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Influence of composition and heat treatment on damping and magnetostrictive properties of Fe–18%(Ga + Al) alloys

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Abstract
The structure, magnetostriction and damping properties of Fe82Ga(18–x)Alx (x = 0, 5, 8, 12) alloys were analyzed. The anelastic response of Fe–18(Ga + Al) alloys was studied as a function of temperature (from 0 to 600 °C), frequency (from 0.01 to 200 Hz) and amplitude (from 0.0004% to 0.2%) of forced vibrations. The origin of the relatively high damping capacity of Fe–Ga–Al alloy at room temperature was determined by applying a magnetic field and different heat treatment regimes. The substitution of Ga by Al in Fe–18% Ga alloys was found to decrease magnetostriction and damping. The heat treatment of alloys influences the damping capacity of alloys more than variations of their chemical compositions. Thermally activated frequency and temperature-dependent anelastic effects in Fe–Ga–Al alloys were analyzed and the corresponding activation parameters for relaxation processes were evaluated. Internal friction effects caused by structural transformations were recorded and were found to be consistent with the A2 → D03 → L12 reaction. The physical mechanisms for all anelastic effects are discussed.

Keywords
Fe–Ga–Al alloys, Damping, Magnetostriction, Structure, Ordering

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Influence of composition and heat treatment on damping and magnetostrictive properties of Fe-18%(Ga+Al) alloys

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Abstract. Structure, magnetostriction and damping properties are analyzed in $Fe_{82}Ga_{(18-x)}Al_x$ (x = 0, 5, 8, 12) alloys. Anelastic response of Fe-18(Ga+Al) alloys is studied as a function of temperature (from 0 to 600°C), frequency (from 0.01 to 200 Hz) and amplitude (from 0.0004% to 0.2%) of forced vibrations. The origin of the relatively high damping capacity of Fe-Ga-Al alloy at room temperature is checked by applying magnetic field and different heat treatment regimes. Substitution of Ga by Al in Fe-18%Ga alloys decreases magnetostriction and damping. Heat treatment of alloys influences the damping capacity of alloys more than variations of their chemical compositions.

Thermally activated frequency and temperature dependent anelastic effects in Fe-Ga-Al alloys are analyzed and corresponding activation parameters for relaxation processes are evaluated. Internal friction effects caused by structural transformations are recorded and are consistent with the $A2 \rightarrow D0_3 \rightarrow L1_2$ reaction. Physical mechanisms for all anelastic effects are discussed.

Key words: Fe-Ga-Al alloys, damping, magnetostriction, structure, ordering

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Introduction

In the last decade, the Fe-Ga 'Galfenol' alloys have been thefocus of a lot of attention due to their good mechanical properties and high magnetostriction in low saturation magnetic fields [1-3] and have the potential to be widely used in magnetostrictive actuators and sensors devices. It is believed that the increase in magnetostriction for Fe-Ga alloys is because of the preferential (110) Ga-Ga pairing in the disordered body centered cubic (b.c.c.) structure. It has been found that dependence of the tetragonal magnetostriction with Ga content, 'λ_{100} *vs* %Ga' exhibits two peaks near 19 at.% of 265×10^{-6} and 27 at.% of 235×10^{-6} [3].

Fe-Ga alloys, being known by their extremely high magnetostriction may be good candidates for damping applications, too. Magnetic domains with sharp 90° and 180° domain walls, which are typical for soft-magnetic materials with positive anisotropy energy [4], are the source for high damping properties even at low amplitudes of vibrations [5]. According to Smith and Birchak theory [6] maximal damping at amplitude dependent internal friction curves is proportional to magnetostriction constant (λ) of ferromagnetic materials: $Q^{-1}_{h\max}\sim\lambda$. The values of λ in Fe-Al alloys are significantly higher than those in α-Fe (e.g., for Fe-16Al[#]: $\lambda_{100} = 85\times10^{-6}$, $\lambda_{111} = -2\times10^{-6}$, $\lambda_{polycr} \approx 35\times10^{-6}$) but, in turn, they are lower than those in Fe-Ga alloys (e.g., for Fe-17Ga: $\lambda_{100} = 207\times10^{-6}$, $\lambda_{111} = -12\times10^{-6}$, $\lambda_{polycr} \approx 76\times10^{-6}$) [5]. Compositions with around 19%Ga have reported strains of up to 400 ppm along <100> direction with low saturation fields of several hundreds Oersteds [3].

