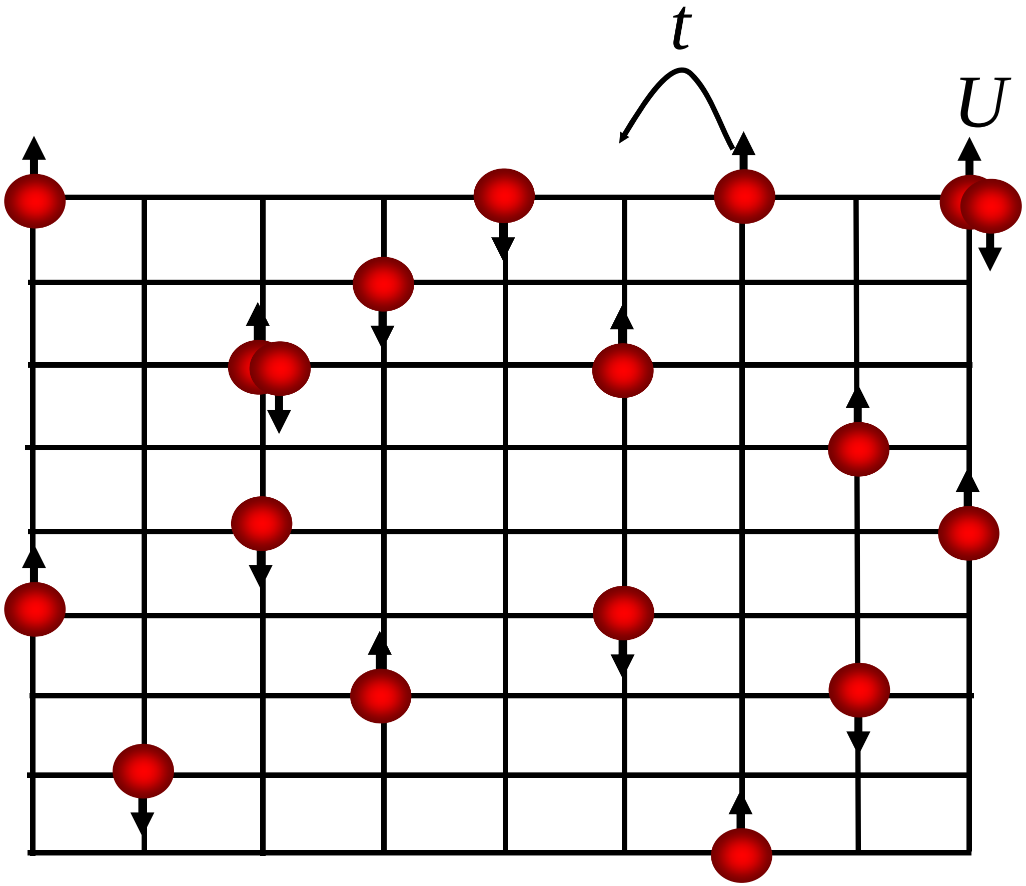
- Identify the spin textures that are accompanied by ordering of charge or electric currents and understand the crucial role of geometric frustration.

- Find unusual physical responses of the charge degrees of freedom in Mott insulators.

- Understand the interplay between electric field and topological defects (domain walls) of certain magnetic orderings.

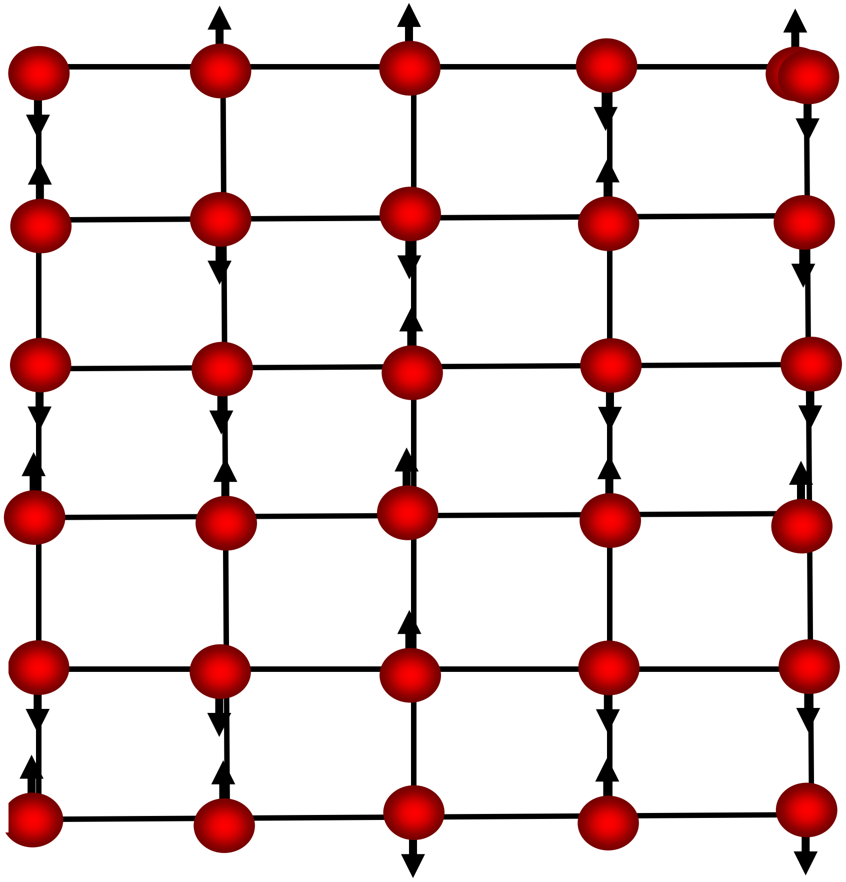
Hubbard Model

$$H = -\sum_{\langle ij\rangle\sigma} t(c^\dagger_{i\sigma}c_{j\sigma} + c^\dagger_{j\sigma}c_{i\sigma}) + \frac{U}{2}\sum_i (n_i - 1)^2,$$



Large U and Half-filling

$$H = -\sum_{\langle ij\rangle\sigma} t(c^{\dagger}_{i\sigma}c_{j\sigma} + c^{\dagger}_{j\sigma}c_{i\sigma}) + \frac{U}{2}\sum_{i}(n_i - 1)^2,$$



$$U/t = \infty$$

2^N degenerate ground states, where N is the number of sites

$$U/t >> 1$$

Spin degeneracy is lifted by processes of order *t^2/U*

$$|\tilde{\psi}_\nu\rangle \rightarrow |\psi_\nu\rangle$$

Excited or polar eigenstates

$$|\tilde{\phi}_\nu\rangle \xrightarrow{\text{Adiabatically connected}} |\phi_\nu\rangle$$

Large U and Half-filling

$$|\psi_\nu\rangle = e^{-S}|\tilde{\psi}_\nu\rangle$$

Where S is an anti-Hermitian, $S = -S^\dagger$, operator so e^{-S} is unitary. The goal is to find S as an expansion in powers of *t/U*.

$$\langle\psi_i|O|\psi_j\rangle = \langle\tilde{\psi}_i|\tilde{O}|\tilde{\psi}_j\rangle$$

$$\boxed{\tilde{O} = \mathcal{P}e^{S}Oe^{-S}\mathcal{P}}$$

Where $\mathcal{P}$ s a projector onto the subspace gSerated by the states . $|\tilde{\psi}_\nu\rangle$efore, the effective operator is a fun$\tilde{O}$on of spin operators.

Determination of S

To obtain S to first order in t/U we impose the condition: must $\tilde{H}$ not have terms that are linear in t.

$$\tilde{H} = \mathcal{P} e^{S} H e^{-S} \mathcal{P}$$

$$\tilde{H} = \mathcal{P}(1 + S + \frac{S^2}{2} + ...)(H_t + H_U)(1 - S + \frac{S^2}{2} + ...)\mathcal{P}$$

$$\tilde{H} = \mathcal{P}(H_U + H_t + [S, H_U] + [S, H_t] - S H_U S + \{\frac{S^2}{2}, H_U\} + ...)\mathcal{P}$$

The cancellation of the linear in t terms implies:

$$\boxed{H_t + [S, H_U] = 0}$$

$$\boxed{\langle\tilde{\phi}_\nu|S|\tilde{\psi}_\nu\rangle = \langle\tilde{\phi}_\nu|\frac{H_t}{U}|\tilde{\psi}_\nu\rangle} \qquad \boxed{\langle\tilde{\psi}_\nu|S|\tilde{\phi}_\nu\rangle = -\langle\tilde{\psi}_\nu|\frac{H_t}{U}|\tilde{\phi}_\nu\rangle}$$

