Origin of large magnetostriction in FeGa alloys

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Using the highly precise full potential linearized augmented plane wave method, the phase stability, magnetism, and magnetostriction of Fe_3Ga alloys were investigated. The magnetostrictive coefficients strongly depend on the atomic arrangement. The B_2-like structure, although it is unstable in the small unit cell chosen here, appears to play a crucial rule for the large positive magnetostriction in the FeGa alloys as observed recently. Electronic origin of enhancement in magnetostriction is discussed in terms of density of states and band structures. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1450791]

Magnetostrictive materials are widely used in various sensor and actuator applications.[1] Large magnetostriction is usually found in rare-earth transition metal compounds, whereas most of magnetic transition metal crystals and alloys show weak magneto-elastic effect. Very recently, it was found that the magnetostriction of bcc Fe is greatly enhanced by the addition of Ga.[2,3] Room temperature magnetostriction (λ_{001}) of $Fe_{1-x}Ga_x$, depending on x and also the quenching condition, can reach to >200 ppm (10^{-6}), a value which corresponds to a tenfold increase above the magnetostriction of the pure bcc bulk Fe (λ_{001} 20 ppm, $\lambda_{111}-16$ ppm).

To understand the mechanism of this unusual enhancement, first principles calculations for magnetostrictive coefficients of several ordered Fe_3Ga alloys are carried out using the full potential linearized augmented plane wave (FLAPW) method.[4] As was applied to many other systems,[5] the torque approach was adopted for the determination of the strain dependence of magneto-crystalline anisotropy energy, $E_{\mathrm{MCA}}(c)$. The magnetostrictive coefficients (λ_{001}) were calculated from the ratio of $dE_{\mathrm{MCA}}(c)/dc$ and $d^2E_t(c)/dc^2$, where E_t stands for the total energy and c is the length of unit cell along the z axis. Three simple structures, namely the DO_3, B_2, and $L1_2$ (as given in Fig. 1) are investigated in the present work.

In the FLAPW approach, no shape approximation is adopted for the charge, potential, and wave function expansions. Augmented plane waves with an energy cutoff of 14 Ry (225 Ry) were used to expand the bases (charge and potential) in the interstitial region. More than 1050 (for the DO_3 structure) and 2100 (for the B_2 and $L1_2$ structures) k points in the irreducible Brillouin zone are used. The generalized gradient approximation (in the PBE96 form[6]) was employed to describe the exchange-correlation interaction.

The lattice constants of the three structures were first optimized through total energy minimizations. The cubic $L1_2$ structure is found to be the ground state (with a lattice constant of $a=c=6.93$ a.u.), while the quasicubic DO_3 structure is a metastable state (with the lattice size of $a=7.742$ a.u., $c=10.949$ a.u.) with the degree of freedom given in the small unit cell. The volume of unit cell appears to conserve well in different geometries. Surprisingly, the B_2-like structure is unstable under the tetragonal distortion mode. As shown in Fig. 2, the total energy decreases monotonically when the B_2-like lattice elongates along the z axis.[7] In reality, however, atoms have much higher degree of freedom with random disordering, and it is still possible to have the B_2-like structure locally in experimental samples. It is thus important to analyze the magnetostrictive behavior of all three geometries.

To determine magnetostrictive coefficients, lattice strain along the z axis (c/c_0) is used as a parameter. The calculated results of E_{MCA} are plotted in Fig. 3. For the $L1_2$ and DO_3 structures, the constant volume mode was adopted in lattice distortion. For the B_2-like structure, however, the lattice size in the x,y plane was fixed (i.e., constant area mode) for the determination of elastic-constants c_{11} and c_{12}.

Interestingly, the calculated slope of E_{MCA} (or equivalently the sign of λ_{001}) for the $L1_2$ and DO_3 structures is negative, while it is positive for the B_2-like structure. Experimentally, λ_{001} for $Fe_{1-x}Ga_x$ is $+150$ to $+200$ ppm, when x is 0.25. This indicates that, although being an unstable structure in the small unit cell, local B_2-like structure plays a key role for the strong positive magnetostriction in FeGa alloys. Indeed, some abnormality was observed very recently by Clark[8] that the magnetostrictive coefficient of $Fe_{1-x}Ga_x$($x=0.241$) drops when temperature (magnetic moment) decreases (increases) to 0.25. This can be interpreted with the current theoretical results as due to the formation of more stable $L1_2$ and DO_3 structures.

