Electric Polarization induced by Magnetic order

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Spin-orbit coupling is known to play an important role in, even a dominant mechanism for,

- Anomalous Hall Effect (->Spintronics)
- Spin Hall Effect (->Spintronics)
- Etc.

- Magnetism-induced Electric Polarization

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Physics of S*L in Magnetic States

If spin orientations are fixed due to magnetic ordering,

$$S*L \sim \langle S\rangle * L$$

acts like Zeeman field and polarize the orbital states.

The results may be polarization of the electronic wave function.

Spin-polarization Coupling via GL Theory

Spin <S> and polarization <R> break different symmetries:

<S> breaks time-inversion symmetry

<R> breaks space-inversion symmetry

Naively, lowest-order coupling occurs at $\langle S \rangle^2 \langle R \rangle^2$.

Lower-order terms involving spatial gradient,

$\langle S \rangle^2 \langle \text{grad } R \rangle$ or $\langle S \rangle \langle \text{grad } S \rangle \langle R \rangle$, are possible.

Spin-polarization coupling via GL theory

Generally one can write down GL terms like

$$F_I \sim \mathbf{P}\cdot\mathbf{M}(\nabla\cdot\mathbf{M}), \quad \mathbf{P}\cdot(\mathbf{M}\cdot\nabla)\mathbf{M}, \quad \mathbf{P}\cdot\nabla(\mathbf{M}^2)$$

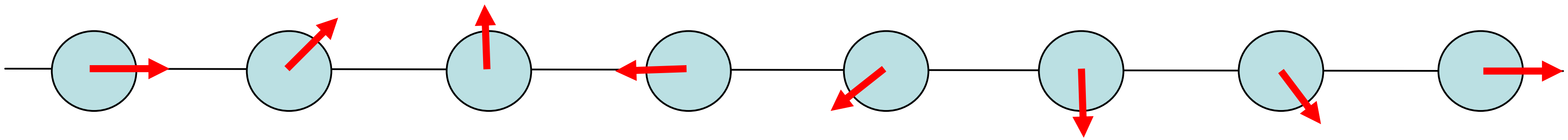
that result in the induced polarization

$$\mathbf{P} \sim \mathbf{M}(\nabla\cdot\mathbf{M}), \quad (\mathbf{M}\cdot\nabla)\mathbf{M}, \quad \nabla(\mathbf{M}^2)$$

in the presence of magnetic ordering

Mostovoy PRL 06

Spin-polarization Coupling via GL Theory



For spiral spins

$$\mathbf{M} = M_1 \hat{x} \cos(\mathbf{k}\cdot\mathbf{r}) + M_2 \hat{y} \sin(\mathbf{k}\cdot\mathbf{r})$$

induced polarization has a uniform component given by

$$\mathbf{P} \sim \mathbf{M}(\nabla\cdot\mathbf{M}), \quad (\mathbf{M}\cdot\nabla)\mathbf{M}, \quad \nabla(\mathbf{M}^2)$$

$$\boxed{\int d^3\mathbf{r}\mathbf{P} \sim M_1 M_2 \mathbf{k} \times (\hat{x} \times \hat{y})}$$

Mostovoy PRL 06

Experimental Evidence of Spin-polarization Coupling

$$\mathbf{M} = M_1 \hat{x} \cos(\mathbf{k} \cdot \mathbf{r}) + M_2 \hat{y} \sin(\mathbf{k} \cdot \mathbf{r})$$

$$\int d^3\mathbf{r}\mathbf{P} \sim \underline{M_1 M_2} \mathbf{k} \times (\hat{x} \times \hat{y})$$

Uniform induced polarization depends on the product M_1 M_2

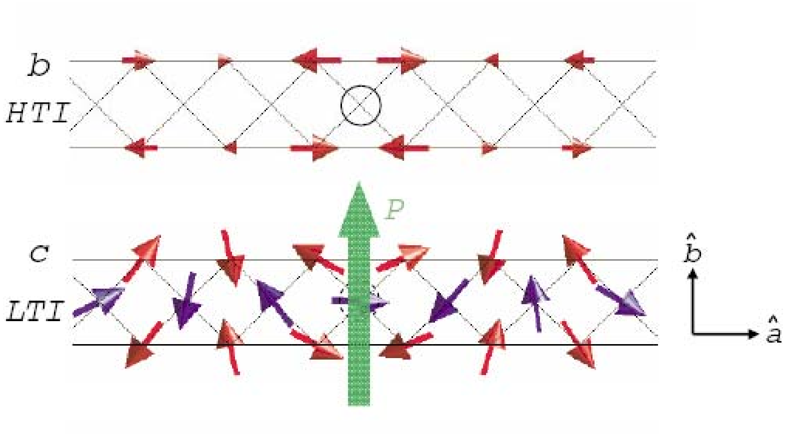
- Collinear (M_1M_2=0) spin cannot induce polarization
- Only non-collinear, spiral spins have a chance