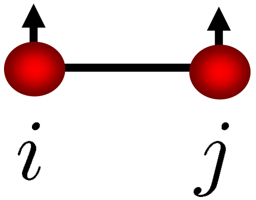
Effective Spin Hamiltonian

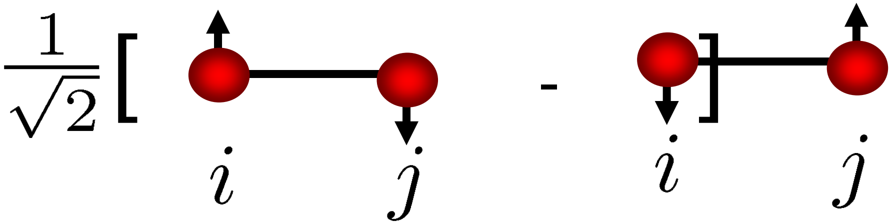
$$\langle\tilde{\phi}_\nu|\tilde{H}^{(2)}|\tilde{\psi}_\nu\rangle = \mathcal{P}([S,H_t] - SH_US)\mathcal{P}$$

Where we used that $H_U\mathcal{P} = 0$

$$\boxed{\tilde{H}^{(2)} = \sum_{ij} J(\mathbf{S}_i\cdot\mathbf{S}_j - 1/4)}$$

Where $S_i^\eta = \sum_{\mu,\nu} c^\dagger_{i\mu}\sigma^\eta_{\mu\nu}c_{i\nu}$ and . $J = 4\frac{t^2}{U}$ (Exercise)





Triplet $E = 0$

Singlet $E = -J$

Current and Charge Density Operators

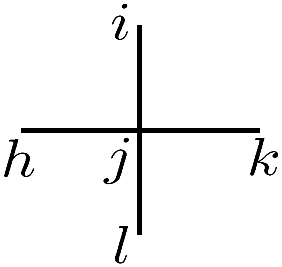
$$\mathbf{I}_{ij} = \frac{iet_{ij}\mathbf{r}_{ij}}{\hbar r_{ij}} \sum_\sigma (c^\dagger_{j\sigma} c_{i\sigma} - c^\dagger_{i\sigma} c_{j\sigma})$$

$$n_i = c^\dagger_i c_i$$

Charge conservation implies:

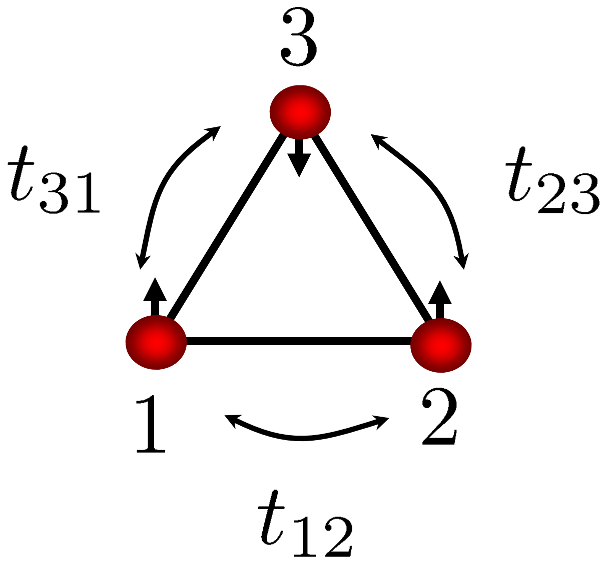
$$e\frac{\partial n_j}{\partial t} + \nabla_{\mathbf{r}_j} \cdot \mathbf{I} = 0$$

Where $\nabla_{\mathbf{r}_j} \cdot \mathbf{I} \equiv \mathbf{I}_{ij} + \mathbf{I}_{kj} - \mathbf{I}_{jh} - \mathbf{I}_{jl}$



(Exercise) Derive continuity eq. by using: $e\frac{\partial n_j}{\partial t} = \frac{ie}{\hbar}[H, n_j]$

Effective Current Density Operator



$$\tilde{\mathbf{I}}_{12,3} = \mathcal{P} e^{S} \mathbf{I}_{12,3} e^{-S} \mathcal{P}$$

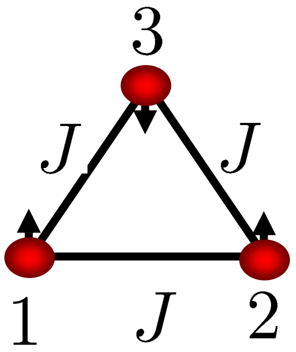
$$\tilde{\mathbf{I}}_{12,3} = \frac{\mathbf{r}_{12}}{r_{12}} \frac{24e}{\hbar} \frac{t_{12}t_{23}t_{31}}{U^2} [\mathbf{S}_1 \times \mathbf{S}_2] \cdot \mathbf{S}_3$$

Scalar spin chirality

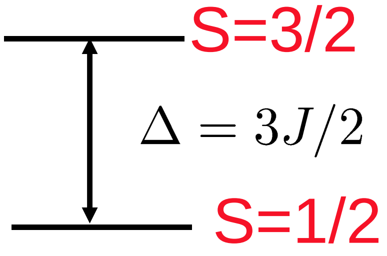
$$\chi_{123} = \mathbf{S}_1 \times \mathbf{S}_2 \cdot \mathbf{S}_3$$

Single Triangle

$$\tilde{H}^{(2)} = J(\mathbf{S}_1 \cdot \mathbf{S}_2 + \mathbf{S}_2 \cdot \mathbf{S}_3 + \mathbf{S}_3 \cdot \mathbf{S}_1 - 3/4)$$



Two S=1/2 doublets (ground states) and one ‼S=3/2 quartet (excited states)



(Exercise)

S=1/2, S^z=1/2 ground states

$$|\tilde{\psi}_{+\uparrow}\rangle = \frac{1}{\sqrt{3}}(c^\dagger_{1\uparrow}c^\dagger_{2\uparrow}c^\dagger_{3\downarrow} + e^{i\frac{2\pi}{3}}c^\dagger_{1\downarrow}c^\dagger_{2\uparrow}c^\dagger_{3\uparrow} + e^{-i\frac{2\pi}{3}}c^\dagger_{1\uparrow}c^\dagger_{2\downarrow}c^\dagger_{3\uparrow})|0\rangle$$

$$|\tilde{\psi}_{-\uparrow}\rangle = \frac{1}{\sqrt{3}}(c^\dagger_{1\uparrow}c^\dagger_{2\uparrow}c^\dagger_{3\downarrow} + e^{-i\frac{2\pi}{3}}c^\dagger_{1\downarrow}c^\dagger_{2\uparrow}c^\dagger_{3\uparrow} + e^{i\frac{2\pi}{3}}c^\dagger_{1\uparrow}c^\dagger_{2\downarrow}c^\dagger_{3\uparrow})|0\rangle$$

$$[\mathbf{S}_1 \times \mathbf{S}_2] \cdot \mathbf{S}_3 |\psi_{\pm\sigma}\rangle = \pm\frac{\sqrt{3}}{4}|\psi_{\pm\sigma}\rangle$$

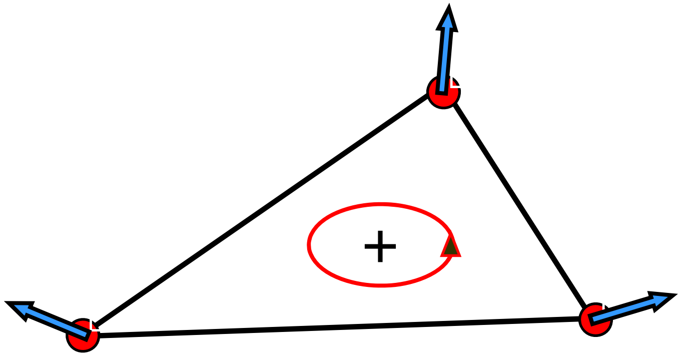
Single Triangle

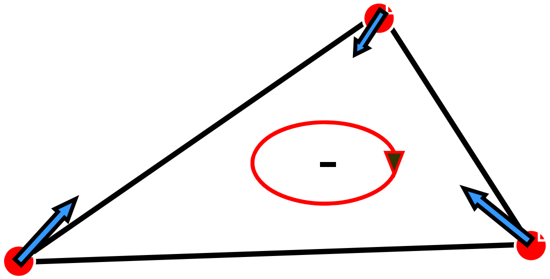
$$[\mathbf{S}_a \times \mathbf{S}_b] \cdot \mathbf{S}_c |\psi_{\pm\sigma}\rangle = \pm \frac{\sqrt{3}}{4} |\psi_{\pm\sigma}\rangle$$

$$\Downarrow$$

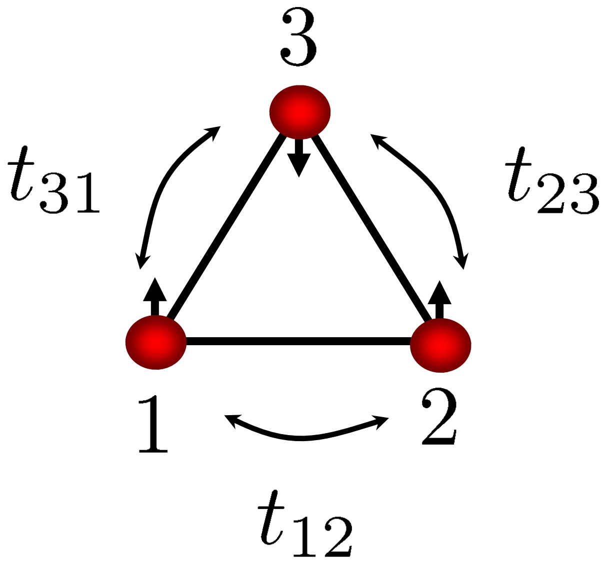
$$\tilde{\mathbf{I}}_{12,3} |\psi_{\pm\sigma}\rangle = \pm \frac{\mathbf{r}_{12}}{r_{12}} \, \frac{6\sqrt{3}e}{\hbar} \, \frac{t_{12}t_{23}t_{31}}{U^2} |\psi_{\pm\sigma}\rangle$$

