- **Ferromagnetic Nanoparticles**
  - Berry Phase
  - Ab initio Calculations

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Motivation

- Ferromagnetic Nanoparticles
  - Potentially useful for high density information storage and spin electronics
- Model
  - Effective Hamiltonian with a single giant spin degree of freedom
- Berry curvature Chern number
  - Specifies the total spin of the system, i.e., the dimension of the Hilbert space
- Ab initio Calculations
  - Realistic many-body calculations which can determine the Hamiltonian of the nanoparticles

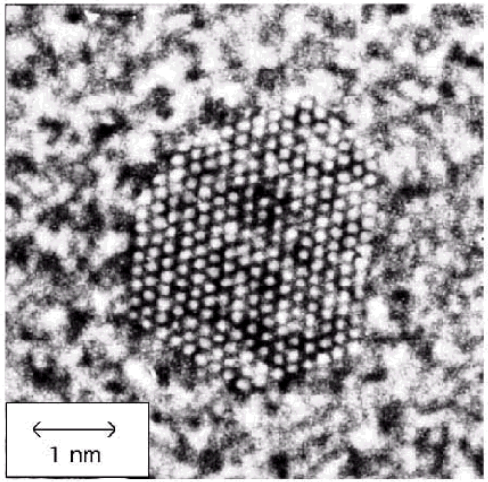
Outline

1. Model
2. Berry Phase
3. Numerical Results
4. Ab initio Calculations
5. Future Work

1. Model

- System
  - Transition metal clusters

  Ex) $^{27}Co : [^{18}Ar]3d^{7}4s^{2}$
- Hamiltonian
  - Tight-binding model with 3d-4s-4p orbitals
  - Local d-orbital exchange interaction treated in mean field approximation



Jamet et al. PRL **86**, 4676 (2001)

$$H = H_{band} + H_{exch} + H_{SO} + H_{Zee}$$

1. Model

1) Band Hamiltonian

$$H_{band} = \sum_{i,j} \sum_{s} \sum_{\mu_1,\mu_2} t^{i,j}_{\mu_1,\mu_2,s} c^{+}_{i,\mu_1,s} c_{j,\mu_2,s}$$

$c^{+}_{i,\mu_1,s}$, $c_{j,\mu_2,s}$: creation, annihilation

$t^{i,j}_{\mu_1,\mu_2,s}$: electron hopping

i, j : atomic site
μ_1, μ_2 : atomic orbital
s : spin

- Eigenvalue problem in non-orthogonal basis

$$H\vec{x} = EO\vec{x} \quad \rightarrow \quad H'\vec{x}' = E\vec{x}'$$

$$H' = O^{-1/2} H O^{-1/2}, \quad x' = O^{1/2} x$$

$$\langle i;\mu_1 \,|\, j;\mu_2 \rangle \neq \delta_{i,j}\delta_{\mu_1,\mu_2}$$

O : overlap matrix
$\vec{x}$: eigenstate

1. Model

2) Exchange Hamiltonian

$$H_{exch} = -2U_{dd}\sum_i \vec{S}_{i,d}\cdot\vec{S}_{i,d}$$

$\Rightarrow$ Largely responsible for magnetic order

- Mean-field approximations

$$\vec{S}_{i,d} = \left\langle \vec{S}_{i,d} \right\rangle + \left(\vec{S}_{i,d} - \left\langle \vec{S}_{i,d} \right\rangle \right)$$

$$g_s \mu_B \vec{h} \equiv 2U_{dd} \left\langle \vec{S}_{i,d} \right\rangle$$

All spins change their orientation coherently

$$H_{exch} = -2g_s\mu_B\vec{h}\cdot\sum_i \vec{S}_{i,d} + (\text{fluctuations})$$

1. Model

3) Spin-orbit interaction

$$H_{SO} = \xi_d \sum_i \sum_{s_1,s_2} \sum_{\mu_1,\mu_2} \left\langle \mu_1, s_1 \left| \vec{L} \cdot \vec{S} \right| \mu_2, s_2 \right\rangle c^{+}_{i,\mu_1,s_1} c_{i,\mu_2,s_2}$$

