Electronic Orbital Currents and Polarization in Mott Insulators

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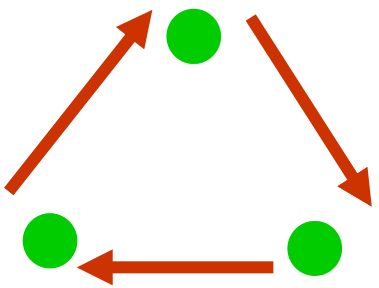
- Introduction: why electrical properties of Mott insulators differ from those of band insulators.
- Magnetic states in the Hubbard model.
- Orbital currents.
- Electronic polarization.
- Low frequency dynamic properties.
- Conclusions.

Difference between Mott and band insulators

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

- Only in the limit $U \to \infty$ electrons are localized on sites.
- At $t/U \neq 0$ electrons can hop between sites.

$$H_S = \frac{4t^2}{U}(\vec{S}_1 \cdot \vec{S}_2 - 1/4).$$



Orbital current

Definition of magnetic states for Hubbard Hamiltonian

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

- $U \to \infty$ ground state $\Psi_{0\nu}$, 2^N spin-degenerate, S-subspace.
- Excited (polar) states are separated by the Hubbard gap, U.
- t_{ij} increases, degeneracy is adiabatically lifted → magnetic states

$$\Psi_\nu = e^{-S(t)}\Psi_{0\nu}.$$

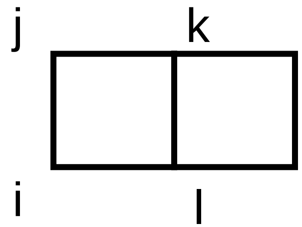
- Magnetic state energies

$$\langle \Psi^*_\nu | H | \Psi_\nu \rangle = \langle \Psi^*_{0\nu} | e^S H e^{-S} | \Psi_{0\nu} \rangle \rightarrow H_S = Pe^S He^{-S}P.$$

- Any other electron operator $\langle \Psi^*_\nu | O | \Psi_\nu \rangle \rightarrow O_S = Pe^S Oe^{-S}P.$
- H_S, O_S are expressed in terms of spin operators $S^\alpha_i = \sum_{\mu\nu} c^{+}_{i\mu}\sigma^\alpha_{\mu\nu}c_{i\nu}.$

Spin Hamiltonian

- For square lattice



$$H_S = \frac{4t^2}{U} \sum_{\langle ij \rangle} (\vec{S}_i \cdot \vec{S}_j - 1/4) + J_{ik} \sum_{\langle\langle ik \rangle\rangle} (\vec{S}_i \cdot \vec{S}_k - 1/4)_k$$

$$+ J_{ij,kl} \sum_{ij,kl} (\vec{S}_i \cdot \vec{S}_j - 1/4)(\vec{S}_k \cdot \vec{S}_l - 1/4), \qquad J_{ij}, J_{ij,kl} \propto t^4 / U^3 .$$

- Only even number of spin operators.

- Some excited magnetic states (collective modes) lay below the Hubbard gap resulting in low-T thermodynamic and low-frequency dynamic properties of Mott insulators.

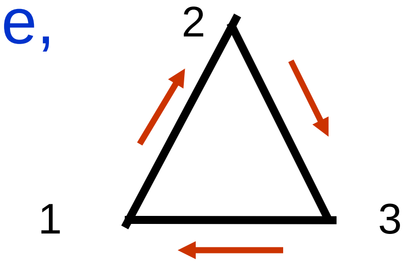
Spin current operator and scalar spin chirality

Bogolyubov, 1949.

- Current operator for Hubbard Hamiltonian on bond ij:

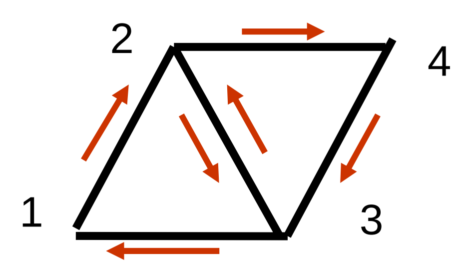
$$\vec{I}_{ij} = \frac{iet_{ij}\vec{r}_{ij}}{\hbar r_{ij}} \sum_{\sigma} (c^{+}_{i\sigma} c_{j\sigma} - c^{+}_{j\sigma} c_{i\sigma}).$$

