Magnetic properties of iron pnictides from spin-spiral calculations

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Abstract. The wave-vector \( q \) and doping dependences of the magnetic energy, iron moment, and effective exchange interactions in LaFeAsO, BaFe\(_2\)As\(_2\), and SrFe\(_2\)As\(_2\) are studied by self-consistent LSDA calculations for co-planar spin spirals. For the undoped compounds, the calculated total energy, \( E(q) \), reaches its minimum at \( q \) corresponding to stripe antiferromagnetic (AF) order. In LaFeAsO, this minimum becomes flat already at low levels of electron-doping and shifts to an incommensurate \( q \) at \( \delta = 0.2 \), where \( \delta \) is the number of additional electrons \( (\delta > 0) \) or holes \( (\delta < 0) \) per Fe. In BaFe\(_2\)As\(_2\) and SrFe\(_2\)As\(_2\), stripe order remains stable for hole doping down to \( \delta = -0.3 \). Under electron doping, on the other hand, the \( E(q) \) minimum shifts to incommensurate \( q \) already at \( \delta = 0.1 \).

Iron pnictides have attracted great interest of both experimentalist and theoreticians after the discovery of superconductivity with \( T_c = 27 \) K in F-doped LaFeAsO\(_{1-x}\)F\(_x\) \[1\] and with even higher \( T_c \) of 38 K in oxygen-free, potassium-doped Ba\(_{1-y}\)K\(_y\)Fe\(_2\)As\(_2\) \[2\]. Both families of iron pnictides have a quasi two-dimensional (2D) tetragonal crystal structure, in which FeAs layers are separated by either LaO or Ba layers. The Fe ions form a square lattice sandwiched between two As sheets shifted so that each Fe is surrounded by a slightly squeezed As tetrahedron. At about 150 K, both stoichiometric parent compounds undergo a structural transition at which the symmetry of the lattice lowers to orthorhombic \[3, 4\]. In BaFe\(_2\)As\(_2\) and SrFe\(_2\)As\(_2\), stripe antiferromagnetic (AF) order of 0.4–0.8 \( \mu_B \) Fe moments \[4, 5\], aligned ferromagnetically (FM) along the shorter \( b \)-axis and antiferromagnetically along the longer \( a \)-axis and the \( c \)-axis \[5, 6\], sets in at the same temperature as the structural transition. Transition to a phase with the same stripe AF order occurs also in LaFeAsO, but at a 20 K lower temperature than the structural transition \[7\]. Electron doping of the FeAs layers in LaFeAsO\(_{1-x}\)F\(_x\) suppresses the structural and magnetic transitions in favor of superconductivity already at \( x = 0.03 \) \[8\]. Also hole doping in Ba\(_{1-y}\)K\(_y\)Fe\(_2\)As\(_2\) suppresses the structural and magnetic transition \[2\] but at much higher doping of \( \sim 0.15 \) holes per Fe \[9\], whereas AF fluctuations are observed up to as high K content as \( y \sim 0.5 \). Electron doping caused by Co substitution in Ba(Fe\(_{1-x}\)Co\(_x\))\(_2\)As\(_2\), on the other hand, suppresses the magnetic transition at \( x \sim 0.06 \).

Since in many of the superconducting iron pnictides the highest \( T_c \) is observed at the doping level at which the magnetic transition is suppressed, the superconductivity seems to be closely related to magnetism and understanding the magnetic interactions between Fe moments in these compounds is of utmost importance. Although the theoretically calculated Fe magnetic moment and the stabilization energies of different magnetic solutions depend strongly on the employed computational method and exchange-correlation functional all band structure calculations that...
the stripe AF order is the magnetic ground state in both parent compounds [10, 11, 12, 13].

Results of spin-spiral calculations and an analysis of effective exchange interactions between Fe moments in electron doped LaFeAsO and hole doped BaFe$_2$As$_2$ were presented in our previous paper [14]. In this paper we show that electron doping of FeAs layers in $M$(Fe$_{1-x}$Co$_x$)$_2$As$_2$ ($M$=Ba, Sr) destabilizes stripe AF order as efficiently as in LaFeAsO.