$\Rightarrow$ Gives rise to magnetocrystalline anisotropy energy

Helps determine orientation of the magnetization

Note) Exchange interaction is isotropic

4) Zeeman Hamiltonian

$$H_{Zee} = \mu_B \sum_i \sum_{s_1,s_2} \sum_{\mu_1,\mu_2} \left\langle \mu_1, s_1 \left| \vec{L} + g_S \vec{S} \right| \mu_2, s_2 \right\rangle \cdot \vec{H}_{ext}\, c^{+}_{i,\mu_1,s_1} c_{i,\mu_2,s_2}$$

$\Rightarrow$ Reorients the magnetization

2.Berry Phase

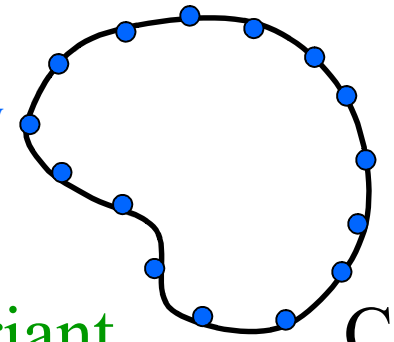
1) Basic concept

- For a parameter-dependent Hamiltonian $H(\vec{R})$

$$e^{i\Delta\gamma_n} = \left\langle n(\vec{R}) \,|\, n(\vec{R}-\Delta\vec{R}) \right\rangle$$

$$\Delta\gamma_n \approx i\left\langle n(\vec{R}) \,|\, \nabla_{\vec{R}} n(\vec{R}) \right\rangle \cdot \Delta\vec{R}$$

pure imaginary



$\Rightarrow$ For closed path C, $\gamma_n(C)$ is gauge invariant

$$\gamma_n(C) = \oint_C \vec{A}_n \cdot d\vec{R} = -\int_S \vec{F}_n \cdot d\vec{a}$$ (Berry Phase)

$$\vec{A}_n \equiv i\left\langle n(\vec{R}) \,|\, \nabla_{\vec{R}} n(\vec{R}) \right\rangle$$ (Berry Connection)

$$\vec{F}_n \equiv -\nabla_{\vec{R}} \times \vec{A}_n$$ (Berry Curvature)

2.Berry Phase

2) Example- Quantum Spinning System

$$H(\vec{R}) = -g_J \mu_B \vec{B} \cdot \vec{J} \equiv \vec{R} \cdot \vec{J}$$

$$H(\vec{R}) \left| k; \vec{R} \right\rangle = kR \left| k; \vec{R} \right\rangle$$

$$\vec{A}_k = i \left\langle k; \vec{R} \mid \nabla_{\vec{R}} k; \vec{R} \right\rangle_{\theta \neq \pi} = \hat{e}_\phi \frac{k(\cos\theta - 1)}{R \sin\theta}$$

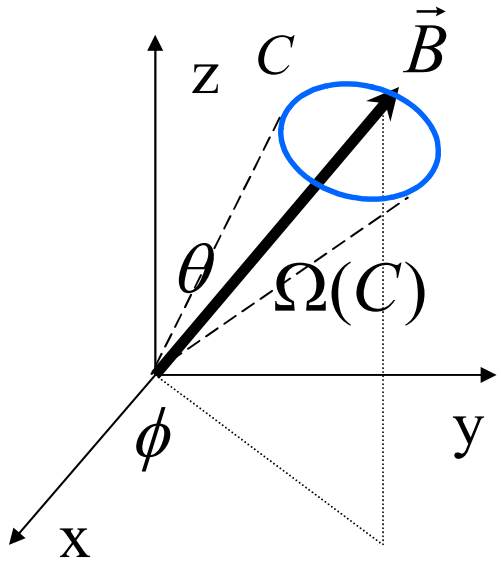
(Berry connection)

$$\vec{F}_k = -\nabla_{\vec{R}} \times \vec{A}_k = \hat{e}_r \frac{k}{R^2}$$

(Berry curvature)

$$\gamma_k(C) = -\int_S \vec{F}_k \cdot d\vec{a} = -k\Omega(C)$$

(Berry phase)



