2024 Feb 20-22 @RIKEN Kobe

Relativistic corrections to strongly correlated electron systems

Department of Physics, Saitama University

Shintaro Hoshino

Acknowledgement :
Michi-To Suzuki (Tohoku Univ), **Hiroaki Ikeda** (Ritsumeikan Univ), **Tatsuya Miki** (D1, Saitama Univ),
H Kusunose (Meiji Univ), **MJS Yang** (Saitama Univ), **S Michimura** (Saitama Univ),
M Fukuda (ISSP), **M Senami** (Kyoto Univ), **M Hirayama** (U Tokyo), **T Naito** (RIKEN)

Outline

1. Introduction

2. Multipoles in strongly correlated electron systems

3. Spherical representation of Coulomb-Breit interaction:
Interaction among multipoles *(Generalized Slater-Condon parameters)*

4. Summary

Relativistic correction in condensed matter

Conventional relativistic correction: Spin-orbit coupling

・Materials with large-atomic number elements:
ex. 5d electrons, f electrons: J-multiplet

・Transition metals including 3d series, organic conductors:
ex. Dzyaloshinskii-Moriya interaction
⇒ small energy scale but can be relevant

For elements with large atomic number, higher-order relativistic corrections can be relevant for low-energy behaviors?

Three relativistic corrections

A. Physical quantities (ex. electron chirality) — SH, M-T Suzuki, H Ikeda, Phys. Rev. Lett. (2023)

B. One-body Hamiltonians ($1/c^3$ corrections) — SH, T. Miki, M-T Suzuki, H Ikeda, in preparation

☞ **C. Interactions (Breit interaction)** — SH, arXiv (2023)

Today's focus

Outline

1. Introduction

2. Multipoles in strongly correlated electron systems

3. Spherical representation of Coulomb-Breit interaction:
Interaction among multipoles (Generalized Slater-Condon parameters)

4. Summary

strongly correlated electron systems

Multipole in SCES

- ☞ Classification scheme for nearly-localized d- and f-electron's degrees of freedom.
- ☞ Candidate order parameter for exotic ordered states.
- ☞ Cluster multipoles, bond orders, multipole expansion in ***k***-space, …

KI Kugel and DI Khomskii, Sov. Phys. JETP Lett. (1972)
FJ Ohkawa, JPSJ (1983)
R Shiina, H Shiba, P Thalmeier, JPSJ (1997)
Y Kuramoto, H Kusunose, JPSJ (2000)
P Santini, G Amoretti, PRL (2000)
A Kiss and Y Kuramoto, JPSJ (2005)
T Takimoto, JPSJ (2005)
H Kusunose, JPSJ (2008)
K Haule and G Kotliar, Nat. Phys. (2009)
H Ikeda, MT Suzuki, R Arita, T Takimoto, T Shibauchi, Y Matsuda, Nat. Phys. (2012)
MT Suzuki, T Koretsune, M Ochi, R Arita, PRB (2017)
S Hayami, M Yatsushiro, Y Yanagi, H, Kusunose, PRB (2018)
and more …

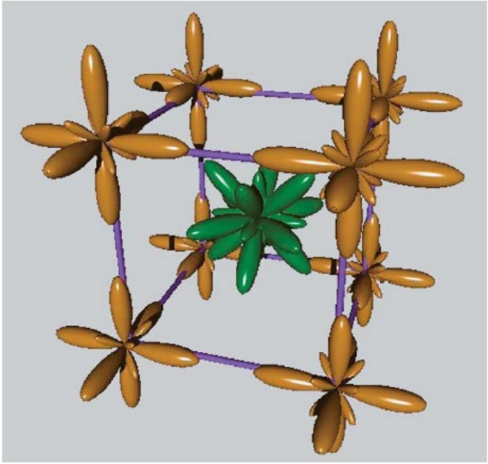
CeB_6

$Ce_{1-x}La_xB_6$

NpO_2

$PrRu_4P_{12}$

URu_2Si_2



Y Kuramoto, PTP Suppl. (2008)