Semi-classical version of the chiral spin states





Effective Charge Density Operator



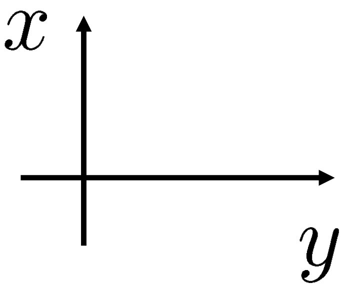
$$\tilde{n}_j = \mathcal{P} e^{S} n_j e^{-S} \mathcal{P}$$

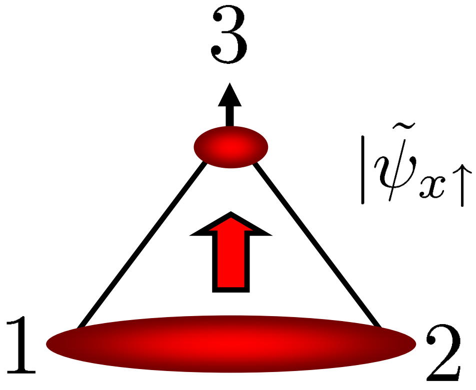
$$\delta\tilde{n}_1 = \tilde{n}_1 - 1 = 8\frac{t_{12}t_{23}t_{31}}{U^3}\left[\mathbf{S}_1\cdot(\mathbf{S}_2+\mathbf{S}_3) - 2\mathbf{S}_2\cdot\mathbf{S}_3\right]$$

$$e\frac{\partial\tilde{n}_j}{\partial t} + \nabla_{\mathbf{r}_j}\cdot\tilde{\mathbf{I}} = 0$$

(Exercise)

Single Triangle





$$|\tilde{\psi}_{x\uparrow}\rangle = \frac{1}{\sqrt{2}}\left[|\tilde{\psi}_{+\uparrow}\rangle - |\tilde{\psi}_{-\uparrow}\rangle\right]$$

$$\tilde{P}_x = \sum_j x_j \tilde{n}_j = 4\sqrt{3}ea(t/U)^3\left[\mathbf{S}_1 \cdot (\mathbf{S}_2 + \mathbf{S}_3) - 2\mathbf{S}_2 \cdot \mathbf{S}_3\right],$$

$$\tilde{P}_y = \sum_j y_j \tilde{n}_j = 12ea(t/U)^3 \mathbf{S}_1 \cdot (\mathbf{S}_2 - \mathbf{S}_3),$$

$$\boxed{\tilde{P}_x|\tilde{\psi}_{x\uparrow}\rangle = 6\sqrt{3}a\frac{t^3}{U^3}|\tilde{\psi}_{x\uparrow}\rangle}$$

Charge transfer from 3 to 1-2: the electronic charge moves to the bond with more singlet character.

Pseudo-spin 1/2 structure of (I, P_x, P_y)

The pseudo-spin operators (T_x, T_y, T_z)

$$\tilde{P}_x = -CT_x, \quad \tilde{P}_y = CT_y, \quad (\hbar a/U)\tilde{I} = CT_z,$$

with $C = 12\sqrt{3}ea(t/U)^3$, the SU(2) commutation relations,

$$\boxed{[T_\eta, T_\mu] = i\epsilon^{\eta\mu\nu}T_\nu}$$

(Exercise)

A state with well defined polarization has zero current and vice-versa.

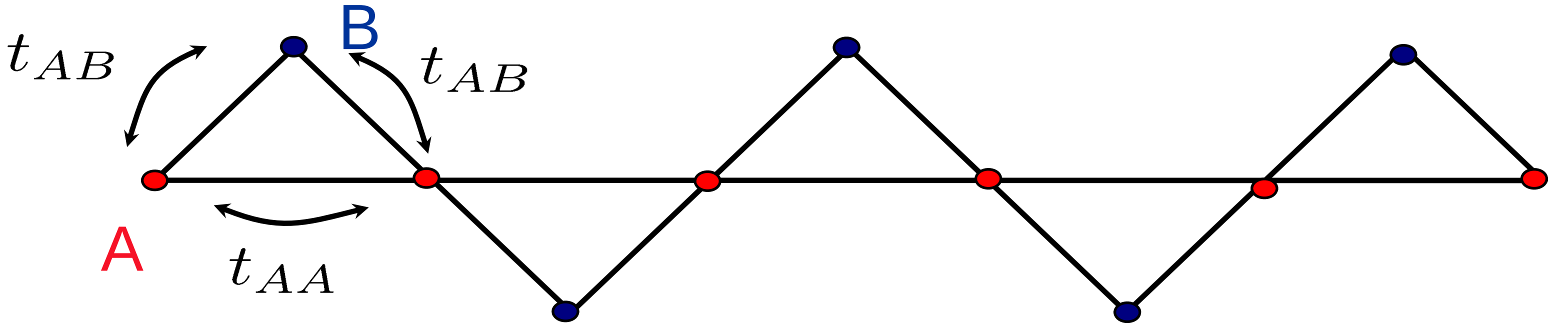
Lattice Systems

In a lattice, the effective local density operator can have contributions from more than one triangular plaquette.

$$\delta\tilde{n}_i = 8\sum_{(j,k)\in i} \frac{t_{ij}t_{jk}t_{ki}}{U^3}\left[\mathbf{S}_i\cdot(\mathbf{S}_j+\mathbf{S}_k) - 2\mathbf{S}_j\cdot\mathbf{S}_k\right]$$

Let us consider simple examples of spin orderings with $\delta\tilde{n}_i \neq 0$

Hubbard model on A-B saw-tooth chain

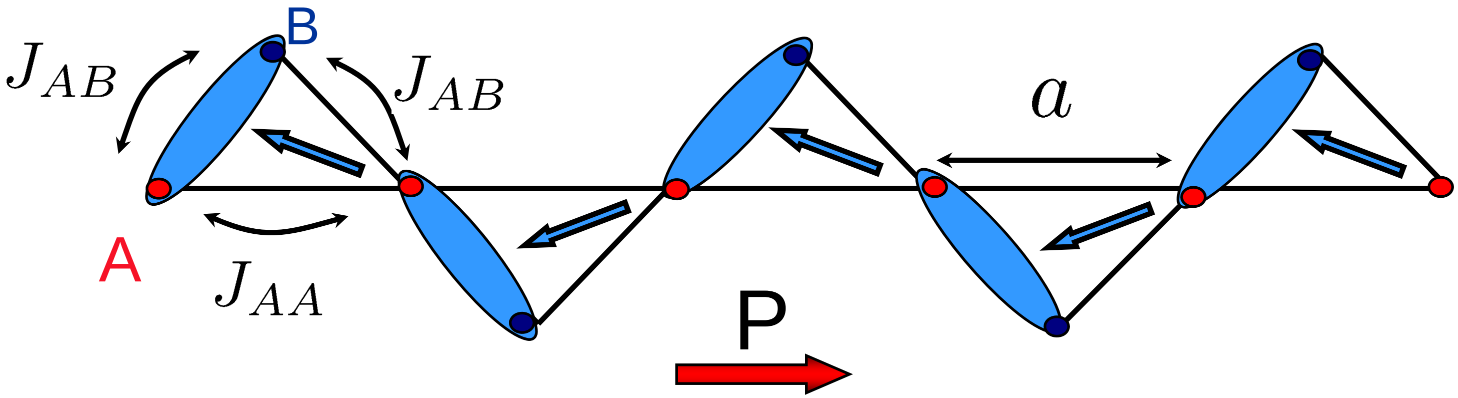


$$\tilde{H}^{(2)} = \sum_j J_{AB}(\mathbf{S}_j\cdot\mathbf{S}_{j+1} - 1/4) + J_{AA}(\mathbf{S}_j\cdot\mathbf{S}_{j+2} - 1/4)$$

