Effect of Crystalline Field Splitting on Orbital Ordering in $KCuF_3$ and its Manifestation in Resonant X-ray Scattering Spectra

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Pseudocubic pervoskite $KCuF_3$ has attracted extensive theoretical interests since 1970s because of its low-dimensional magnetism, cooperative Jahn-Teller effect and orbital ordering[1]. It is recognized that the antiferro-orbital order, i.e. alternative x^2-y^2 and $3z^2-r^2$ orbital occupations in different Cu sites, which arises from the superexchange coupling of Cu 3d electrons and the Jahn-Teller effect, stabilizes the unusual magnetic structure. Recently Khomskii and Mostovoy[2] pointed out that both the pure electronic superexchange coupling and the Jahn-Teller phonon-mediated orbital coupling lead to strong orbital frustrations. The conventional mean-field solutions underestimated the effect of quantum spin-orbital fluctuations on the ground state of $KCuF_3$ and neglected the role of the crystalline field splitting in suppressing the orbital frustration.

In this work, utilizing the Cluster Self-Consistent Field approach developed recently, we study the role of the crystalline field splitting on the realistic ground state of $KCuF_3$ and its manifestation in the resonant X-ray scattering intensity. We first showed that due to the frustration, the system with the pure electronic superexchange coupling and the Jahn-Teller phonon-mediated orbital coupling is an orbital liquid phase. Then we demonstrated that the crystalline field splitting induced by the Jahn-Teller distortion stabilizes the orbital order observed in recent resonant x-ray scattering experiments[3,4] and the A-type antiferromagnetic order. Considerably reduced magnetic moment of Cu, about $0.55\mu_B$, is in agreement with the experimental value $0.49\mu_B$. In contrast, the theoretical result by the first-principles electronic structure calculations predicted about $0.91\mu_B$[5]. And in the orbital ordered ground state, the orbital occupation is about 42% in the x^2-y^2 orbit and 58% in $3z^2-r^2$ orbit. Such orbital configuration leads to strong anisotropic spin correlations, the ratio of magnetic coupling strengths along the c-axis and in the ab-plane, J_c/J_{ab}, is about 26. The azimuthal dependence of resonant x-ray scattering intensity calculated for the $1s-3d$ excitation event with relevant orbital ordering provides the clue for further resonant x-ray scattering experiment.

These theoretical results consistently explain most of experimental data in $KCuF_3$, suggesting that the crystalline field splitting plays a crucial role in stabilizing the orbital ordered ground state in $KCuF_3$.

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