Electronic degrees of freedom

☞ **Spin degrees of freedom**

$|\sigma\rangle \quad (\sigma = \uparrow, \downarrow)$ $\qquad \boldsymbol{s} = \frac{1}{2}$

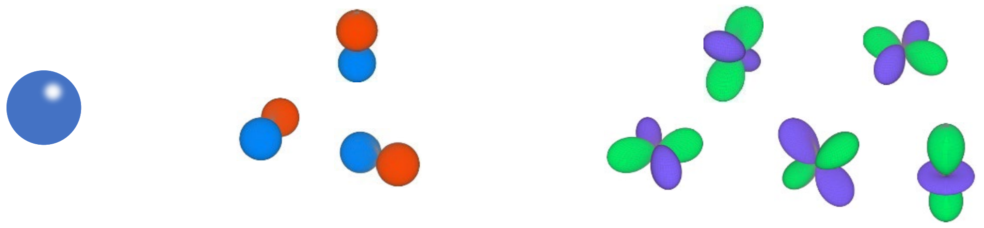
(in terms of multipole)

Operators $\quad n = \sum_{\sigma} |\sigma\rangle\langle\sigma|, \quad \boldsymbol{s} = \frac{1}{2}\sum_{\sigma\sigma'} |\sigma\rangle \boldsymbol{\sigma}_{\sigma\sigma'} \langle\sigma'|$

Charge (TR+) *Spin (TR-)*

$\boldsymbol{S} = \frac{1}{2} \times \frac{1}{2} = 0 + 1$ **(monopole + dipole)**

"rank"



☞ **Orbital degrees of freedom** (ex. p electron)

$|m\rangle \quad (m = 0, \pm 1)$ $\qquad \boldsymbol{\ell = 1}$

Figs from **H Kusunose & Y Kuramoto, KOTAI BUTSURI (2006)**

Classification of operators ($|m\rangle\langle m'|$) $\quad \boldsymbol{L = 1 \times 1 = 0 + 1 + 2}$ **(monopole + dipole + quadrupole)**

Charge (TR+), Magnetic orbital (TR-), Electric orbital (TR+)

Even/Odd of rank corresponds to Even/Odd of time-reversal.

Spin-orbit coupled case

☞ Classification of $|m\sigma\rangle\langle m'\sigma'| \sim c^\dagger_{m\sigma} c_{m'\sigma'}$ with L-S coupled basis (T^{JM}_{LS})
(Spatial inversion is always even for a fixed ℓ)

Rank $p = 2q$ or $2q + 1$
SI: Spatial Inversion / TR: Time-reversal

Multipole	Type	SI/TR
Electric Toroidal	$(2q+1)_d$	$+/+$
Magnetic Toroidal	$(2q)_d$	$+/-$
Electric	$(2q)_{a,b,c}$	$+/+$
Magnetic	$(2q+1)_{a,b,c}$	$+/-$

Example 1: $L = 1$ (TR=odd) and $S = 1$ (TR=odd)

$$J = 1 \times 1 = 0_+ + 1_+ + 2_+$$

Electric toroidal (ET) dipole (Odd-rank,TR=even)

Example 2: $L = 2$ (TR=even) and $S = 1$ (TR=odd)

$$J = 2 \times 1 = 1_- + 2_- + 3_-$$

Magnetic toroidal (MT) quadrupole (Even-rank,TR=odd)

Examples of dipoles:
M dipole: $\boldsymbol{L}$, $\boldsymbol{S}$
ET dipole: $\boldsymbol{G} = \boldsymbol{L} \times \boldsymbol{S}$

Multipole operator $$X^\gamma(p_\eta) = \sum_{mm'\sigma\sigma'} c^\dagger_{m\sigma} \mathcal{O}^\gamma_{m\sigma,m'\sigma'}(p_\eta) c_{m'\sigma'}$$