Fe-Ga alloys are known by their ordering of Ga atoms in bcc iron: the type of order depends on temperature and concentration of Ga atoms [7, 8]. Ordering decreases mobility of magnetic domain walls and dislocations, leads to low ductility [9] and decreases damping capacity [10]. In contrast to systematic study of magnetic characteristics, very little is known yet on internal friction in Fe-Ga alloys [11]. After the paper [5], several studies of anelastic effects in Fe-Ga alloys were carried out [10, 12-17].

Taking into account that the Fe-Ga alloys structure is rather similar to other b.c.c. iron based alloys with tendency to ordering (e.g., Fe-Al alloys), it is not surprising that Fe-Ga and Fe-Al alloys have several common features of magnetic and amplitude- and temperature dependent anelastic effects. The ground-state electron configurations of nonmagnetic elements Al and Ga are $1s^2 2s^2 2p^6 3s^2 3p^1$ and $1s^2 2s^2 2p^6 3s^2 3p^6 3d^{10} 4s^2 4p^1$. Their

[#] in this paper we use atomic per cent

outer-shell electron configurations are similar because the d-shell electrons of Ga are filled, and the d-shell electrons of Al are absent. Both Al and Ga can enhance the magnetostriction of b.c.c. iron giving a magnetoelastic contribution to damping capacity of these alloys. Atomic ordering in both systems will decrease damping due to additional pinning of magnetic domain walls at antiphase boundaries [18]. Thermally activated damping in these alloys is due to point defects relaxation, dislocations and point defects interaction, grain boundaries anelastic sliding [10-17].

The formation of the equilibrium face-centered cubic (fcc)-based $L1_2$ ordered phase below 650°C (according to the *equilibrium* Fe-Ga diagrams [7,8]) is so slow that in most cases the decomposition and ordering develop in accordance with the *metastable* phase diagram and at room temperature it is presented by mixture of A2 and $D0_3$ phases [19-21]. Formation of transient nonstoichiometric nanosized B2 phase may take place prior to nearly equilibrium $D0_3$ phase [21]. Quenching suppresses the formation of $D0_3$ structure in favor of a disordered supersaturated A2 structure and creates freeze-in vacancies. Annealing may produce a two-phase mixture of A2 + $D0_3$ for alloys with 14-20%Ga [22].

In this paper we have studied three ternary Fe-Ga-Al alloys with actual amount of Ga+Al ≈ 17÷18 at.% and, for comparison reasons, several tests were also done with Fe-18Ga alloy. Aluminum, which is less expensive than Ga by several orders, is added to substitute for Ga [23]. The situation with ordering type and kinetics in ternary Fe-Ga-Al alloys is not clear enough yet. Based on the metastable Fe-Ga diagram, $D0_3$ ordering may also take place in the ternary composition. In this paper we have focused on amplitude dependent and amplitude independent, temperature dependent damping caused by different thermally activated relaxations and structure induced transitions. Measurements of magnetostriction, heat flow, hardness as well as XRD and microscopy studies are used to understand the anelastic effects of these alloys.

Experimental technique

The alloys of nominal composition $Fe_{80}Ga_{(18-x)}Al_x$ (x = 0, 5, 8, 12) were prepared from iron, gallium and aluminum (with 99.95% metals basis) under an argon atmosphere in a vacuum induction furnace. The as-cast ingots were approximately 10 mm in diameter and were placed in quartz tubes. The ingots were then directionally solidified by using a directional solidification furnace. After confirming the completion of the melting process, the rods were grown with a controlled rate of 8mm/min. Actual compositions of the alloys were obtained by both energy dispersive spectrometry: Fe-11.9%Ga-5.1%Al, Fe-9.0%Ga-8.0%Al, Fe-5.2%Ga-11.9%Al and chemical analyses: Fe-12.2%Ga-5.1%Al-0.05%C, Fe-9.2%Ga-8.0%Al-0.04%C and

Fe-5.0%Ga-12.2%Al-0.04%C. These ternary alloys are denoted in this paper as Fe-12Ga-5Al, Fe-9Ga-8Al and Fe-5Ga-12Al alloys, respectively. Binary Fe-18%Ga alloy, was used in several cases for comparison with ternary alloys.