Recent examples (partial)

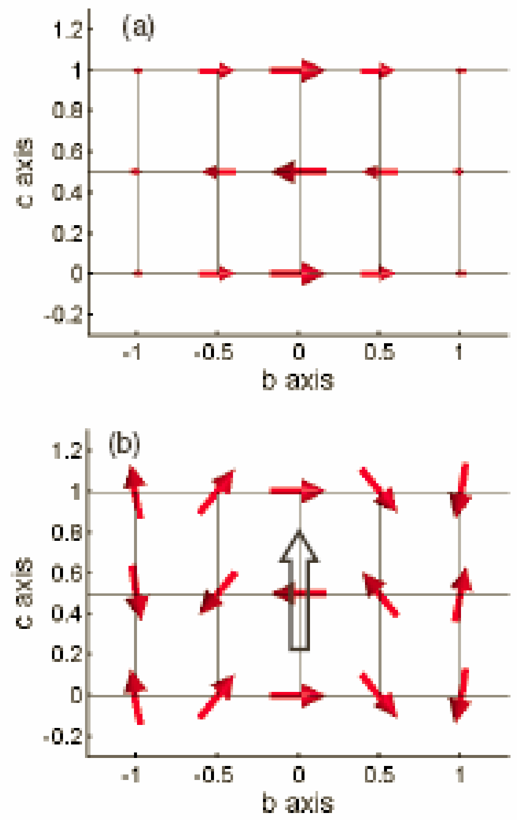
$Ni_3V_2O_8$ – PRL 05
$TbMnO_3$ – PRL 05
$CoCr_2O_4$ – PRL 06

$Ni_3V_2O_8$	$TbMnO_3$

Collinear to non-collinear spin transition accompanied by onset of polarization with P direction consistent with theory



