Temperature-induced martensite in magnetic shape memory Fe_2MnGa observed by photoemission electron microscopy

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Abstract

The magnetic domain structure in single crystals of a Heusler shape memory compound near the composition Fe_2MnGa was observed during phase transition by photoelectron emission microscopy at Beamline 11.0.1.1 of the Advanced Light Source. The behavior is comparable with recent observations of an adaptive martensite phase in prototype Ni_2MnGa, although the pinning in the recent work is an epitaxial interface and in this work the effective pinning plane is a boundary between martensitic variants that transform in a self-accommodating way from the single crystal austenite phase present at high temperatures. Temperature dependent observations of the twinning structure give information as to the coupling behavior between the magnetism and the structural evolution.

I. INTRODUCTION

The shape memory effect is the ability of a material to repeatedly recover large plastic deformation and has applications including medicine, robotics, and active or passive vibration damping [1]. The first known and best studied magnetic shape memory (MSM) compound is Heusler Ni_2MnGa [2], and further examples are being demonstrated: Ni_2FeGa [3], Mn_2NiAl [4], and others. Magnetic field actuation overcomes some of the speed and response drawbacks of thermally actuated shape memory so the coupled magnetostructural transitions in Ni_2MnGa have led many researchers to focus on it as a prototypical MSM compound.

Several important differences manifest in the electronic structure and transport behavior of Fe_2MnGa. Stoichiometric Ni_2MnGa exhibits magnetically as well as thermally driven shape memory based on a self-accommodating tetragonal to cubic transition at 202 K with $c/a = 0.94$ associated with a very slight (about 1%) shrinking of the unit cell. In contrast the c/a ratio of the equilibrium low temperature phase in stoichiometric Fe_2MnGa is 0.927 and the transformation is associated with a volume change of +1.35% [6]. Annealed off-stoichiometric polycrystals ($Fe_{50}Mn_{22.5}Ga_{27.5}$) were recently shown to have a dramatic

magnetic-field induced strain with a theoretical performance up to $(c-a)/c = 33.5\%$ [7]. This dwarfs the theoretical strain in Ni_2MnGa (about 10%, [8]) but the magnetic structure and predicted half-metallic behavior have not yet been investigated.

Recent scanning tunneling microscopy on sputter cleaned surfaces of Ni_2MnGa epitaxial films led to the proposal of a theory of adaptive martensite in that material, changing the earlier conception of "modulated" phases [9]. This work demonstrates the applicability of the theory for other MSM compounds despite electronic and structural differences and shows that geometric corrugations associated with twinning in the proposed adaptive martensite phase [5] are thermally actuated.

II. EXPERIMENTAL DETAILS

A single crystal of $Fe_{48}Mn_{24}Ga_{28}$ with faces parallel to $\langle 100 \rangle_{aus}$ directions, martensitic start and finish temperatures $M_s = 213\ K$ and $M_f = 162\ K$, austenitic start and finish temperatures $A_s = 231\ K$ and $A_f = 294\ K$ (thermal hysteresis of about 80 K) was obtained by grain growth and annealed for 168h as 1273 K as previously described [6]. The sample surface is polished *ex situ* with diamond paper down to 0.1 μm and transferred to UHV.

Photoemission electron microscopy (PEEM) was performed on soft x-ray elliptically polarized undulator (EPU) beamline 11.0.1.1 at the Advanced Light Source. Element-specific magnetic contrast in a PEEM image is found by dividing two images taken with opposite helicities of incident photons. Surface domains reflect the distribution of volume domains in materials with high coupling between the magnetization and the magnetocrystalline anisotropy as in this case [10]. Therefore, surface-sensitive domain observation is informative regarding the volume behavior.

III. RESULTS AND DISCUSSION

Ambient temperature (300 K) magnetic domains are displayed before cooling in Figure 1 where the field of view of 36 μm was chosen to encompass several variants. Purely topographic information was relatively featureless. Region boundaries are well defined by

