Anharmonic effect on lattice distortion, orbital ordering and magnetic properties in Cs$_2$AgF$_4$

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Abstract
We develop the cluster self-consistent field method incorporating both electronic and lattice degrees of freedom to study the origin of ferromagnetism in Cs$_2$AgF$_4$. After self-consistently determining the harmonic and anharmonic Jahn–Teller distortions, we show that the anharmonic distortion stabilizes the staggered $x^2-z^2/y^2-z^2$ orbital and ferromagnetic ground state, rather than the antiferromagnetic one. The amplitudes of lattice distortions, $Q_2$ and $Q_3$, the magnetic coupling strengths, $J_x, J_y$, and the magnetic moment are in good agreement with the experimental observations.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Recently, the layered perovskite compound Cs$_2$AgF$_4$ containing the spin-1/2 4$d^9$ Ag(II) ion has attracted great interest, because it is isomorphic to the high-$T_c$ 3$d^9$ Cu(II) cuprates. Cs$_2$AgF$_4$ was first refined to the tetragonal structure with the space group $I4/mmm$ in 1974 by Odenthal et al [1]. Very recently, however, McLain et al [2] found that the crystal structure is orthorhombic, with the space group $Bbcm$. The magnetic susceptibility and the inelastic neutron scattering experiments showed that this compound is well described as a two-dimensional (2D) ferromagnet (FM) with $T_c \sim 15$ K [2]. Further, McLain et al suggested that Cs$_2$AgF$_4$ is orbitally ordered at all temperatures of measurement, and the orbital ordering (OO) is responsible for the FM. These properties are in sharp contrast to the high-$T_c$ parent La$_2$CuO$_4$ that is an antiferromagnetic (AFM) insulator. The microscopic origin of the unusual FM and OO in Cs$_2$AgF$_4$ attracts a lot of attention.

In Cs$_2$AgF$_4$, the basal plane consists of a 2D lattice of Jahn–Teller (JT) distorted AgF$_6$ octahedra with a pattern of alternating short and long Ag–F bonds. These analogous compounds with orbital degeneracy, such as Cs$_2$AgF$_4$ and K$_2$CuF$_4$, turn out to be 2D FM [2], while other compounds, such as K$_2$NiF$_4$ and Rb$_2$MnF$_4$, are AFM [3, 4]. Notice that the former with active JT ions leads to an orthorhombic structure, while the latter with nondegenerate orbitals only has a tetragonal structure. Although the difference between these two structures seems small, i.e. the main change is the position of the fluorine atoms in the basal plane, such a difference is of the utmost importance in leading to the distinct properties [5]. From early studies [6–9] in perovskite compounds, it is known that the anharmonic JT effect is a decisive factor for the orthorhombic crystal structure. To date, a lot of studies have been done for Cs$_2$AgF$_4$ utilizing the density functional theory [10–15]; however, few authors focus on the anharmonic JT effect on the lattice, OO and FM ground-state properties in Cs$_2$AgF$_4$.

Previous studies in the analogous compound K$_2$CuF$_4$ have provided us with two distinct scenarios for the FM and OO ground state. On the one hand, Kugel and Khomskii showed that the cooperative JT effect, especially the anharmonic JT effect [16], plays a key role in stabilizing the OO ground state in K$_2$CuF$_4$. On the other hand, to resolve the difficulty of the Kugel–Khomskii (KK) electronic superexchange (SE) coupling model [17], which is usually suitable for the Mott–
Hubbard insulators with $U_\parallel < \Delta$ (here $U_\parallel$ is the on-site Coulomb repulsion and $\Delta$ is the charge-transfer energy), and to address the FM and OO state, Mostovoy and Khomskii [18] proposed a modified SE coupling model, which is suitable for the charge transfer insulating K$_2$CuF$_4$ with $U_\parallel > \Delta$. Obviously, to understand the unusual ground-state properties, such as the lattice, orbital and magnetic properties in K$_2$CuF$_4$ and Cs$_2$AgF$_4$, one should incorporate the cooperative JT effect [19] and the anharmonic JT effect into the charge-transfer SE interactions.