The internal friction samples were sectioned from the directional solidification rod into shapes of 60×3×0.9 mm^3, then sealed under vacuum into quartz tubes, annealed at 1000˚C for one hour followed by a water quench. Internal friction (IF, or Q^{-1}), i.e. tanφ at forced vibrations, where φ is the phase lag between the applied cyclic stress and the resulting strain ($Q^{-1} \cong \tan\varphi$), has been measured on a dynamical mechanical analyser DMA Q800 TA Instruments. These measurements were conducted as a function of temperature between 0 and 600°C, using forced bending vibrations in the range between 0.1 and 30 Hz with $\varepsilon_0 = 7\times10^{-5}$ with a heating and cooling rate of 1 K/min (temperature dependent IF and elastic modulus: TDIF and TDEM). Additional measurements were conducted as a function of amplitude between 4×10^{-6} and 2×10^{-3} at frequency 3 Hz and room temperature (amplitude dependent IF and elastic molulus: ADIF and ADEM, correspondingly).

The test conditions are illustrated in Fig. 1, where typical frequency dependent curve (amplitude of deformation $\varepsilon_0 = 7\times10^{-5}$) at room temperature is presented. Dynamical mechanical analyser DMA Q800 TA Instruments operates in the frequency range between 0.01 and 200 Hz but it has its own resonance in the vicinity of 100 Hz. Thus, damping measurements at f > 30 Hz (Range II in Fig. 1) are not reliable, at least from the viewpoint of absolute damping values. Weak increase in damping with decrease in frequency below 10 Hz (Range I) is a normal consequence of increase of time for different relaxation processes. Nevertheless, measurements at frequencies below 0.3 Hz are time consuming: one experimental point is measured by averaging tanφ over seven cycles of loading, and already at f = 0.1 Hz it takes more than 1 min. This is critical for temperature dependent tests with a typical heating and cooling rate 1 K/min because at low frequencies density of experimental points decreases drastically. Thus, we have chosen five frequencies, namely 0.3, 1, 3, 10 and 30 Hz (marked by blue arrows), in the reliable frequency range for our temperature dependent tests and f = 3 Hz for amplitude dependent tests for a fixed temperature.

The structural analysis was carried out by D/MAX-RB X-ray diffraction. The longitudinal magnetostriction values were measured through strain gauge using the JDAW-2011 magnetostriction measurement equipment. Thermal analysis measurements were carried out using Labsys Setaram equipment with heating rates from 20 to 40 K/min.

A Buehler IndentaMet 1105 Microindentation Hardness Tester device and a Wolpert Wilson 402MVD equipment were used for the microhardness measurements. The indentation force and the dwell time were set to 1 kgf and 10 sec, respectively. An average value of at least 20 individual measurements, distributed over the whole polished surface of the sample, was used to characterize the given microhardness level.

Results and discussion

Amplitude dependent damping. Typical amplitude dependent internal friction and elastic modulus (ADIF and ADEM, correspondingly) curves for two Fe-Ga-Al alloys after water quenching from 1000°C have a well pronounced extreme: maximum for IF at $\varepsilon_0 = (1\text{-}2)\times10^{-4}$ and minimum for elastic modulus values (Fig. 2). The nature of this peak-like effect is magnetomechanical damping: This is easy to prove by applying external magnetic field. In the external saturated magnetic field (MF) all magnetic domains inside a sample are fixed by MF and do not contribute to energy dissipation during vibrations in the elastic range of cyclic loading. Construction of DMA Q800 specimen holder does not allow placing a specimen inside electrical coil in order to create saturated magnetic field. Consequently, we used four strong NdFeB magnets (Br = 1.2-1.3 kGs, Hci = 12-15 kOe, $(B_H)_{max}$ = 30-35 MGOe) attached to both wings of the sample mounted in double cantilever configuration in the dynamical mechanical analyser. This method does not allow to fix well all magnetic domains in the magnetic field of attached magnets but even this simple experiment clearly demonstrates that total damping of studied samples supplied with NdFeB magnets is two-three times lower compared to measurements without magnets. The same considerations apply to the relation between modulus defect with and without magnetic field (Fig. 2).

