

Monte Carlo studies of directional pair ordering in disordered binary and ternary ferromagnetic BCC crystalline alloys

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Magnetic field annealing can be used to induce uniaxial magnetic anisotropy in ferromagnetic alloys, which is often desirable, for example, to reduce core losses in materials of interest for high frequency inductive components in power conversion applications. An understanding of the compositional dependence of the uniaxial induced anisotropy constant K_U is important to develop materials for such applications. In this work, Monte Carlo studies of directional pair ordering in bcc crystalline alloys using the Néel pair model and a Metropolis-type algorithm are reported. The compositional dependence of K_U in disordered binary alloys is compared with analytical results and extended to the calculation of K_U for ternary alloys. An approximate $X_B^2(1-X_B)^2$ functional dependence of K_U is observed for binary alloys, where X_B is the atomic fraction of one of the atomic species. For ternary alloys a more complicated functional dependence is observed. These results are used to critically evaluate the potential for increasing K_U through ternary alloying additions to binary ferromagnetic alloys. © 2007 American Institute of Physics. [DOI: 10.1063/1.2711389]

INTRODUCTION

The directional pair ordering mechanism of field induced anisotropy first proposed by Néel¹ and investigated in detail by Chikazumi² and others³⁻⁵ has been used to explain the composition and temperature dependences of field induced anisotropies in binary ferromagnetic alloys including polycrystalline Ni-Fe.^{6,7} Recently, directional pair ordering in a crystalline phase has been used to explain magnetic field induced anisotropy in the Fe-rich (Fe,Co)-based magnetically soft nanocrystalline/amorphous nanocomposite materials such as Finemet and Nanoperm,^{8,9} where the crystalline phase is a bcc-derivative structure. To investigate the predictions of Néel's anisotropic pair ordering model for these alloys, Monte Carlo calculations have been used to simulate the formation of an anisotropic distribution of atom pairs in bcc-based binary and ternary ferromagnetic crystalline alloys through magnetic field annealing.

MODEL DESCRIPTION AND ASSUMPTIONS

A code using a Metropolis-type^{10,11} algorithm, suitable for simulating chemical ordering in binary alloys and considering only nearest neighbor interactions, has been modified to include anisotropic bond energies. A code for use with the commercially available program MATHEMATICA® was written by one of the authors (Ohodnicki). In all simulations, the system Hamiltonian and the transition probability governing the evolution of the ordering during field annealing were

$$E = \frac{1}{2} \sum_{j=1}^8 \sum_{i=1}^N (V_{ij} + \ell_{ij} \cos^2 \theta_j), \quad (1)$$

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$$W = \frac{e^{-(\Delta E^{n \leftrightarrow m}/kT)}}{[1 + e^{-(\Delta E^{n \leftrightarrow m}/kT)}]}, \quad (2)$$

where the sums with i and j indices are taken over the atomic positions in the simulated crystals and the eight possible near neighbor directions, respectively. E is the overall bond energy of the crystal, including both isotropic (V_{ij} 's) and anisotropic (ℓ_{ij} 's) contributions. $\Delta E^{n \leftrightarrow m}$ is the change in internal energy associated with swapping the atom at the n th lattice position and the m th lattice position. W is a transition probability¹⁰ calculated using $\Delta E^{n \leftrightarrow m}$. The magnetization is assumed to be saturated along a particular direction, and θ_j is the angle between the assumed magnetization direction and the j th bond direction. V_{ij} and ℓ_{ij} , respectively, are the isotropic bond energy and anisotropic pseudodipolar interactions between the atom at the i th position and the nearest neighbor atom oriented along the j th bond direction. The inclusion of both chemical and pseudodipolar nearest neighbor interactions between atoms for long-range and short-range chemical ordering and anisotropic directional pair ordering in binary alloys has been carried out by Iwata in a statistical mechanics framework using the Bethe approximation.¹² This work motivated our choice of the Hamiltonian of Eq. (1), although here the isotropic part of the pseudodipolar contribution is absorbed into the V_{ij} 's.

In all simulations, a disordered binary or ternary crystal of $16 \times 16 \times 16$ conventional unit cells (9826 atoms) was generated on a bcc lattice with a specified composition. Simulated field annealing was carried out at a fixed temperature by choosing a magnetization orientation and a corresponding set of ℓ_{ij} 's and V_{ij} 's. Using the pseudorandom number generator function in MATHEMATICA®, pairs of atoms were chosen randomly, and the values of $\Delta E^{n \leftrightarrow m}$ and W were calculated. Atoms were swapped if W was larger than a random number generated between 0 and 1. Periodic boundary conditions were employed. Attention is focused here on ideal