Saw-tooth chain

$$\tilde{H}^{(2)} = \sum_j J_{AB}(\mathbf{S}_j \cdot \mathbf{S}_{j+1} - 1/4) + J_{AA}(\mathbf{S}_j \cdot \mathbf{S}_{j+2} - 1/4)$$

$$J_{AB} = 4\frac{t_{AB}^2}{U} \qquad J_{AA} = 4\frac{t_{AA}^2}{U}$$



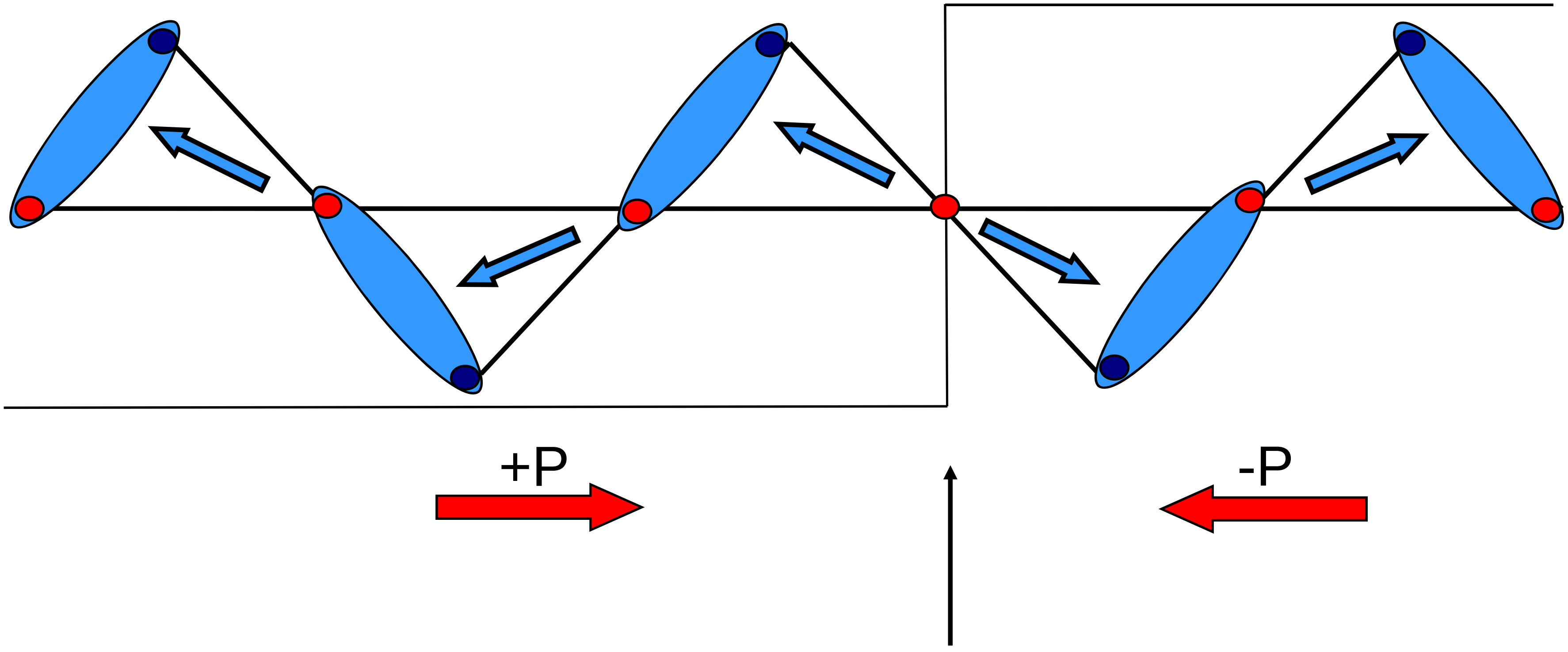
$$\langle \mathbf{S}_n \cdot \mathbf{S}_{n+1} \rangle = \alpha + \beta(-1)^n \quad \text{for} \quad \gamma = \frac{J_{AA}}{J_{AB}} > \gamma_c$$

$$\boxed{\tilde{P} = 24ea(t_{AB}^2 t_{AA}/U^3)\beta}$$

Ferroelectric state!

Saw-tooth chain

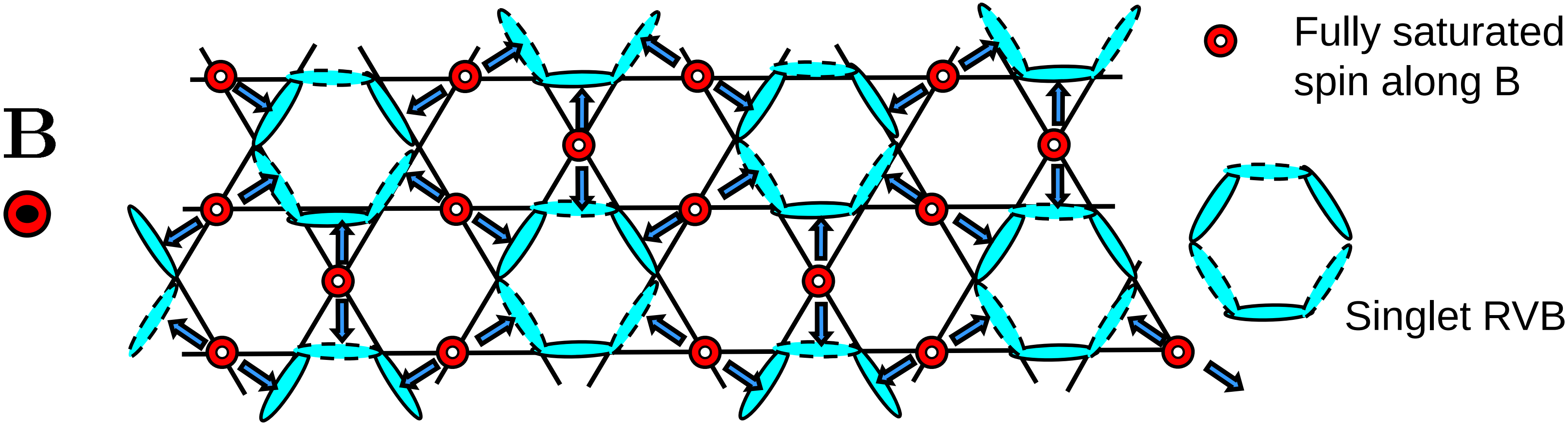
Charged soliton



$$Q = 2P/a$$

Kagome Lattice

The following ground state has been found for a Heisenberg model in an applied magnetic field when $M=M_{sat}/3$.



Fully saturated spin along B

Singlet RVB

The resulting charge-density wave does not break inversion symmetry (**P**=0).

1) There is a subclass of bond-ordered spin states in which charge ordering occurs as an epiphenomenon.

Bipartite lattices (unfrustrated)

2) Geometric frustration is a crucial requirement.

Half-filled Hubbard model on a bipartite lattice:

$$H = -\sum_{\langle ij\rangle\sigma} t(c_{i\sigma}^{\dagger}c_{j\sigma} + c_{j\sigma}^{\dagger}c_{i\sigma}) + \frac{U}{2}\sum_{i}(n_i - 1)^2,$$

Under a particle-hole transformation: $\bar{c}_{j\sigma}^{\dagger} = c_{j\sigma},$

the Hamiltonian becomes:

$$\bar{H} = \sum_{\langle ij\rangle\sigma} t(\bar{c}_{i\sigma}^{\dagger}\bar{c}_{j\sigma} + \bar{c}_{j\sigma}^{\dagger}\bar{c}_{i\sigma}) + \frac{U}{2}\sum_{i}(\bar{n}_i - 1)^2,$$

$$\bar{H}(\bar{c}_j^{\dagger}, \bar{c}_j) = H(-t, \bar{c}_j^{\dagger}, \bar{c}_j)$$

Bipartite lattices (unfrustrated)

(Exercise)

Since
$$\bar{\mathbf{S}}_j = -\mathbf{S}_j$$

we get
$$\tilde{H}(t, \mathbf{S}_j) = \tilde{\bar{\tilde{H}}}(t, \bar{\mathbf{S}}_j) = \tilde{H}(-t, -\mathbf{S}_j) = \tilde{H}(-t, \mathbf{S}_j)$$

Time reversal invariance of H

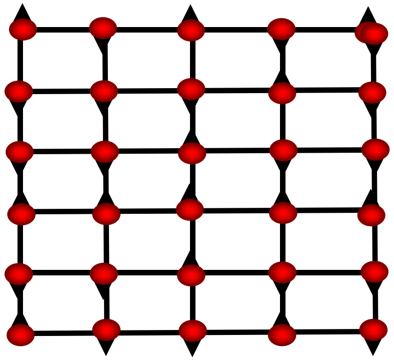
In the same way, from
$$\mathbf{I}_{jk} = \frac{iet_{jk}\mathbf{r}_{jk}}{\hbar r_{jk}} \sum_\sigma (c^\dagger_{k\sigma} c_{j\sigma} - c^\dagger_{j\sigma} c_{k\sigma}),$$

we obtain
$$\tilde{\mathbf{I}}_{jk}(t, \mathbf{S}_j) = \tilde{\mathbf{I}}_{jk}(-t, -\mathbf{S}_j),$$

which implies
$$\tilde{\mathbf{I}}_{jk}(t) = -\tilde{\mathbf{I}}_{jk}(-t).$$

Bipartite lattices (unfrustrated)

On a bipartite lattice we always need an even number of hopping processes to close a loop. This implies,



$$\boxed{\tilde{\mathbf{I}}_{jk}(t) = \tilde{\mathbf{I}}_{jk}(-t) = 0,}$$

on bipartite lattices.

