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Magnetic properties of $MgCNi_{3-x}Fe_x$ by the first-principles study

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Abstract

The first-principles electronic structure calculations are performed for the nonoxide perovskite $MgCNi_{3-x}Fe_x$ compounds by FP-LAPW approach. A small local magnetic moment for Fe in $MgCNi_{3-x}Fe_x$ is always obtained in the cases condsidered. The consequence on spin moment of a distorted structure (*Pnma*) as well as bond length variation is discussed. The spin moment of Fe decreases significantly when the bond length of Fe–C reduces 5%.

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Keywords: Electronic structure; Spin moment; Distortion

The discovery of superconductivity (SC) ($T_c \sim 8$ K) [1] in the nonoxide perovskite $MgCNi_3$ has initiated great interests in recent years [2–9]. Considering the high concentration of magnetic element Ni atoms, the most surprising of this intermetallic compound is the observation of SC rather than ferromagnetism which generally breaks down SC. It is expected that magnetic interaction may play important role in the very existence of SC in this material [2–4]. A number of studies have been performed aimed at elucidating the nature of this superconducting phase [5,6]. Band structure calculations display a characteristic very narrow and high peak in the DOS of $MgCNi_3$ just below the Fermi level and it is dominated by 3d orbitals of Ni [2–4]. Naturally, it is suggested that the DOS at E_F should be greatly changed by 3d elements substitutions at Ni sites [7,8] and this will result in considerable changes of relevant properties.

There is an evidence of observing phase transition when applying Fe doping above 60% [9]. And also it needs an explanation for the increase of T_c with Fe doping [8] disregarded the fact that Fe induces the magnetic moment in $MgCNi_{3-x}Fe_x$. This work intends to investigate the magnetic properties in Fe-doped $MgCNi_3$ as well as the distortion effects. The first-principles density-functional electronic structure calculations are performed for $MgCNi_{3-x}Fe_x$ by using FP-LAPW of WIEK2k code [10].

In our calculation, the GGA [11] correction is taken into account when solving Kohn–Sham equation. The lattice parameter is fixed at the experimental value of 7.2 a.u. for both the doped and undoped cases. The Muffin–Tin radii are taken as 1.8, 1.7 and 1.9 a.u. for Mg, C and magnetic elements, respectively.

Table 1 lists the calculated magnetic moments. With $x = 0$, the spin moment of Ni is almost 0, which is consistent with the results in literatures. When Fe is doped in $MgCNi_3$, the polarization of Ni is noticeable. In the case of $x = 1$, Ni is opposite polarized comparing to Fe. There is always a considerable local magnetic moment for Fe but are much reduced from their pure bulk values. A strong hybridization between body-centered C and face-centered Ni or Fe is responsible for the reduction of their magnetic moments. The broad band of Ni 3d is kept approximately unchanged with the Fe doping, while the exchange splitting increases as the

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Table 1
Spin moments (μ_B) of Mg, C, Ni, Fe in $MgCNi_{3-x}Fe_x$

	Mg	C	Ni	Fe
$x = 0.0$[a]	0.0	0.0	0.0014	
$x = 1$[a]	−0.006	−0.06	−0.19	0.63
$x = 2$[a]	−0.02	0.008	0.11	1.25
$x = 3$[a]	−0.03	−0.13	−0.13	1.42
$x = 1$[b]	−0.006	−0.06	−0.18	0.62
$x = 1$[c]	0.0	−0.03	0.0	0.65

[a] $Pm\bar{3}m$ or $P4/mmm$ symmetry structure with lattice constant $a = 7.20$ a.u.

[b] $P4/mmm$ symmetry with $a = 7.19$ a.u. after structure optimization.

[c] *Pnma* structure.

doping concentration increases, which gives rise to the net spin moment on Ni, seen from Fig. 1. The band of Fe 3d is quite narrow with $x = 1$ (Fig. 1a), and the width increases consequently with $x = 2$ and 3. Apart from the hybridization between body-centered C and face-centered Fe, the interaction between Fe and Ni is also importantly responsible for the polarization change. The 3d-band character, for example, either narrow of Fe or broad of Ni, should make sense when applying the element substitution for improving the physical performance.