To treat these strongly correlated systems more precisely, based on the cluster self-consistent field approach developed previously [20], we explicitly take into account the orthorhombic JT distortions and the charge-transfer SE interactions, in which spin order, OO and lattice distortion are determined self-consistently. We show that, by a strong anharmonic effect and Hund’s coupling, Cs$_2$AgF$_4$ has a much more stable ferromagnetic ground state. The theoretical amplitudes of lattice distortions, $Q_2$ and $Q_3$, the magnetic coupling strengths, $J_1$, and, and the magnetic moment are in good agreement with the experimental observation in Cs$_2$AgF$_4$.

This paper is organized as follows: an effective Hamiltonian and the cluster self-consistent field (SCF) approach are described in section 2; then the results and discussions of the lattice structure and distortions, orbital ordering and magnetic properties in Cs$_2$AgF$_4$ are presented in section 3; section 4 is devoted to remarks and the summary.

2. Model Hamiltonian and method

An effective 2D Hamiltonian in Cs$_2$AgF$_4$ including both the SE and the JT couplings is written

$$H = H_{QJT} + H_{SE} \quad (1)$$

where $H_{SE}$ describes the effective SE couplings; $H_{QJT}$ describes the JT couplings associated with the electron and the lattice distortion. Note that spin, orbital and lattice degrees of freedom couple to each other in Hamiltonian (1). The JT effect associated with one hole per Ag$^{2+}$ site in the AgF$_4$ sheets reads [19]

$$H_{QJT} = g \sum_i (Q_{i2} \tau_i^x + Q_{i3} \tau_i^z) + \frac{K}{2} \sum_i (Q_{i2}^2 + Q_{i3}^2) + G \sum_i \left[ (Q_{i3}^2 - Q_{i3}^2) \tau_i^z - 2Q_{i2}Q_{i3} \tau_i^x \right] \quad (2)$$

where both the linear and the quadratic vibronic coupling terms have been included. The first and second terms describe the linear harmonic JT effect. The third one, i.e., the quadratic coupling, arises from the anharmonic JT effect and contributes to the anisotropic energy [16, 19]. Here $Q_{i2}$ and $Q_{i3}$ are the normal vibration coordinates, defined as $Q_{i2} = (-X_i + X_2 + Y_i - Y_4)/2$ and $Q_{i3} = (-X_i + X_2 - Y_i + Y_4 + 2Z_i - 2Z_4)/\sqrt{2T}$ [21], with $X, Y$ and $Z$ being the coordinates of the $i$th F ions. $g$ is the linear JT coupling strength, $G$ is the coefficient of quadratic coupling, and $K$ is the elastic constant. For Cs$_2$AgF$_4$, we fix $K = 10$ eV Å$^{-2}$ throughout this paper.

Since Cs$_2$AgF$_4$ is a strongly correlated charge-transfer insulator [11], the 2D SE interactions between 4d orbitals of Ag$^{2+}$ ions through 2p orbitals of F atoms are described [18]:

$$H_{SE} = \sum_{i,a,x,y} \left[ \left( J_1 + J_2 I_1^a + J_3 I_1^a I_3^a \right) \vec{S}_i \cdot \vec{S}_i \right] + J_4 I_3^a + J_5 I_3^a I_3^a, \quad (3)$$

where the operator $\vec{S}_i$ denotes the $S = 1/2$ spin at site $i$, and $I_3^a = \cos(2\pi m_a/3)\tau_i^z - \sin(2\pi m_a/3)\tau_i^x$, which is a combination of the components of the orbital operators $\tau$. $a = x, y$, and $(m_x, m_y) = (1, 2)$, denote the direction of a bond in the AgF$_4$ sheets. In equation (3), the coupling coefficients $J_n (n = 1–5)$ read

$$\begin{align*}
J_1 & = \tau^2 \left[ 1/4U_\parallel + 2/4(2\Delta + U_\parallel) - J_{SE}/(2U_\parallel^2) \right], \\
J_2 & = 4\tau^2 \left[ 1/8U_\parallel + 2/(2\Delta + U_\parallel) \right], \\
J_3 & = 4\tau^2 \left[ 1/4U_\parallel + 2/(2\Delta + U_\parallel) + J_{SE}/(2U_\parallel^2) \right], \\
J_4 & = \tau^2 \left[ 2/\Delta - 1/4U_\parallel - 2/(2\Delta + U_\parallel) \right], \\
J_5 & = \tau^2 \left[ 1/4U_\parallel + 2/4(2\Delta + U_\parallel) + 3J_{SE}/(2U_\parallel^2) \right]
\end{align*}$$