Lawes et al PRL 05

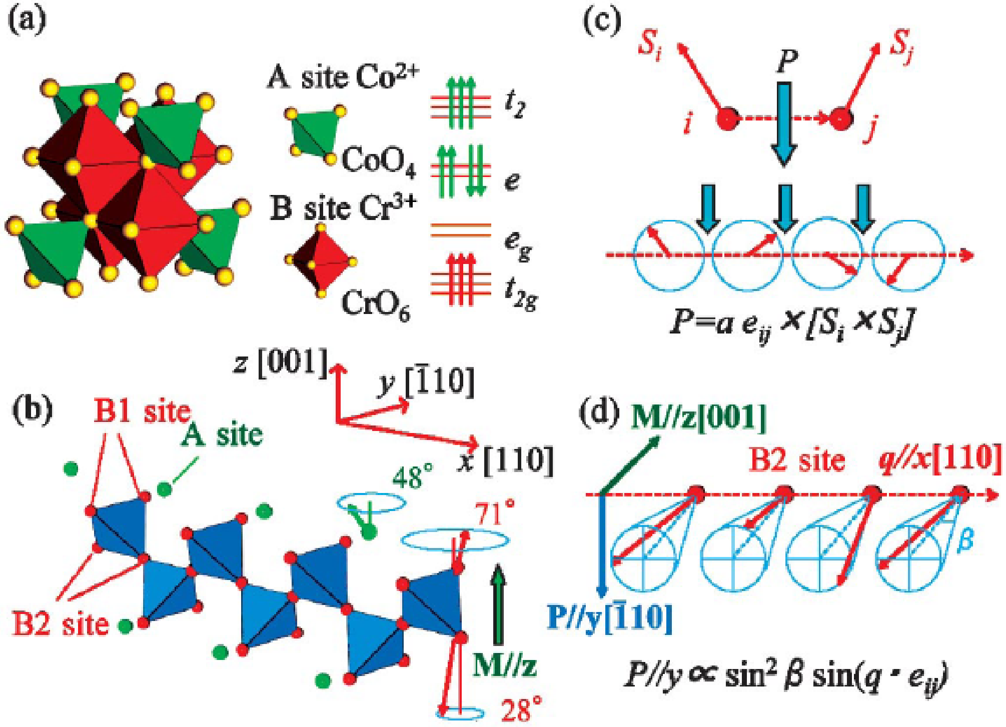


Kenzelman et al PRL 05

$CoCr_2O_4$

Co spins have ferromagnetic + spiral (conical) components

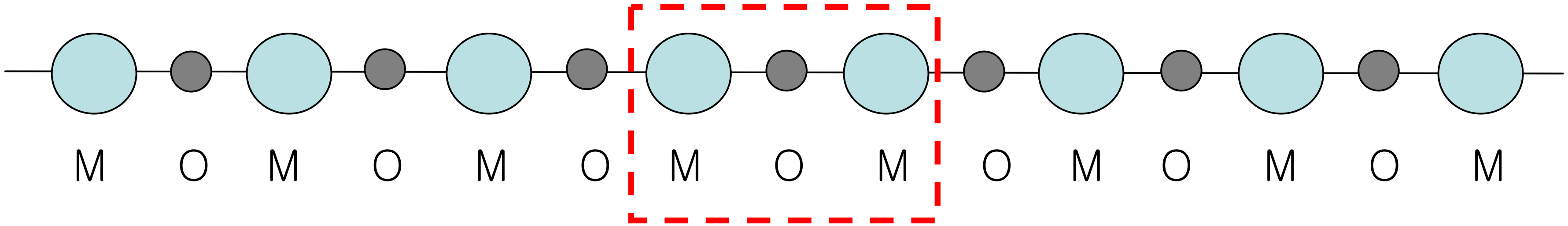
Emergence of spiral component accompanied by P



Tokura group PRL 06

Microscopic Theory of Spin-induced Polarization

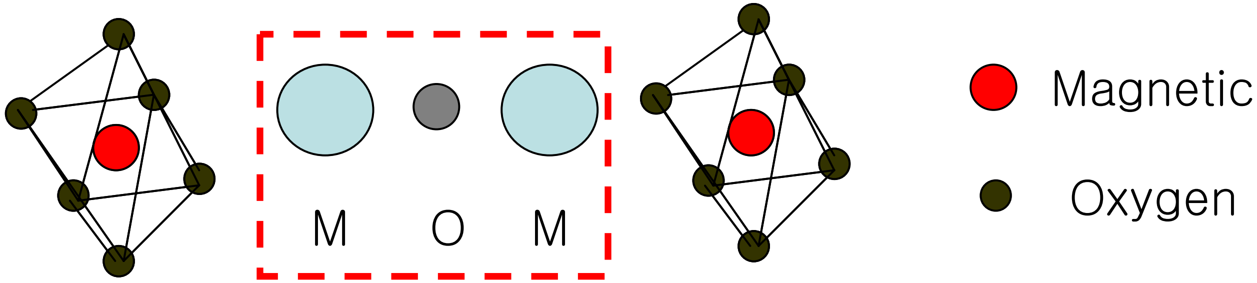
A linear chain consisting of alternating M(agnetic) and O(xygen) atoms is a reasonable model for magneto-electric insulators.



The building block is a single M-O-M cluster.

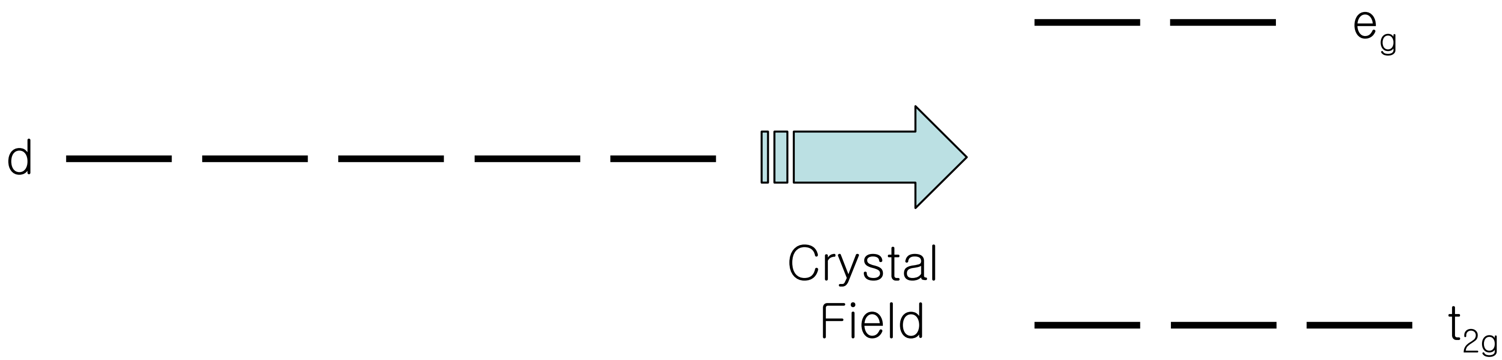
One tries to solve this model as exactly as possible to see if noncollinear-spin-induced polarization can be understood.