- Projected current operator: odd # of spin operators, scalar in spin space. For smallest loop, triangle,



$$\vec{I}_{S,12}(3) = \frac{\vec{r}_{ij}}{r_{ij}} \frac{24et_{12}t_{23}t_{31}}{\hbar U^2} [\vec{S}_1 \times \vec{S}_2] \cdot \vec{S}_3.$$

- Current via bond 23



$$I_{S,23} = I_{S,23}(1) + I_{S,23}(4).$$

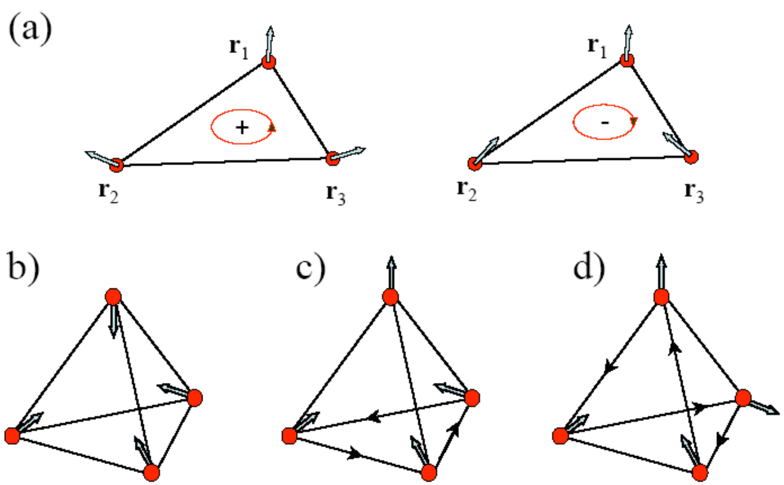
- On bipartite nn lattice I_S is absent.

Orbital currents in the spin ordered ground state $\langle \vec{S}_i \rangle \neq 0$

- Necessary condition for orbital currents is nonzero average chirality

$$\chi_{12,3} = [\vec{S}_1 \times \vec{S}_2] \cdot \vec{S}_3, \qquad \langle \chi_{ij,k} \rangle \neq 0.$$

- It may be inherent to spin ordering or induced by magnetic field



Triangles with $\pm$ chirality

On tetrahedron chirality may be nonzero but orbital currents absent.

Chirality in the ground state without magnetic ordering

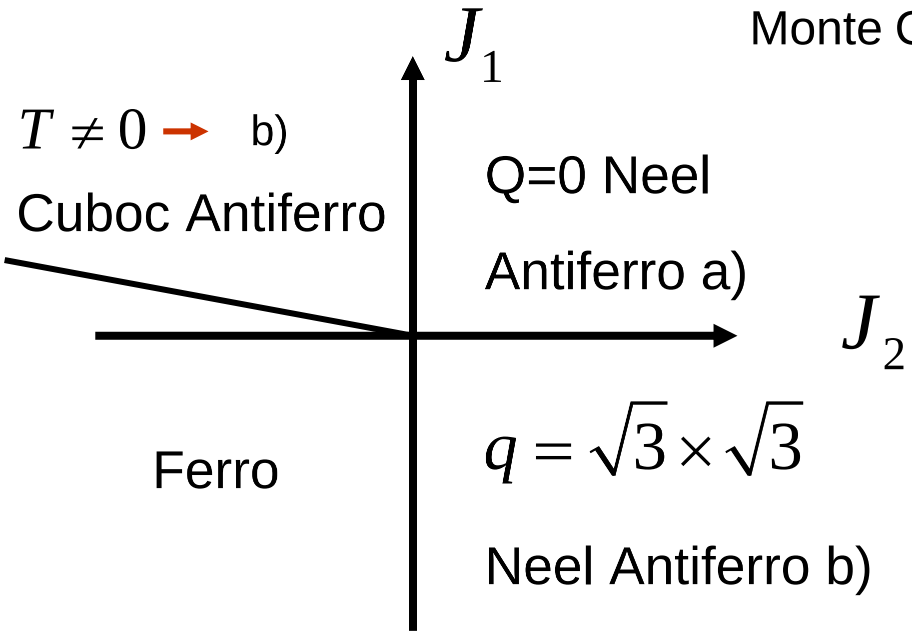
- $\langle \chi_{12,3} \rangle = \langle [\vec{S}_1 \times \vec{S}_2] \cdot \vec{S}_3 \rangle \neq 0, \qquad \langle \vec{S}_i \rangle = 0.$