$\Rightarrow$ Same form of a magnetic monopole $\frac{e_M e}{\hbar c} = \frac{1}{2} \times \text{integer}$

$\Rightarrow$ k is a half-of-integer

2.Berry Phase

3) Ferromagnetic cluster

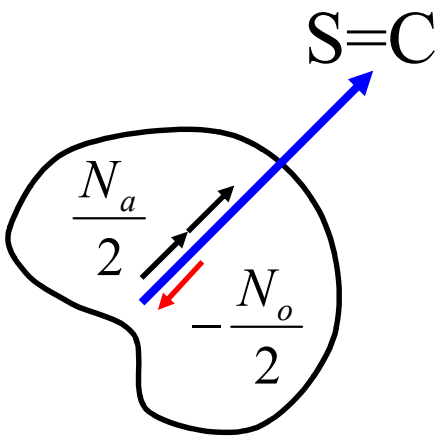
- Without spin-orbit interaction

$$\vec{F} = \frac{\hat{e}_r}{R^2}\left(\frac{N_a - N_o}{2}\right) \quad (\vec{R} \sim \vec{h})$$

(Berry curvature)

$$C \equiv \frac{1}{4\pi}\oiint_{R=1} \vec{F}\cdot d\vec{a} = \frac{N_a - N_o}{2}$$

(Chern number)



N_a, N_o : number of ½ spin aligned with, opposed to S

- When spin-orbit interaction is present

$\Rightarrow$ Chern number C is still a half-of-integer and considered as the sum over contributions from each orbital

$\Rightarrow$ Chern number specifies the total spin of the system

4) Numerical methods

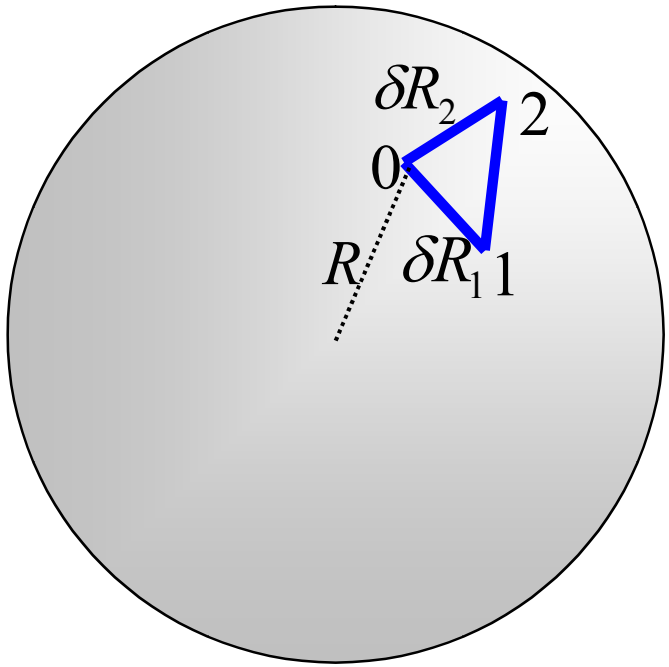
$$\gamma(C) = \text{phase of } \langle 0|2\rangle\langle 2|1\rangle\langle 1|0\rangle$$

(Berry phase)

$$= -\int_{R=1} \vec{F}\cdot d\vec{a} \approx -\frac{1}{2} F_0\, \delta R_1\, \delta R_2$$