Microscopic Theory – Further Details

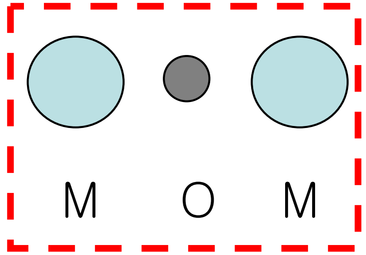


In magnetic atoms, d-orbital electrons are responsible for magnetism.
Keep the outermost d-orbitals and truncate out the rest.

Five-fold d-orbitals are further split into 3 t_{2g} and 2 e_g orbitals with a large energy gap of a few eV due to crystal field effects. Keep the t_{2g} or e_g levels only.



Microscopic Theory – Further Details



Electrons can "hop" between M and O sites as represented by a hopping integral V.

KEY ELEMENT: SPIN-ORBIT INTERACTION

Each magnetic site is subject to spin-orbit interaction.
If the spin state is polarized, so is the orbital state.

Theory of Katsura, Nagaosa, Balatsky (KNB)

The cluster Hamiltonian

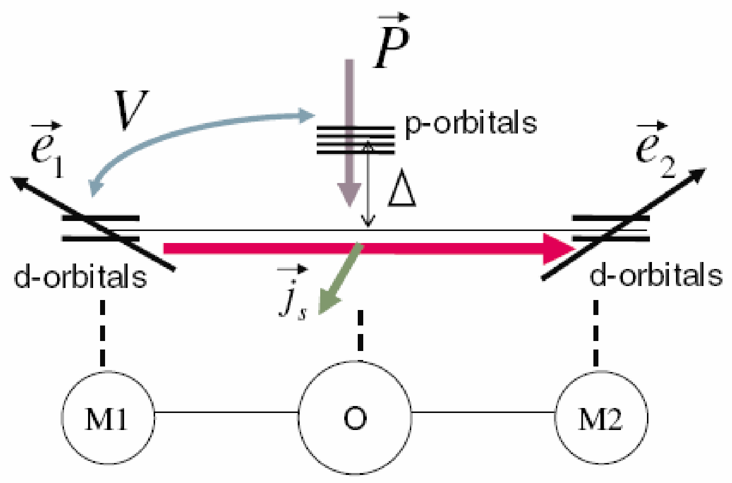
$$H = H_{SO} + H_M + H_O + H_V$$

$$H_{SO} = \lambda S \bullet L$$

$$H_M = -U \sum_{a=r,l} m_a \bullet \left(\sum_{l=xy,yz,zx} S_{a,l} \right)$$

$$H_O = E_p \sum_{b=x,y,z} \sum_{\sigma} p^{+}_{b\sigma} p_{b\sigma}$$

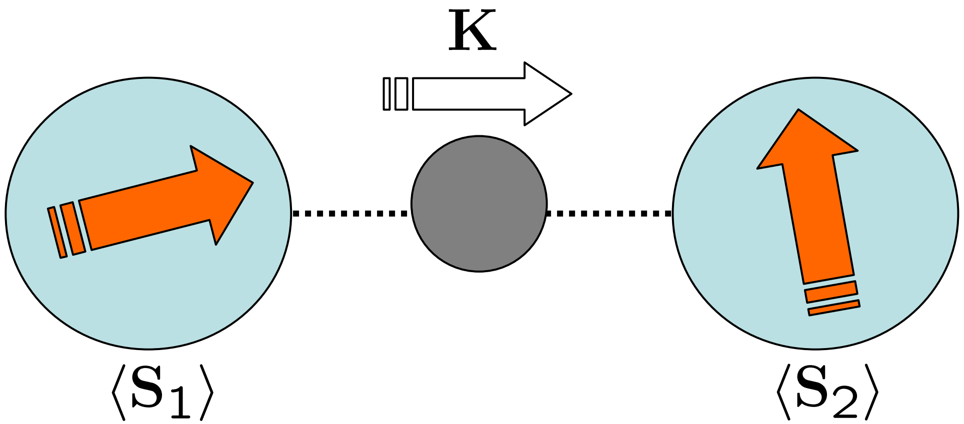