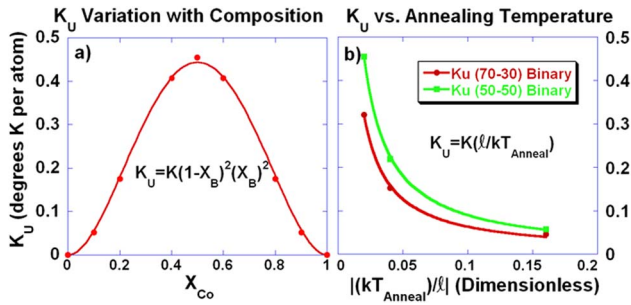


FIG. 1. (Color online) (a) Induced anisotropy constant (K_U) as a function of composition for an ideal binary solid solution and (b) as a function of annealing temperatures illustrating agreement with the expected compositional and temperature trend. The plotted points are actual simulation results, while the curves are fitted to $K_U \propto X_B^2(1-X_B)^2$ and $K_U \propto 1/(kT_{\text{anneal}})$, respectively.

solid solutions where the V_{ij} 's are all equal and/or zero.

Most simulations were carried out for $\sim 10^6$ attempted atomic swaps, though several longer simulations ($\sim 10^7$) were conducted to verify that a metastable state was not approached. The magnitude of the induced anisotropy was calculated according to the following expression (M_{anneal} corresponds to the magnetization direction assumed during annealing):

$$K_U = \left[\frac{1}{2} \sum_{j=1}^8 \sum_{i=1}^N (\ell_{ij} \cos^2 \theta_j) \right] \Bigg|_{M \perp M_{\text{anneal}}} - \left[\frac{1}{2} \sum_{j=1}^8 \sum_{i=1}^N (\ell_{ij} \cos^2 \theta_j) \right] \Bigg|_{M \parallel M_{\text{anneal}}}. \quad (3)$$

Equation (3) is only truly valid in the case where M_{anneal} is along one of the high symmetry directions of the cubic crystal; otherwise the induced easy axis is expected to deviate from the direction of M_{anneal} .²

DISCUSSION OF MODEL RESULTS

Figure 1(a) plots values of K_U calculated according to Eq. (3) as a function of the composition for a binary alloy using:

$$\frac{\ell_{AB}}{kT_{\text{anneal}}} = -\frac{\ell_{AA}}{kT_{\text{anneal}}} = -\frac{\ell_{BB}}{kT_{\text{anneal}}} = \frac{\ell}{kT_{\text{anneal}}} = -0.1. \quad (4)$$

The magnetization was assumed to be oriented along [111], which results in the maximum K_U for binary bcc single crystals according to directional pair ordering theory.^{2,12} For the chosen parameters of Eq. (4), the composition dependence of K_U for a binary ideal solid solution is well fitted to Iwata's expression $K_U \propto X_B^2(1-X_B)^2$.¹² Because $\ell_{ij}/kT \ll 1$ is typically true for realistic annealing temperatures, care must be taken in choosing the values of ℓ_{ij} to be significantly less than kT_{anneal} to obtain good agreement with experiment and Iwata's theory. Actually, the necessary condition for a binary alloy is $\ell_0^{AB}/kT \ll 1$, where ℓ_0^{AB} defined below in Eq. (5). However, $\ell_{ij}/kT \ll 1$ for all ℓ_{ij} 's implies $\ell_0^{AB}/kT \ll 1$. For smaller assumed values of ℓ_{ij}/kT , K_U values are more difficult to estimate reproducibly since the fluctuations in K_U become comparable to the magnitude of K_U . Values of

$\ell_{ij}/kT_{\text{anneal}}$ were chosen according to Eq. (4), so that the model reasonably reproduced the expected variation of K_U in the binary system without making estimation of the K_U value prohibitively difficult.

The extended model has also been used to investigate the dependence of K_U on the annealing temperature with fixed values of ℓ_{ij} . Here, the values of ℓ_{ij} were assumed independent of temperature (a good approximation for alloys with $T_{\text{Curie}} \gg T_{\text{anneal}}$), and K_U is expected to vary as $K_U \propto 1/(kT_{\text{anneal}})$ if the expression $\ell_{ij}/kT \ll 1$ holds.^{2,12} Figure 1(b) shows that the observed values of K_U for several compositions of the binary alloy using the ℓ_{ij} 's of Eq. (4) are well fitted by expressions of this form.

