Multipole ordering in $f$-electron systems

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Abstract

In order to investigate multipole ordering in $f$-electron systems from a microscopic viewpoint, we study the so-called $\Gamma_8$ models on three kinds of lattices, simple cubic (sc), bcc, and fcc, based on a $j-j$ coupling scheme with $f$-electron hopping integrals through $(ff\sigma)$ bonding. From the $\Gamma_8$ model, we derive an effective model for each lattice structure by using the second-order perturbation theory with respect to $(ff\sigma)$. By further applying mean-field theory to the effective model, we find a $\Gamma_{3u}$ antiferro-quadrupole transition for the sc lattice, a $\Gamma_{3u}$ antiferro-octupole transition for the bcc lattice, and a longitudinal triple-$q$ $\Gamma_{3u}$ octupole transition for the fcc lattice.

Key words: multipole ordering, $j-j$ coupling scheme, $\Gamma_8$ crystalline electric field ground state

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In recent decades, various kinds of magnetic and orbital ordering have been found in $f$-electron systems. In particular, it has been recognized that cubic systems with $\Gamma_8$ crystalline electric field ground states frequently exhibit higher-order multipole ordering due to their high symmetry. Indeed, octupole ordering has been proposed to reconcile experimental observations for Ce$_3$La$_{1-x}$B$_6$ [1,2,3,4,5] and NpO$_2$ [6,7,8,9]. To understand the origin of such multipole ordering from a unified viewpoint, it is important to analyze a simple microscopic model with correct $f$-electron symmetry.

In this paper, we study the so-called $\Gamma_8$ models on three kinds of lattices, simple cubic (sc), bcc, and fcc, based on a $j-j$ coupling scheme. For the description of the model, we define annihilation operators in the second-quantized form for $\Gamma_8$ electrons with $\alpha$ and $\beta$ orbitals as $f_{\alpha\uparrow} = \sqrt{5/6}a_{\alpha\uparrow} + \sqrt{1/6}a_{\alpha\downarrow}$, $f_{\beta\uparrow} = \sqrt{5/6}a_{\beta\uparrow} + \sqrt{1/6}a_{\beta\downarrow}$, where $a_{\alpha\beta}$ is the annihilation operator for an electron with the $z$-component $j_z$ of the total angular momentum $j=5/2$ at site $r$.

In the tight-binding approximation, the model Hamiltonian is given by [10]

$$H = \sum_{r,\mu,\tau,\sigma,\tau',\sigma'} t^{\mu}_{\tau\sigma,\tau',\sigma'} f_{\tau\sigma}^{\dagger} f_{\tau'\sigma'} + U \sum_{r,\tau} n_{\tau\uparrow} n_{\tau\downarrow} + U' \sum_{r,\tau} n_{\tau\alpha} n_{\tau\beta} + J \sum_{r,\sigma,\sigma'} f^{\dagger}_{\tau\sigma} f_{\tau'\sigma'} f^{\dagger}_{\tau'\sigma'} f_{\tau\sigma} + J' \sum_{r,\tau,\sigma,\tau',\sigma'} f^{\dagger}_{\tau\sigma} f^{\dagger}_{\tau'\sigma'} f_{\tau'\sigma'} f_{\tau\sigma},$$

where $\mu$ is a vector connecting nearest-neighbor sites, $t^{\mu}_{\tau\sigma,\tau',\sigma'}$ is the hopping integral of an electron with $(\tau',\sigma')$ at site $r+\mu$ to the $(\tau,\sigma)$ state at $r$ through $(ff\sigma)$ bonding [11], $n_{\tau\sigma} = f^{\dagger}_{\tau\sigma} f_{\tau\sigma}$, and $n_{\tau\sigma} = \sum_{\sigma} n_{\tau\sigma}$. The coupling constants $U$, $U'$, $J$, and $J'$ denote the intra-orbital, inter-orbital, exchange, and pair-hopping interactions, respectively. Note that the form of $t^{\mu}_{\tau\sigma,\tau',\sigma'}$ characterizes the lattice structure.

By using the second-order perturbation theory with respect to $(ff\sigma)$ including only the lowest energy $\Gamma_8$ triplet among the intermediate states, we obtain effective multipole interactions for each lattice structure. The detail of the effective models will be reported elsewhere, and here we report the ordered state obtained in the mean-field theory.

For sc lattice, a $\Gamma_{3u}$ antiferro quadrupole (AFQ) transition occurs at a finite temperature, and as lowering temperature further, we find another transition to
Fig. 1. Ordered states in the sc lattice under a magnetic field along [001]. (a) The $\Gamma_{3g}$ AFQ state. (b) The FM state with the $\Gamma_{3g}$ AFQ moment.

Fig. 2. Ordered states in the bcc lattice under a magnetic field along [001]. (a) The $\Gamma_{2u}$ AFO state. (b) The FM state with the $\Gamma_{2u}$ AFO moment.

Fig. 3. The triple-$q$ $\Gamma_{5u}$ octupole state in the fcc lattice.

In summary, we have derived the multipole interaction model from the microscopic $\Gamma_8$ Hamiltonian. By analyzing the effective model, we find a $\Gamma_{3g}$ AFQ state for the sc lattice, a $\Gamma_{2u}$ AFO state for the bcc lattice, and the longitudinal triple-$q$ $\Gamma_{5u}$ octupole state for the fcc lattice.

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