$$H_V = V \sum_{\sigma} \left[\left(d^{+}_{l,xy\sigma} p_{y\sigma} + d^{+}_{l,zx\sigma} p_{z\sigma} \right) - \left(d^{+}_{r,xy\sigma} p_{y\sigma} + d^{+}_{r,zx\sigma} p_{z\sigma} \right) \right] + h.c.$$



KNB PRL 05

KNB Hamiltonian is solved assuming λ (spin-orbit) > U (Hund)

Results of KNB

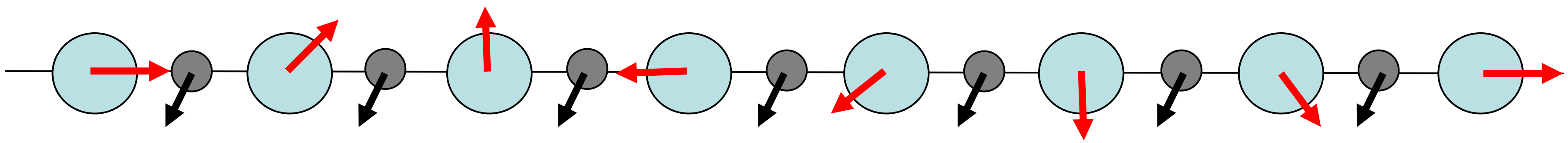


$$\mathbf{P} \propto \mathbf{K} \times \langle \mathbf{S}_1 \rangle \times \langle \mathbf{S}_2 \rangle$$

Polarization orthogonal to the spin rotation axis and modulation wave vector develops

Results of KNB

The results may be generalized to the lattice case; consistent with phenomenological theories

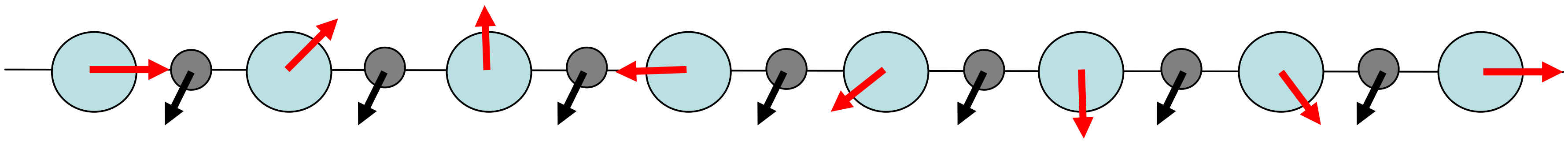


RED: spin orientation
BLACK: polarization

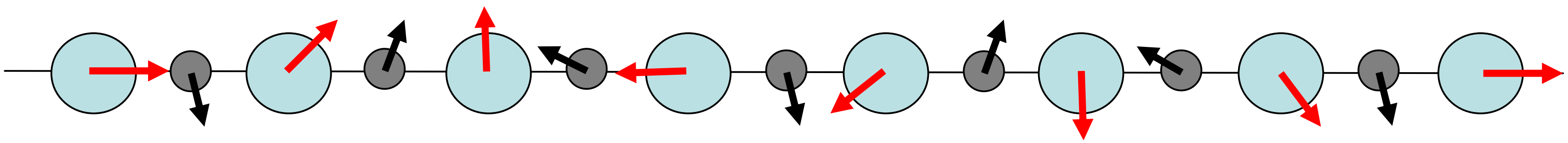
Our Recent Results

We revisited the KNB Hamiltonian in the limit of large Hund coupling U and small spin-orbit interaction λ, which is presumably more realistic.

A new (longitudinal) component of the polarization is found which was absent in past theories which only predicted transverse polarization.