with $t = \sqrt{\tau^2/\Delta}$, respectively. Here $\Delta$ is the electron-transfer energy between silver 4d and fluorine 2p orbitals, $U_\parallel$ and $U_\parallel$ are the Coulomb repulsion energies on Ag and F, respectively, and $J_{SE}$ is the Hund’s coupling on Ag. Note that the coefficients may be positive and negative, thus the AFM and the FM components coexist in $J_n$. Since the 4d orbitals of Ag$^{2+}$ ions are less localized than the 3d orbitals of Cu$^{2+}$ ions in K$_2$CuF$_4$, the Coulomb repulsion $U_\parallel$ of the Ag$^{2+}$ ion is about 3–5 eV, and $J_{SE}$ ranges from 0.1 to 0.5 eV, in accordance with the LDA+$U$ results [10, 11, 15]. In general, the Coulomb repulsion $U_\parallel$ is about 5 eV in F$. The charge-transfer energy $\Delta$ is roughly estimated from the difference of the centers of gravity between the 4d and 2p levels in band structures, which also allows us to estimate $t_{pd}$ from the bandwidth of the electronic structures available [4, 10, 15], $\Delta = U_\parallel - 2$ eV, and $t_{pd} \approx 0.6$ eV.

In such a strongly correlated spin–orbital–lattice system, according to the Feynman–Hellman theorem, the ground state energy is minimized with respect to $Q_{i2}$ and $Q_{i3}$, i.e.,

$$\left. \frac{\partial H}{\partial Q_{i2}} \right|_{Q_{i3}} = 0, \quad \left. \frac{\partial H}{\partial Q_{i3}} \right|_{Q_{i2}} = 0.$$

From this one could find that the strength of the normal modes and the lattice distortion critically depend on the orbital polarization through the following equations:

$$\langle Q_{i2} \rangle = g \frac{K(\tau_i^x)^2 + 4G((\tau_i^z)^2)}{4G^2((\tau_i^x)^2 + (\tau_i^z)^2) - K^2}, \quad \langle Q_{i3} \rangle = g \frac{K(\tau_i^z)^2 + 2G((\tau_i^x)^2 - (\tau_i^z)^2)}{4G^2((\tau_i^x)^2 + (\tau_i^z)^2) - K^2} \quad (4)$$

Notice that, in the absence of the anharmonic JT effect ($G = 0$), $\langle Q_{i2} \rangle \sim -(g/K)(\tau_i^x)$ and $\langle Q_{i3} \rangle \sim$
consisting of four Ag$_2$ ions. The exact treatment of the interactions inside the cluster, and the orbital Hamiltonian (1). The cluster-SCF approach includes the self-consistent field treatment of the interactions between the cluster and the surrounding environment. The main procedure is briefly outlined as follows. First, we choose a cluster consisting of four Ag$_2$ ions. The $i$th site is surrounded by two Ag sites inside the cluster and two Ag sites outside the cluster, which provide two internal interactions and two external SCF fields, respectively. Thus, the effective Hamiltonian of the cluster reads

$$
h_{\text{cluster}} = \sum_{\alpha=a,\gamma} \left[ \left( J_1 + J_2 \right) t_{ij}^\alpha \right] \tilde{S}_i \cdot \tilde{S}_{i+\alpha} + J_4 t_{ij}^{\alpha \gamma} + g \sum_i (Q_{13}^\alpha \tau^x_i + Q_{12}^\alpha \tau^y_i) + G \sum_i \left[ (Q_{31}^\alpha - Q_{32}^\alpha) \tau^z_i - 2 Q_{12} Q_{31}^\alpha \tau^x_i \right] + \frac{K}{2} \sum_i (Q_{12}^2 + Q_{33}^2) + \sum_i h_i^{\text{scf}} \tag{5}\nonumber$$