Fe doping may bring either local distortion or global changes of lattice structure. Firstly, the structure optimization is performed to simulate the global lattice changes. As an example, for the $MgCNi_2Fe$ case, the lattice constant is relaxed to be 7.19 from 7.2 a.u. The magnetic moment of Fe is reduced to $0.62\mu_B$ from $0.63\mu_B$ and to $0.18\mu_B$ from $0.19\mu_B$ for Ni, after structure optimization. This result means the lattice parameters hardly change with Fe doping and the spin moments both for Fe and Ni also change very little. The phase transition is also possible with the Fe doping so we consider the distorted structure *Pnma*, which is typical in perovskites. The spin moment of Fe in $MgCNi_2Fe$ with *Pnma* is obtained as $0.65\mu_B$, not very distinguishable from that of the $P4/mmm$ symmetry structure. However there are also interesting results with *Pnma* and will be discussed elsewhere.

Local environment distortion results in the bond length change. Table 2 presents the spin moment of every element in $MgCNi_2Fe$. It produces a shrinking chain. When the bond length reduces to 3.41 a.u., all spin moments in $MgCNi_2Fe$ are suppressed. The suppression of magnetic moment will benefit the superconductivity so that the probing of local distortion is very much expected.

In summary, the magnetic properties of $MgCNi_{3-x}Fe_x$ compounds are investigated. The local

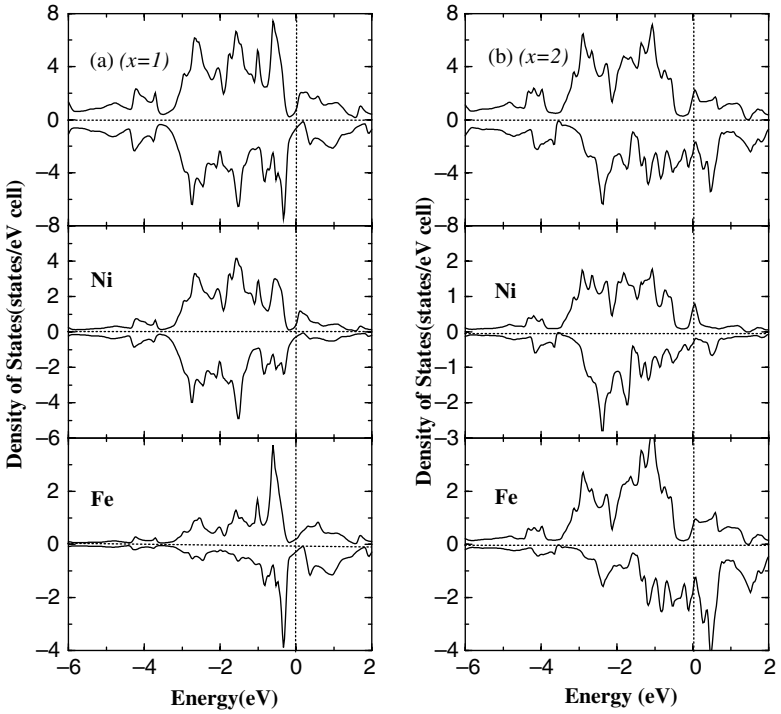

Fig. 1. Density of states of $MgCNi_{3-x}Fe_x$ ($x = 1, 2$) and partial density of states of Ni 3d and Fe 3d by FP-LAPW, the energy is respected to Fermi energy.

Table 2
Spin moments (μ_B) of Mg, C, Ni, Fe in $MgCNi_2Fe$ with Fe–C bond length changing

d_{Fe-C} (a.u.)	Mg	C	Ni	Fe
3.41	0.0	0.0	−0.012	0.02
3.48	−0.004	−0.04	−0.099	0.36
3.60	−0.006	−0.06	−0.19	0.63
3.72	−0.001	−0.07	−0.06	2.10

environment distortion of Fe is the crucial factor to explain the suppression of spin moment on Fe in these compounds although thermal effect is another reason. There is noticeable spin splitting on Ni with Fe doping, which is accounted for by the interaction between Fe and Ni.

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