Phenomenologically, the energy loss, ΔW ($=2\pi W\times Q^{-1}$), due to magnetic domains for a vibration stress (σ_0) below some critical stress (σ_c) is given by the Rayleigh law: $\Delta W = D\sigma^3$ ($\sigma_0 < \sigma_c$; D = const). For larger stresses ΔW saturates, so the energy losses can be satisfactory described by the equation $\Delta W = k\lambda_S\sigma_c$ ($\sigma_0 > \sigma_c$). According to the Smith and Birchak model [6] the magnetomechanical damping in ferromagnetic materials has its source in the stress-driven irreversible movement of the magnetic domain walls. The maximum damping due to magnetomechanical effect ($Q_{h.max}^{-1}$) is proportional to $\lambda_S E/\sigma_i$, where λ_S is the saturation magnetostriction, E is Young's modulus, and σ_i is the average internal stress opposing domain boundary motion.

For a Maxwell distribution of internal stresses the value of maximal hysteretic internal friction is described by Smith and Birchak as: $Q_{h.max}^{-1} = 0.34\ k\ \lambda_S\ E\ /\ \pi\ \sigma_i$ (at $\sigma_0 \approx \sigma_{max}$), where $Q_{h.max}^{-1}$ is the maximum value of Q_h^{-1} in the ADIF curve, σ_i is the average residual internal stress, k=1 is a constant characteristic of the shape of the hysteresis loop and E is Young's modulus.

When the maximal value of applied periodic deformation ($\varepsilon_0 = E^{-1}\sigma_0$) is small, one would expect that domain walls execute small motions in the vicinity of their equilibrium position, defined largely by the internal stress distribution. This is the amplitude range where the Smith and Birchak model [6] works. When ε_0 is large, the domain walls are moving considerably further and so the effect of internal stress is averaged over a considerably larger region [23]. For such long-range domain wall motion it is necessary to add pinning effects of domain walls by different nonmagnetic obstacles (which was done in general in the Kërsten theory [24]) as well as magnetic inhomogeneities and the inhomogeneities of the structure. That is why the testing of the Smith and Birchak theory by Astie and Deqauque [26, 27] gave unsatisfactory agreement in the vicinity of the damping peak (frequency f≈1 Hz) in contrast to the satisfactory situation in the Rayleigh region.

In contrast with the Smith and Birchak model [6] and in agreement with the Astie and Deqauque experiments there is no direct proportionality between our experimental data for Fe-Ga-Al alloys: maximal damping level at ADIF curves and magnetostriction of the alloys if we consider different compositions (Table 1).

The situation is different if we consider one composition after different treatments. Heat treatment of the Fe-Ga-Al samples plays an important role in the total damping. In Fig. 3 one can see ADIF curves for four different states of our alloys: as-cast, water quenched from 1000°C, annealed at 1000°C cooled down in furnace to 730°C (3 hrs) and water quenched, and water quenched from 1000°C plus heated to 400°C during TDIF tests.

In the as-cast state total damping has mainly nonmagnetic sources: there is no magnetomechanical peak at ADIF curves. After water quenching from both 1000°C and 730°C, the ADIF curves have clear damping peaks due to magnetomechanical damping. There is no well pronounced difference between the absolute values of ADIF curves following quenching from either temperatures. In contrast, annealing (heating to 400°C with heating rate 2 K/min in furnace) decreases damping significantly. Hardness of the studied samples in these four states are collected in Table 2. Maximal damping (damping index $\Psi = 2\pi Q_m^{-1} \approx 15\%$) in Fe-18(Ga+Al) alloys in as-quenched state ($Q_m^{-1} \approx 0.03$) is slightly lower than that is in the binary Fe-18Ga alloy

($Q_m^{-1} \approx 0.04$) measured under the same conditions. Since amplitude dependent damping is dependent not only chemical composition and heat treatment but also on processing route, we do not present in this paper direct comparison between our ternary alloys and Fe-18Ga alloy.