- Y Wang, H Weng, L Fu, X Dai, PRL (2017)
- H Kusunose, R Oiwa, S Hayami, JPSJ (2020)
- N Chikano, SH, H Shinaoka, PRB (2021)
- S Hayami, R Oiwa, H Kusunose, JPSJ (2022)
- SH, MT Suzuki, H Ikeda, PRL (2023)

Outline

1. Introduction

2. Multipoles in strongly correlated electron systems

3. Spherical representation of Coulomb-Breit interaction:
Interaction among multipoles (Generalized Slater-Condon parameters)

4. Summary

Relativistic correction to SCES: Motivation

Exotic behaviors in uranium-based materials:
☞ Hidden order (URu2Si2)
☞ Spin-triplet superconductor candidates (UBe13, UPt3, UGe2, URhGe, UCoGe, UTe2)

Why uranium (atomic number = 92) material is so special?
☞ Large atomic number → Relevance of **relativistic corrections**
☞ Localized f orbital → Relevance of **Coulomb interactions**

⇨ Possible relevance of **relativistic correction to Coulomb interaction**

Most general Hamiltonian (Coulomb gauge)

$$\mathscr{H} = \mathscr{H}_0 + \frac{1}{2}\int \mathrm{d}\boldsymbol{r}\mathrm{d}\boldsymbol{r}' \frac{\rho(\boldsymbol{r})\rho(\boldsymbol{r}')}{|\boldsymbol{r}-\boldsymbol{r}'|} + \sum_{\boldsymbol{k}\alpha} \hbar\omega_{\boldsymbol{k}} a^{\dagger}_{\boldsymbol{k}\alpha} a_{\boldsymbol{k}\alpha} - \frac{1}{c}\int \mathrm{d}\boldsymbol{r}\, \boldsymbol{j}(\boldsymbol{r})\cdot\boldsymbol{A}(\boldsymbol{r})$$

Coulomb-Breit interaction

Tracing out photons and keep leading-order term in 1/c expansion

T Itoh, Rev. Mod. Phys. (1965)

$$\mathscr{H}_C = \frac{1}{2}\int \frac{\rho_1\rho_2}{r} - \frac{\pi\lambda\!\!\!^{-2}}{2}\int \delta(\boldsymbol{r})\rho_1\rho_2 - \int \frac{\boldsymbol{r}\cdot\boldsymbol{P}_1\,\rho_2}{r^3}$$ (Coulomb)

($\lambda\!\!\!^{-} = \frac{\hbar}{mc}$: reduced Compton length)

$$\mathscr{H}_B = -\frac{1}{4c^2}\int \frac{1}{r}\left[\boldsymbol{j}_1\cdot\boldsymbol{j}_2 + \frac{(\boldsymbol{j}_1\cdot\boldsymbol{r})(\boldsymbol{j}_2\cdot\boldsymbol{r})}{r^2}\right]$$
$$- \frac{1}{c}\int \frac{(\boldsymbol{r}\times\boldsymbol{M}_1)\cdot\boldsymbol{j}_2}{r^3}$$
$$+ \frac{1}{2}\int \left[-\frac{8\pi}{3}\delta(\boldsymbol{r})\boldsymbol{M}_1\cdot\boldsymbol{M}_2 + \left(\frac{\boldsymbol{M}_1\cdot\boldsymbol{M}_2}{r^3} - \frac{3(\boldsymbol{M}_1\cdot\boldsymbol{r})(\boldsymbol{M}_2\cdot\boldsymbol{r})}{r^5}\right)'\right]$$ (Breit)

$[\boldsymbol{r} = \boldsymbol{r}_1 - \boldsymbol{r}_2,\ A_1 = A(\boldsymbol{r}_1)]$

Cf. in quantum chemistry / atomic physics context

Ex.) E Eliav et al., Nucl. Phys. A (2015)

T Naito, R Akashi, H Liang, S Tsuneyuki, J. Phys. B (2020)

SCES: Slater-Condon parameters (F0,F2,…) describe U_{ijkl} effectively for conventional Coulomb part.
How about for relativistic corrections?