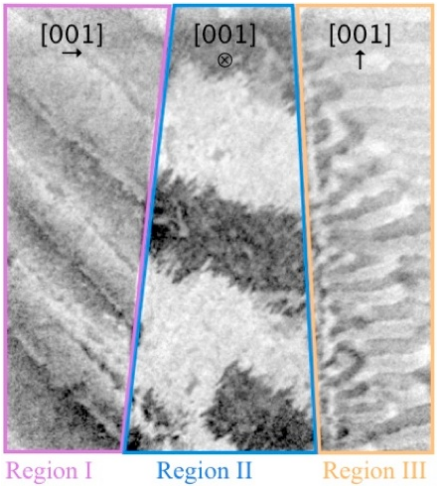

FIG. 1: Photoemission micrograph of the polished crystal surface. The field of view is 36 μm and the contrast comes from dividing two images taken at the Fe L_3-edge (707.5 eV) in positive and negative circular polarization. Variants can be assigned based on morphology.

abrupt changes in the domain structure across the twins. All observable magnetic contrast is carried by the iron and none by the manganese, superficially opposite to what is predicted from analogy with Ni_2MnGa where it is known that Mn^{3+} carries the majority of the total highly localized bulk moment of 3-4 μ_B [11]. However, PEEM is a surface-sensitive technique and the higher reactivity of the manganese atoms and the *ex situ* polishing together mean that the electronic structure of the top 2 nm of manganese has been altered. Indeed, (equally surface sensitive) absorption spectroscopy indicates strongly localized character as in Mn^{2+}. The less reactive iron preserves the surface expression of the deep magnetic domain structure.

We refer to the Region I as having "band" morphology and to Region II as being "maze-like" in order to be consistent with the language of Lai and colleagues [10]. This coincides with structural variants with the local c-axis ([001]-direction) pointing in-plane (left-right) and out-of-plane, respectively, with the magnetization axis M aligned at all times with the axis of tetragonal distortion. The morphology of Region III is consistent with in-plane (up-down) domain alignment.

Upon cooling (Figure 2), the banded Region I expands at the expense of maze-like Region II, so there must be a symmetrically compatible plane between the two twinned variants, allowing the plane to be mobile with the driving force of temperature. Re-heating to an

intermediate temperature shows a hysteresis in the regrowth of Region II by motion of the same wall, consistent with both a large structural deformation and thermal hysteresis. The nearly square area enclosed by the curve indicates the large energy consumed in a cycle.

The region of interest for the heating transition is boxed in Figure 2 and several frames of the thermal evolution of this region are arranged in Figure 3. A magnetic line feature (marked with an arrow) with no associated topographic contrast was observed to nucleate in Region III and travel toward the heretofore immobile boundary with Regions II. The result of this is a doubling of the line frequency of the affected magnetic domains from approximately one repeat per 4 μm to one repeat per 2 μm and leaving Region III in a wrinkled state as in Figure 4. This surface-specific "branching" reduces the magnetostatic energy and is consistent with the formalism for an adaptive martensite outlined for a pinned, twinned microstructure near a rigid interface [5]. In the original publication the rigid interface was an epitaxial substrate but here it is a variant plane that is immobile due to the broken symmetry of the surface, indicating that the presence of the surface changes the nature of the boundary between the martensitic variants. No further evolution above 231 K was observed.

The evolution of the magnetic domain structure in single crystals of an MSM Heusler compound near the composition Fe_2MnGa supports the proposed adaptive martensite phase formerly only discussed for Ni_2MnGa despite significant electronic and structural differences between the compounds.

Acknowledgements

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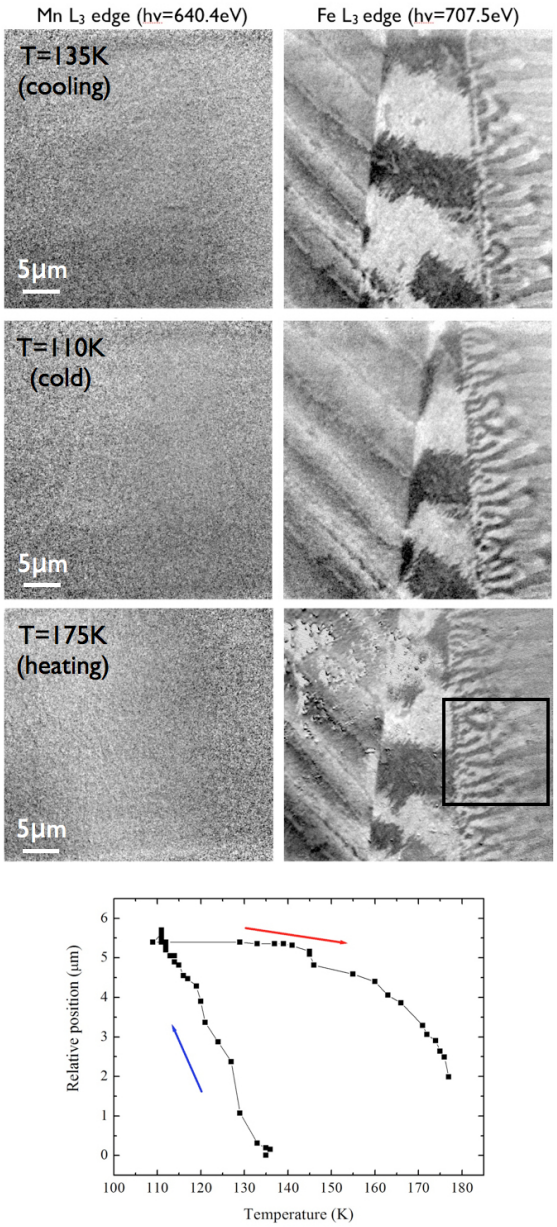