Self-consistent calculations for co-planar spin spirals in LaFeAsO and $M$Fe$_2$As$_2$, with $M$=Ba or Sr, were performed within the LSDA using the linear muffin-tin orbital (LMTO) method in the atomic sphere approximation [15]. The effect of doping was simulated by using the virtual crystal approximation, with the doping level $\delta$ defined as the deviation of the number of valence electrons from corresponding values for undoped compounds normalized to the number of Fe atoms in the unit cells. Positive and negative values of $\delta$ correspond to electron and hole doping, respectively. Calculations for LaFeAsO were performed for $\delta$=0.1, 0.2, and 0.3, which corresponds to F content of $x=\delta$ in LaFeAsO$_{1-x}$F. Potassium doping of $y=0.2, 0.4, 0.6$ in Ba$_{1-y}$K$_y$Fe$_2$As$_2$ and Sr$_{1-y}$K$_y$Fe$_2$As$_2$ was modeled by $\delta=-0.1, -0.2, -0.3$ ($y=2|\delta|$). Finally, electron doping of FeAs layers in $M$(Fe$_{1-x}$Co$_x$)$_2$As$_2$ caused by Co substitution was studied for $\delta=0.1$.

The calculations for all doping levels were carried out for experimental room temperature crystal structures of the undoped compounds. The tetragonal ($P4/nmm$) unit cell with $a=4.0353$ Å, $c=8.7409$ Å, $z_{\text{La}}=0.14154$, and $z_{\text{As}}=0.6512$ was used for LaFeAsO [1]. MFe$_2$As$_2$ band structures were calculated for the body centered tetragonal ($I4/mmm$) unit cell with $a=3.9625$ Å, $c=13.0168$ Å, and $z_{\text{As}}=0.3545$ for $M$=Ba [4] and $a=3.9243$ Å, $c=12.3644$ Å, and $z_{\text{As}}=0.36$ for $M$=Sr [16].

As explained in details in [14], LMTO calculations for LaFeAsO place Fe $d_{xy}$ bands too close to $d_{yz,zx}$-derived bands as compared to results obtained with the full potential linear augmented plane wave (LAPW) method [17]. Better agreement between two band structures can be obtained by adding in LMTO calculations an on-site shift of $-150$ meV to the Fe $d_{xy}$ states. In the following, the results calculated for LaFeAsO with the shifted Fe $d_{xy}$ states are shown.

![Figure 1](image_url)

**Figure 1.** The $\mathbf{q}$ dependencies of the Fe magnetic moment (a) and the total energy (b) calculated for LaFeAsO with $\delta=0$ (red •), 0.1 (magenta ▼), and 0.2 (green ■). Spin spiral calculations were performed on the base of the generalized Bloch theorem [18]
under the assumption that the direction of the magnetization is constant within each atomic sphere. In these calculations the angle \( \theta \) between Fe magnetic moments and the \( c \) axis is set to 90 degree, i.e., all Fe moments lie in the \( ab \) plane. The angle \( \phi \) between the Fe moment and the \( a \) axis depends on the wave vector \( q \) of a spiral and is given by \( \phi_i = \phi_0^i + q \cdot R \), where \( R \) is the translation vector. The initial phases \( \phi_0^i \) for two Fe sites at the positions \( t_i \) in the tetragonal unit cell are fixed by \( \phi_0^i = q \cdot t_i \).