SH, arXiv (2023)

Stratonovich-Hubbard transformation

Effective Lagrangian ($\alpha = 1,2$: transverse photons, $\alpha = 0$: "scalar photon")

$$\mathscr{L}_{\rm eff} = \sum_{\boldsymbol{k}} \sum_{\alpha=0}^{2} \hbar\omega_{\boldsymbol{k}} a^*_{\boldsymbol{k}\alpha} a_{\boldsymbol{k}\alpha}$$

$$- \sqrt{\frac{2\pi\hbar}{cV}} \int \mathrm{d}\boldsymbol{r} \sum_{\boldsymbol{k}} \sum_{\alpha=0}^{2} \frac{1}{\sqrt{k}} j_\alpha(\boldsymbol{r};\hat{\boldsymbol{k}}) (\mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}} a_{\boldsymbol{k}\alpha} + \mathrm{c.c.})$$

$$j_0(\boldsymbol{r};\hat{\boldsymbol{k}}) = \mathrm{i}\rho(\boldsymbol{r})$$

$$j_{1,2}(\boldsymbol{r};\hat{\boldsymbol{k}}) = \boldsymbol{j}(\boldsymbol{r}) \cdot \boldsymbol{\epsilon}_{1,2}(\hat{\boldsymbol{k}})$$

Spherical wave expansion of photons

*Berestetskii-Lifshitz-Pitaevski, textbook
*Igi-Kawai, textbook (Japanese)

$$a^{(\lambda)}_{JM}(k) = \sqrt{\frac{V}{(2\pi)^3}}\, k \sum_{\alpha=1}^{2} \int \mathrm{d}\hat{\boldsymbol{k}}\, \boldsymbol{Y}^{(\lambda)*}_{JM}(\hat{\boldsymbol{k}}) \cdot \boldsymbol{\epsilon}_\alpha(\hat{\boldsymbol{k}}) a_{\boldsymbol{k}\alpha}$$

($\lambda = 0,1$)

$$a^{(-1)}_{JM}(k) = \sqrt{\frac{V}{(2\pi)^3}}\, k \int \mathrm{d}\hat{\boldsymbol{k}}\, \boldsymbol{n} \cdot \boldsymbol{Y}^{(-1)*}_{JM}(\hat{\boldsymbol{k}}) a_{\boldsymbol{k}0}$$

(Vector spherical harmonics $\boldsymbol{Y}^{(1,0,-1)}_{JM}$)

$\lambda = 1$: electric photon: parity $(-1)^J$
$\lambda = 0$: magnetic photon : parity $(-1)^{J+1}$
$\lambda = -1$: scalar photon: parity $(-1)^J$

SH, arXiv (2023)

Multipole representation

Angular momentum tensor (for electrons with fixed n, l)

$$T_{LS}^{JM} = \sum_{mm'} \sum_{\sigma\sigma'} \left(O_{LS}^{JM}\right)_{m\sigma,m'\sigma'} c_{m\sigma}^{\dagger} c_{m'\sigma'}$$

Interaction in spherical basis

$$\mathscr{L}_{\mathrm{int}} = -\sum_{\lambda=-1}^{1} \sum_{J=0}^{\infty} \sum_{M=-J}^{J} \sum_{LS} \int_0^{\infty} \mathrm{d}k \sqrt{\hbar c k} \times \left[g_{JLS}^{(\lambda)}(k) a_{JM}^{(\lambda)}(k) T_{LS}^{JM} + s_\lambda (\mathrm{c.c.}) \right]$$

