Pergamon

Scripta Metallurgica et Materialia, Vol. 30, No. 6, pp. 683–688, 1994

0956-716X/94 $6.00 + .00

VIEWPOINT SET No. 22

THE STRUCTURE AND PROPERTIES OF $L1_0$ ORDERED FERROMAGNETS: Co-Pt, Fe-Pt, Fe-Pd AND Mn-Al

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(Received July 28, 1993)

Introduction

An interesting class of ferromagnets from the structure-properties point of view derives from the formation of the $L1_0$ superstructure in ordering systems such as the Co-Pt, Fe-Pt, Fe-Pd family and the MnAl-base alloys. In the Co-Pt family the $L1_0$ phase shown in Figure 1 derives from an A1(fcc)$\rightarrow L1_0$ ordering transformation which occurs via a nucleation and growth process (1) whereas in the MnAl-type alloys the $L1_0$ phase stems from a shear or displacive transformation within an ordered (B19) orthorhombic phase (2). In both instances the resultant $L1_0$ (CuAuI-type) superstructure exhibits a strong uniaxial magnetocrystalline anisotropy with $K_1 \sim 10^7$-10^8 ergs/cm^3 (3). The ferromagnetic (τ) phase in the MnAl-base materials is metastable and the stability of the τ-phase is a major concern in the thermomechanical processing and heat treatment of permanent magnets based on this system. The addition of carbon is very effective as a stabilizer of the ferromagnetic τ phase against decomposition to the equilibrium γ_2 and β phases. Thus, commercial permanent magnets are essentially based on the Mn-Al-C ternary system (4).

Importantly, the uniaxial $L1_0$ phase with an "easy" c-axis forming in the Co-Pt, Fe-Pt, Fe-Pd, and Mn-Al-C permanent magnet materials inherits a defect structure comprised primarily of a high density of extended planar faults, viz. twins, APB's, and stacking faults (5,6). The microtwins modulate the easy direction of magnetization and crystallographically impose a spin transition across the twin plane. In the Co-Pt family the transformation twins are conjugated along the {110} planes whereas in the Mn-Al-C ferromagnets the twins are along the {111} planes of the τ-phase using a conventional fcc reference cell (5,6,7). It has been shown that the easy axis rotates by 90° in going from one microtwin to another in the magnetically modulated structure which forms in the CoPt-type alloys (10). The APB's represent a special situation magnetically in the MnAl-base alloys because the perturbation of the atomic order in the vicinity of the fault alters the nearest-neighbor distances of the Mn atoms producing a local antiferromagnetic coupling (8,9). This causes the APB's to act primarily as nucleation centers for reverse domains during magnetization and demagnetization. The APB's which develop in Co-Pt, Fe-Pt, and Fe-Pd alloys result from the impingement of the growing $L1_0$ particles within the disordered parent phase during the ordering transformation and these APB's appear to act primarily as pinning centers for migrating domain walls within the microtwin ensembles characteristic of these materials (10). However, in both classes of $L1_0$ ferromagnets, the interaction of the planar faults with the processes of magnetization and demagnetization is at the core of understanding the relationship between material structure and magnetic properties.

In this short paper a simple framework based on domain wall pinning is discussed to provide a semi-quantitative basis for understanding the relationship between structure and properties in the Co-Pt, Fe-Pt, Fe-Pd, and Mn-Al-C permanent magnet materials. In addition, some recent Lorentz microscopy studies of the unique domain configurations which emerge in the polytwinned Fe-Pt and Fe-Pd alloys will be presented which relate to the mechanism of coercivity in these ordered ferromagnets. Although the Co-Pt family of alloys will never become large volume permanent magnet materials they still have great potential for thin film applications including perhaps

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magnetic recording as well as in the expanding area of miniaturization. The Mn-Al-(C) permanent magnets still have great potential commercially but various problems related to processing as well as a need to enhance intrinsic properties curtail their employment in technical applications (3,4).

The experimental details are contained in the various papers cited in the analysis and discussion.