BEFORE



AFTER

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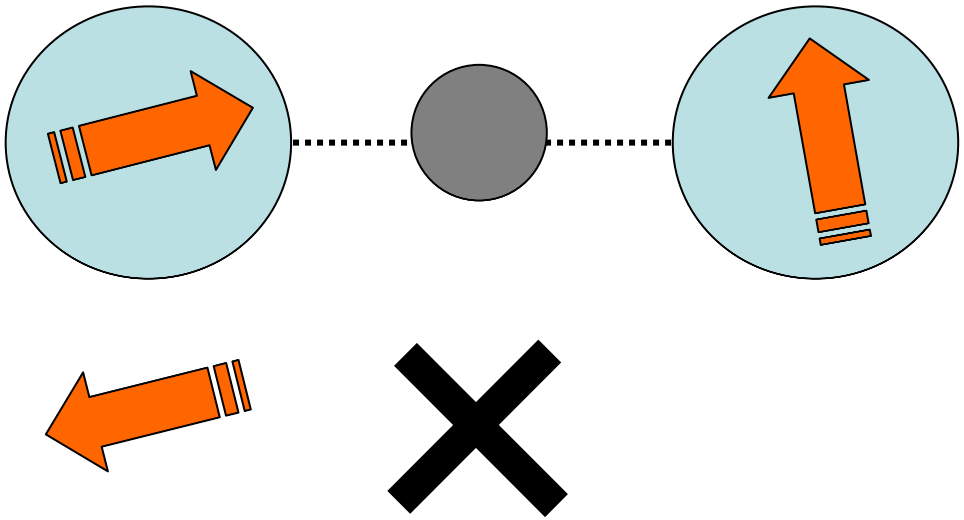
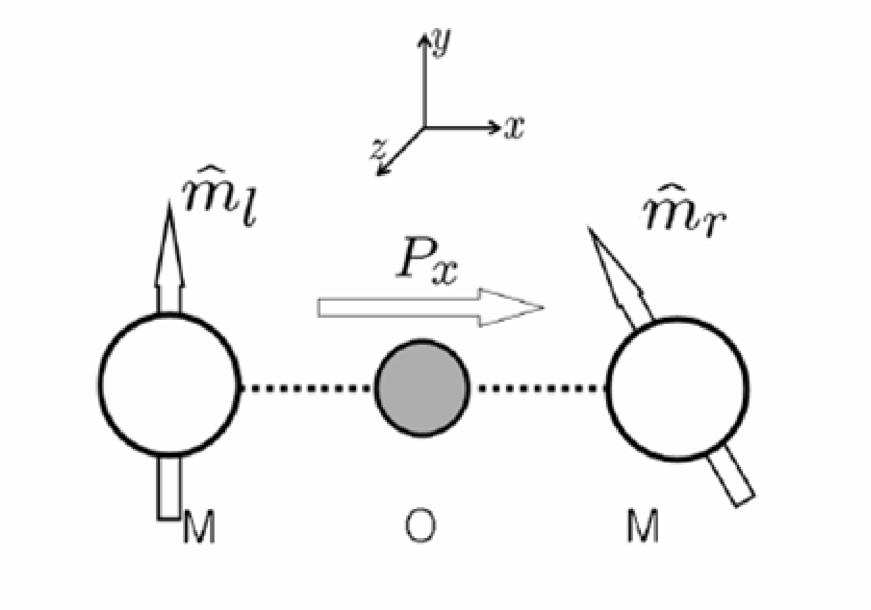
Technical Comment

Large-U offers a natural separation of spin-up and spin-down states for each magnetic site. All the spin-down states (antiparallel to local field) can be truncated out. This is similar to double-exchange physics.



Our Results



Spontaneous polarization exists ALONG the bond direction – LONGITUDINAL.

No transverse polarization was found.

(NB: KNB's theory in powers of U/λ, our theory in powers of λ/U)

Numerical Approach

KNB and our results probe different regions of parameter space.

