Materials and Design 140 (2018) 1–6

Contents lists available at ScienceDirect

Materials and Design

journal homepage: www.elsevier.com/locate/matdes

Evidence for lattice softening of the Fe-Ga magnetostrictive alloy: Stress-induced local martensites

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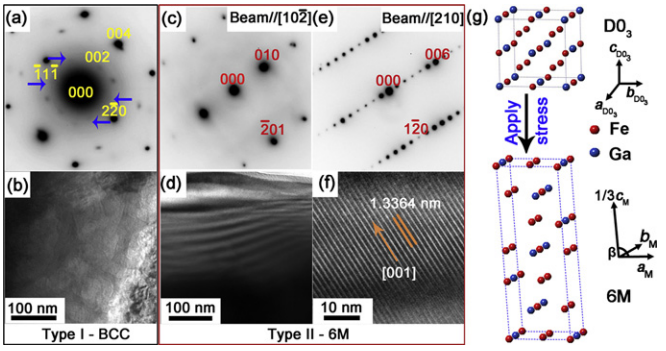
HIGHLIGHTS

- Local monoclinic martensite with six-layer modulation (6 M) is found in polycrystalline Fe-Ga.
- The low energy barrier between $D0_3$ and 6 M Fe-Ga allows martensitic transformation by applying stress.
- As a new evidence of lattice softening, stress-induced 6 M martensite helps to understand large magnetostriction in Fe-Ga.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 10 July 2017
Received in revised form 17 October 2017
Accepted 17 November 2017
Available online 20 November 2017

Keywords:
Fe-Ga alloy
Magnetostriction
Martensites

ABSTRACT

The over tenfold magnetostriction enhancement by soluting nonmagnetic Ga into body centered cubic (BCC) Fe has been ascribed to crystal lattice softening, which indicates that the Fe-Ga solid solution lies in the crossover of martensitic transformation. Identifying the martensitic phase then becomes essential. Here we report the stress-induced local six-layer modulated monoclinic (6 M) martensites in a $Fe_{79}Ga_{21}$ alloy solution-treated at 1373 K. Although X-ray diffraction (XRD) and magnetic measurements reveal that the bulk sample exhibits BCC average structure, the transmission electronic microscopy (TEM) characterizations however show that the mechanically grinded foil sample contains local 6 M phase. First principal calculations suggest that low formation energy barrier between the ordered $D0_3$ and the 6 M phases are responsible for the stress-induced structural transformation. Our work provides evidence for the lattice softening of Fe-Ga solid solution, adding important insight into understanding the origin of its extraordinary magnetostriction enhancement.

1. Introduction

Magnetostrictive materials that can generate elastic strain under external magnetic fields are attractive in applications of sensors, actuators and transducers. The search for ductile and highly magnetostrictive material with low switching field has stimulated considerable efforts over time. A landmark material is Fe-Ga solid solution, for which the

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https://doi.org/10.1016/j.matdes.2017.11.036
0264-1275/

magnetostriction along [001] can be as high as 3/2 $\lambda_{001} = 400$ ppm at low saturation fields [1,2], being tenfold higher than the non-soluting host, BCC Fe. Previous studies [3–14] of $Fe_{100-x}Ga_x$ (at.%) alloys mainly focus on the compositions with x in the vicinity of 19 and 27, at which two anomalous magnetostriction peaks have been observed [2], respectively. The sensitivity of magnetostriction to thermal history suggests that this material is structurally heterogeneous [3–8] and it has been recently reported that the magnetostriction is even non-volume-conservation [14]. These facts suggest that the large magnetostriction in Fe-Ga is difficult to explain by domain rotation mechanism for a conventional homogeneous system. Consequently, understanding the origin of such extraordinary magnetostriction enhancement by soluting nonmagnetic Ga has attracted extensive interest since its discovery in 2000.

Despite of the controversy between the intrinsic electronic structure [15,16] and the extrinsic nanoscale structural heterogeneity [6–8, 17–19] caused by Ga addition, macroscale measurements including continuous-wave [2,3,20] and inelastic neutron scattering [21,22] have shown that Fe-Ga system possesses large magnetoelastic coupling constant b_1 and small shear elastic constant $1/2(C_{11}–C_{12})$. Therefore, the large magnetostriction can be understood in terms of the crystal lattice softening and an associated variation of the magnetoelastic coupling, as suggested by Wuttig et al. [3]. The fact that $1/2(C_{11}–C_{12})$ decreases linearly with the increase of Ga content ($0 < x < 26$) is analogues to the scenario of martensitic transformation, shear elastic constant decreases when cooling approaching the martensitic transformation start temperature, M_S [23]. It indicates that the Fe-Ga solid solution contains nanosized embryos of a martensitic phase, i.e. this system could be within a crossover region that is very close to the martensitic transition. Yasuda et al. reports the 14 M martensite by applying tensile stress to the $D0_3$-structured $Fe_{76.2}Ga_{23.8}$ single crystal [24]. An early paper on $Fe_{81}Ga_{19}$ single crystal also reports that extra tetragonal reflections appear in the XRD pattern, which has been ascribed to the distortion of the $D0_3$ phase [25]. Although the tetragonal phase could be caused by surface damage during mechanical grinding [5], it indeed demonstrates that lattice of the $D0_3$ phase is soft, indicating that it could be distorted into a lower symmetry phase by applying mechanical stress. The martensitic transformation may happen locally in polycrystalline Fe-Ga alloys due to the fact that they contain high concentration defects. For instance, disordered A2 phase always coexists with the $D0_3$ phase even at the quenched state [5,11,12,26–28], anti-phase domain boundaries (APBs, two-dimensional defects) could also exist even within $D0_3$ single crystals [29]. The defects will suppress the spontaneous structural transformation suppose it could happen for the $D0_3$ phase. In addition, the reported structure of martensite is still in controversy. Consequently, identifying the martensites within polycrystalline samples is a fundamental question because it would help to understand the soft lattice nature as well as the large magnetostriction of Fe-Ga alloys.