In as-quenched samples hardness slightly decreases with substitution of Ga atoms by Al atoms. Influence of annealing is not systematic with respect to Al content: annealing (heating to 400°C) increases hardness in Fe-18Ga, Fe-9Ga-8Al and Fe-5Ga-12Al samples while it surprisingly does not influence in the case of Fe-12Ga-5Al alloy. Hardness of annealed samples is practically the same with hardness of as-cast alloys underlying that in the as-cast and annealed states samples are nearly in the same structural state.

We suppose that decrease in damping and increase in hardness may be a result of short-range ordering of the sample structures. The X-ray structural studies identify texture in as cast samples: after solidification $Fe_{80}Ga_{12}Al_6$ and $Fe_{80}Ga_9Al_8$ samples have better <110> orientation, $Fe_{80}Ga_5Al_{12}$ have <211> orientation. The X-ray peak intensity changes after annealed at 1000°C. The $Fe_{80}Ga_9Al_8$ sample got strong <110> orientation, $Fe_{80}Ga_{12}Al_5$ and change to <100> and $Fe_{80}Ga_5Al_{12}$ change to <110> orientation but we did not find clear evidences of ordering.

The results reported above clearly demonstrate the complicated behavior of the damping capacity of Fe-Ga-Al ferromagnetic materials possessing a high level of magnetomechanical damping. The understanding of this complex behavior requires to account for all the changes in both the fine crystalline structure and in the magnetic domain structure. It is also clear that heat treatment influences damping greatly. Consequently, this has lead us to study the temperature dependent effects.

Temperature dependent effects. Temperature dependent internal friction and elastic modulus (TDIF and TDEM) curves for all three Fe-Ga-Al samples with a heating rate of 1 K/min at maximal deformation $\varepsilon_0 = 7\times10^{-5}$ and at five different frequencies from 0 to 600°C, are shown in Fig. 4. Additionally, TDIF and TDEM curves for Fe-18Ga binary alloy are presented for the same measuring conditions for comparison. After samples were heated to 600°C, they were cooled down under the same measuring conditions to 0°C (Fig. 5).

Three peaks are observed on heating: for water-quenched sample, the first peak is around 100-200°C (denoted as the P1 peak), the second peak is around 300-400°C (denoted as the P_{Tr} peak), and the third peak is at 450-600°C (denoted as the P2 peak). The P1 and P2 peaks are accompanied by a decrease in modulus which is also frequency dependent within each relaxation process sometimes referred to as the Kronig–Kramers function [11]. On cooling the P1 peak is not observed anymore, and in contrast, the P2 peak is clearly recorded. An increase in damping

background with lowering temperature below roughly 400°C takes place on cooling. On subsequent heating/cooling cycles (Fig. 6a), the alloys demonstrate TDIF and TDEM spectra very similar to those presented in the Fig. 5 for the first cooling cycle. The frequency dependent internal friction measurements in the temperature range of the P2 peak (450-575°C) are shown in Fig. 6b for the sample which was water quenched from 1000°C and then twice measured to 600°C: these tests clearly confirm the thermally activated nature of the P2 relaxation process.

Analysing results presented for all four Fe-Ga-Al samples in figures 4-6 one can distinguish on heating two different temperature ranges with normal modulus decrease with increase in temperature: range I corresponds to as-quenched state and range II corresponds to the annealed state of the studied samples. Irreversible structural transformation (IST), characterised by an increase in modulus with temperature, takes place between these two different states in Fe-Ga samples during first heating as schematically shown in Fig. 7a.

Internal friction background (dotted line) decreases in range I and increases in range II. In both ranges, there is at least one thermally activated relaxation effect denoted as the P1 and P2 peaks, correspondingly. These peaks exhibit clear dependencies of peak temperature on measuring frequency. This allows calculation of activation parameters of corresponding relaxation effect ($\overline{H}$ – mean value for activation energy, τ_0 – inverse frequency factor in Arrhenius equation, and β - parameter of relaxation time distribution). Method and details of the method of determination are presented in our earlier papers [28, 29], and can be shortly summarize for the resulting loss spectrum as:

$$Q^{-1} = Q_m^{-1} \cosh^{-1}\left[\frac{\overline{H}}{k_B r_2(\beta_\tau)}\left(\frac{1}{T} - \frac{1}{T_m}\right)\right]$$

Here k_B is the Boltzmann factor and $r_2(\beta_\tau)$ represents the relative peak width, i.e. the peak width with respect to the single Debye peak with $\beta_\tau = 0$. The relaxation time distribution (b_τ) may originate from distributions in both H and τ_0: $b_\tau = \left| b_{\tau 0} \pm b_H/k_B T \right|$, values $b_{\tau 0}$ and $b_{\tau H}$ can be obtained by plotting experimental data in axes b_τ vs 1/T.