with the SCF $h_i^{\text{scf}}$ contributing from the $j$th external site interacting with the $i$th internal site through $H_{ij}$. $h_i^{\text{scf}} = \text{Tr}_j (\rho_j H_{ij})$, where $\rho_j$ denotes the reduced density matrix of the $j$th site, and $i$ runs over all sites inside the cluster. We first substitute the spin coupling $\tilde{S}_i \cdot \tilde{S}_{i+\alpha}$ into the cluster Hamiltonian with the initial spin correlation function $(\tilde{S}_i \cdot \tilde{S}_{i+\alpha})$, and diagonalize the orbital part of the cluster Hamiltonian (5) in the presence of the orbital SCF. The orbitalization $(\tilde{\tau})$ and the orbital correlation functions $(\tilde{\tau}_i \cdot \tilde{\tau}_{i+\alpha})$ are thus obtained. Then, we substitute the orbital operator and the orbital couplings with $(\tilde{\tau})$ and $(\tilde{\tau}_i \cdot \tilde{\tau}_{i+\alpha})$ into $h_{\text{cluster}}$ and diagonalize the spin part of the cluster Hamiltonian in the presence of the spin SCF. Hence, we obtain a set of new averaged spin $\tilde{S}_i$ and spin correlation functions $\tilde{S}_i \cdot \tilde{S}_{i+\alpha}$. We repeat the above steps until the ground-state energy and the spin and orbital correlation functions converge to the accuracies. From the stable spin–orbital ground state, one could get the magnetic coupling strengths $J_k$ and $J_l$ through $J_0 = J_1 + J_2 (t_{ij}^\alpha) + J_3 (t_{ij}^{\alpha \gamma})$; here $\alpha = x, y$. The advantage of our approach over the traditional mean-field method is that the short-range spin and orbital correlations as well as the quantum fluctuations are taken into account properly, especially in the low-dimensional systems.

3. Results and discussion

In this section, we present the numerical results of the ground state of the charge-transfer insulator Cs$_2$AgF$_4$ within the cluster-SCF approach. We mainly discuss the role of the anharmonic effect on lattice structure and distortions, orbital ordering and magnetic properties in the ground state.

Figure 1. Phase diagrams of JT coupling $g$ versus Hund’s coupling $J_H$ in Cs$_2$AgF$_4$ for different Coulomb repulsions, (a) $U_d = 3$ eV, (b) $U_d = 4$ eV and (c) $U_d = 5$ eV. The solid circles, dashed line, solid triangles and solid squares denote the FM/AFM phase boundaries for the anharmonic coupling $G = −0.75g$, 0, 0.25g and 0.75g, respectively. $G$ is in the unit of eV A$^{-2}$.

3.1. Ground-state phase diagram

To clarify the roles of the SE coupling and the JT effect in the origin of the FM, we first perform numerical calculations with equation (5) and obtain the $g$–$J_H$ phase diagram, as shown in figure 1. Since the on-site Coulomb interaction $U_d$ in Cs$_2$AgF$_4$ is not well defined, we present the $g$–$J_H$ phase diagram for $U_d = 3, 4$ and $5$ eV in figures 1(a), (b) and (c), respectively. Accordingly, we find that the anharmonic distortion strength $G$ plays a very important role in the phase diagram. With the variation of $G$ from negative to positive via zero, the FM–AFM phase boundaries exhibit critical changes. As we see in figure 1(a), at $G = −0.75g$, the stable ground-state phase in regions I–IV is AFM, while that in region V is FM; at $G = 0$, a vertical line separates the AFM ground state in regions I–III from the FM one in regions IV–V; when the anharmonic distortion strength $G$ becomes positive, more regions become FM ordering; at $G = 0.75g$, only the ground state in region I is AFM. Figures 1(b) and (c) qualitatively resemble figure 1(a).