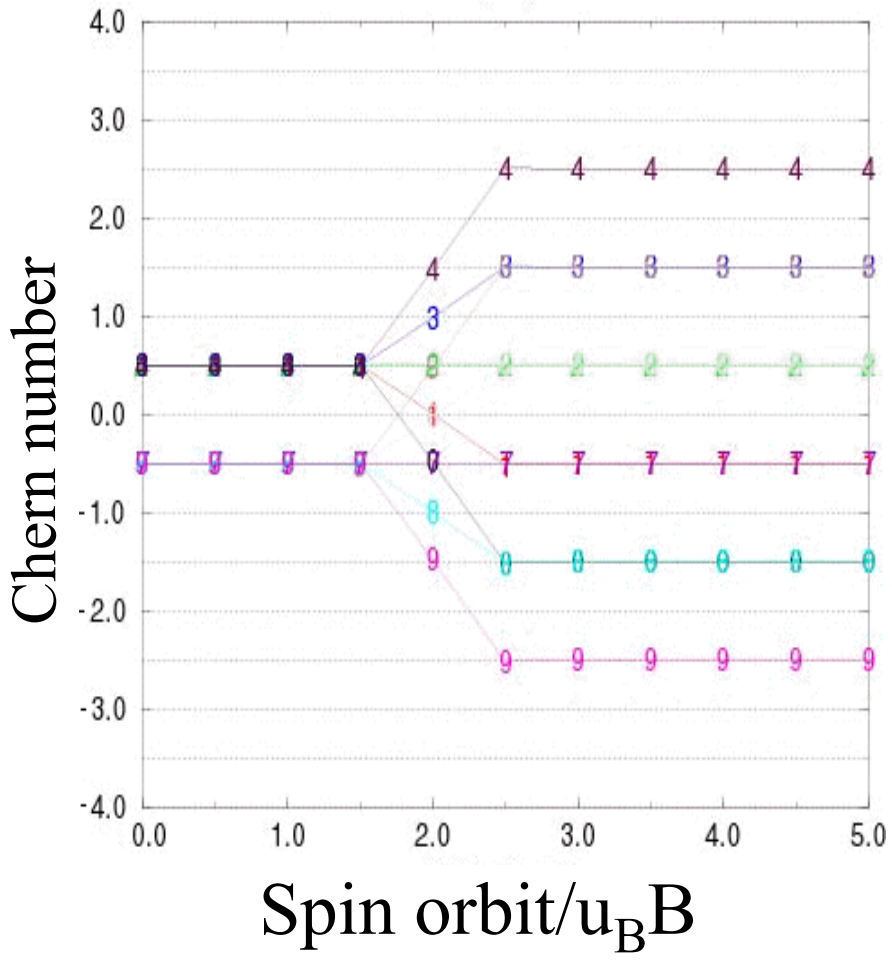
$$F_0 = -\frac{\text{phase of } \langle 0|2\rangle\langle 2|1\rangle\langle 1|0\rangle}{1/2\, \delta R_1\, \delta R_2}$$

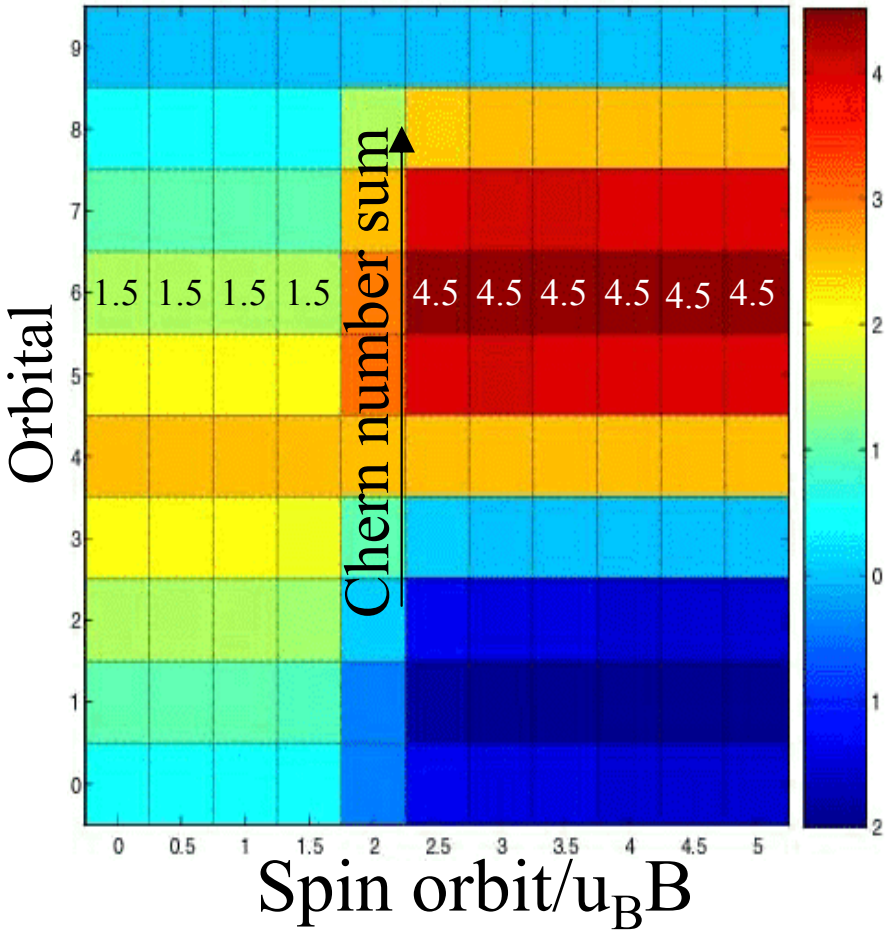
(Berry curvature)