A similar conclusion is obtained for the effective charge density operator:

$$\boxed{\tilde{n}_j = 1,}$$

on bipartite lattices by using that

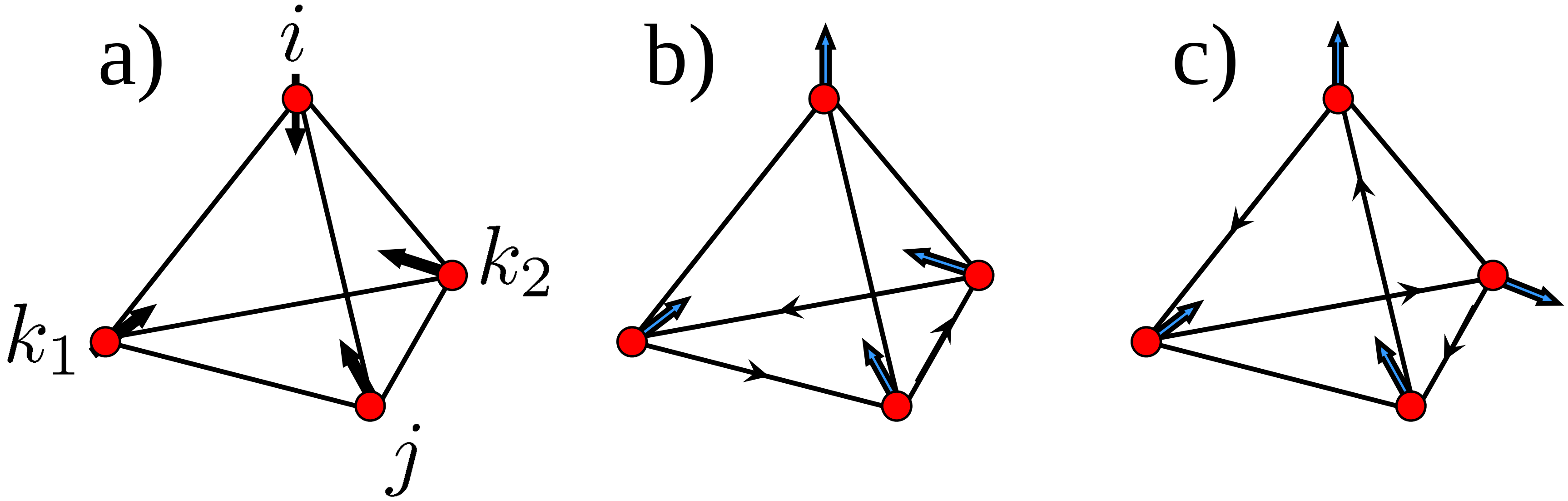
$$n_j - 1 = 1 - \bar{n}_j .$$

(Exercise)

Orbital Currents and Scalar Spin Chirality

3) $\langle \chi_{ijk} \rangle \neq 0$es not imply the existence of nonzero orbital currents because contributions from different triangles can cancel each other.

$$\tilde{\mathbf{I}}_{ij,k} = \frac{\mathbf{r}_{ij}}{r_{ij}} \frac{24e}{\hbar} \sum_{k\in(i,j)} \frac{t_{ij}t_{jk}t_{ki}}{U^2} \left[\mathbf{S}_i \times \mathbf{S}_j\right] \cdot \mathbf{S}_k$$

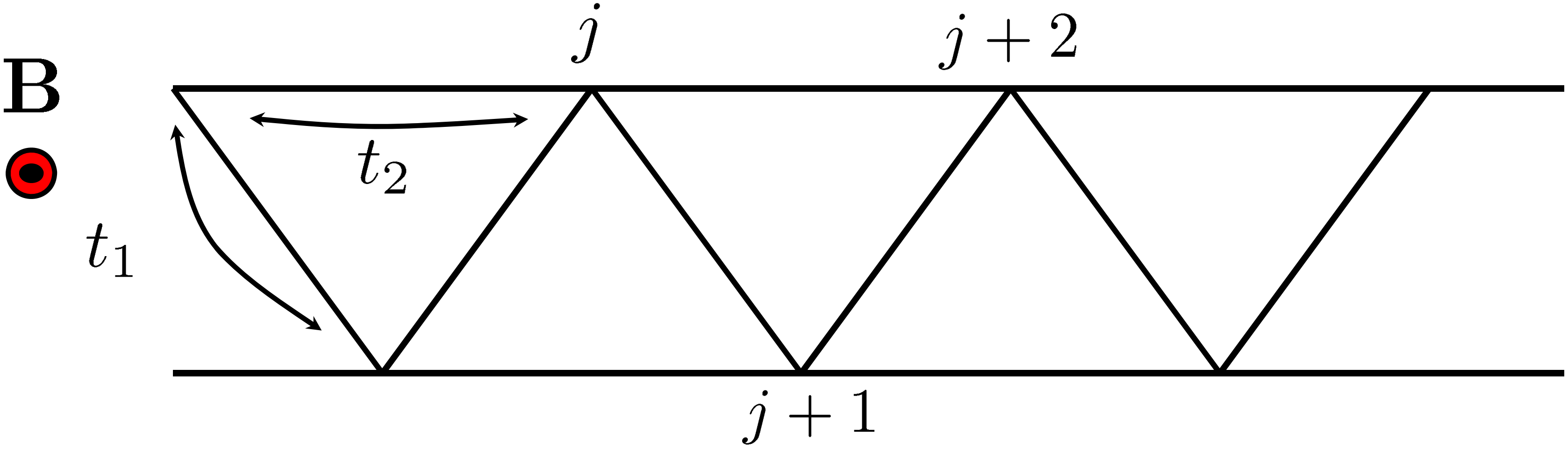


No orbital currents!

Orbital Currents in Lattice Systems

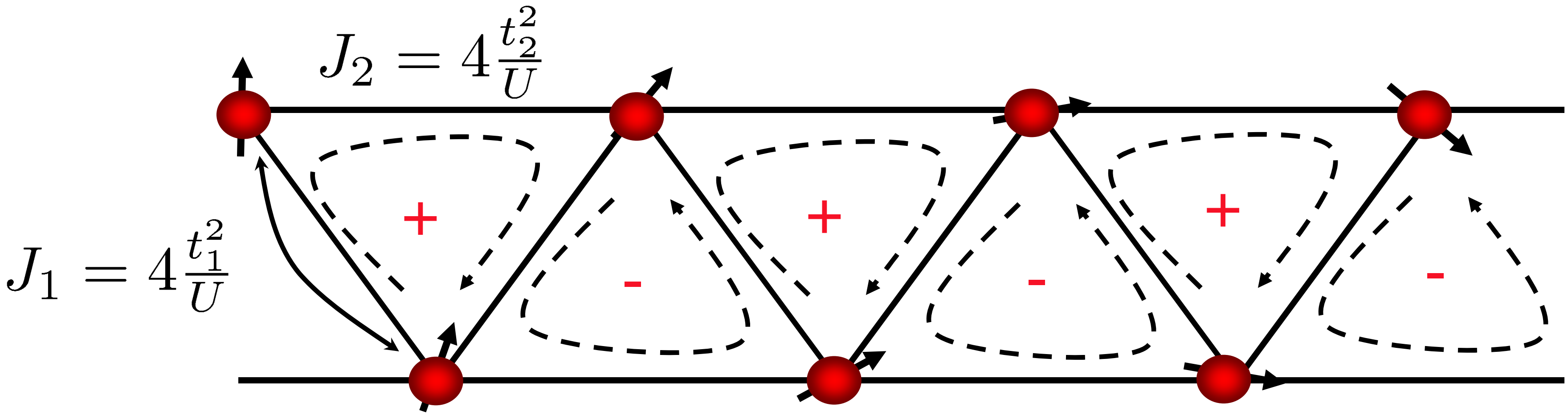
4) There is a subclass of scalar chiral spin states that are accompanied by ordering of orbital electric currents.