These equations were used to evaluate the parameters of the peak ($\overline{H}$, T_m, Q_m^{-1}, β_τ) by fitting the experimental curves. The mean values for the activation energy $\overline{H}$ and the pre-exponential factor τ_0 were calculated from the frequency and/or temperature shift of the peak using the Arrhenius equation. Parameters of the P1 and P2 peaks both at heating and cooling are presented

in the Table 3.

In contrast with the P1 and P2 peaks, a peak denoted as the P_{Tr} peak, the temperature of which is independent of measuring frequency, is recorded in range II (around 360°C in Fig. 7). The P_{Tr} peak height is irreversibly proportional to measuring frequency: $Q_{Tr}^{-1} = A \times f^{-1}$, this dependence is typical for transient effects for displacive transformations [30]. Values A, which is proportional to volume fraction of transition $\partial n/\partial T$, are provided in the Table 3: there is no pronounce influence of Ga/Al ratio on this coefficient. If heating of a quenched Fe-Ga-Al sample is stopped at the temperature of the P_{Tr} peak maximum, a decrease of IF takes place. In case of Fe-5Ga-12Al alloy, the P_{Tr} peak vanishes within 1.5-2 hrs of annealing at 360°C (f = 0.3 Hz).

The IST effect (at 175-275°C in Fig. 7) is due to a structural transition: its parameters can be estimated from position exothermal peak at heat flow tests performed with different heating rates (Fig. 8a). Activation energy is calculated by Kissinger's method [31], temperature T_0 was estimated by extrapolation of the corresponding heat flow peak to zero heating rate, the Curie temperature T_C was estimated by the same method (Table 4). Values of T_0 (temperature of the heat flow peak) correspond to the excess of the interval where elastic modulus increases with increase in temperature, i.e. they corresponds to the IST range. In the same temperature range an increase in hardness takes place in Fe-18Ga (Fig. 8b), Fe-9Ga-8Al and Fe-5Ga-12Al alloys (Table 2).

During cooling, the modulus of elasticity increases over the whole testing interval and there is no effect on P_{Tr} on the internal friction. In contrast with the P1 effect, the P2 relaxation peak on cooling is more pronounced, the P2 peaks for low frequency broaden and become slightly higher. In the next heating-cooling tests, the activation parameters of the P2 peak remain fairly constant (Table 3), while the peak height during cooling is always a little bit higher (Fig. 6a). In TDIF tests, on heating (especially on the first heating of as-quenched samples) or on cooling, the parameters of the P2 peak are influenced by changes in structure as well as their accuracy depends on the temperature difference between the thermocouple and the relatively massive sample. Frequency dependent tests (Fig. 6b), run at a fixed temperature, help to avoid these problems and give more representative activation parameters of $\overline{H}$ = 2.53 eV and $\tau_0 = 10^{-17}$ s.

An increase in internal friction background with lowering temperature is a result of an increasing magnetomechanical contribution to damping according to Curie-Weiss law known for many b.c.c. iron-based alloys (e.g. for Fe-Ca [32] or Fe-Al [33] alloys). This contribution can be estimated as the difference between experimental curves in Fig. 7b and dotted line which roughly

represents nonmagnetic damping. This behaviour of internal friction background has some hysteresis between heating and cooling tests in the subsequent runs (Fig. 6a): it may occur as a result of ordering – disordering effects during heating/cooling cycles at a cooling rate of 1 K/min. The P1 peak during cooling either disappears completely after heating to 600°C or significantly decreases if the sample is annealed at lower temperatures between 200 and 400°C. It is not observed in subsequent heating-cooling cycles with heating to 600°C.