3.Numerical Results

1) Chern number vs Spin orbit interaction for a single atom





3.Numerical Results

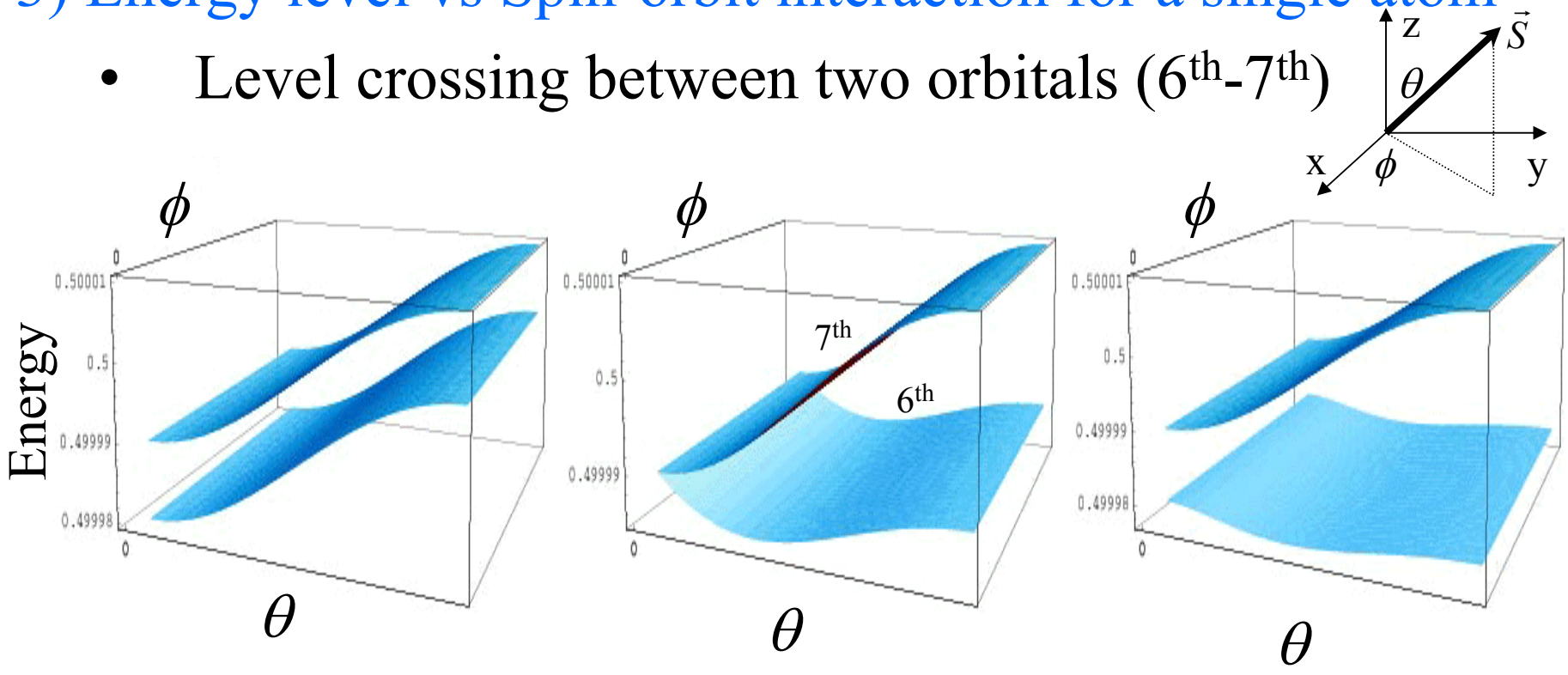
2) Hund's rules and Chern number in d-shells