Hubbard model on zig-zag ladder in a magnetic field



$$H = \sum_{j\sigma\eta=1,2} t_\eta \left(c^\dagger_{j\sigma} c_{j+\eta\sigma} + c^\dagger_{j+\eta\sigma} c_{j\sigma}\right) + \frac{U}{2}\sum_j (n_j - 1)^2 - g\mu_B B S^z_j$$

Orbital Currents in Lattice Systems



$$\tilde{H}^{(2)} = \sum_j J_1(\mathbf{S}_j \cdot \mathbf{S}_{j+1} - 1/4) + J_2(\mathbf{S}_j \cdot \mathbf{S}_{j+2} - 1/4) - g\mu_B B S_j^z$$

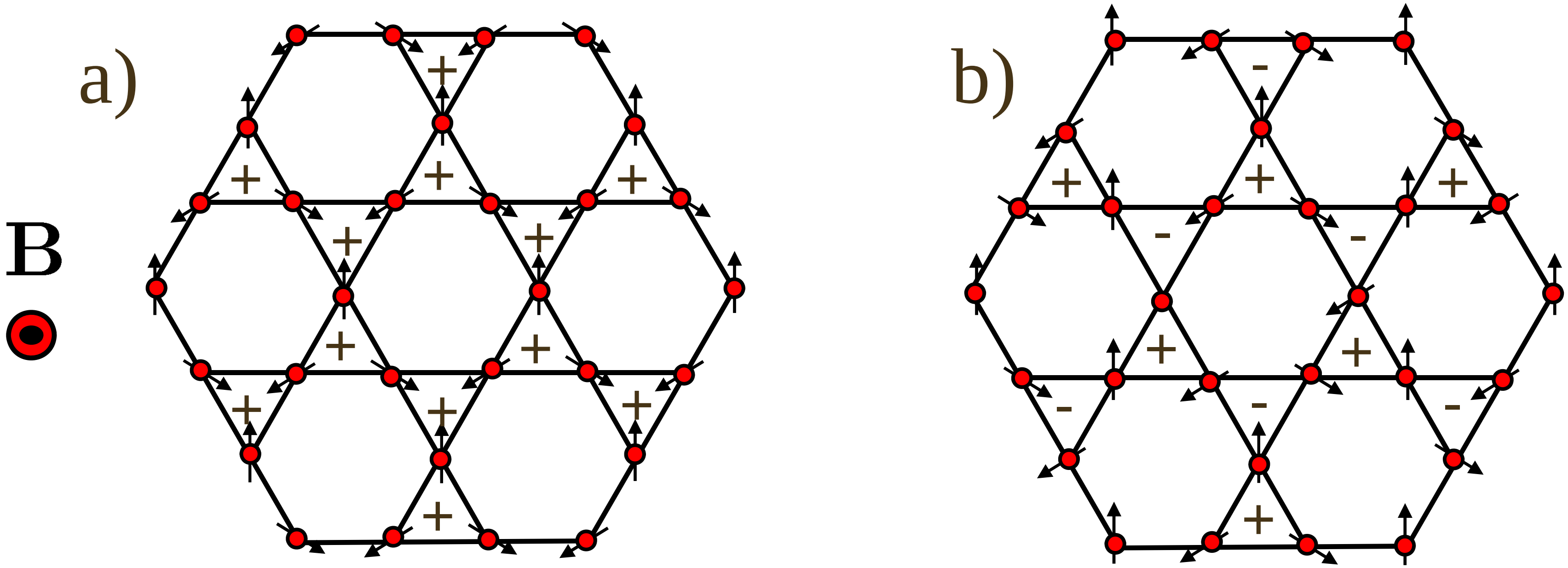
The ground state of this spin Hamiltonian is chiral:

$$\langle \mathbf{S}_j \rangle = 0 \quad \langle \mathbf{S}_j \times \mathbf{S}_{j+1} \rangle \neq 0 \quad \mathbf{S}_j \cdot \langle \mathbf{S}_{j+1} \times \mathbf{S}_{j+2} \rangle \neq 0$$