Quantitatively, the calculated values of λ_{001} are -107 ppm (10^{-6}), -298 ppm, and $+380$ ppm for the DO_3, $L1_2$ and B_2-like structures, respectively. Overall, the magnitude of λ_{001} for all three structures is much larger than that for the pure bcc bulk Fe (20 ppm), owing to the stronger magneto-elastic coupling as well as to the smaller Young's modulus.

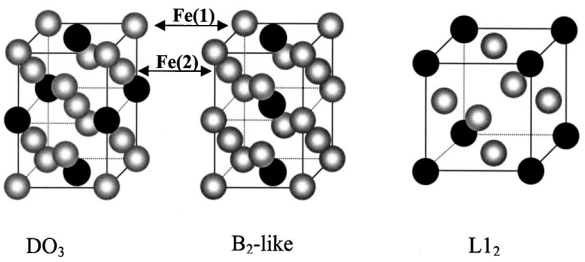

FIG. 1. Schematic model for the Fe_3Ga in DO_3, B_2-like and $L1_2$ structures. Gray (black) balls are for Fe (Ga) atoms.

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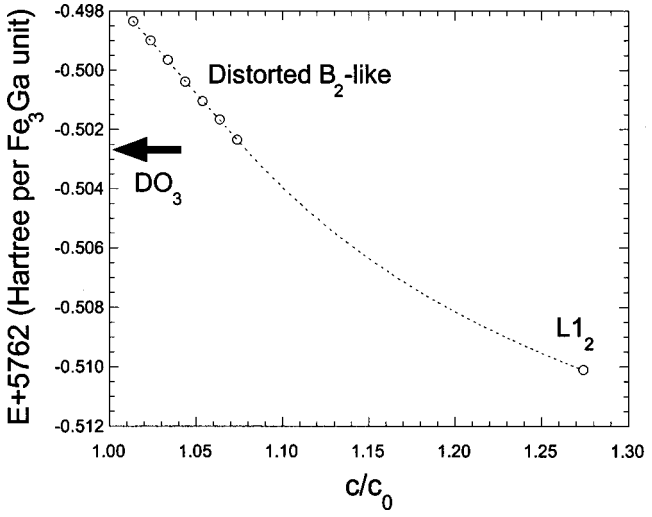

FIG. 2. Calculated total energies of the B_2-like Fe_3Ga lattice as a function of lattice distortion. Total energies for the DO_3 and $L1_2$ structures are for the optimized lattice sizes.

The change in sign of λ_{001} from the DO_3 structure to the B_2-like structure is fascinating, since only the second neighbor arrangement is altered. The calculated magnetic moments in the B_2-like structure [2.48 μ_B for Fe(1), 2.08 μ_B for Fe(2), and -0.08 μ_B for Ga] are slightly larger than those in the DO_3 structure [2.41 μ_B for Fe(1), 1.96 μ_B for Fe(2), and -0.07 μ_B for Ga].

Analyses in electronic properties of Fe_3Ga indicate that the spin-orbit coupling interactions among states localized in the Fe(2) layer are of key importance for the enhancement in magnetostriction. In the DO_3 structure, the negative magnetostriction is associated with strain dependence of Fermi surface of the Fe(2)-$d_{xz,yz}$ state (majority spin) around the center of Brillouin zone. For the B_2-like structure, as shown in Fig. 4 (top panel), the strain-induced change in $E_{MCA}(k_z)$ (obtained by integrating contributions in the k_x, k_y planes) occurs merely in the top 2/5 portion of the three-dimensional

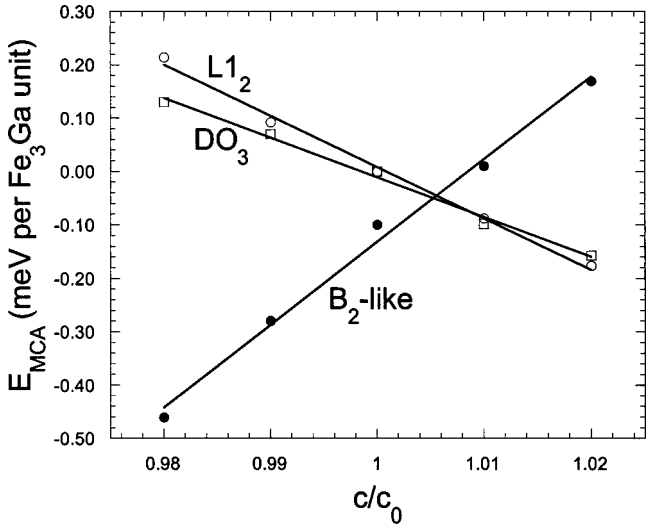

FIG. 3. Calculated strain-induced magneto-crystalline anisotropy energies of Fe_3Ga.