FIG. 2: Manganese (left) and iron (right) L_3-edge contrast images from cooling and reheating. Mn exhibits zero magnetic contrast in this surface-sensitive technique. The boundary between Regions I and II is highly mobile and the boundary between Regions II and III is pinned by symmetry. The lower plot is the relative position of the mobile domain wall with cooling (blue arrow) and then heating (red arrow).

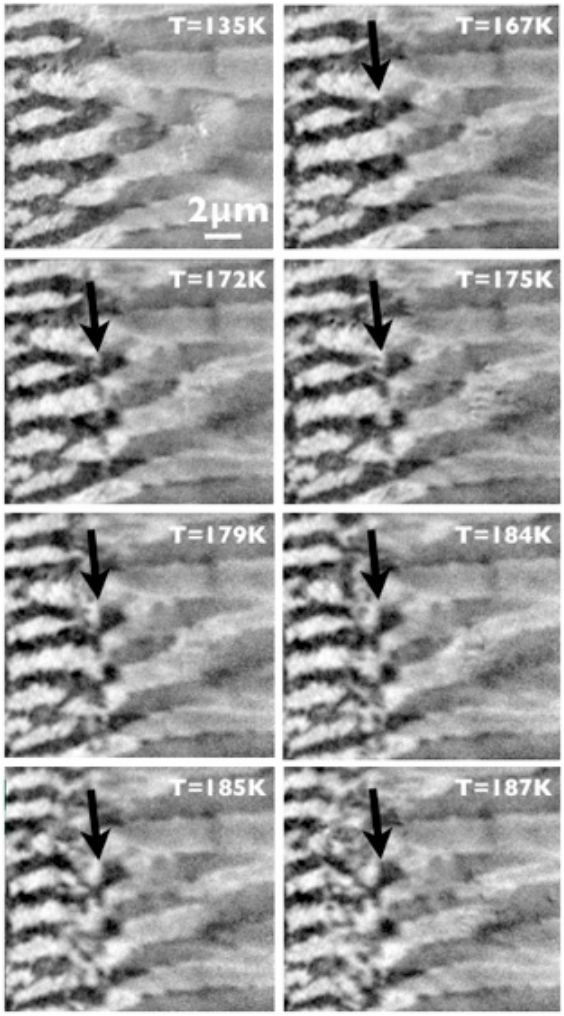

FIG. 3: With heating, a magnetic domain is nucleated which reduces the magnetostatic energy at the surface as it propagates. The original formalism was a martensite in contact with an epitaxial substrate but here the rigid boundary condition is a pinned twin boundary.

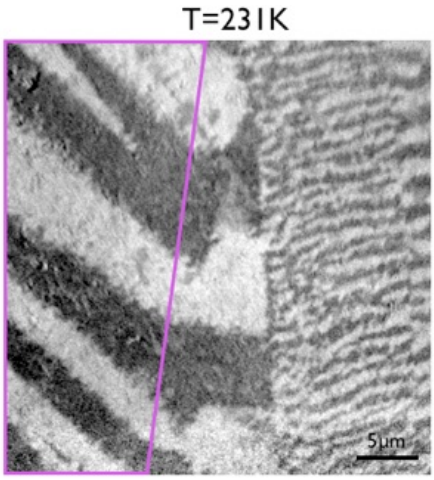

FIG. 4: After heating, Region I has transformed from band domains to maze domains in a swept first order transition. Region III has undergone line frequency doubling (branching) to minimize the magnetostatic energy.