Noncollinear magnetism in substitutionally disordered face-centered-cubic FeMn

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We use first principles electronic structure techniques to study the magnetic structure of γ-FeMn using the Korringa–Kohn–Rostocker multiple-scattering approach in conjunction with an extension of the single site coherent potential approximation to noncollinear magnetic structures. Our results show that the noncollinear $3Q$ and $2Q$ structures are both stable solutions with the former being slightly lower in energy. The collinear solutions could only be converged in a traditional spin-polarized calculation and are unstable in a noncollinear treatment. © *1999 American Institute of Physics.* [S0021-8979(99)59008-7]

I. INTRODUCTION

The magnetic structure in γ-FeMn has been the subject of numerous studies over the last 4 decades. Three models for the spin structure have been proposed[1] and are displayed in Fig. 1. However, up to now, it has not been possible to unambiguously determine the ground state magnetic structure or even to exclude one of the three models. Neutron diffraction, the method that is usually used to determine magnetic order, cannot distinguish between the noncollinear $3Q$ states (see Fig. 1) and a collection of domains which individually are in a collinear $1Q$ state.[1] Mössbauer transmission spectra indicate that the magnetic structure is either $3Q$ or $2Q$.[2] Inelastic neutron scattering measurements, however, support the collinear $1Q$ spin model.[3] The difficulty common to all these experiments is the rather involved and indirect data evaluation. A better theoretical understanding of the magnetic structure in FeMn would therefore be desirable to clarify some the of these ambiguities.

The theoretical situation, however, is not much clearer. Work by Cade and Young[4] indicates that collinear spin structures in γ-Mn are unstable. Recent first principles calculations[5] for face-centered-cubic (fcc) Fe show that the $3Q$ state is the ground state for small lattice constants but for large volumes, such as the experimental one, the lowest energy states are collinear. For the alloy, Hirai and Jo[6] argued within the rigid band approximation, that the magnetic structure should be collinear for Fe concentrations above 20 at. %.

In the present article we report on a first principles investigation of γ-FeMn. The calculations are performed within the local spin density approximation to density functional theory and we use the layer version of the Korringa–Kohn–Rostoker multiple scattering approach (LKKR).[7] We show how the spin-polarized version of the coherent potential approximation (CPA),[8] which successfully reproduces configurationally averaged quantities such as magnetic moments in random binary alloys, can be extended to noncollinear magnetic systems. The method is applied to solid solution Fe–50 at. % Mn on an fcc lattice to investigate stability and relative energies of the proposed magnetic structures.

II. CPA FOR ARBITRARY MAGNETIC STRUCTURES

The essence of the KKR–CPA theory is the replacement of the alloy by an effective material (CPA medium) with the following property: If one of its atoms is replaced by either one to the alloy components, no further scattering occurs on average. While the substitutionally disordered alloy is not periodic, the CPA medium is, which in turn reduces the computational complexity of the problem. Mathematically this requires the solution of the following equations in spin and angular-momentum representation,[9]

$$\sum_{\alpha} \tau_{i,\alpha}(\epsilon) c_{i,\alpha} = \tau_{i,\mathrm{CPA}}(\epsilon), \tag{1a}$$

$$\tau_{i,\alpha}(\epsilon) = [\tau_{i,\mathrm{CPA}}^{-1}(\epsilon) + m_{i,\alpha}(\epsilon) - m_{i,\mathrm{CPA}}(\epsilon)]^{-1}, \tag{1b}$$

$$\tau_{i,\mathrm{CPA}}(\epsilon) = \frac{1}{\Omega} \int_{\Omega} [m_{i,\mathrm{CPA}}(\epsilon) - G(\mathbf{k},\epsilon)]^{-1} d\mathbf{k}, \tag{1c}$$

where α and i label the species and the lattice site in the unit cell, respectively. $m = t^{-1}$ denotes the inverse single-site t

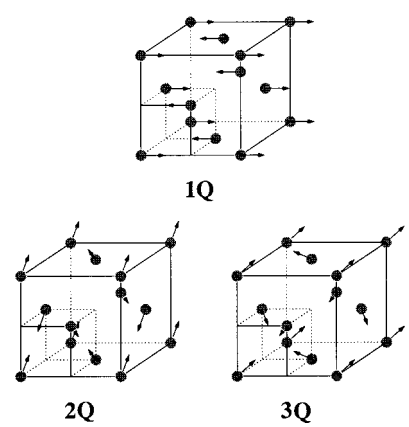

FIG. 1. Three simplest antiferromagnetic configurations in an fcc lattice.