The model was extended to an ideal ternary solid solution where four additional ℓ_{ij} 's are required to specify the interactions of the system. To include isotropic chemical interactions. For binary alloys, the development of K_U is governed entirely by a single parameter defined by Eq. (5), which is analogous to the well-known chemical interaction parameter:

$$V^{AB} = V_{AB} - \frac{1}{2}(V_{AA} + V_{BB}),$$

$$\ell_0^{AB} = \ell_{AB} - \frac{1}{2}(\ell_{AA} + \ell_{BB}). \quad (5)$$

A positive or negative V^{AB} indicates a tendency for formation of less or more AB pairs, respectively, as compared to an ideal solid solution. The generalization to anisotropic bond energies predicts AB pairs to orient parallel to the magnetization direction during annealing for a binary alloy when ℓ_0^{AB} is negative. When ℓ_0^{AB} is positive, AA and BB pairs tend to orient parallel to the magnetization direction. If ℓ_0^{AB} does not change sign between measurement and annealing temperatures, the induced easy axis lies along the magnetization direction during annealing if it is a high symmetry direction of the crystal. K_U is expected to increase monotonically up to $X_B=0.5$ following the relationship $K_U \propto X_B^2(1-X_B)^2$. These predictions are independent of the absolute and relative values of ℓ_{AA} , ℓ_{BB} , ℓ_{AB} , and hence ℓ_0^{AB} , though the magnitude of K_U does depend on the magnitude of ℓ_0^{AB} .

Figure 2 represents a binary crystal, a (110) planar section, and the magnetic anisotropy energy surface (neglecting magneto-crystalline anisotropy) for the parameters chosen in Eq. (4) ($\ell_0^{AB}/kT_{\text{anneal}} = -2\ell = -0.2$) and also after replacing ℓ with $-\ell$ ($\ell_0^{AB}/kT_{\text{anneal}} = +0.2$) for comparison. Since the predicted compositional dependence of K_U does not depend on the values of the ℓ_{ij} parameters, a common technique used to increase field induced anisotropy in ferromagnetic materials has been to add a second ferromagnetic transition metal element to allow directional pair ordering between the atomic species ($K_U \propto X_B^2$ for dilute binary alloys).⁹

In ternary alloys, one can define an ℓ_0 parameter for each of the unlike bonds (ℓ_0^{AB} , ℓ_0^{AC} , and ℓ_0^{BC}) and several new configurations can arise. Thus, the analysis is more complicated and the compositional dependence of K_U is a richer problem than for the corresponding binary alloys. To investigate these compositional effects, two different sets of ℓ_{ij} 's were used. The first set is chosen to be the same as the parameters used in Eq. (4) to generate Fig. 1. These param-

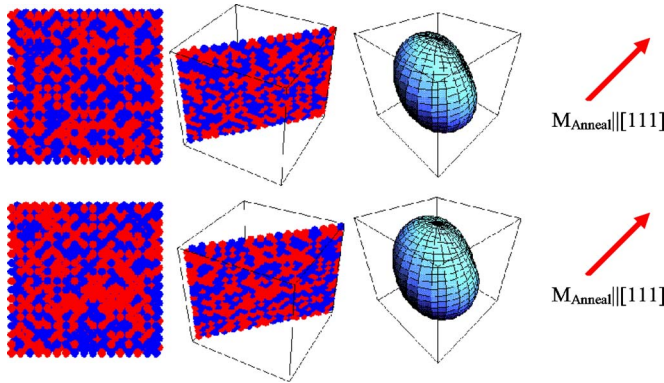


FIG. 2. (Color online) Graphical illustrations of binary bcc crystals and corresponding (110) sections and magnetic anisotropy energy surfaces (neglecting magnetocrystalline) after simulated field annealing for $\ell_0^{AB} = +0.2kT_{\text{anneal}}$ (top row) and $\ell_0^{AB} = -0.2kT_{\text{anneal}}$ (bottom row).

eters are listed in Eq. (6) and result in values of ℓ_0^{AB} , ℓ_0^{AC} , and $\ell_0^{BC} = -0.2kT_{\text{anneal}}$. Here all unlike atom pairs prefer to lie parallel to the magnetization, and the same composition dependence of K_U is expected in the corresponding binary systems,

$$\frac{\ell_{AB}}{kT_{\text{anneal}}} = \frac{\ell_{AC}}{kT_{\text{anneal}}} = \frac{\ell_{BC}}{kT_{\text{anneal}}} = \frac{\ell}{kT_{\text{anneal}}} = -0.1,$$

$$\frac{\ell_{AA}}{kT_{\text{anneal}}} = \frac{\ell_{BB}}{kT_{\text{anneal}}} = \frac{\ell_{CC}}{kT_{\text{anneal}}} = -\frac{\ell}{kT_{\text{anneal}}} = +0.1. \quad (6)$$

A second set of parameters assumed $\ell_{BC}/kT_{\text{anneal}} = +0.15$ with all other interaction parameters defined as in Eq. (6). Here the magnitudes of the ℓ_0^{AB} , ℓ_0^{AC} , and ℓ_0^{BC} parameters are identical, but the B - C pairs would prefer to lie perpendicular to the magnetization direction in the corresponding binary alloy while the A - B and A - C pairs prefer to lie parallel ($\ell_0^{AB} = \ell_0^{AC} = -0.2kT_{\text{anneal}}$ and $\ell_0^{BC} = +0.2kT_{\text{anneal}}$).

