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Phase transitions as a tool for tailoring magnetostriction in intrinsic Fe-Ga composites

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ABSTRACT

Fe-Ga alloys exhibit unique functional properties such as magnetostriction that can be varied from the highest positive values among iron-based alloys to negative values including zero magnetostriction, if proper compositions and heat treatments are chosen. This remarkable behavior is related to rather complex phase transformation sequences in this alloy family that are still unresolved. In earlier studies, the phase transformations in Fe-Ga alloys were studied by X-ray diffraction, which provides structural information limited to the near-surface sample area. In this paper, we use electron back-scatter diffraction and *in situ* neutron diffraction to characterize the $D0_3$ and $L1_2$ phases that originate from the fcc and bcc phases in the Fe-27Ga type bulk alloy, respectively. Different ratios between these phases, characterized by magnetostriction values of different signs, were achieved using an isothermal annealing treatment that produces an intrinsic composite in the alloy. Depending on the relative fraction of the $D0_3$ and $L1_2$ phases, the magnetostriction values of the alloy, λ_S, vary from +100 to −50 ppm, including the value of $\lambda_S = 0$ for the alloy with $L1_2$:$D0_3$ = 2:1 achieved after 600 min annealing at 400 °C, thus demonstrating the controlled adjustment of magnetostriction in these advanced alloys.

1. Introduction

Magnetostrictive materials play an increasingly important role in applications ranging from active vibration control, controlled surface deployment, actuators, damping devices, linear motors, positioning devices, and energy harvesting to stress and torque sensing [1,2]. In materials science, galfenol [3] is the general term for an alloy of iron and gallium. Galfenol exhibits rather high magnetostriction (350 ppm) under very low magnetic fields of 100 Oe (8 kA/m) [4]. This alloy also shows a very low hysteresis [5] and demonstrates a high tensile strength (500 MPa) [6,7] and limited variation in magnetomechanical properties for temperatures between −20 and 80 °C [8,9]. Fe-Ga alloys possess a high permeability ($\mu_r > 100$) [3], a high Curie temperature ($T_C > 650$ °C) [10], and are corrosion resistant [11]. Such unique engineering capabilities make galfenol a sustainable alternative to conventional Terfenol-D that suffers extremely from brittleness and low yield stress under shock and tensile loads [12–14].

Previous investigations [15–19] proved that the structure of Fe-Ga alloys is very sensitive to heat treatments. Various structures including disordered bcc A2, ordered bcc B2 and $D0_3$, ordered fcc $L1_2$, and hcp $D0_{19}$ can be obtained when the amount of Ga is ~27 (~26–29) at.%. The interplay between the metastable $D0_3$ and the stable $L1_2$ phases in as-cast Fe-Ga alloys plays a crucial role for the formation of functional properties of Galfenol. Both phases are ferromagnetic but they have different physical characteristics. For example, they show magnetostriction of different signs, different coercive forces, and magnetization [17,20]. Recently, a potential route has been suggested to obtain zero-magnetostriction in Fe-Ga alloys by establishing a controlled ratio between the $D0_3$ and $L1_2$ phases, which is essential for the design of soft magnets [20,21]. For Fe-Ga alloys, samples with A2 and $D0_3$ structures possess positive magnetostriction; whereas samples with $L1_2$ and $D0_{19}$ structures exhibit negative magnetostriction. Compared to the bcc and hcp phases, the $L1_2$ phase has a larger magnetization [16,17]. Following this structure-property relation, one could achieve an intrinsic Fe-

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Ga composite containing both, a bcc phase with positive magnetostriction and an fcc phase with negative magnetostriction [20,21].

The kinetics of the $D0_3$ to $L1_2$ transition in Fe-27Ga under constant heating- and cooling conditions was studied using *in situ* XRD [22–24] and most recently – using neutron diffraction [25–27]. It was shown that during heating with a rate of 2 K/min, the $D0_3$ to $L1_2$ transition occurs in the temperature range of about 50 °C [27]. Obviously, it is nearly impossible in practice to produce controlled amounts of the co-existing phases using linear heating. In order to solve the problem, i.e., to create an alloy with a given ratio between $D0_3$ and $L1_2$ phases, we decided to use isothermal annealing at temperatures slightly below the interval at which this transition occurs during continuous heating. To study the phase transitions, several techniques were involved: scanning electron microscopy equipped with EBSD, *in situ* XRD, and neutron diffraction.