While analysing activation parameters of the P1 and P2 peaks determined by standard Arrhenius plot (Table 3), it is easy to see that the parameters of the P1 peak are out of the physical meaning ($\tau_0 < 10^{-34}$) as they are too fast for any processes in metals. At least one reason for this result is that the P1 peak overlaps with the structural transition (IST), which significantly affect the P1 peak parameters, and standard Arrhenius treatment cannot be applied properly. Nevertheless, a thermally activated behaviour (i.e. dependence of IF on measuring frequency) is clearly seen below 150-170°C (Fig. 4). Three thermally activated processes in Fe-based alloys have been reported in the literature in this temperature-frequency range [11]: the Snoek-type relaxation due to stress induced jumps of interstitial atoms, the Hasiguti and the γ relaxation due to different aspects of dislocation motion in applied stress. In our earlier papers [10, 12, 15-17, 34], we always added ~0.05 at.%C in our Fe-Ga alloys to study Snoek-type relaxation, nearly the same amount of carbon (~0.04 at.%) contain in these Fe-18(Ga+Al) alloys. Taking into account huge width of this P1 effect, it is possible that several different relaxation effects may contribute in this temperature range. The combination of several relaxation processes in the same frequency-temperature range lead to unacceptable values of τ_0. Heating above 150°C leads to structural processes in the alloys, which suppress these relaxation effects either by trapping C atoms, or decrease in vacancy concentration or ordering processes.

As it concerns elastic and anelastic effects, which take place in both the IST and P_{Tr} range, we offer the following hypothesis based on experimental structural studies and the results of simulations of Fe-Ga alloys summarized in papers [21, 35-40]. First transition takes place in as-quenched, i.e. disordered, Fe-Ga alloys at 150-300°C: in presence of as-quenched vacancies a short-range $D0_3$-type ordering takes place. In binary Fe-Al alloys this state is the well-known K-state: volume fraction of ordered domains is 10^{-2}-10^{-3}, their size is around 5 nm, volume density is 10^{-18} cm^{-3} [36]. The vacancies stabilize the K-phase by migrating to the phase boundary and relieving mismatch strain between the K-phase and the bcc matrix.

In Fe-Ga alloys, including Fe-18%Ga composition, the A2 → B2 → $D0_3$ transition with the formation of B2 type nanoprecipitates with sizes from 3 to 10 nm at the intermediate stage in

reported [21, 40]. This ‘disorder – short-range order’ transition is accompanied by increase in elastic modulus, hardness and exothermic events during in the thermal analysis curves reported in this paper. Dilatometry results show a significant arrest in linear expansion within these temperature ranges [16].

The second step of transition is a phase transition of the b.c.c. ordered $D0_3$ phase to f.c.c. ordered $L1_2$ phase (300-400°C). The kinetics of this transition has been reported to be kinetically inhibited, taking up to several months for completion. Even if this transition is, in general, diffusion controlled, elements of a displacive shear transformation may be involved; namely due to loss of stability or buckling {110}<110> of the $D0_3$ phase with formation of metastable closed packed structure. A similiar viewpoint about displacive transformation or a stress induced reorientation of tetragonal phase microdomains in Fe-Ga [35] has been recently discussed by A. Khachaturyan [21, 37, 38]. The Bain strain is required for the diffusionless part of cubic to tetragonal displacive transformation that brings the structure closer to an equilibrium fcc-based $L1_2$ ordered phase. As mentioned above, the features of the P_{Tr} peak are typical for shear transformations and, thus, it can be associated only with this transformation in studied alloys.

At present stage of our work, this interpretation of the IST and P_{Tr} effects is based on indirect data (TDIF, TDEM, DSC, HV) and analyses of literature. As yet we do not have direct confirmation for this hypothesis by XRD or TEM in our alloys.