Ground state	S	L	J	C_i	$\lvert\sum C_i\rvert$
↓	0.5	2.0	1.5	-1.5	1.5
↓ ↓	1.0	3.0	2.0	-0.5	2.0
↓ ↓ ↓	1.5	3.0	1.5	0.5	1.5
↓ ↓ ↓ ↓	2.0	2.0	0.0	1.5	0.0
↓ ↓ ↓ ↓ ↓	2.5	0.0	2.5	2.5	2.5
↓↑↓ ↓ ↓ ↓	2.0	2.0	4.0	1.5	4.0
↓↑↓↑↓ ↓ ↓	1.5	3.0	4.5	0.5	4.5
↓↑↓↑↓↑↓ ↓	1.0	3.0	4.0	-0.5	4.0
↓↑↓↑↓↑↓↑↓	0.5	2.0	2.5	-1.5	2.5
↓↑↓↑↓↑↓↑↓↑	0.0	0.0	0.0	-2.5	0.0

3.Numerical Results

3) Energy level vs Spin-orbit interaction for a single atom

- Level crossing between two orbitals (6^{th}-7^{th})

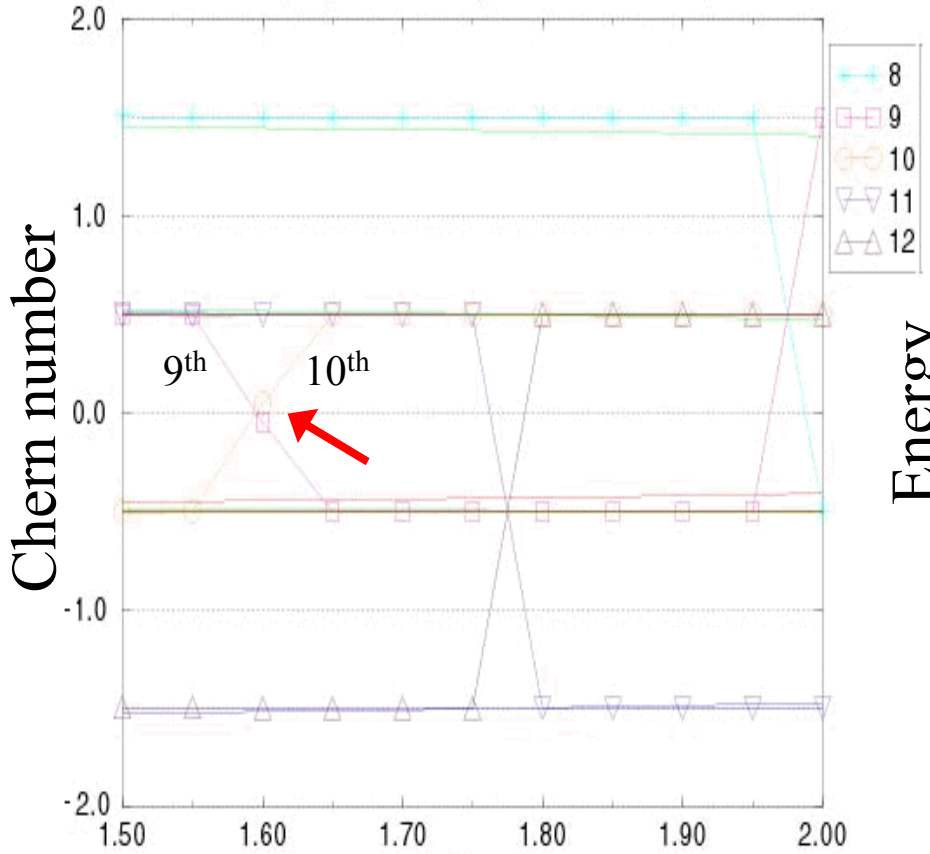


Spin orbit/u_BB=0 Spin orbit/u_BB=2 Spin orbit/u_BB=4

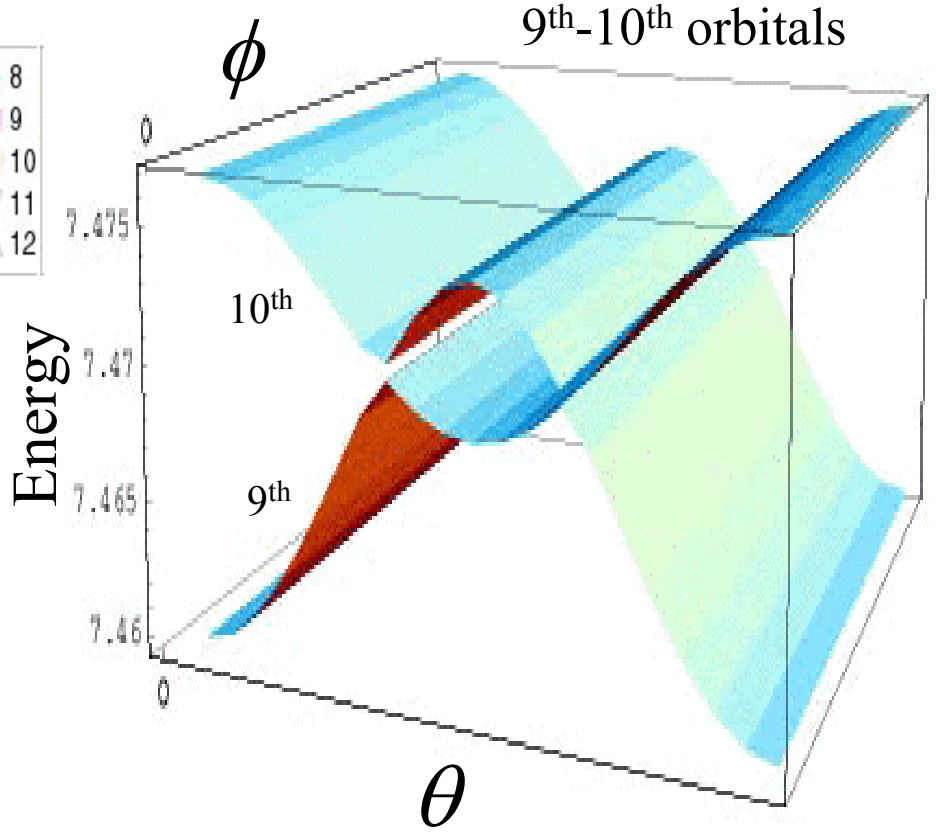
$\Rightarrow$ Chern numbers change only when level crossing occurs

3.Numerical Results

4) Chern number vs Hopping for 2-atoms along z-axis



Hopping ratio



Hopping ratio~1.6

4. Ab initio calculations

1) Approximations in many-body problems

- Ion-ion interactions

$\Rightarrow$ Born-Oppenheimer approximation

- Electron-electron interactions

$\Rightarrow$ Density functional theory

- Electron-ion interactions

$\Rightarrow$ Pseudopotential theory

4. Ab initio calculations

2) Electron-electron interactions : Density functional theory

$$\left(-\frac{\hbar^2}{2m}\nabla^2 + \underbrace{V_{ion}[n] + \int \frac{e^2 n(\vec{x}')}{|\vec{x}-\vec{x}'|} d\vec{x}' + V_{xc}[n]}_{V_{eff}[n]} \right) |\phi_i\rangle = \varepsilon_i |\phi_i\rangle$$

$$n(\vec{x}) = \sum |\phi_i(\vec{x})|^2$$

V_{xc} : exchange-correlation potential

Self-consistent method : $n^{(1)} \rightarrow \phi^{(1)} \rightarrow n^{(2)} \rightarrow \phi^{(2)} \rightarrow \cdots$