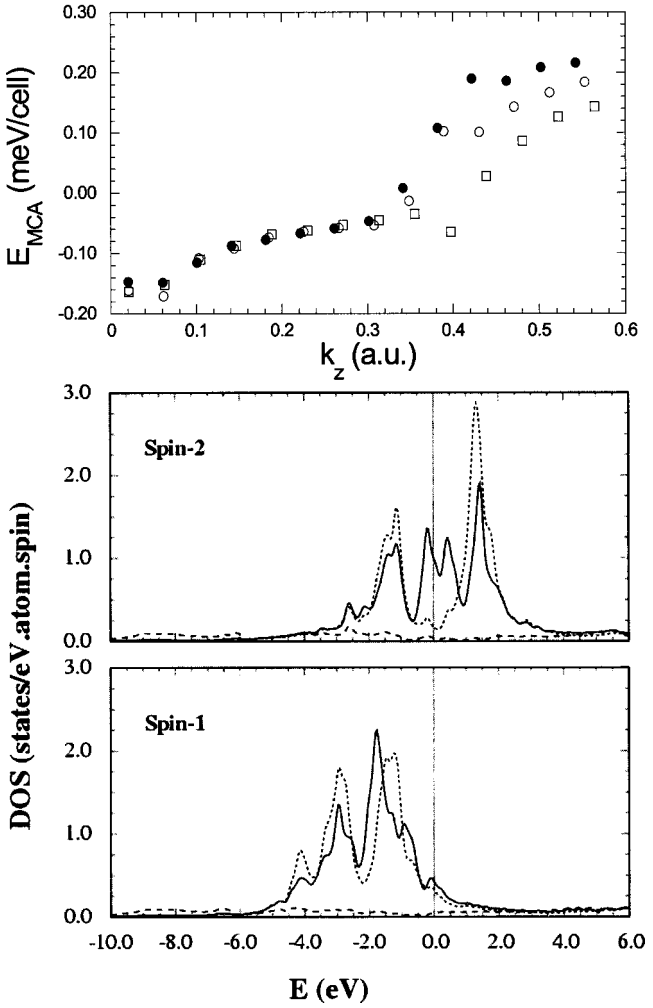

FIG. 4. Calculated density of states for Fe(1) (dotted lines), Fe(2) (solid lines), and Ga (dashed lined) of B_2-like Fe_3Ga separated in the two spins. Data points in the top-panel are for the distribution of E_{MCA} along the k_z axis in the 3D Brillouin zone (solid circles, open circles, and open squares are for the 2%, 0%, and -2% stretched lattice, respectively).

(3D) Brillouin zone. The calculated density of states (DOS) for the B_2-like Fe_3Ga lattice is also shown in Fig. 4 for the two spins. Similar to results for the DO_3 structure, the Fe(2)-t_{2g} bands are narrowed owing to the weakness of Fe–Ga hybridization, and strong Fe(2)-DOS peaks (in solid lines) form across the Fermi level. The leading contribution to the magneto-elastic coupling in the top plane of the Brillouin zone (mainly at the edge in the k_x, k_y plane) is from the spin-orbit coupling interaction between the nondegenerate Fe(2)-d_{xz} and Fe(2)-d_{yz} states in the minority spin channel, which leads to positive magnetostriction in the B_2-like structure. These two states, however, are degenerate for the DO_3 structure because of the higher symmetry. Clearly, the magnitude and even sign of λ_{001} sensitively depend on subtle changes in atomic arrangement. Due to the limit in space here, extensive discussions on this aspect will be published elsewhere.[9] Clearly, a large portion of the B_2-like structure in a sample is necessary to obtain strong positive magnetostriction as observed in experiments. In fact, the B_2-like structure can be stabilized if (1) the local Fe/Ga

ratio is not 75/25 or (2) it mixes with other structures in a random way. Calculations with larger unit cells are under way to investigate these possibilities.

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