Coercivity of the Faulted and Twinned $L1_0$ Ordered Ferromagnets

These permanent magnet materials typically exhibit coercivities in the range 0.5-5.0 kOe and as cited above the nature of the planar faults and twins characteristic of these alloys almost certainly govern their properties. In the Co-Pt, Fe-Pt, and Fe-Pd alloys, APB pinning within the faulted twin lamellar appear to control the magnetic hardness, whereas in the Mn-Al-(C) ferromagnets the coercivity most likely stems from the interaction of moving domain walls with the array of stacking faults which develop in the τ phase. The interaction of migrating 180° Bloch walls with the APB's and stacking faults can be estimated from the following equation:

$$H_c^{max} \sim \frac{K\omega}{2M_s\delta}\left[\frac{A}{A'}-\frac{K'}{K}\right] \tag{1}$$

Where K is the anisotropy constant, M_s is the saturation magnetization, and A is the exchange constant of the host material: δ is the characteristic wall thickness given by $\delta = \sqrt{A/K}$ for a uniaxial ferromagnet. The parameter ω is the effective thickness of the planar defect or magnetic heterogeneity ($\omega \ll \delta$) and K' and A' are the perturbed values of the anisotropy and exchange constants, respectively, within the fault. This equation has been derived by several investigators (8,11-13) using somewhat different approaches over the years and essentially represents an upper bound to the coercivity attainable through pinning by the extended faults to within a factor of about 2 or better. the coercivity of a real material, H_c, is expected to include a statistical factor, α_f, as follows:

$$H_c \sim \alpha_f \frac{K\omega}{2M_s\delta}\left[\frac{A}{A'}-\frac{K'}{K}\right] \tag{2}$$

where α_f is essentially the average areal fraction of the unit wall area which contains faulted material as it moves through the array of obstacles under the influence of the applied field. In the $L1_0$ ferromagnets under discussion it is a good approximation to take $K' \ll K$ in Equations [1] and [2].

Taking values of the various parameters from the extensive literature on these materials and assuming $A' \sim A$ and $\omega \sim 4\times10^{-8}$cm gives for APB and stacking fault pinning the values of H_c^{max} shown in Table I.

TABLE I
Estimated Values of the Coercivity (Upper Bound)

	K(ergs/cm^3)	M_s(emu/cm^3)	δ(Å)	H_c^{max}
Mn-Al-(C)	1x10^7	500	100	0.5
Co-Pt	5x10^7	555	70	3.5
Fe-Pt	7x10^7	450	40	8.5
Fe-Pd	3x10^7	1100	60	1.2

The relatively low value of H_c^{max} for Mn-Al-(C) predicted by Equation [1] is interesting. This value is lower by a factor of 5 or more than the observed coercivities in Mn-Al-(C) permanent magnets (4). It is suggested here that the term A/A' in Equation [1] and [2] is more important in these materials than in the others and contributes significantly to the strength of the pinning by extended planar faults in MnAl-type materials. The relatively high value of H_c^{max} for the Fe-Pd alloys compared to the measured coercivities almost certainly is rooted in the α_f term (10). The Fe-Pd ferromagnets generally show coercivities ~ 500 Oe, less than 50% the value H_c^{max}. The Co-Pt and Fe-Pt ferromagnets generally exhibit values of Hc ~ 1-5kOe (3).

It is concluded here that the coercivity of the $L1_0$ ordered ferromagnets is generally controlled by wall pinning by planar faults or extended magnetic heterogeneities and that Equations [1] and [2] are useful for semi-quantitatively understanding the relationship between structure and properties in these materials.