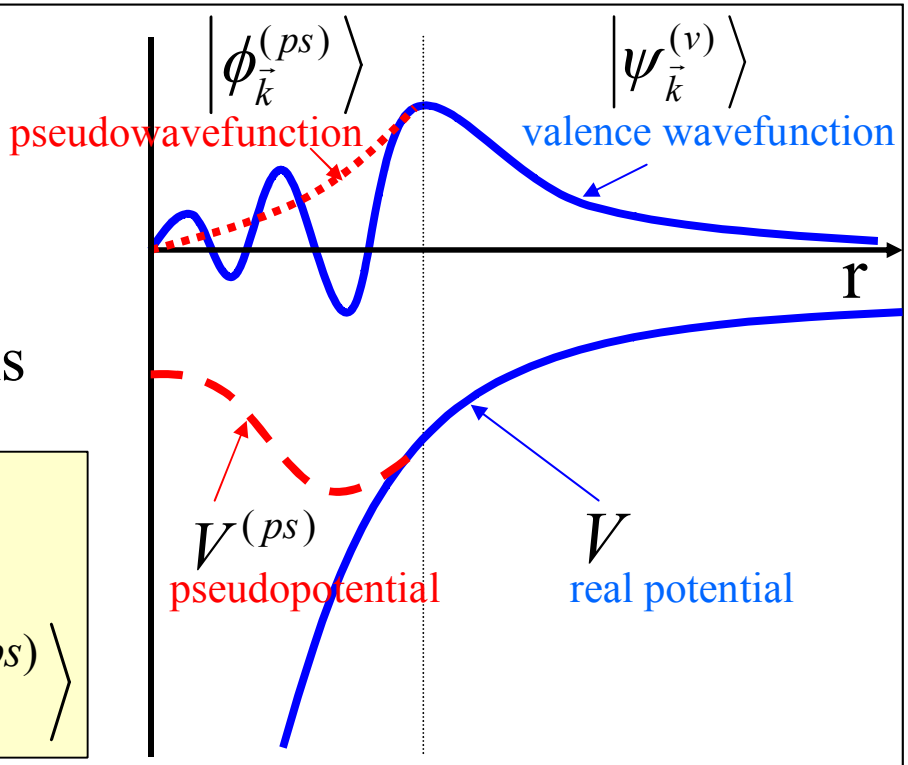
$\Rightarrow$ Mapping of the interacting many-electron system onto a system of non-interacting electrons moving in an effective potential due to all the other electrons

4. Ab initio calculations

3) Electron-ion interactions

: Pseudopotential theory

Physical properties of solids are dependent on the valence electrons



valence

$$\Rightarrow \left(H_0 + V \right) \left| \psi_{\vec{k}}^{(v)} \right\rangle = \varepsilon_{\vec{k}}^{(v)} \left| \psi_{\vec{k}}^{(v)} \right\rangle$$

pseudo

$$\left(H_0 + V^{(ps)} \right) \left| \phi_{\vec{k}}^{(ps)} \right\rangle = \varepsilon_{\vec{k}}^{(v)} \left| \phi_{\vec{k}}^{(ps)} \right\rangle$$

$\left| \psi_{\vec{k}}^{(v)} \right\rangle$ = Sum of a large number of plane waves

$\left| \phi_{\vec{k}}^{(ps)} \right\rangle$ = Sum of a small number of plane waves

cf) $V^{(ps)}$ less singular $\Rightarrow$ Nearly free-electron

4. Ab initio calculations

4) Current work : I'm learning VASP

- VASP (Vienna Ab-initio Simulation Package)
  - Pseudopotential
  - Local density approximation
  - Plane-wave basis set

$\Rightarrow$ In VASP, it is available to do constrained magnetic moment calculations changing its orientation

5. Future work

- Calculate Berry phase for Co clusters (N = 2 ~ 100)
- Compare results with those of ab initio calculations
- Calculate other magnetic properties
  - Energy level statistics
  - Anisotropy energy
  - Magnetization dependence on magnetic fields
  - …

References

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