- Geometrically frustrated 2d system ⟶ Mermin-Wagner theorem ⟶ $\langle \vec{S}_i \rangle = 0.$
- State with maximum entropy may be with broken discrete symmetry $\langle \chi_{12,3} \rangle \neq 0.$
- Example: $J_1 - J_2$ model on kagome lattice:

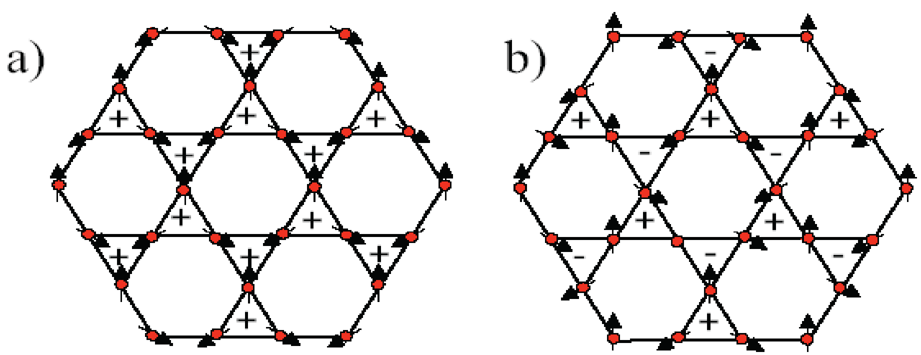
$$H_S = J_1 \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j + J_2 \sum_{\langle\langle ij \rangle\rangle} \vec{S}_i \cdot \vec{S}_\mathrm{k}.$$

Ordering in $J_1 - J_2$ model on kagome lattice at T=0

Monte Carlo results by Domenge et al., 2007.

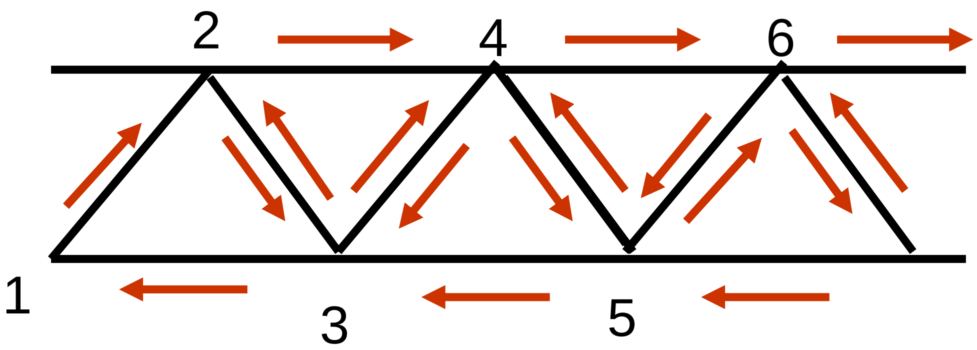


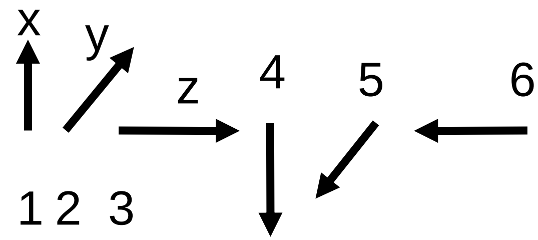
Phase diagram for classical order parameter



For S=1/2 for low T cuboc is chiral with orbital currents b) and without spin ordering.