Domain Structures and Polytwinned Fe-Pt and Fe-Pd Alloys

Several interesting studies of the magnetic domain structure of Mn-Al-(C) ferromagnets have appeared in the literature over the past decade or so (14,15). However, in spite of the extensive discussion of the domain structures expected in the Co-Pt, Fe-Pt, and Fe-Pd equiatomic ordered alloys particularly in the former Soviet Literature (16-18), no comprehensive experimental study of the magnetic domain structures had been carried out until recently (10) and some of these new Lorentz microscopy results will be presented here. Figure 2 shows an electron micrograph revealing the profuse microtwinning and macrotwinning characteristic of a fully ordered Fe-Pd or (Fe-Pt) ferromagnet. Also, a dense array of APB's within the macrotwin plates show diffraction contrast. This assembly of large macrotwin plates subdivided into finer microtwins conjugated along the {110} planes of the parent cubic matrix (A1) is typical of the polytwinned structures exhibited by the Co-Pt family in the fully ordered state. This fine-scale twinning is expected to have a profound effect on the magnetic domain structure which develop in these ferromagnetic materials. The APB's within the microtwins are shown more clearly in Figure 2b. After prolonged aging the polytwinned structure coarsens under the mutual influence if the elastic stain energies of the coherent twin aggregates and large regions dominated by two variants emerge. Also, during aging there is a coarsening and slight reduction of the APB density within the plates (1).

Figure 3 shows a characteristic array of macrodomains cutting across a macrotwin plate in the polytwinned $L1_0$ phase of an equiatomic Fe-Pd alloy. These macrodomains traverse many microtwin plates contained within the large plate. This is revealed clearly in Figures 5a-d which exhibit the salient features of the magnetic domain structures which emerge in the polysynthetically twinned microstructures. In Figures 4a-d, four macrotwin boundaries are observed; two are "frozen" on the macrotwin boundaries and two serrated walls are seen cutting across the assembly of microtwin plates comprising the macrotwin twin plates or bands. The thin lines of dark and light contrast are coincident with the traces of the (101) and (011) microtwin boundaries. The thick lines of contrast are walls associated with the crystallographical imposed spin transition. The serrated or kinked walls are the 180° macrodomain walls cutting across the array of microtwin lamellae which are mobile and expected to play the primary role in magnetization reversal. Various models including mixtures of Néel and Bloch segments have been proposed to describe the nature of the "frozen" walls at the macrotwin boundaries (16-18,19).

As discussed above, magnetization reversal in the polytwinned Co-Pt, Fe-Pt, and Fe-Pd polytwinned ferromagnets is most likely controlled by the motion of 180° macrodomain walls located within the macrodomain plates and their interaction with the array of APB's within the plates. These mobile wall segments extending across an assembly of microtwins are clearly shown in Figures 5a-d using the Fresnel and Foucault imaging modes. Figures 6a and b show a similar set of walls crossing the twin lamellae.

The domain wall thickness characteristic of the fully ordered state in the Fe-Pd alloy was measured within the microtwin plates using the divergent wall thickness method (20) giving a value of ~ 60 Å which is in excellent agreement with the calculated wall thickness $\delta = \sqrt{A/K}$.

Summary

1. The analysis of the Co-Pt, Fe-Pt, Fe-Pd, and Mn-Al-(C) alloys presented here suggests that the salient technical magnetic properties and the relationship between structure and properties of these permanent magnet materials can be understood semi-quantitatively in terms of pinning by extended planar defects or magnetic heterogeneities.
2. The APB's in the Mn-Al-(C) ferromagnets act primarily as centers for nucleation of reverse domain structure and domain wall pinning in these materials stems from the stacking faults which arise in the τ-phase.
3. The change in the exchange parameter within the stacking faults of the Mn-Al-(C) ferromagnets most likely contributes significantly to the pinning strength of these magnetic heterogeneities.
4. The APB's in the Co-Pt, Fe-Pt, and Fe-Pd alloys act as pinning sites which control the magnetic hardness and hysteresis of these polytwinned ferromagnets.
5. Lorentz microscopy has revealed the characteristic domain structures which develop in the Co-Pt, Fe-Pt, and Fe-Pd ferromagnets. The magnetic domain structure is consistent with the model of coercivity controlled by

Acknowledgements

This work is supported by the U.S. Department of Energy (DOE), Office of Basic Energy Sciences, Division of Materials Science through the Department of Materials Science and Engineering at the University of Pittsburgh.