J=even; SI=+; TR=+: Electric
J=odd; SI=+; TR=-: Magnetic
J=odd; SI=+; TR=+: Electric Toroidal
J=even; SI=+; TR=-: Magnetic Toroidal

magnetic photon

electric photon

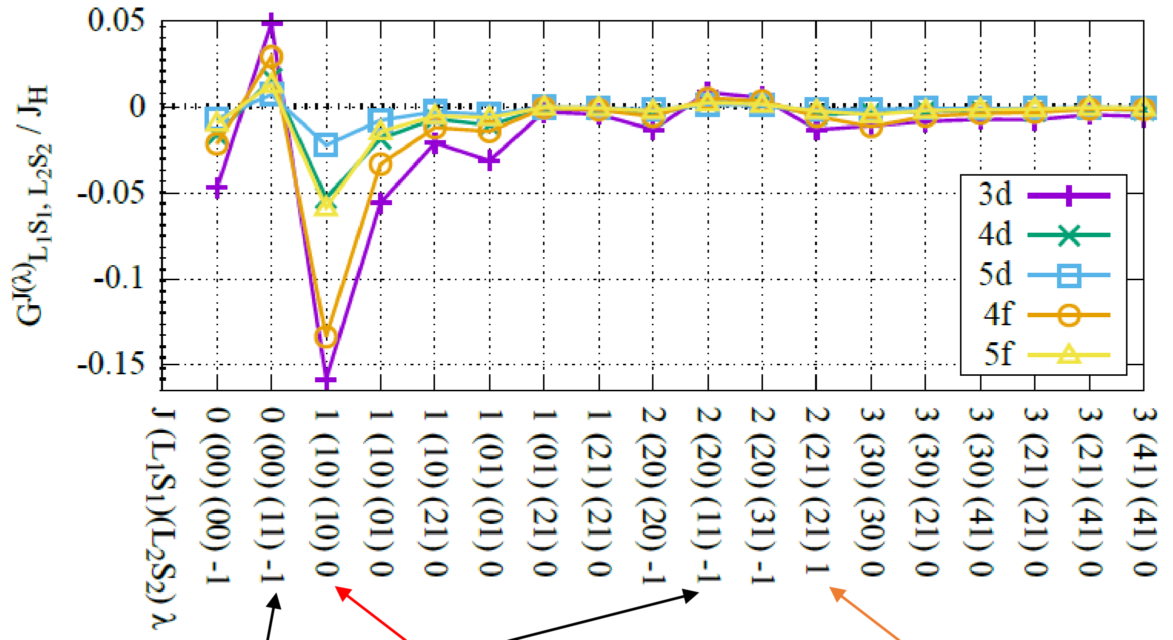
[Figs from N Papasimakis et al., Nat. Mater. (2016)]

By parity, magnetic photons couple to magnetic multipoles, and electric photons couple to magnetic toroidal multipoles.

SH, arXiv (2023)

Generalized Slater-Condon parameters

$$\mathscr{L}_{\mathrm{eff,int}} = \sum_{\lambda=-1}^{1}\sum_{JM}\sum_{L_1S_1L_2S_2} I^{J\lambda}_{L_1S_1,L_2S_2}\, T^{JM}_{L_1S_1}\, T^{JM*}_{L_2S_2}$$



Elec. polarization in Coulomb

Mag. multipoles - Mag. photons

Mag. Toroidal multipoles - Elec. photons

(Elec. Toroidal multipoles appear in higher-order Coulomb)

Cf. No odd-rank multipoles in conventional Coulomb interaction even in solids (cRPA)
S Iimura, M Hirayama SH, PRB Lett (2021)

Rough estimation:

☞ Order of Breit itnerction
$H_B \sim (Z\alpha)^2\, H_C$

☞ Hund's coupling for uranium: $J_H \sim 0.5$ eV

☞ Effective Nuclear charge for *f* electrons: $Z \sim 30$

$\therefore H_B \sim 0.1 \times J_H (Z\alpha)^2 \sim$ 100 K

Cf. Heavy-electron state: effective band width becomes 10^2-10^3 times smaller than usual metal.

Summary

SH, arXiv (2023)

Relativistic corrections to interactions in condensed matter

☞ Possible relevance of Coulomb-Breit interactions in SCES.

☞ Atomic representation, i.e. generalized Slater-Condon parameters, is derived, which can be combined with Hubbard model, Anderson lattice, LDA+*U*, LDA+DMFT, etc.

☞ Foundation for further exploring low-energy physics (ex. Amperean pairing).