The competitive FM and AFM couplings in the SE Hamiltonian result in the FM phase for large $J_H$ and the AFM phase for small $J_H$. As we expect in figures 1(a)–(c), strong Coulomb repulsion favors the AFM phase, hence the AFM regime becomes large with the increase of $U_d$. Besides the fact that strong Hund’s coupling $J_H$ favors the FM phase, the large anharmonic JT effect also favors the FM
ground state. Therefore, the complete consideration of the realistic interactions in equation (1) most probably leads to the FM ground state in Cs$_2$AgF$_4$. Moreover, from the recent first-principles electronic calculations, the Coulomb repulsion between Ag 4d electrons is about 3 eV [10]. We expect that the Hund’s coupling $J_{\text{H}}$ lies in the range 0.1–0.3 eV, so that the interaction parameters of Cs$_2$AgF$_4$ fall in the FM region in the phase diagram.

Experimentally, it is hard to determine the sign of the anharmonic coupling strength $G$ in Cs$_2$AgF$_4$. From figure 1, it is seen that when the positive anharmonic effect is very large the ground state stabilizes in the FM phase even in the absence of Hund’s coupling. Therefore, the positive anharmonic coupling most favors the FM ground state. In contrast, for $G < 0$, due to the SE coupling, the ground state is FM when the harmonic JT coupling strength $g$ is small; however, when $g$ becomes large, the anharmonic coupling favors the AFM ground state. Thus, considering the strong JT effect in Cs$_2$AgF$_4$, one expects that $G$ is most probably positive, which will be further confirmed in the lattice distortion, the orbital ordering and the magnetic properties in what follows.

### 3.2. Lattice structure and distortions

Different from La$_2$CuO$_4$, the unusual FM and the orthorhombic structure of Cs$_2$AgF$_4$ is the result of the interplays of the spin, orbital and lattice degrees of freedom, especially the contribution of the anharmonic JT effect. The normal coordinates $Q_2$ and $Q_3$ of the JT distortions in Cs$_2$AgF$_4$ are obtained self-consistently through equations (4) and (5). We find that for positive and large $G$ the structural distortion in the ground state is antiferro-type ($Q_2$, $Q_3$; $-Q_2$, $Q_3$), corresponding to the alternative short and long bonds in the AgF$_4$ plaquette. However, the ground-state energy of the ferro-type ($Q_2$, $Q_3$; $Q_2$, $Q_3$) distortion is higher than that of the antiferro-type distortion, as shown in figure 2, and the ferro-type distortion only appears in the strong JT region because of its instability in this system.

Notice that the experimentally observed lattice structure has the antiferro-type distortion with the long and short Ag–F bond lengths of 2.441 and 2.111 Å in the AgF$_4$ plaquette [2]; one can deduce the corresponding distortions $Q_2 \approx 0.33$ Å and $Q_3 \approx -0.19$ Å. Theoretically, through the self-consistent numerical calculations, we obtain the amplitudes of the $Q_2$ and the $Q_3$ distortions in Cs$_2$AgF$_4$ with the variation of the JT coupling for different anharmonic coupling parameters, as shown in figure 3. When the linear JT coupling strength $g$ is about 4.8 eV Å$^{-1}$ at $G = 0.75$ g, the theoretical distortions are $|Q_2| \approx 0.33$ Å and $Q_3 \approx -0.19$ Å (see the dashed vertical line in figure 3), in good agreement with the corresponding experimental observation in Cs$_2$AgF$_4$ [2]. Meanwhile, we find that a positive $G$ leads to a negative $Q_3$, giving rise to the correct distortion $c/a = 2/(l + s) < 1$ in orthorhombic Cs$_2$AgF$_4$. However, a negative $G$ gives a positive $Q_3$, resulting in a wrong distortion $c/a = 2/(l + s) > 1$. All the other analogous substances without JT ions, e.g. K$_2$NiF$_4$, K$_2$MnF$_4$ [3, 4], are tetragonal, rather than the orthorhombic symmetry in Cs$_2$AgF$_4$ with JT ions. In fact, it is the positive anharmonic effect that lowers the lattice symmetry, leading to the compression of the ligand octahedron along the c-axis [22, 23].