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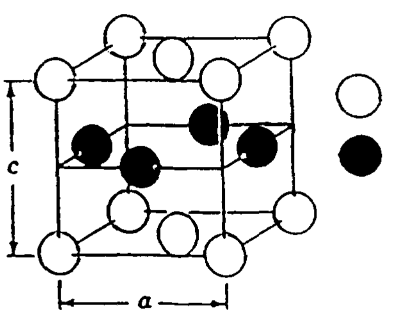

FIG. 1. The unit cell of the ordered tetragonal $L1_0$ phase.

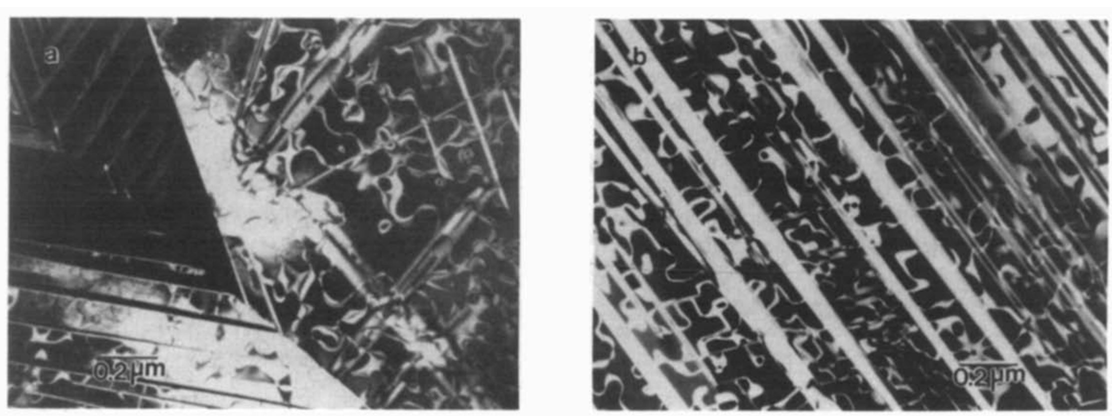

FIG.2. Microtwinning and macrotwinning and APB's in an Fe-Pd alloy aged 61 hrs. at 500°C.

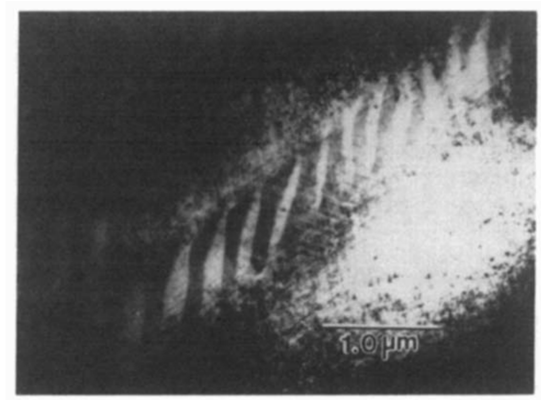

FIG. 3. Macrodomains terminating on the twin band boundaries.

