Electronic and magnetic structure of LaOAsFe: similarities and differences between pnictides and cuprate superconducting parent compounds

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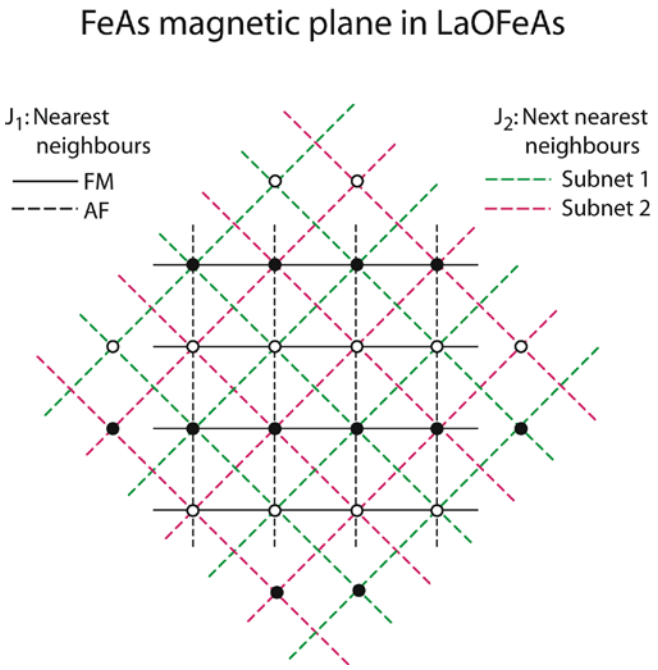
After almost ten years of relative stagnation, the search for superconductors with high critical temperature is experiencing revitalization due to recent discovery of superconductivity in iron-arsenide layered materials.[1] The new superconducting materials involve iron as transition metal rather than copper as in well-known superconducting cuprates. The parent material LaOFeAs exhibits an intriguing magnetic behaviour and become superconducting after (electron) doping with F or (hole) doping with Sr leading to a phase diagram closely related to that of superconducting cuprates. Neutron scattering experimentes show that undoped LaOFeAs exhibits a complex magnetic behaviour with an evident change in both χ versus T curve and conductivity, and undergoes a structural distortion from tetragonal to monoclinic symmetry at 155 K.[2] This relationship poses intriguing questions about similarities, differences, and ultimately on the underlying mechanisms of the superconductivity in both types of materials.

To better understand the similarities and differences of these two families of materials requires a careful analysis of their electronic structure. However, this is not simple as revealed from the literature dealing with cuprates. Several years and many publications have been necessary to establish that LDA and GGA incorrectly predict La_2CuO_4 (and other similar systems) to be a non-magnetic metal and hence they are unable to describe the electronic structure of these materials even at a qualitative level. For instance, LDA and GGA approaches predict a too small band-gap for cuprates indicating either a metallic or, in the most favourable cases, a semiconducting behaviour and a strong d character of the bands above and below Fermi level. This is in a clear contradiction with the experimentally measured properties such as the charge transfer insulating gap of 1-2 eV, antiferromagnetic ordering derived from the leading $J_1 \sim -140$ meV nearest neighbour exchange interaction or the strong mixing between Cu d and O p states for most of the valence bands. Clearly, proper description of the electronic structure of LaOFeAs requires as well going beyond the LDA and GGA methods thus avoiding the erroneous interpretations one can find in the literature

dealing with the theoretical description of cuprates [3]

In this work we show that the magnetic structure of LaOFeAs and superconducting cuprates parent compounds are more similar than imagined. In fact, accurate hybrid-DF and GGA+*U* periodic density functional calculations consistently describe the ground state of LaOFeAs as a complex insulator/semiconductor with a physical nature intermediate between Mott-Hubbard and charge-transfer limiting situations and a band gap at finite T perturbed by spin excitations.[4] The electronic structure involves Fe^{2+} cations in a distorted tetrahedral environment which can be effectively considered as particles with S=2 spin moments localized at Fe^{2+} sites with a local $3d^6$ electronic configuration in a quasi-tetragonal $FeAs_4$ structural unit (see Figure below).



Calculated magnetic ordering of the electronic ground state of LaOFeAs in terms of the two dominant magnetic coupling constants showing two interconnected square networks. B3LYP results suggest that LaOFeAs involves S=2 effective magnetic moments coupled by $J_1 \in [-18,-12]$ meV and with $J_1/J_2 \approx 1$.

The figure illustrates that the electronic ground state involves an intricate magnetic structure with two nearly equal antiferromagnetic nearest-neighbours and next nearest-neighbours magnetic coupling constants which gives rise to a highly frustrated magnetic ground state which cannot be easily anticipated from the crystal structure. Hence, the present study provides unbiased, independent confirmation of the magnetic order suggested by de la Cruz et al.[2] from neutron diffraction experiments on polycrystalline LaOFeAs samples and has is in agreement with subsequent studies using similar theoretical techiques.[5]

References

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