### 3.3. Orbital ordering

The unusual FM and distinct orthorhombic structure in Cs$_2$AgF$_4$ are in fact associated with the formation of the long-range OO, as shown by a few authors recently utilizing first-principles calculations [10, 11, 13, 15]. Our study also confirms this point. Furthermore, we demonstrate that the anharmonic effect plays an essential role in the OO properties of Cs$_2$AgF$_4$. In general, one can describe the orbital occupied

![Figure 2](image-url)  
**Figure 2.** Dependence of ground-state energy on the JT coupling strength with $G = 0.75$ g in the antiferro-type (a) and the ferro-type (b) distortions in the AgF$_4$ plaquette. The dashed vertical line indicates the appearance of the ferro-type distortion. Theoretical parameters: $U = 3$ eV and $J_{\text{H}} = 0.3$ eV.

![Figure 3](image-url)  
**Figure 3.** The amplitudes of $|Q_2|$ and $Q_3$ as a function of the JT coupling with different $G$. Two dashed horizontal lines correspond to the experimental values of $|Q_2|$ and $Q_3$; the vertical dashed line indicates the linear JT coupling $g = 4.8$ eV Å$^{-1}$ for $G = 0.75$ g in Cs$_2$AgF$_4$. The other parameters are the same as figure 2.
state of each site in terms of \( |\phi\rangle = \cos \frac{\phi}{2} |3z^2 - r^2\rangle \pm \sin \frac{\phi}{2} |x^2 - y^2\rangle \) with the orbital angle \( \theta \). Here ‘\pm’ refers to the two sublattices of the antiferro-distortion in the AgF\(_4\) plaquette. Figure 4 shows the orbital angle \( \theta \) as a function of the linear JT coupling strength \( g \) with different anharmonic couplings \( G \). The role of the anharmonic coupling in the orbital angle, hence the OO, is clearly seen. For strong JT distortion and \( G = 0.75g \), the OO is staggered \( z^2 - x^2/z^2 - y^2 \); see inset (b) in figure 4. The orbital angle obtained is about 61.8° for \( g = 4.8 \text{ eV Å}^{-1} \), as indicated by the dashed vertical line in figure 4, which is consistent with the experimental suggestion for Cs\(_2\)AgF\(_4\) [2]. This is different from the case in La\(_2\)CuO\(_4\), where the distance between apical O and Cu is much larger than that in the \( a-b \) plane, resulting in the pure \( 3z^2 - r^2 \) hole orbitals. Notice that the orbital angle is also close to 60° in the ferro-type solution. However, it is the ferro-oralbital \( z^2 - x^2/z^2 - x^2 \) or \( z^2 - y^2/z^2 - y^2 \) symmetry rather than the staggered antiferro-orbital \( z^2 - x^2/z^2 - y^2 \) symmetry in the antiferro-type solution.

As a comparison, for \( G < 0 \), the orbital angle \( \theta > \pi/2 \); as \(|G|\) becomes very large, \( \theta \to 2\pi/3 \), and the holes occupy the \( d_{xy}, d_{xz}, d_{yz} \) orbitals in an orderly way, as seen in inset (a) in figure 4. For \( G = 0 \), however, the orbital angle is nearly constant with the increase of the linear JT coupling \( g \), giving rise to \( \theta = 68° \). Note that in the absence of the anharmonic effect the SE couplings contribute to the same OO as the harmonic JT effect does. In this situation, the orbital configuration is characterized by orbital angle \( \theta_0 \): \( \tan \theta_0 = Q_{y3}/Q_{b3} \) [19]. Then \( Q_{y3}/Q_{b3} \sim r^4/r^6 \), which is almost independent of the linear coupling \( g \), indicating that the linear JT coupling has little influence on the orbital angle in the absence of the anharmonic effect. This also shows the key role of the anharmonic effect in the OO from the other aspect. In addition, we find that the orbital angle is nearly unchanged with the Hund’s coupling \( J_H \). This result can be easily understood, since the orbital angle is mainly determined by the ‘orbital field’ corresponding to the \( J_d \) term in \( H_{SE} \), which is independent of \( J_H \) [18].