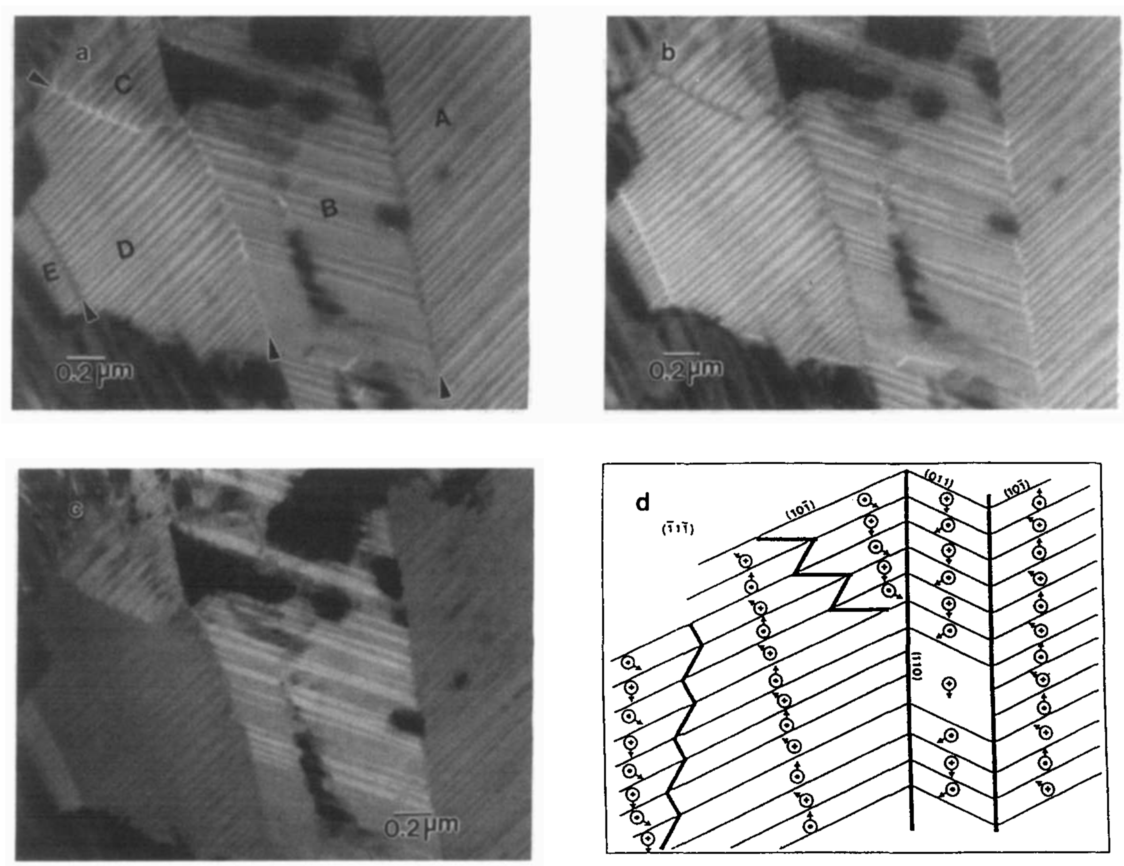

FIG. 4. Macrodomain and microdomain structures and the domain walls: a) Fresnel under-focus; b) Fresnel over-focus; c) Foucault in-focus images; d) the schematic analysis of the domain configuration revealed in a), b) and c).