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matrix, τ is the scattering path operator, and G the usual KKR structure constant.[9] Note that, in contrast to the spin-polarized case, the general noncolliear treatment involves operators in Eqs. (1a)–(1c) which are not block diagonal in the spin indices. The underlying assumption for the treatment of noncollinear magnets within the atomic sphere approximation[10] is that inside an atomic sphere the magnetization direction is constant. A local frame of reference can therefore be found, in which the charge density is block diagonal in the spin index. Thus, if $u(\theta,\phi)$ is the transformation in spin space that corresponds to the real space transformation between local and global frame of reference, we have

$$\begin{pmatrix} \rho_{11} & \rho_{12} \\ \rho_{21} & \rho_{22} \end{pmatrix} = u(\theta,\phi)^{\dagger} \begin{pmatrix} \rho'_{\uparrow} & 0 \\ 0 & \rho'_{\downarrow} \end{pmatrix} u(\theta,\phi),$$

where the prime denotes quantities in the local frame of reference. We are now ready to formulate the procedure for calculating the electronic structure of substitutionally disordered alloys with possibly noncollinear orientation of magnetic moments. We start with a set of local frames, determined by their polar angles $(\theta_{i,\alpha},\phi_{i,\alpha})$, and spherically symmetric potentials given in these local frames as up- $(\uparrow)$ and down-spin $(\downarrow)$ contributions, $u'^{s=(\uparrow,\downarrow)}_{i,\alpha}(r)$. With these potentials the regular and irregular solutions, $Z'^{s,l}_{i,\alpha}(r)$ and $J'^{s,l}_{i,\alpha}(r)$, respectively, and the single-site t matrices $t'^{s,l}_{i,\alpha}$ are determined from the atomic Schroedinger equation in the local frame. The t matrices are then transformed into the global frame, in which Eqs. (1a)–(1c) are solved and the resulting scattering path operators are transformed back into the local frames using

$$\tau'_{i,\alpha} = u(\theta_{i,\alpha},\phi_{i,\alpha})\tau_{i,\alpha}u(\theta_{i,\alpha},\phi_{i,\alpha})^{\dagger}.$$

Note that in general $\tau'_{i,\alpha}$ is not block diagonal in spin indices. The charge density is calculated in these local frames in the usual way, i.e., from the trace of the imaginary part of the Green function, $G(\mathbf{r},\epsilon)$,

$$[\rho'_{i,\alpha}(\mathbf{r})]_{s_1,s_2} = -\frac{1}{\pi}\int_{\epsilon}[G'_{i,\alpha}(\mathbf{r},\epsilon)]_{s_1,s_2}\,d\epsilon, \tag{2}$$

with

$$\begin{aligned}[G'_{i,\alpha}(r,\epsilon)]_{s_1,s_2} = &-\sum_{L} Z'^{s_1,L}_{i,\alpha}(\mathbf{r},\epsilon)J'^{s_1,L}_{i,\alpha}(\mathbf{r},\epsilon)\delta_{s_1,s_2} \\ &+\sum_{L_1,L_2}\{Z'^{s_1,L_1}_{i,\alpha}(\mathbf{r},\epsilon) \\ &\times[\tau'_{i,\alpha}(\epsilon)]_{L_1,s_1,L_2s_2}Z'^{s_2,L_2}_{i,\alpha}(\mathbf{r},\epsilon)\},\end{aligned}$$

where we have introduced $L=(l,m)$ and

$$F^{s,L}_{i,\alpha}(\mathbf{r},\epsilon) = F^{s,l}_{i,\alpha}(r,\epsilon) * Y_L(\mathbf{r}/r)$$

for $F=Z$ and $F=J$, respectively.