The total energy of spin-spirals was calculated for \( q \) varying along \( \Gamma (0,0) - X (\pi/d,0) - M (\pi/d,\pi/d) - \Gamma \) lines in the two dimensional magnetic Brillouin zone (BZ), where \( d \) is the distance between Fe nearest-neighbors in the \( ab \) plane. At \( X \) and \( M \) points collinear AF stripe- and checkerboard-like spin structures are formed. The calculations were performed for FM (\( q_z=0 \)) and AF (\( q_z = \pi/d_\perp \)) alignments of Fe moments in adjacent Fe layers, were \( q_z \) is the \( z \) component of the wave vector and \( d_\perp \) is the interlayer distance.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure2.png}
\caption{The \( q \) dependencies of the Fe magnetic moment (a, c) and the total energy (b, d) calculated for BaFe\(_2\)As\(_2\) (a, b) and SrFe\(_2\)As\(_2\) (c, d) with AF alignment of Fe moments along the \( c \) axis (\( q_z = \pi/d_\perp \)) and \( \delta=-0.1 \) (blue ▲), 0 (red ●), and 0.1 (magenta ▼). The corresponding curves calculated for undoped compounds (\( \delta=0 \)) with \( q_z=0 \) are also shown (red ○).}
\end{figure}

By comparing the total energies calculated for \( q_z=0 \) and \( q_z = \pi/d_\perp \) we found that the interlayer magnetic coupling in LaFeAsO is extremely weak. The \( q \) dependencies of the Fe magnetic moment and the total energy per Fe, \( E(q) \), calculated for LaFeAsO with \( q_z=0 \) and \( \delta=0, 0.1, \) and 0.2 are shown in figure 1. The minimum of \( E(q) \) for the undoped compound is found at the \( X \) point, i.e., for stripe AF order. This, together with the appearance of a local minimum along the \( M-\Gamma \) line, suggests that effective interactions between the nearest (\( j_1 \)) and next-nearest (\( j_2 \)) Fe neighbours are antiferromagnetic with \( j_2 > j_1/2 \) [14]. The energy of spin spirals rapidly increases with the decrease of \( |q| \) and at short wave vectors the self-consistent solution becomes nonmagnetic. As a FeAs layer in LaFeAsO is doped with electrons (\( \delta > 0 \)) to simulate Fe doping in LaFeAsO\(_{1-x}\)Fe\(_x\), another local minimum develops at an incommensurate
wave vector $\mathbf{q}_{\text{min}}$ along the $\bar{X}-\bar{M}$ line. $E(\mathbf{q}_{\text{min}})$ becomes lower than $E(\bar{X})$, i.e., the collinear stripe-like ordered AF order becomes unstable, at $\delta \gtrsim 0.1$.

In contrast to LaFeAsO, the energy of spin spirals calculated for $MFe_2As_2$ with AF alignment of Fe moments along the $c$ direction is significantly lower than for FM one. The corresponding curves for the undoped compounds are compared in figure 2. The change of magnetic order in Fe chains running along $c$ from AF to FM costs at the $\bar{X}$ point 3.5 meV/Fe in BaFe$_2$As$_2$ and 8.6 meV/Fe in SrFe$_2$As$_2$. The $\mathbf{q}$ dependencies of the total energy for both undoped $MFe_2As_2$ compounds are similar to $E(\mathbf{q})$ for LaFeAsO, with the minimum at $\bar{X}$ and a local one along the $\bar{M}-\Gamma$ line. Although the stabilization energy of the stripe-ordered solution, $|E(\bar{X}) - E(0)|$, is somewhat lower in SrFe$_2$As$_2$ than in BaFe$_2$As$_2$, the energy difference $E(M) - E(\bar{X})$ of 38 meV/Fe in the former is larger than in the latter (29 meV/Fe). This indicates that also $j_1$ and $j_2$ couplings within a FeAs layer are stronger in SrFe$_2$As$_2$ than in BaFe$_2$As$_2$.

As it was shown in [14], when BaFe$_2$As$_2$ is doped with holes, stripe AF order remains stable up to the doping level of $\delta \sim -0.3$, which corresponds to K content of about 0.6 in Ba$_{1-x}$K$_x$Fe$_2$As$_2$. The stabilization energy of the stripe-ordered solution is, however, strongly reduced by hole doping. The same tendency is also obtained from spin spiral calculations for SrFe$_2$As$_2$ with $\delta < 0$, which is illustrated by $\delta = -0.1$ curves in figure 2. Calculations performed for electron-doped $MFe_2As_2$ show that the total energy minimum shifts from $\bar{X}$ to incommensurate $\mathbf{q}$ already for $\delta = 0.1$. These results agree with the experimental phase diagram of Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ according to which the structural and magnetic transition at $T \sim 140$ K is suppressed at $x \approx 0.06$ [19].