for $\gamma = \frac{J_2}{J_1} > \gamma_c$ nea$\mathbf{B}$ $\mathbf{B}_{sat}$

Orbital Currents in Lattice Systems

Kagome Lattice

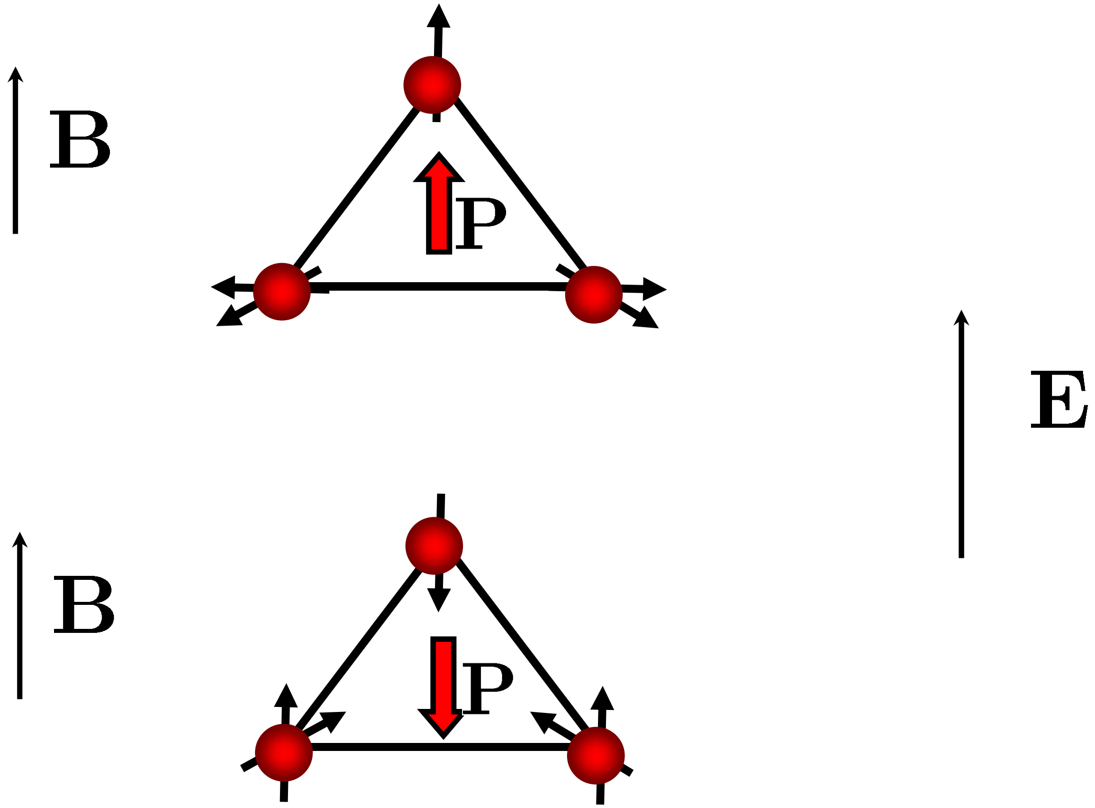


Field Induced

Uniform scalar chirality

Staggered scalar chirality

Magneto-electric Effects

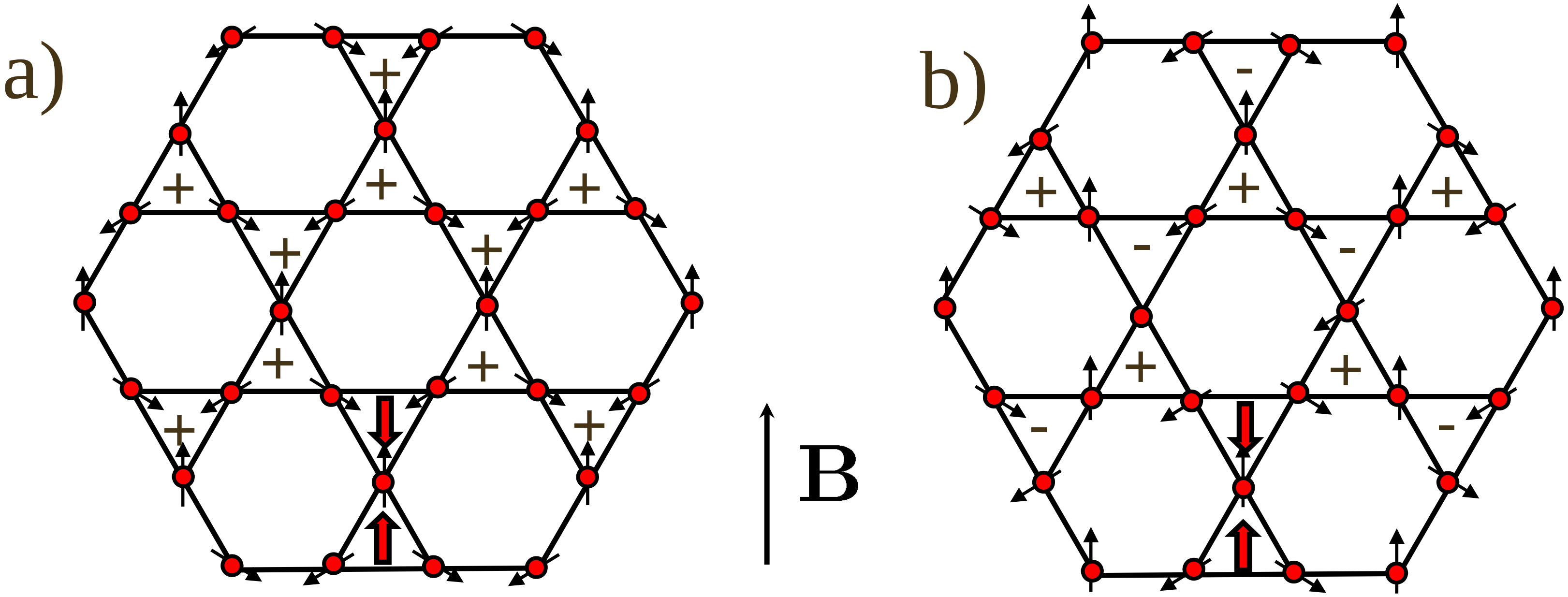


In the continuum:

$$\nabla \cdot \mathbf{M} \quad \mathbf{B} \cdot \mathbf{P}$$

$$\nabla \cdot \mathbf{M} \quad \mathbf{E} \cdot \mathbf{M}$$

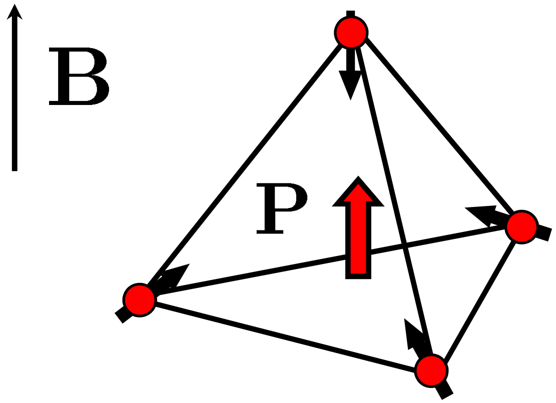
Magneto-electric Effects

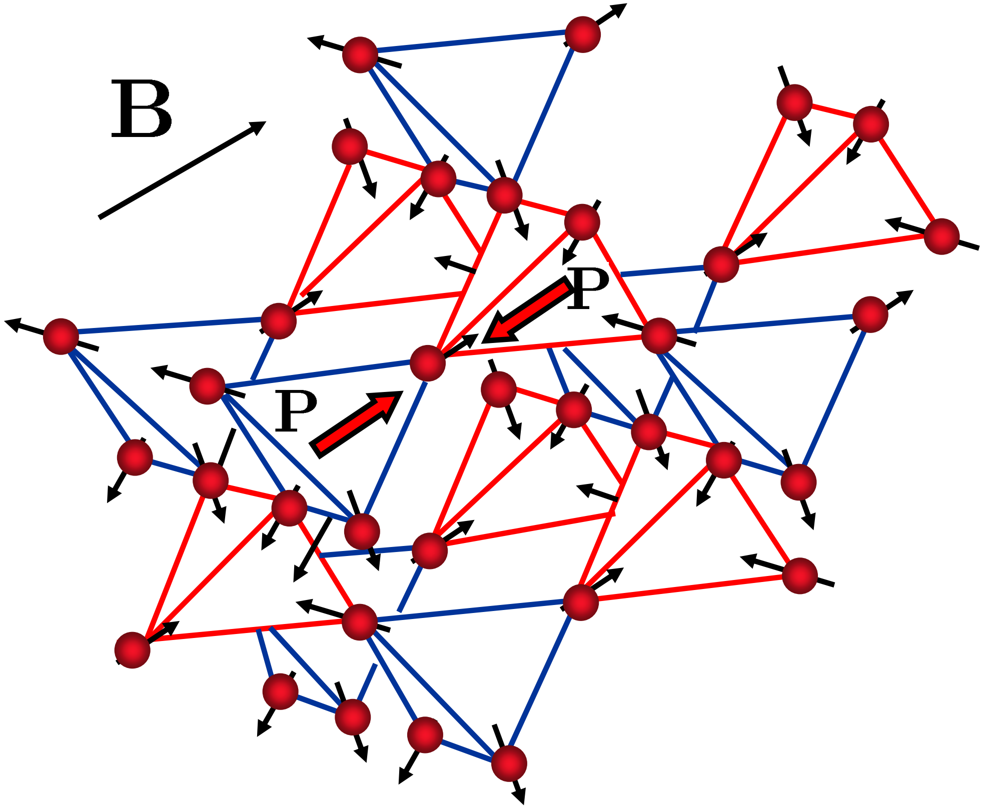


Field induced CDW but no net electric diople moment.

Magneto-electric Effects

AFM state of Pyrochlore lattice

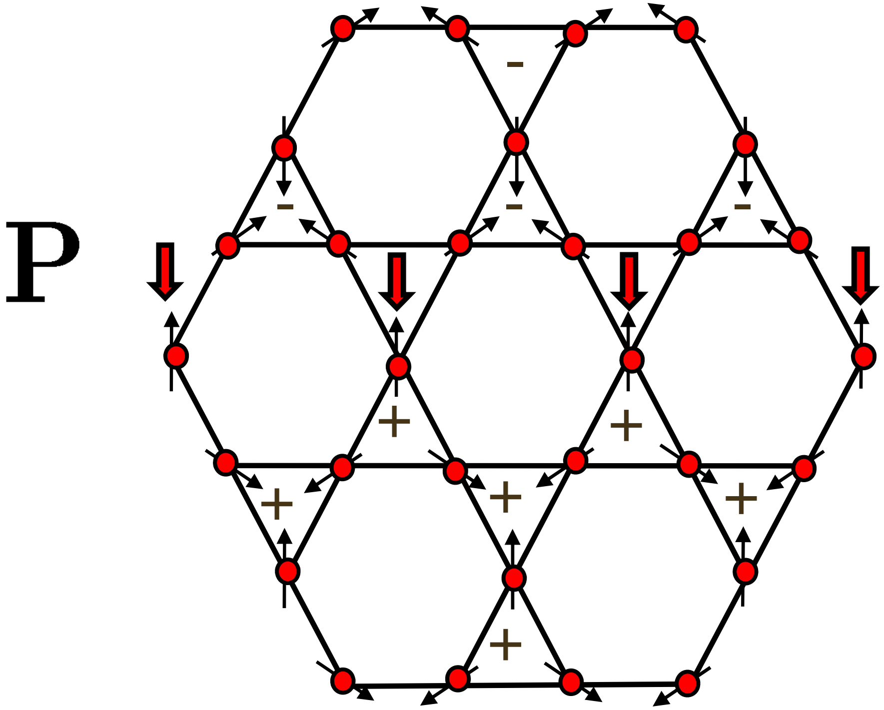




Field induced CDW but no net electric dipole moment.

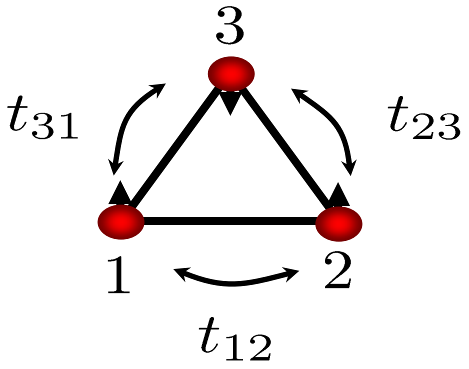
AFM Domain Walls

There is a net electric dipole moment on the domain wall. Therefore, it should be possible to move this AFM domain wall by applying a gradient of electric field.



Dynamical Properties

Let's get back to the single triangle to learn something about the dynamical consequences of our previous observations.