3.4. Magnetic properties

The magnetic properties are also strongly influenced by the anharmonic effect, as shown in figure 5. In the present situation, we find that the difference between \( J_x \) and \( J_y \) is negligible. From figure 5, one finds that in the absence of the anharmonic effect \( (G = 0) \), due to the SE coupling, the magnetic couplings between Ag spins are FM, and nearly remain unchanged with the increasing of \( g \), while for \( G < 0 \), due to the SE coupling, the magnetic coupling between Ag ions is FM at small \( g \). With the increase of the JT effect, the magnetic couplings between Ag ions, \( J_{x,y} \), vary from FM to AFM, which is in contradiction with Cs\(_2\)AgF\(_4\). For \( g = 4.8 \text{ eV Å}^{-1} \) and \( G = 0.75g \), as denoted by the dashed vertical line in figure 5, the magnetic couplings in the AgF\(_4\) plaquette, i.e. \( J_{x,y} \), are about \(-3.3\) meV, very close to the experimental values, \(-3.793 \) to \(-5.0\) meV [2]. The agreement between the present magnetic coupling strengths and the experimental observation is much better than those inferred from the first-principles electronic structure calculations [13, 14].

This result, combining those of the lattice distortion and the orbital angle, is in accordance with the Goodenough–Kanamori–Anderson rules [24]. Also in figure 5, one notices that in the presence of strong anharmonic coupling \( G \) the FM coupling strength increases with the linear JT coupling \( g \). Therefore, the FM couplings in Cs\(_2\)AgF\(_4\) further verify the key role of the anharmonic effect in the ground-state properties. In addition, the calculated magnetic moment is about 1 \( \mu_B \), which approaches the classical value and is comparable to the experimental data observed in a muon-spin relaxation experiment [25]. This is a classical value of the Ag ion,
is that the electron or hole doping in La$_2$CuO$_4$ leads to strong
of Cs$_2$AgF$_4$ is unusual, FM insulating with the orthorhombic
integrals, the other hand, once the JT distortion occurs, the hopping
covalency effect [12]. One consequence of such a difference
the possibility of the FM ground state originating from the
covalency effect [12]. One consequence of such a difference
is that the electron or hole doping in La$_2$CuO$_4$ leads to strong
AFM fluctuations, which may contribute to the Cooper-pairing
for the high-T$_c$ superconductivity. We anticipate that the
electron doping in Cs$_2$AgF$_4$ will lead to weak FM fluctuation,
though it will not contribute to the same SC mechanism as the
cuprate superconductors.

Notice that in the present intermediate correlated and
charge-transfer insulator the 2D effective SE interactions
incorporating the JT couplings underestimate the covalency
effect between Ag and F 4d and F 2p orbitals. The covalency effect
between Ag and F ions can be well considered within the first-
principles electronic structure calculations [10, 12, 14, 15]. On
the other hand, once the JT distortion occurs, the hopping
integrals, $t_{xy}$, between Ag ions are slightly different in
different crystallographic axes. We expect that this change will
not significantly modify our results quantitatively.

In summary, an effective model Hamiltonian with spin,
orbital and lattice degrees of freedom coupling to each other
allows us to self-consistently describe the lattice structure
change, orbital ordering and magnetic coupling properties, etc.
Driven by the JT distortion, especially by the anharmonic
effect, Cs$_2$AgF$_4$ stabilizes in the 2D FM ground state. The
correct lattice structure, orbital ordering and magnetic coupling
observed experimentally can also be addressed in the present
theoretical framework.

4. Remarks and summary

As we have shown in the preceding sections, the anharmonic
JT effect incorporating the SE coupling drives the 4d orbitals
of Cs$_2$AgF$_4$ into the d$_{2-x^2}$/$d_{z^2-x^2}$ ordering, rather than the
d$_{3z^2-r^2}$ orbital ordering in La$_2$CuO$_4$. Consistent with the
Goodenough–Kanamori–Anderson rules [24], the ground state
of Cs$_2$AgF$_4$ is unusual, FM insulating with the orthorhombic
structure, different from the Néel AFM rules insulating ground state
with the tetrahedral structure in La$_2$CuO$_4$. Our results preclude
the possibility of the FM ground state originating from the
covalency effect [12]. One consequence of such a difference
is that the electron or hole doping in La$_2$CuO$_4$ leads to strong
AFM fluctuations, which may contribute to the Cooper-pairing
glue for the high-$T_c$ superconductivity. We anticipate that the
electron doping in Cs$_2$AgF$_4$ will lead to weak FM fluctuation,
though it will not contribute to the same SC mechanism as the
cuprate superconductors.

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