Another important problem intensively discussed in the literature is the mechanism of the transition from the ordered bcc ($D0_3$) to the ordered fcc ($L1_2$) phase. Several atomic mechanisms were proposed:

- A diffusionless displacive bcc → fcc reconstruction was proposed by A.G. Khachaturyan [28,29]: in that model, the atomic order inherited from the $D0_3$ phase corresponds to a transformed tetragonal ($D0_{22}$ structure) lattice and the corresponding Bain strain transforms the bcc lattice further into an fct lattice bringing Fe and Ga atoms to their new positions in an fcc-based lattice without an atomic interchange between these sites.
- In the work by Yin-Chih Lin and Chien-Feng Lin [30], an $Fe_{73}Ga_{27}$ alloy was as-quenched and aged at 700°C for 24 h, the $D0_3$ nanoclusters underwent a phase transformation to an intermediate tetragonal phase ($L1_0$-like martensite) via Bain distortion, and finally $L1_2$ structures precipitated, as demonstrated by TEM and XRD analyses. The $L1_0$-like martensite and the $L1_2$ structures in the aged $Fe_{73}Ga_{27}$ alloy drastically decreased the magnetostriction from positive to negative values, as also confirmed by experimental magnetostriction measurements.
- Mingming Li et al. [31] found that according to the analysis of the results of *in situ* heating, XRD and TEM measurements, the structure transitions of $D0_3$ → A1 → $L1_2$ were identified in the temperature range of 20–500 °C, which correspond exactly to the changes of Young's modulus in as cast and directionally solidified samples.
- In our recent paper [32], it was independently suggested that the $D0_3$ → $L1_2$ transition in bulk and powder-shaped Fe-27Ga alloys is of the order-disorder-order type, i.e., an intermediate disorder state exists between the two order states.

Thus, in this paper, our goal is twofold: first, the type and the kinetics of the $D0_3$ → $L1_2$ transition are analyzed and for the first time explained in detail, and secondly, a new pathway is elucidated to adjust an intrinsic composite structure with a controlled ratio of bcc and fcc phases and, consequently, with controlled magnetostriction, allowing to tune the magnetostriction in accordance with the requirements of a given specific application.

2. Experimental procedure

An alloy with a nominal composition of Fe-27Ga was produced by directional solidification using 99.99% pure Fe and 99.999% pure Ga by induction melting under protective high-purity argon gas using an Indutherm MC–20 V mini furnace. Using energy dispersive spectroscopy, the real chemical composition of the cast button was revealed as Fe-27.8Ga within accuracy of ±0.2% (denoted as Fe-27Ga throughout the text). The crystallographic structure of the samples was examined by X-ray diffraction (XRD) using a Bruker D8 Advanced diffractometer with Cu K_α radiation. The microstructure was characterized using a FEI Nova NanoSEM 230 SEM device. The grain size was estimated from the EBSD maps that were determined with a scan step size of 1 μm and included 1236 grains. Electron back-scatter diffraction was used to identify the bcc (bcc-derivative $D0_3$) and fcc (fcc-derivative $L1_2$) phases and to provide a measure of the residual strain within the grains.

The neutron diffraction (ND) patterns were measured at the high-resolution Fourier diffractometer (HRFD) at the IBR-2 pulsed reactor (JINR, Dubna). The description of the diffractometer can be found in the paper of A.M. Balagurov [33] and with more details in the work of A.M. Balagurov et al. [34]. HRFD is a correlation TOF spectrometer with several detectors placed at fixed scattering angles from 30 to 152°. One of the HRFD features is its possibility to switch easily between high-intensity and high-resolution modes of operation, which is important for *in-situ* studies of changes in both crystal structure and microstructure. If a correlation analysis is used, the HRFD $\Delta d/d$ resolution is determined by the maximum rotational speed of a fast Fourier chopper. In routine operation ($V_{max} = 4000$ rpm), "high-resolution" patterns ($\Delta d/d \approx 0.001$ for $d = 2$ Å) are measured with back-scattering detectors ($2\theta = 152°$, d-spacing range is 0.6–3.6 Å). In parallel, the conventional (without correlation analysis) TOF-patterns of "medium" resolution ($\Delta d/d \approx 0.01$–0.02 up to $d_{hkl} = 4.5$ Å) are measured, with their intensity being about 10 times higher than that of high-resolution patterns.

High-resolution diffraction patterns were measured at room temperature. During isothermal annealing at 410 °C, the medium resolution patterns were measured continuously with 2 min exposure time.

A DualScope C26 magnetic force microscope, MFM (DME Company, Copenhagen, Denmark) was used to obtain the magnetic force gradient image with a typical lift height of 250 nm and a high-moment Co-coated tip magnetized normal to the sample surface. The MFM facility used for this study is equipped with an optical microscope with a magnification of 300×.

The magnetization was recorded using a vibrating sample magnetometer VSM-130 up to 750 °C with a heating rate of 6 K/min and applied magnetic field of ~800 kA/m, and magnetostriction was measured using a custom-designed experimental setup based on a strain gauge method up to a saturation magnetic field value of 40 kA/m, respectively.

3. Experimental results and discussion

The phenomenon of a concomitant transformation of the microstructure and the evolution of ordering was studied using different characterization techniques: scanning electron and magnetic forced microscopy, XRD, and neutron diffraction.

3.1. Scanning electron (SEM) and magnetic forced (MFM) microscopy

The results of energy-dispersive X-ray spectroscopy (EDX) for Fe and Ga show that the distribution of chemical elements in the sample is sufficiently homogeneous for all samples. For all three samples, observations using three different magnifications (the scanned areas with the edges of 30, 100 and 500 μm) were performed:

1) An overview measurement to determine the misorientation angle and grain size distributions;
2) A more detailed measurement to determine the ratio of the fcc/bcc phases;