The electric dipole moment has nonzero matrix elements between states with opposite chirality χ_{123}.

$$\tilde{P}_x = -CT_x, \quad \tilde{P}_y = CT_y, \quad (\hbar a/U)\tilde{I} = T_z,$$

$$[T_\eta, T_\mu] = i\epsilon^{\eta\mu\nu} T_\nu$$

$$T_z|\psi_{\pm\sigma}\rangle = \pm\tfrac{1}{2}|\psi_{\pm\sigma}\rangle \quad \Rightarrow \quad \begin{aligned} \langle\psi_{+\sigma}|T_x|\psi_{-\sigma}\rangle &= \tfrac{1}{2} \\ \langle\psi_{+\sigma}|T_y|\psi_{-\sigma}\rangle &= \tfrac{i}{2} \end{aligned}$$

Dynamical Properties

The T=0 dielectric function is given by:

$$\boxed{\epsilon_{\eta\mu}(\omega) = \epsilon_0\delta_{ik} - \frac{8\pi}{V}\sum_n \frac{\omega_{no}\langle 0|\tilde{P}_\eta|n\rangle\langle n|\tilde{P}_\mu|0\rangle}{(\omega^2-\omega_{n0}^2-i\delta)},}$$

where $\tilde{H}|n\rangle = E_n|n\rangle$ $\hbar\omega_{n0} = E_n - E_0$, $\eta,\mu = \{x,y\}$

ϵ_0 is the contribution from all other modes and V is the volume.

Since

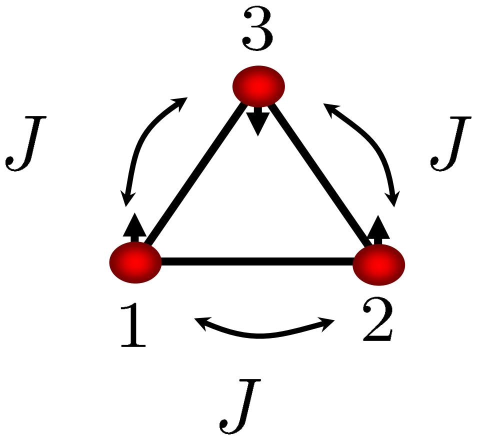
$$T_z|\psi_{\pm\sigma}\rangle = \pm\tfrac{1}{2}|\psi_{\pm\sigma}\rangle \quad \Longrightarrow \quad \begin{aligned} \langle\psi_{+\sigma}|T_x|\psi_{-\sigma}\rangle &= \tfrac{1}{2} \\ \langle\psi_{+\sigma}|T_y|\psi_{-\sigma}\rangle &= \tfrac{i}{2} \end{aligned}$$

$$\boxed{\epsilon_{xy}(\omega) \neq 0}$$

A chiral ground state induces circular dichroism

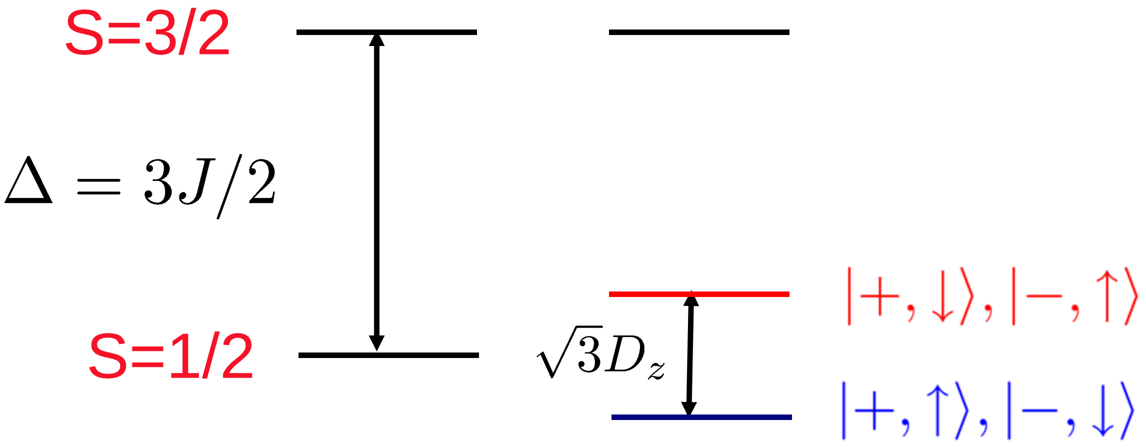
Dynamic Properties

In some real systems such as V15 the lattice symmetry allows for a nonzero DM coupling:



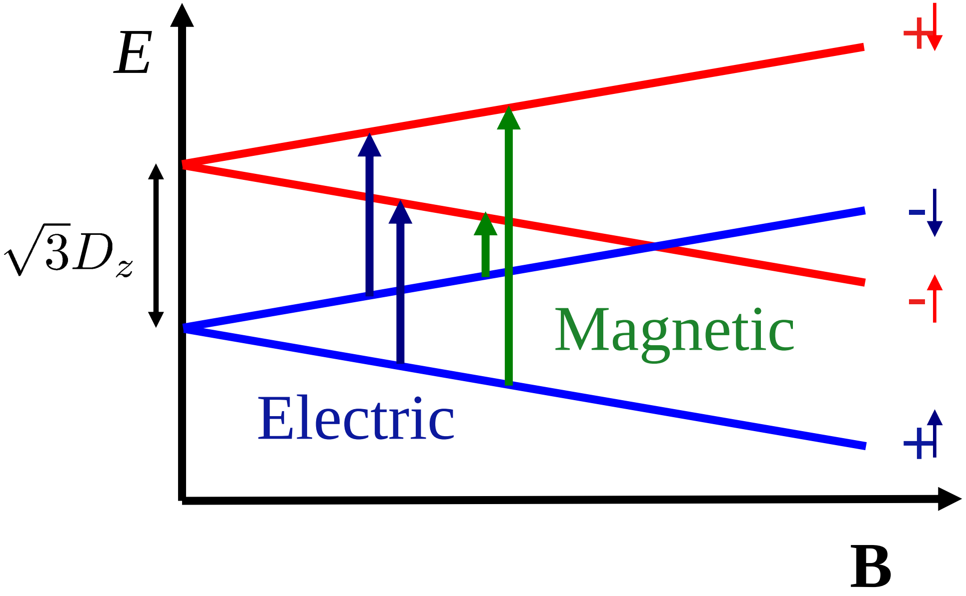
$$H_{\mathrm{DM}} = \sum_{ij} \mathbf{D}_{ij} \cdot [\mathbf{S}_i \times \mathbf{S}_j]$$

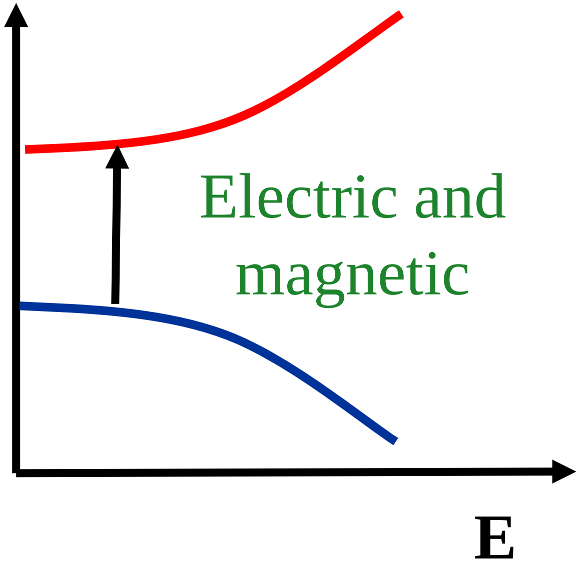
$$D = D_z \hat{\mathbf{z}}$$



Dynamic Properties

Low energy levels of the single triangle with nonzero DM coupling D_z as a function of magnetic and electric fields. Blue (green) arrows show transitions induced by ac electric (magnetic, ESR) field.





An AC electric field induces transitions between states with opposite chiralities. Thus we have here a new dipole active “ESR” transition caused by the electric component of the electromagnetic field.

Conclusions

- Spin ordering in Mott insulators can drive **orbital electric currents** or charge-density-waves of purely electronic origin.

- There is a clear connection between the notion of “scalar spin chirality” and orbital currents or magnetic moments.

- Charge ordering appears for a subclass of bond orderings that make the different sites nonequivalent.

- Geometric frustration plays a crucial role: the effect disappears in bipartite lattices.

- An electric field gradient can be used to move certain AFM domain walls because they have a net electric dipole moment.

- Interesting magneto-electric effects emerge as a consequence of this electronic charge redistribution.

Conclusions

- The coupling between spin and charge degrees of freedom leads to unusual physical responses such as circular dichroism for chiral spin states.

- An AC electric field induces transitions between states with opposite chiralities. Thus we have here a new dipole active "ESR" transition caused by the electric component of the electromagnetic field.

- The electric field causes transitions between states with the same total spin. Therefore, $\epsilon_{\eta\mu}(\omega)$s with the ground state magnetization until the contribution of magnetic states vanishes when all spins become aligned in the field direction