In this work, we selected polycrystalline $Fe_{79}Ga_{21}$ within the metastable (A2 + $D0_3$) two-phase region [26] to study. Although a BCC average structure for the bulk sample is detected by X-ray diffractometer, local micro-sized 6 M domains are observed in the mechanically grinded foil sample by TEM. First-principle calculations reveal that the monoclinic phase has quite low formation energy difference from the ordered $D0_3$ phase. The stress-induced martensites demonstrate the soft lattice nature of Fe-Ga solid solution, adding important insight into understanding the microstructural origin of the extraordinary magnetostriction enhancement.

2. Experimental procedures

Polycrystalline $Fe_{79}Ga_{21}$ sample was prepared by induction melting and subsequent hot rolling. Sheet specimen (~2 mm thick) was solution-treated for 3 h at 1373 K and finally quenched into ice-water. After careful mechanical polishing and etching with Nital solution, the specimen was subjected to a Rigaku X-ray diffractometer with Cu *K*α radiation to detect the average crystal structure. Small bulk with the size of $2 \times 2 \times 2$ mm^3 was cut from the sheet after XRD characterization and subjected to magnetic measurements using a Lakeshore 7400 vibrating sample magnetometer (VSM) and a physical property measurement system (Quantum Design, PPMS-9T), respectively. The specimen for TEM observation was also cut from the same sheet. Foil was prepared by standard grinding and mechanical polishing (thickening to 50 μm), followed by twin-jet electropolishing at temperatures below 243 K. Before setting to the JEM-2100F microscope, the foil was further ion-milled for 15 min (beam glancing angle = 8°, beam voltage = 3 kV, and beam current = 5 mA) to remove surface contaminations. To evaluate the total energy/unit-cell of different structures, first-principles calculations were performed using density functional theory (DFT) from Vienna ab initio simulation package with a plane wave basis [30]. We employed generalized gradient approximation (GGA) for exchange-correlation function [31]. The core electrons are represented by the projector-augmented-wave (PAW) potential [32]. Kinetic energy cutoff is set above 400 eV and k-point sampling on the unit cell is $8 \times 8 \times 8$. The structure optimization process is performed with force convergence criteria at 0.01 eV/Å.

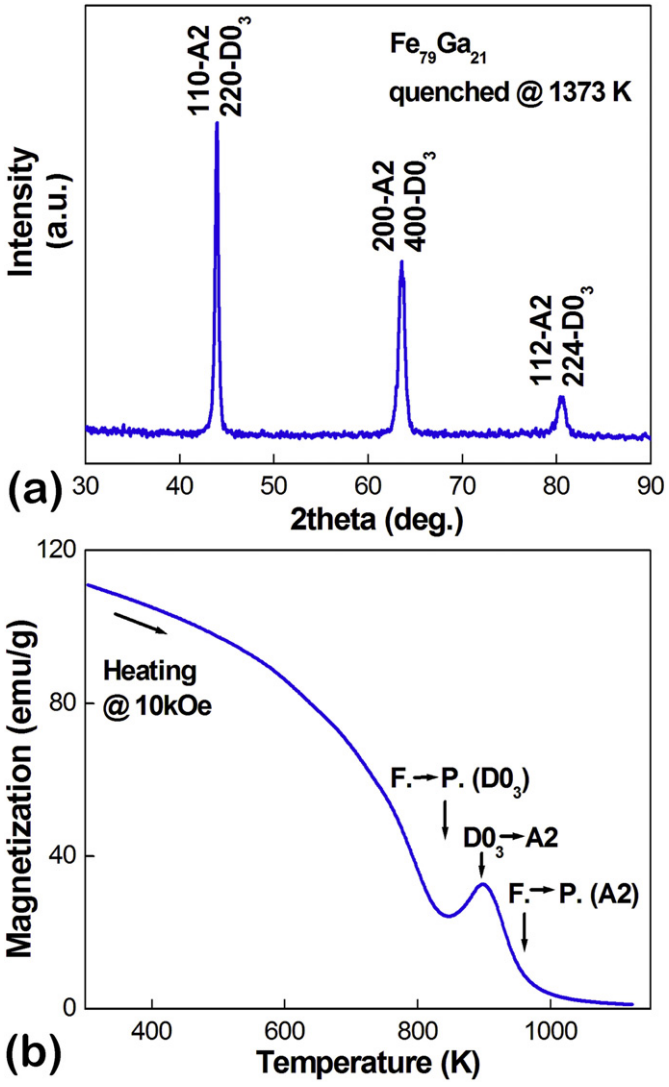

Fig. 1. Characterizations of $Fe_{79}Ga_{21}$ bulk sample. (a) XRD profiles obtained from the polished surface after Nital etching, revealing an average BCC structure. (b) *M*–*T* curve measured upon heating under a constant field of 10 kOe, exhibiting two ferromagnetic (F.) to paramagnetic (P.) transitions and an order to disorder transition, respectively.