At least three relaxation processes were reported in Fe-based alloys in similar temperature – frequency range with the P2 peak: Zener relaxation due to reorientation of substitute atoms, grain boundary relaxation due to elastic sliding of grain boundaries and medium temperature dislocation effects [41]. Activation parameters of the P2 peak and, in particular the τ_0 term, are not consistent with a dislocation dominated relaxation process. Taking into account (i) the activation energy of the P2 peak, (ii) the weak dependence if any, of the peak parameters on ratio between Ga and Al atoms, and (iii) our previous experiments on Fe-Ga alloys with different grain size [12], it is reasonable to assume that the relaxation process takes place at grain boundaries by local elastic reorientation of atoms at the boundaries. The distribution of relaxation time (β_τ) comes from both distribution in activation energies (β_H) and frequency factor ($\beta_{\tau 0}$) and these values vary from heating to cooling (Table 3). It is notable, that the P2 peak width during first heating for the low frequencies used in this study is always smaller than that during subsequent cooling and it is levelled off in the next heating-cooling cycles (values for β_τ measured for f=1 Hz are given in the Table 3). Thus in the first heating test of as-quenched samples, the effect of quenching on the structure of studied alloys is inherited up to 450-500°C

and an overlap between Zener-type and grain boundary relaxation processes is not completely excluded [10]. This observation is also supported by presence of the P_{Tr} effect at first heating TDIF curves.

Conclusions

Anelastic response of Fe-18(Ga+Al) alloys is studied as a function of temperature (from 0 to 600°C), frequency (from 0.01 to 200 Hz) and amplitude (from 0.0004% to 0.2%) of forced vibrations. Quenching stabilizes and supersaturates the b.c.c. Fe solid solution. Main contribution to total damping in as-quenched Fe-Ga-based alloys at room temperature comes from magnetomechanical damping. According to the damping index $\Psi = 2\pi Q_m^{-1} \approx 15\%$ all studied ternary alloys belong to high damping materials (i.e., $\Psi > 15\%$). Nevertheless, in contrast with Smith and Birchak theory, we did not find direct proportionality between saturation magnetostriction and maximal damping of Fe-Ga-Al alloys by varying their chemical composition. This fact may be a result of short range ordering in the alloys. Hypothesis about sequence of ordering processes at heating of quenched alloys: formation short-range $D0_3$ ordered domains at the first stage, followed by the $D0_3 \rightarrow L1_2$ transition at the second stage is in an agreement with new experimental data on internal friction and elastic modulus behaviour obtained in this paper. With regards to the non-magnetic contribution, two thermally activated relaxation processes are detected above room temperature and their activation parameters are analysed by means both temperature and frequency dependent tests. Low temperature effect (P1) is a combination of several relaxation processes including Snoek-type relaxation while the high temperature effect (P2) is attributed mainly to atomic motion at grain boundaries.

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Figures:

Fig. 1. Frequency dependent internal friction curve for forced vibrations (DMA Q800): choice for frequencies for further tests.

Fig. 2. ADIF and ADEM curves for water quenched Fe-12Ga-5Al (a) and Fe-5Ga-12Al (b) alloys: measurements were carried out first without magnetic field (MF), then in magnetic field. Dotted line – approximation of nonmagnetic IF background. In all tests f = 3 Hz.

Fig. 3. ADIF curves for Fe-12Ga-5Al (a), Fe-9Ga-8Al (b) and Fe-5Ga-12Al (c) alloys in three different states: as-cast, as-quenched (two different annealing temperatures before quenching) and as annealed. All tests without magnetic field.

Fig. 4. TDIF and TDEM curves at heating with 1 K/min for studied alloys after water quenching from 1000°C and for chosen frequencies: Fe-18Ga (a), Fe-12Ga-5Al (b), Fe-9Ga-8Al (c), Fe-5Ga-12Al (d).

Fig. 5. TDIF and TDEM curves at cooling (1 K/min) from 600°C for studied alloys Fe-18Ga (a), Fe-12Ga-5Al (b), Fe-9Ga-8Al (c), Fe-5Ga-12Al (d).

Fig. 6. Fe-12Ga-5Al alloy: second TDIF and TDEM run (a); FDIF and FDEM curves at temperatures from 475 to 575°C (P2 peak) (b)

Fig. 7. Schemes of TDIF and TDEM curves for quenched Fe-Ga-Al alloys at heating (a) and cooling (b).

Fig. 8. Influence of annealing of water quenched samples on heat flow of water quenched from 1000°C Fe-9Ga-8Al alloy (a) and hardness of water quenched from 730°C Fe-18Ga alloy (b). Inset to (a) – dependence of peak temperature on heating rate