We decided to compute polarization numerically without ANY APPROXIMATION

Exact diagonalization of the KNB Hamiltonian (only 16 dimensional) for arbitrary parameters (λ/V,U/V)

$$H = H_M + H_{SO} + H_O + H_V$$

$$H_M = -U \sum_{a=r,l} m_a \bullet \left(\sum_{l=xy,yz,zx} S_{a,l} \right)$$

$$H_{SO} = \lambda S \bullet L \qquad H_O = E_p \sum_{b=x,y,z} \sum_{\sigma} p^+_{b\sigma} p_{b\sigma}$$

$$H_V = V \sum_{\sigma} \left[\left(d^+_{l,xy\sigma} p_{y\sigma} + d^+_{l,zx\sigma} p_{z\sigma} \right) - \left(d^+_{r,xy\sigma} p_{y\sigma} + d^+_{r,zx\sigma} p_{z\sigma} \right) \right] + h.c.$$

Both longitudinal and transverse polarization types were found !

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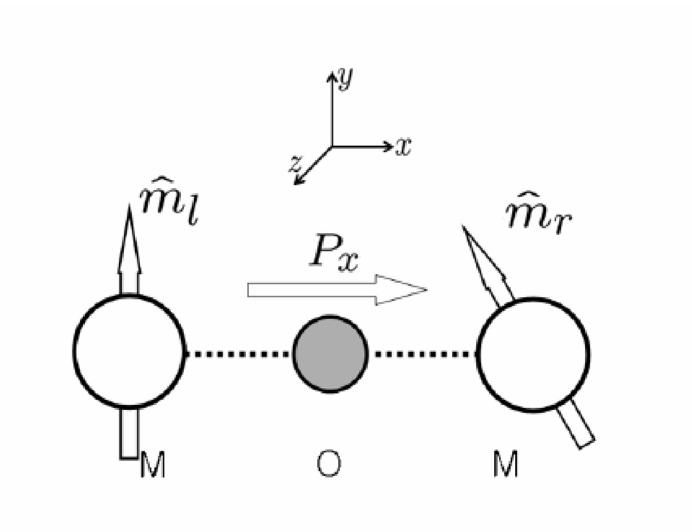
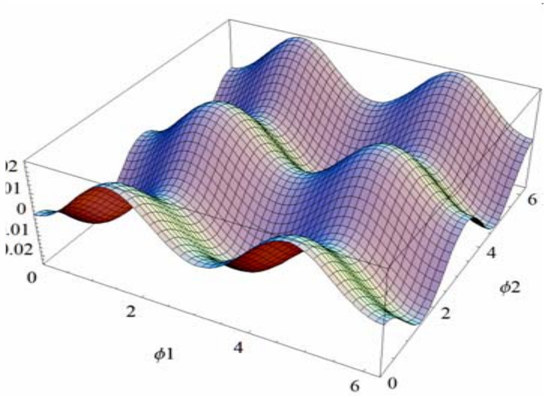
Numerical Results for Polarization

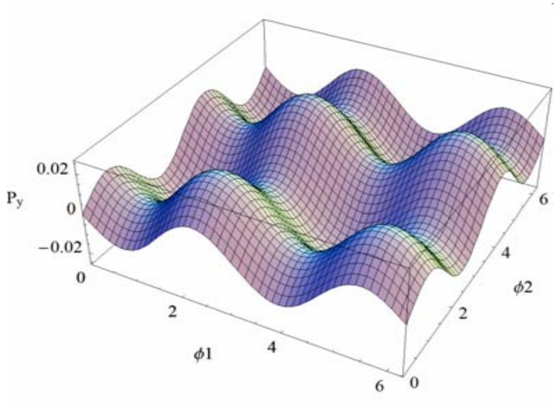
Transverse and longitudinal components exist which we were able to fit using very simple empirical formulas:



Px (longitudinal)



Py (transverse)



$$P_x/L = A\left[\cos(2\phi_r) - \cos(2\phi_l)\right]$$

$$P_y/L = -B_1 \sin(\phi_r - \phi_l) + B_2\left[\sin(2\phi_r) - \sin(2\phi_l)\right]$$

KNB

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Uniform vs. non-uniform

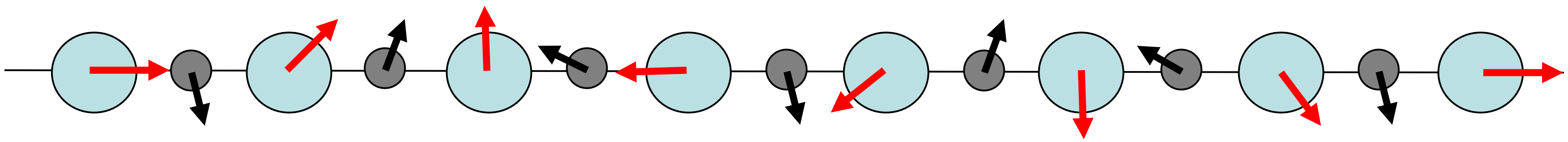
$$P_x/L = A\left[\cos(2\phi_r) - \cos(2\phi_l)\right]$$ NON-UNIFORM

$$P_y/L = -B_1 \sin(\phi_r - \phi_l) + B_2\left[\sin(2\phi_r) - \sin(2\phi_l)\right]$$

UNIFORM NON-UNIFORM

When extended to spiral spin configuration, P_x gives oscillating polarization with period half that of spin.

