Half-metallic antiferromagnetism in LaOCr$_{0.5}$Fe$_{0.5}$As as a probe of single spin superconductivity

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Abstract. The normal ground state of LaOFeAs is an antiferromagnet (AFM) with magnetically stripe-ordered FeAs planes. By using a full-potential augmented plane wave method within density functional theory, we find LaOCr$_{0.5}$Fe$_{0.5}$As to be a quasi-2D half-metallic (HM) AFM, where two component magnetic ions in a unit cell have antialigned local moments that cancel exactly because of the integer filling of an insulating channel. Both striped and checkerboard structures in CrFeAs$_2$ planes show stable HM-AFM behaviors in the ground state.

1. Introduction

Half metals (HMs) [1, 2] have a collinear magnetic arrangement with two different types of spin-dependent band structures: One spin channel is metallic while the other is insulating. Since the electrons responsible for the metallic behavior share the same spin, the conduction electrons are fully spin polarized in principle. The Pauli spin susceptibility vanishes because there are no low energy spin-flip transitions available to the system. As a result of having a band gap at the Fermi energy ($E_F$) in the insulating channel, the spin magnetic moment per unit cell is quantized in units of Bohr magneton ($\mu_B$). Half-metallic antiferromagnets (HM-AFMs) [3], or rather fully compensated half-metallic ferrimagnets, are an unusual case in which the net moment per unit cell corresponds to the integer zero; the material possesses no macroscopic magnetization, yet its conduction electrons are fully spin polarized. Conventional antiferromagnets (AFMs), however, cannot fulfill the requirements of being half-metallic, because conduction electrons in each spin channel must be equivalent by symmetry.

Zinc-blende (ZB) transition-metal pnictides and chalcogenides form a class of HMs with tetrahedral coordination [4]. The origin of this behavior is the $p$-$d$ hybridization due to the symmetry of crystal fields. So far the theoretical attempt to find this class of HM-AFMs has been quite successful [5]. Besides conceivable applications to an ideal tip in a spin-polarized STM and a stable spin-polarized electrode in a junction device, HM-AFMs provide a possibility of “single spin superconductivity” (SSS) due to the triplet (spin-parallel) Cooper pairs in the metallic channel [6]. The lack of macroscopic magnetization would prevent the competition between superconducting order and pre-existing magnetic field.

The newly discovered oxypnictide LaOFeAs has a layered crystal structure containing apparently distinct LaO and transition-metal pnictide layers [7]. The FeAs layers consist of a square lattice sheet of Fe atoms coordinated As above and below the Fe plane to form face sharing FeAs$_4$ tetrahedra, which are squeezed along the $c$ axis. Three different long-range magnetic
2. First-principles results

2.1. Computational method

Self-consistent band-structure calculations are based on the WIEN2k package [8] for the scalar relativistic hybrid full-potential augmented plane-wave plus local-orbitals method (FP-APW++) within the framework of spin-polarized density-functional theory (DFT). We employ the semirelativistic approximation and the core levels are treated fully relativistically. The electrons exchange-correlation potential is described with the generalized gradient approximation (GGA) using the Perdew-Burke-Ernzerhof functional parametrization [9]. The plane-wave cutoff is determined by $R_{MT}K_{\text{max}} = 8.0$ (where $R_{MT}$ is the muffin-tin radius, and $K_{\text{max}}$ is the maximum modulus for the reciprocal lattice vector). The Brillouin zone (BZ) is sampled on a uniform mesh with $16 \times 16 \times 7 \ k$ (in $P \bar{4}m2$) and $11 \times 11 \times 7 \ k$ points (in $Pccm$). The muffin-tin radii $R_{MT}$ are chosen as touching spheres to keep as much core charge inside the muffin-tin sphere as possible.

2.2. Half-metallicity

Like LaOFeAs, the striped and checkerboard structures are possible in LaOCr$_{0.5}$Fe$_{0.5}$As. Figure 1 presents the spin-resolved total and partial densities of states (DOSs) for striped LaOCr$_{0.5}$Fe$_{0.5}$As, consisting of alternating Cr and Fe stripes along the [100] direction, whose lattice parameters are optimized. Since the total energy and the magnetic moments depend on the As position anomalously strongly, the optimization is performed in the AFM states (not in paramagnetic states as usual). We choose a spin direction such that spin-polarized conduction electrons have spin-up (positive) states. Evidently, the electronic structure is characteristic of an HM-AFM; the spin-up channel is metallic and hence gapless while the spin-down channel is insulating, having a gap ($E_g$) for spin-diagonal excitations of spin-down states. Comparing the partial DOSs, we can readily realize that the exchange splitting is of opposite sign for the Cr 3$d$ and Fe 3$d$ states. This behavior makes the local magnetic moments of the two transition-metal cations antiparallel. Quite similar behavior is observed also in DOSs for checkerboard LaOCr$_{0.5}$Fe$_{0.5}$As, consisting of interpenetrating Cr and Fe square sublattices, as shown in figure 2. Unlike the stripe-ordered case ($Pccm$ with four formula units per cell), the checkerboard structure ($P \bar{4}m2$ with two formulae) does not possess inversion symmetry.