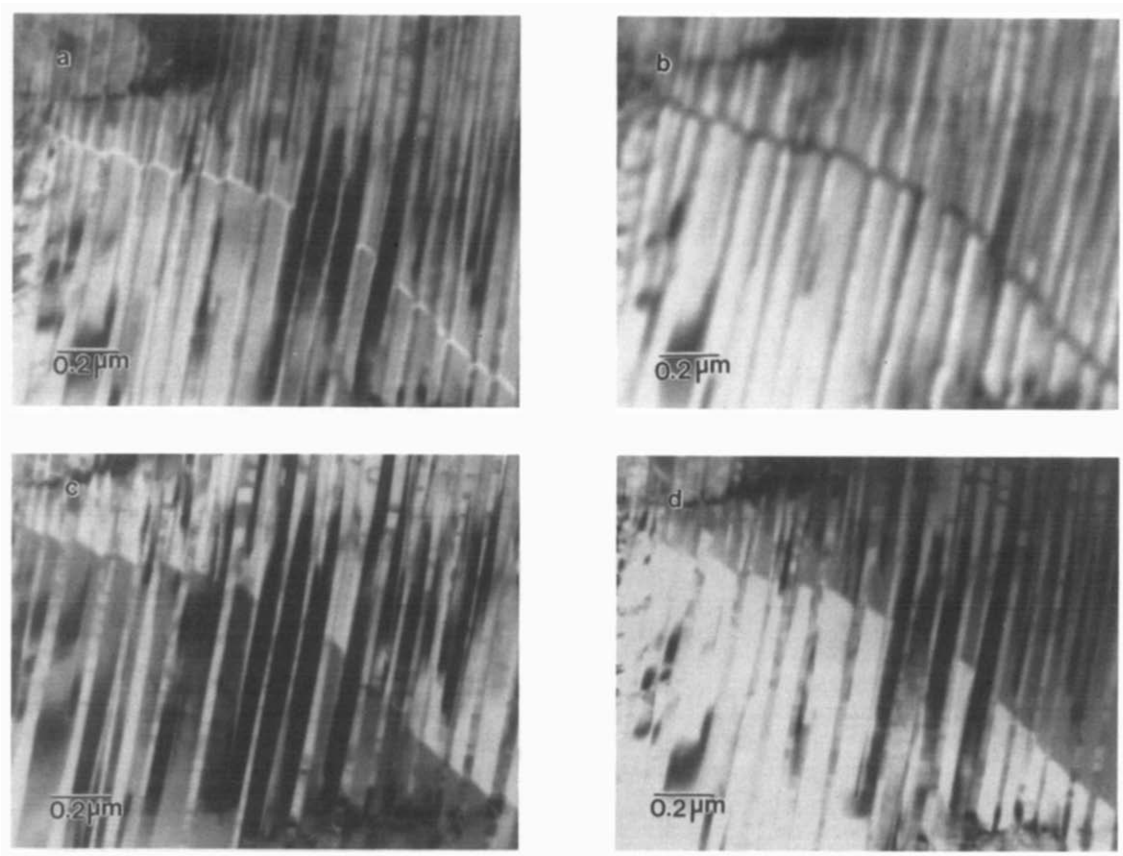

FIG. 5. Macrodomain wall cutting across the microtwin plates and showing the kinked feature: a) Fresnel under-focus; b)Fresnel over-focus; c) and d) in-focus Foucault images with the aperture displaced in opposite directions.

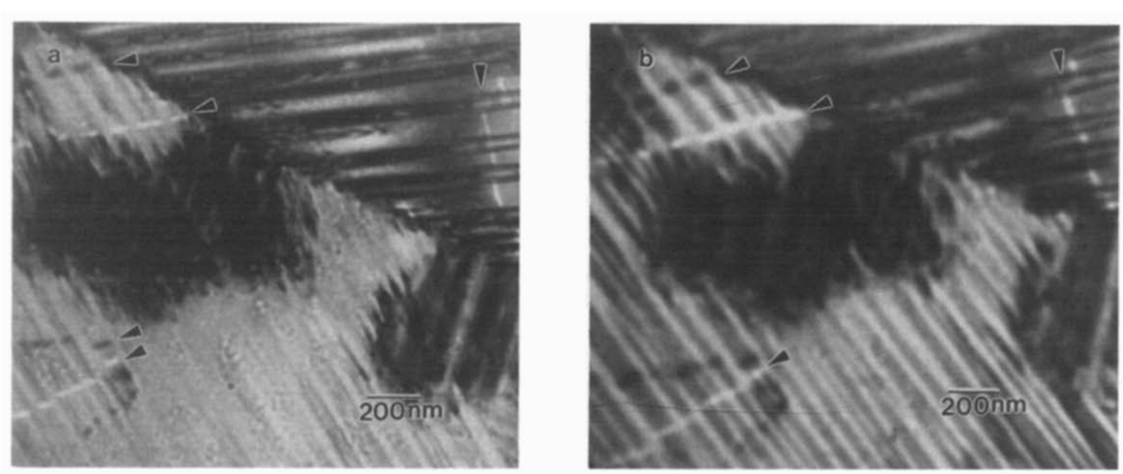

FIG. 6. Macrodomain walls cutting across the microtwins moved from a) to b) due to the change of local magnetic field.