Boundary and persistent current



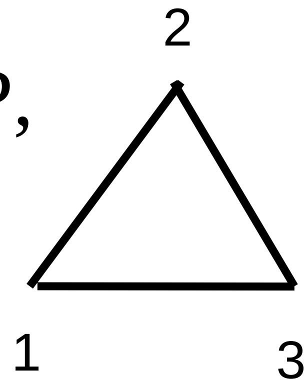


$$\chi = const$$

Boundary current in gaped 2d insulator

Spin dependent electronic polarization

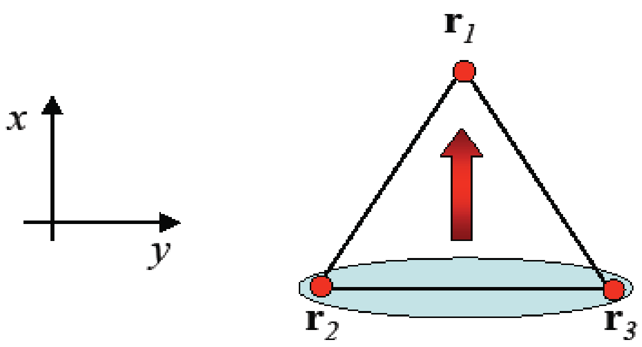
- Charge operator on site i: $$Q_i = e\sum_{\sigma} c_{i\sigma}^{+} c_{i\sigma}.$$
- Projected charge operator $n_{S,i} = P e^{S} n_i e^{-S} P,$



$$n_{S,1}(2,3) = 1 - \frac{8t_{12}t_{23}t_{31}}{U^2}[\vec{S}_1 \cdot (\vec{S}_2 + \vec{S}_3) - 2\vec{S}_2 \cdot \vec{S}_3].$$

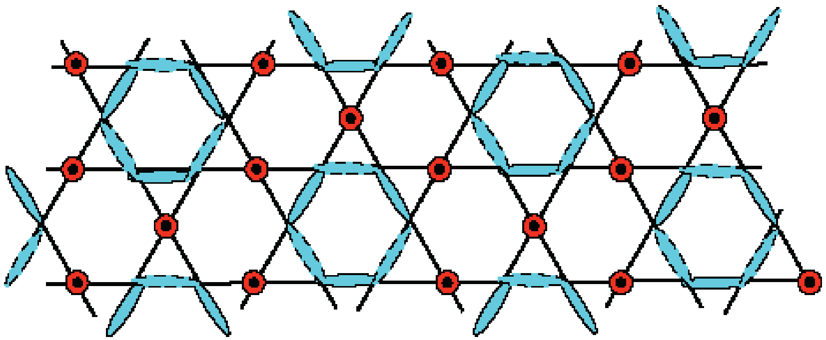
- Polarization on triangle $$\vec{P}_{123} = e\sum_{i=1,2,3} n_{S,i}\vec{r}_i, \qquad \sum_i n_{S,i} = 3.$$
- Charge on site i is sum over triangles at site i.

Electronic polarization on triangle



- Magnitostriction results in similar dependence of polarization on spins.

Charges on kagome lattice



Charge ordering for spins 1/2 in magnetic field at $\langle S_z \rangle = S/3$.

Low frequency dynamic properties: circular dichroism

- Responses to ac electric and ac magnetic field are comparable for $J \sim 100$ K:

$$\varepsilon_{ik}(\omega) = \varepsilon_0 \delta_{ik} + \frac{8\pi}{V} \sum_n \frac{\omega_{n0} \langle 0 | P_{S,i} | n \rangle \langle n | P_{S,k} | 0 \rangle}{\omega^2_{n0} - \omega^2 + i\delta},$$

$$\hbar\omega_{n0} = E_n - E_0, \qquad H_S |n\rangle = E_n |n\rangle.$$

- Compare $P \sim 8eat^3/U^3,$ and $M \sim g\mu_B S.$

- Nonzero orbital moment $L_z \propto \partial/\partial\varphi$

$$\langle 0 | P_x | n \rangle, \langle 0 | P_y | n \rangle \propto \cos\varphi, \sin\varphi \neq 0$$

- Rotation of electric field polarization in chiral ground state.

Low frequency dynamic properties: negative refraction index

- Responses to ac electric and ac magnetic field are comparable for J □100 K:

$$\varepsilon_{ik}(\omega) = \varepsilon_0 \delta_{ik} + \frac{8\pi}{V} \sum_n \frac{\omega_{n0} \langle 0 | P_{S,i} | n \rangle \langle n | P_{S,k} | 0 \rangle}{\omega_{n0}^2 - \omega^2 + i\delta},$$

- Spin-orbital coupling may lead to common poles in $\varepsilon_{ik}(\omega)$ and $\mu_{ik}(\omega)$ →

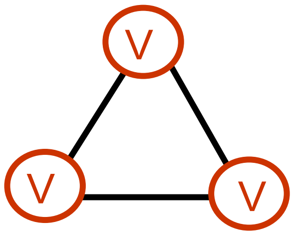
- Negative refraction index if dissipation is weak.