The optimized lattice parameters ($a$, $b$, $c$, $z_{\text{La}}$, and $z_{\text{As}}$) and the total energy difference between the HM-AFM and FM states ($\Delta E \equiv E_{\text{HM-AFM}} - E_{\text{FM}}$ in eV per formula unit), which are a measure of the magnetic coupling between Cr and Fe spins, are listed in table 1. The GGA calculations indeed lead to HM-AFM solutions at the equilibrium lattice parameters. Of course, the paramagnetic state is even (around 0.25 eV/f.u.) higher than the FM state, having a reduced optimum $z_{\text{As}}$ of 0.649.

Although local magnetic moments are ill-defined quantities in band-structure calculations, table 2 gives the total moments per formula unit cell and local moments within muffin-tin spheres of striped and checkerboard LaOCr$_{0.5}$Fe$_{0.5}$As. According to the notation of DOSs with spin-up positive and spin-down negative, it is convenient to choose the moment of Cr to be positive and then that of Fe to be negative. Negative moments at the anion sites $m_{\text{As}}$ arise from the $p$-$d$ hybridization effect [5]. As expected, the total moments are exactly zero within computational accuracy by virtue of the integer filling of the spin-down channel.
Figure 1. Spin-resolved total and partial densities of states (DOSs) per formula unit for striped LaO(Cr\textsubscript{0.5}Fe\textsubscript{0.5})As at the theoretical equilibrium lattice parameter, with spin-up positive and spin-down negative.

Figure 2. Spin-resolved total and partial densities of states (DOSs) per formula unit for checkerboard LaO(Cr\textsubscript{0.5}Fe\textsubscript{0.5})As at the theoretical equilibrium lattice parameter, with spin-up positive and spin-down negative.

Table 1. Calculated equilibrium lattice parameters (a, b, c, z\textsubscript{La}, and z\textsubscript{As}) of striped and checkerboard LaO(Cr\textsubscript{0.5}Fe\textsubscript{0.5})As.

| Structure   | a (Å)  | b (Å)  | c (Å)  | z\textsubscript{La} | z\textsubscript{As} |
|-------------|--------|--------|--------|----------------------|----------------------|
| Stripe      | 5.746  | 5.761  | 8.935  | 0.140                | 0.663                |
| Checkerboard| 4.075  | 4.075  | 9.002  | 0.139                | 0.662                |

Table 2. Calculated total spin magnetic moments per cell (m\textsuperscript{cell}), local moments within muffin-tin spheres at cation, anion, and the other interstitial sites (m\textsuperscript{Cr}, m\textsuperscript{Fe}, m\textsuperscript{As}, and m\textsuperscript{int} in µB), and the total energy differences between AFM and FM states (ΔE ≡ E\textsubscript{HM-AFM} − E\textsubscript{FM} in meV per formula unit) of striped and checkerboard LaO(Cr\textsubscript{0.5}Fe\textsubscript{0.5})As.

| Structure   | m\textsuperscript{cell} | m\textsuperscript{Cr} | m\textsuperscript{Fe} | m\textsuperscript{As} | m\textsuperscript{int} | ΔE  |
|-------------|-------------------------|-----------------------|-----------------------|-----------------------|-----------------------|-----|
| Stripe      | 0.00                    | 2.74                  | -2.71                 | -0.07                 | 0.06                  | -137|
| Checkerboard| 0.00                    | 2.78                  | -2.75                 | -0.07                 | 0.06                  | -128|

2.3. Normal conductivity
The locally near-tetrahedral environment for Fe ions is reminiscent of the AFM ZB FeX [5]. The squeeze of the FeAs\textsubscript{4} tetrahedra along the c axis rearranges the t\textsubscript{2g} and e\textsubscript{g} crystal-field energy levels to bring all d orbitals into a close proximity of E\textsubscript{F} and thus to diminish the direct overlaps between Fe d and As p orbitals. Orbital-resolved contributions to DOSs in figures 3 and 4 indicate that conduction electrons belong mainly to the d\textsubscript{xz} (and also d\textsubscript{yz} in the checkerboard structure) orbitals, suggesting that the p–d hybridization is still essential to the HM-AFM behavior.
Fe and Cr $d_{xz}$ bands broaden around $E_F$ in the striped structure (see figure 3), whereas the Fe $d_{xz}$ and $d_{yz}$ bands are well localized in the checkerboard one (see figure 4).

3. Conclusion

We conclude that both striped and checkerboard LaOCr$_{0.5}$Fe$_{0.5}$As are indeed HM-AFMs in the normal ground states. These HM-AFM states are the precursor states of the SSS phase with broken time-reversal symmetry that has a pairing instability. The striped structure might be preferable to the checkerboard one. Especially in noncentrosymmetric metals such as the checkerboard case, the spin degeneracy of the electronic bands is lifted by spin-orbit coupling. A 2D superconducting system with a significant spin-orbit coupling induced by the lack of inversion symmetry would display a mixed singlet-triplet superconducting state. This means that the superconducting order parameter would possess the exotic feature of having no definite parity.

References

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