P_y has oscillating (B_2) as well as uniform (B_1,KNB) component



Macroscopic polarization only proportional to B_1
So far the only polarization component detected experimentally.

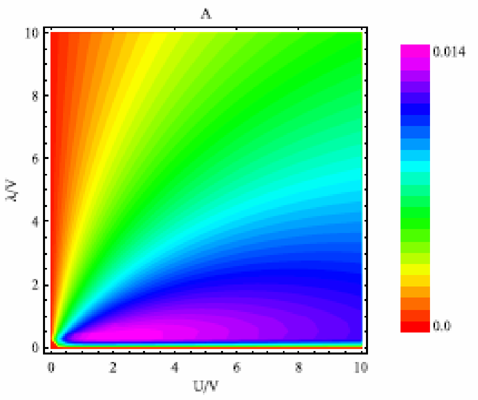
Uniform vs. non-uniform

What people normally detect is macroscopic (uniform) polarization but that may not be the whole story. Non-uniform polarization, if it exists, is likely to lead to some modulation of atomic position which one can pick up with X-rays.

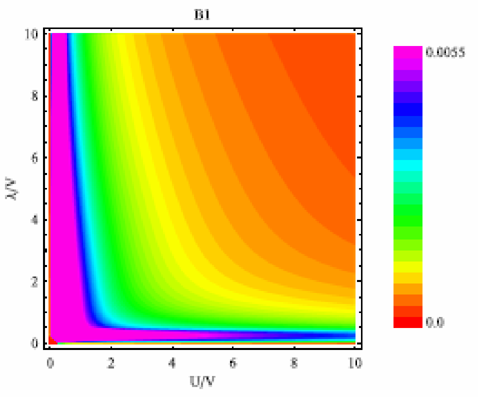
How big is the non-uniform component locally?

Coefficients

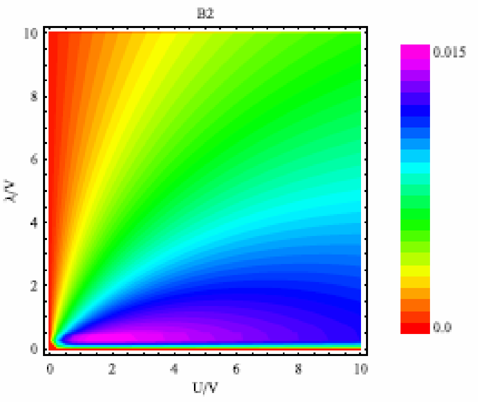
A



B_1



B_2



$$P_x/L = A\left[\cos(2\phi_r) - \cos(2\phi_l)\right]$$

$$P_y/L = -B_1 \sin(\phi_r - \phi_l) + B_2\left[\sin(2\phi_r) - \sin(2\phi_l)\right]$$

The uniform transverse component B_1 is significant for small U (KNB limit).

A and B_2 (non-uniform) are dominant for large U (our limit).

$$A \sim B_2 \sim 200 \; nC / cm^2$$

$$B_1 \sim 80 \; nC / cm^2$$

WORTH A TRY !

Summary

Spin-orbit coupling leads to interesting magnetism-induced polarization of electronic wave function as observed in a class of magnetic materials.

Induced polarization has longitudinal and transverse, uniform and non-uniform components with non-trivial dependence on spin orientations.

Detecting such local ordering of polarization will be interesting.

Relevance to Nanosystems (SPECULATIONS)

The theory may be applicable to quantum dot arrays consisting of two kinds of dots

$$-D_1-D_2-D_1-D_2-D_1-D_2-D_1-D_2-$$

Or a long molecule with several magnetic sites in it.

Transport (I–V) through such a system may be susceptible to local electric polarization profile.