Low frequency dynamic properties of isolated triangles

- Trinuclear spin complex V15 $K_6[V_{15}^{IV}As_6O_{42}(H_2O)]\cdot 8H_2O.$

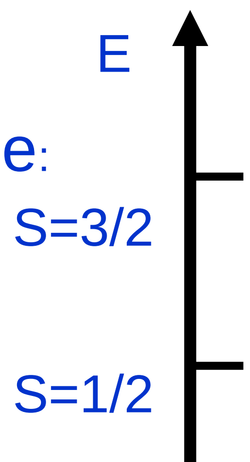
- Energy levels of the Heisenberg Hamiltonian:

$$H_S = J(\vec{S}_1\cdot\vec{S}_2 + \vec{S}_2\cdot\vec{S}_3 + \vec{S}_1\cdot\vec{S}_3 - 3/4):$$



☆ ground state: quartet S=1/2, $\chi = T_z = \pm 1,$ (pseudospin), $S_z = \pm 1/2,$

☆ quartet S=3/2 separated by 3J/2.



- Polarization and orbital current in low-energy subspace:

$$P_{S,x} = -CT_x, \qquad P_{S,y} = CT_y, \qquad (\hbar a/U)I_S = T_z,$$

$$C = 12\sqrt{3}ea(t/U)^3.$$

Isolated triangle: accounting for DM interaction

- DM coupling: $H_{DM} = \sum_{ij} D_{ij} \vec{S}_i \times \vec{S}_j .$

- For V15 $H_{DM} \approx D_z L_z S_z .$

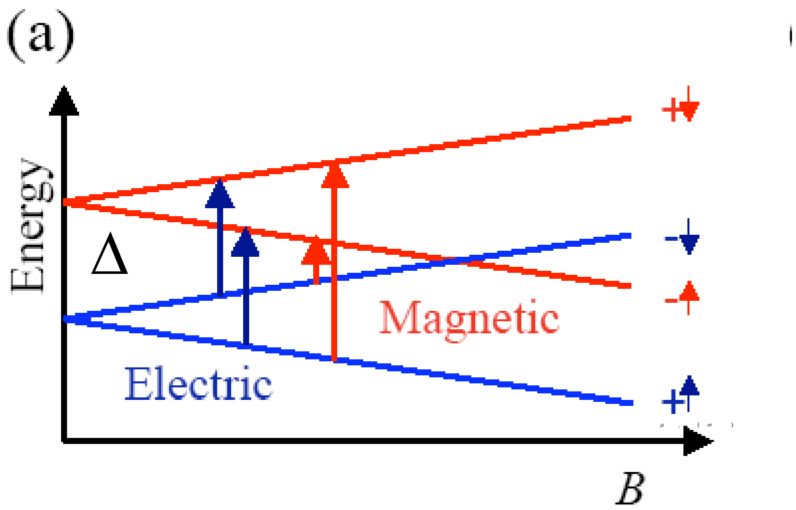
- Splits lowest quartet into 2 doublets $\left|+\uparrow\right\rangle, \left|-\downarrow\right\rangle$ and $\left|+\downarrow\right\rangle, \left|-\uparrow\right\rangle$ separated by energy $\Delta = D_z .$

- Ac electric field induces transitions between $\chi = \pm 1.$

- Ac magnetic field induces transitions between

$S_z = \pm 1/2.$

Isolated triangle with DM interaction



- Frequnces of EPR and microwave absorption differ in field.
- Near resonance strong rotation of electric field polarization.
- Below resonance at B=0 both $\varepsilon_{ik}(\omega)$ and $\mu_{ik}(\omega)$ are negative if dissipation is low enough.

Conclusions

- Dissipationless orbital microscopic currents, macroscopic boundary currents, macroscopic current around the ring like in superconductor.

- Spin-dependent electronic polarization and multiferroic behavior.

- Chirality results in circular dichroism, option to probe chirality when magnetic ordering is absent.

- Crystal with negative refraction index ?