The new averaged direction of the magnetization of species α on site i is determined by the local moment,

$$\boldsymbol{\mu}_{i,\alpha} = \int \sum_{s_1,s_2}[\rho'_{i,\alpha}]_{s_1,s_2}\boldsymbol{\sigma}_{s_1,s_2}\,dr,$$

TABLE I. Magnetic moments in μ_B and energies in meV of FeMn in different magnetic configurations. The minus sign of μ_{Fe} indicates that Fe and Mn have opposite magnetic moments on the same sublattice. FM and AFM refers to, respectively, the ferro and antiferromagnetic alignment of the average moments on different sites.

	$1Q$-FM	$1Q$-AFM	$2Q$	$3Q$
μ_{Mn}	1.80	1.58	1.85	1.91
μ_{Fe}	−1.23	−1.48	2.00	2.05
Energy	73.7	85.2	5.6	0.0

where $\boldsymbol{\sigma}=(\sigma_x,\sigma_y,\sigma_z)$ are the Pauli matrices. This defines the angles of new local frames $(\theta^{\mathrm{new}}_{i,\alpha},\phi^{\mathrm{new}}_{i,\alpha})$ in which the charge density is taken to have the form $\rho^{\mathrm{new}}_{i,\alpha,s=(\uparrow,\downarrow)}$ that allows the determination of the new potential to start the next iteration. This procedure can be repeated until self-consistency is attained, i.e., until the output potentials and angles are equal to their respective input values.

Similarly to the case of pure systems, the procedure described above implicitly assumes that the local moments are aligned with the local magnetic fields. The algorithm will therefore only give correct answers for states in which the directions of the input potentials and the output charge density coincide and is therefore only adequate to determine fixed points in magnetic configuration as is the case in the states studied below.

III. RESULTS FOR fcc FeMn

With the method described in Sec. II, we are now in a position to investigate the different magnetic states that have been proposed for fcc FeMn. The calculations are performed at the experimental lattice constant ($a=3.63$ Å). Within the LKKR–CPA formalism which we use here, the magnetic unit cell of the noncollinear $2Q$ and $3Q$ structures can be composed from two (001) layers each having two sites in its two-dimensional (2D) periodic unit cell. These layers are then repeated in the (001) direction using the layer doubling algorithm described in Ref. 7. In order to determine energy differences accurately, all calculations are performed with the same basis.[11]

The first important result of the calculations is concerned with the stability of the self-consistent solutions. The collinear solution could only be converged in a conventional spin-polarized mode where the moments are actually constrained to be collinear. When the resulting self-consistent potentials are introduced into the noncollinear calculation, small deviation from the collinear alignment of the moments, caused for example by numerical noise, would drive the system away from its initial state into either the $3Q$ or the $2Q$ state. Both noncollinear states, however, are stable in the sense that the moment could be perturbed away from the orientation of the self-consistent field (SCF) solution and then would converge back to the original state.

The magnetic moments and relative energy differences between different magnetic configurations are summarized in Table I. Two collinear states could be converged. They differ in the relative orientation of the average magnetic moments per site, one being ferromagnetic while the other is antifer-

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romagnetic. The most important difference between collinear and noncollinear solutions with respect to the moments is that for both collinear states the Fe and Mn moments align in opposite directions on the same sublattice while in the noncollinear states the Fe and Mn moments are parallel. Note that this is a result of the calculation, not a constraint, since the noncollinear CPA formalism does not require moments of different species on a given sublattice to be collinear.

Clearly the relative energy difference indicate that the collinear solution is not the ground state. More importantly the issues of stability and the ferromagnetic alignment in the collinear state with lower energy indicate strongly that the FeMn is in a noncollinear magnetic state. The energy difference between the two noncollinear states is relatively small with the $3Q$ being more favorable. The difference, however, which corresponds to $\approx$60 K, is small enough that the systems may actually occupy both states. This would explain why the Mössbauer study of Kennedy and Hicks[2] could not uniquely identify the ground state configuration.

IV. CONCLUSIONS

We have extended of the KKR–CPA method to noncollinear magnetic systems. The application of the technique to FeMn clearly indicates the the magnetic ground state of this alloy is noncollinear. In fact, collinear states could only be found in a conventional spin-polarized calculations with the lowest energy spin-polarized state having a finite average magnetization which clearly contradicts the experiment. Two stable noncollinear states are found, where the $3Q$ has slightly lower energy than the $2Q$. The energy difference is however small enough so that at room temperature both states may be occupied.

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