DFT+DMFT Derivation of Multipolar Susceptibilities and Interactions in $4f^1$- and $4f^2$-Electron Materials

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Some rare-earth compounds exhibit a variety of phase transitions arising from the localized nature and diverse internal degrees of freedom of the $4f$ electrons. The corresponding order parameters can be described in terms of multipoles, which provide a unified representation of charge, magnetic, and orbital degrees of freedom. Multipolar ordering spontaneously breaks the symmetry of the electronic state and thereby gives rise to characteristic responses. Therefore, identifying the multipolar order parameter is essential for understanding phase transitions and responses in strongly correlated electron systems.

Density functional theory with dynamical mean-field theory (DFT+DMFT) provides first-principles descriptions of strongly correlated materials [1,2]. Based on DFT+DMFT, we address multipolar fluctuations as follows. First, we calculate the electronic structure using DFT and construct a low-energy tight-binding model that reproduces the DFT bands. Next, a DMFT treatment yields the single-particle Green's function incorporating local correlations among the $4f$ electrons. Using this Green's function, we obtain the momentum-dependent susceptibility $\chi(\vec{q})$ and effective interaction $I(\vec{q})$ by solving the Bethe–Salpeter equation in the strong-coupling limit [3]. Projecting the resulting $\chi(\vec{q})$ and $I(\vec{q})$ onto a symmetry-adapted multipole basis [4] labeled by γ yields the multipolar susceptibilities $\chi_\gamma(\vec{q})$ and interactions $I_\gamma(\vec{q})$, respectively. By examining the tendency of $\chi_\gamma(\vec{q})$ to diverge, we can predict the order parameter. Indeed, we have successfully reproduced multipolar orders in several Ce compounds with localized $4f$ electrons [5-7]. For general $4f^n$-electron systems with $n \geq 2$, a susceptibility represented in a many-particle basis *M* is required. Recently, we have extended this method to $4f^n$-electron systems by introducing a $4f^1$-to-$4f^n$ mapping that transforms the conventional susceptibility $\chi_{m_1 m_2, m_3 m_4}$ expressed in a single-particle basis *m* into the many-particle susceptibility $\mathcal{X}_{M_1 M_2, M_3 M_4}$ [8]. Evaluating the extended susceptibility $\mathcal{X}_\gamma(\vec{q})$ thus enables us to discuss possible order parameters not only in Ce compounds but also in other rare-earth compounds, such as Pr compounds.

In this presentation, we focus on applications of this method to $CeRh_2As_2$ and $PrCdNi_4$, two materials exhibiting multipolar ordering. For $CeRh_2As_2$, our susceptibility analysis identifies checkerboard antiferromagnetic M_z order at $\vec{q} = (\pi, \pi, 0)$ as the leading order-parameter candidate [7]. We further show that the anisotropy of the ordered phase arises from quadrupolar fluctuations induced by an in-plane magnetic field. For $PrCdNi_4$, we propose a previously unreported $Q_{3z^2-r^2}$-type antiferroquadrupolar order at $\vec{q} = (2\pi, \pi, 0)$ [8]. This order originates from the competition between nearest- and fourth-nearest-neighbor quadrupolar interactions.

[1] A. Georges *et al.*, Rev. Mod. Phys. **68**, 13 (1996).
[2] G. Kotliar *et al.,* Rev. Mod. Phys. **78**, 865 (2006).
[3] J. Otsuki, K. Yoshimi, H. Shinaoka, and Y. Nomura, Phys. Rev. B **99**, 165134 (2019).
[4] H. Kusunose, R. Oiwa, and S. Hayami, Phys. Rev. B **107**, 195118 (2023).
[5] J. Otsuki, K. Yoshimi, H. Shinaoka, and H. O. Jeschke, Phys. Rev. B **110**, 035104 (2024).
[6] S. Itokazu, A. Kirikoshi, H. O. Jeschke, and J. Otsuki, Commun. Mater. **6**, 269 (2025).
[7] K. Numa, E. Matsuda, A. Kirikoshi, and J. Otsuki, Phys. Rev. B **113**, 115141 (2026).
[8] K. Numa and J. Otsuki, in preparation.