Recently, strongly anisotropic nearest neighbour couplings along AF ($j_{1a}$) and FM ($j_{1b}$) Fe chains have been obtained from linear response calculations [20]. We estimated $j_{1a}$, $j_{1b}$, and $j_2$ from a list-square fit to $E(\mathbf{q})$ curves calculated using the magnetic force theorem. Preliminary results show that the anisotropy of $j_{1a}$ and $j_{1b}$ is much weaker than reported in Ref. [20]. However, the anisotropy strongly increases if the fit is performed in a small region of $\mathbf{q}$-space around the $\bar{X}$ point. The results of these calculations will be published elsewhere.

References

[1] Kamihara Y, Watanabe T, Hirano M and Hosono H 2008 J. Am. Chem. Soc. 130 3296
[2] Rotter M, Tegel M and Johrendt D 2008 Phys. Rev. Lett. 101 107006
[3] Nomura T, Kim S W, Kamihara Y, Hirano M, Sushko P V, Kato K, Takata M, Shluger A L and Hosono H 2008 Supercon. Sci. Technol. 21 125028
[4] Rotter M, Tegel M, Johrendt D, Schellenberg I, Hermes W and Poettgen R 2008 Phys. Rev. B 78 020503(R)
[5] Huang Q, Qiu Y, Bao W, Green M A, Lynn J W, Gasparovic Y C, Wu T, Wu G and Chen X H 2008 Phys. Rev. Lett. 101 257003
[6] Zhao J, Wu W R, Lynn J W, Chen G F, Luo J L, Wang N L, Hu J and Dai P 2008 Phys. Rev. B 78 140504
[7] de la Cruz C, Huang Q, Lynn J W, Li J, Ratchtiff W I, Zarestky J L, Mook H A, Chen G F, Luo J L, Wang N L and Dai P 2008 Nature (London) 453 899-902
[8] Dong J, Zhang H J, Xu G, Li Z, Li G, Hu W Z, Wu D, Chen G F, Dai X, Luo J L, Fang Z and Wang N L 2008 EPL 83 27006
[9] Chen H, Ren Y, Qiu Y, Bao W, Liu R H, Wu G, Wu T, Xie Y L, Wang X F, Huang Q and Chen X H 2009 EPL 85 17006
[10] Yin Z P, Lebegue S, Han M J, Neal B P, Savrasov S Y and Pickett W E 2008 Phys. Rev. Lett. 101 047001
[11] Yildirim T 2008 Phys. Rev. Lett. 101 057010
[12] Mazin I I, Singh D J, Johannes M D and Du M H 2008 Phys. Rev. Lett. 101 057003
[13] Opahle I, Kandpal H C, Zhang Y, Gros C and Valenti R 2009 Phys. Rev. B 79 024509
[14] Yaresko A N, Liu G Q, Antonov V N and Andersen O K 2009 Phys. Rev. B 79 144421
[15] Andersen O K 1975 Phys. Rev. B 12 3060-3083
[16] Tegel M, Rotter M, Weiss V, Schappacher F M, Poettgen R and Johrendt D 2008 J. Phys.: Condens. Matter 20 452201
[17] Mazin I I, Johannes M D, Boeri L, Koepernik K and Singh D J 2008 Phys. Rev. B 78 085104
[18] Sandratskii L M 1991 J. Phys.: Condens. Matter 3 8565-8585
[19] Chu J H, Analytis J G, Kuchareczyk C and Fisher I R 2009 Phys. Rev. B 79 014506
[20] Han M J, Yin Q, Pickett W E and Savrasov S Y 2009 Phys. Rev. Lett. 102 107003