K_U versus composition for three pseudobinary systems corresponding to different fixed atomic ratios of $A:B$ (50:50, 70:30, and 90:10) is shown in Fig. 3. The variation with addition of a ternary alloying element is more complicated, and even the qualitative variation of K_U depends on the interaction parameters in the system. K_U decreases with addition of element C to the 50:50 and 70:30 pseudobinary systems for the first parameter set. K_U increases for all

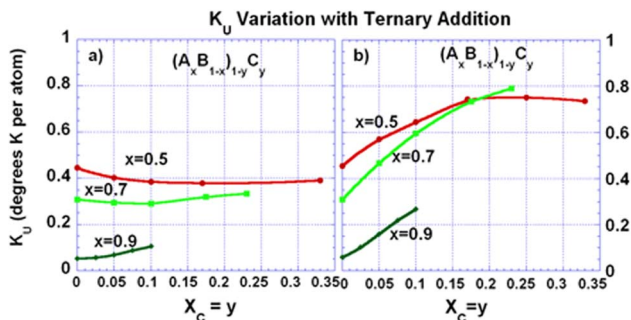


FIG. 3. (Color online) K_U as a function of addition of ternary alloying element C to a binary alloy of fixed $A:B$ atomic ratio for (a) $\ell_0^{AB} = \ell_0^{AC} = \ell_0^{BC} = -0.2kT_{\text{anneal}}$ and (b) $\ell_0^{AB} = \ell_0^{AC} = -\ell_0^{BC} = -0.2kT_{\text{anneal}}$. In all cases the predicted values of K_U for each of the binary systems are identical according to directional pair ordering theory.

pseudobinary systems presented with addition of element C for the second parameter set. For both parameter sets, K_U increases with C content for the 90:10 pseudobinary. This result is reasonable as in the dilute limit of both the second and third elements, the binary alloy relationship $K_U \propto X_C^2$ is expected due to the low probability of interacting B - C pairs. According to these results, one should not necessarily expect K_U to increase in proportion to X_C^2 unless the solid solution is dilute in both the second and third alloying elements. In ternary alloys, where the field induced anisotropy is primarily due to directional pair ordering, prediction of the variation in K_U with third element alloying additions is not so straightforward and the strategy to increase K_U is not obvious without detailed information regarding the system's interaction parameters.

CONCLUSIONS

Monte Carlo simulations reproduce results of the Néel theory of directional pair ordering during magnetic field annealing for ideal binary alloys. The extension to ternary systems shows that even for an ideal ternary solid solution, K_U does not necessarily increase with ternary element alloying additions. Unlike the case for binary alloys, the variation of K_U with atomic fraction of a third alloying element depends on the relative values of ℓ_{ij} 's in the system. However, when both the second (element B) and third (element C) alloying elements are dilute, the compositional dependence of K_U is still expected to vary as $K_U \propto X_C^2$ because of the low probability of finding B - C pairs in the alloy. We conclude that the common strategy of adding an additional atomic species to enable directional pair ordering through formation of a binary alloy does not straightforwardly extend to the general ternary case.

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- ¹L. Néel, J. Phys. Radium **15**, 225 (1954).
- ²S. Chikazumi, *Physics of Magnetism* (Wiley, New York, 1964).
- ³E. T. Ferguson, J. Appl. Phys. **29**, 252 (1958).
- ⁴M. Takahashi and T. Kono, J. Phys. Soc. Jpn. **15**, 926 (1960).
- ⁵H. Katada, T. Shimatsu, H. Watanabe, I. Watanabe, H. Muraoka, Y. Nakamura, and Y. Sugita, IEEE Trans. Magn. **38**, 2664 (2002).
- ⁶S. Chikazumi and T. Oomura, J. Phys. Soc. Jpn. **10**, 842 (1955).
- ⁷E. A. Nesbitt and R. D. Heidenreich, J. Appl. Phys. **30**, 1000 (1959).
- ⁸G. Herzer, Mater. Sci. Eng., A **181-182**, 876 (1994).
- ⁹K. Suzuki, N. Ito, J. S. Garitaonandia, and J. D. Cashion, J. Appl. Phys. **99**, 08F114 (2006).
- ¹⁰K. Binder and D. W. Heermann, *Monte Carlo Simulation in Statistical Physics* (Springer-Verlag, Berlin, 1992).
- ¹¹D. P. Landau and K. Binder, *A Guide to Monte Carlo Simulations in Statistical Physics* (Cambridge University Press, Cambridge, 2005).
- ¹²T. Iwata, Sci. Rep. Res. Inst. Tohoku Univ. A **A13**, 137 (1961).