

Order Evolution in Iron-Based Alloys Viewed through Amplitude Dependent Damping Studies

Osvaldo Agustín Lambri^{1,*}, José Ignacio Pérez-Landazábal², Damián Gargicevich¹, Vicente Recarte², Federico Guillermo Bonifacich¹, Gabriel Julio Cuello³ and Vicente Sánchez-Alarcos²

¹Laboratorio de Materiales, Escuela de Ingeniería Eléctrica, Centro de Tecnología e Investigación Eléctrica, Facultad de Ciencias Exactas, Ingeniería y Agrimensura, Universidad Nacional de Rosario—CONICET. Avda. Pellegrini 250, (2000) Rosario, Argentina

²Departamento de Física, INAMAT, Universidad Pública de Navarra, Campus de Arrosadía 31006 Pamplona, Spain

³Institute Laue-Langevin, 71 Av. des Martyrs, B.P. 156, 38042 Grenoble Cedex 9, France

Amplitude dependent damping and neutron thermo-diffraction studies were performed in Fe-Al-Si, Fe-Al-Cr and Fe-Si alloys. Results show that the behaviour of the strength of the amplitude dependent damping as a function of temperature is a suitable tool for determining the order changes as a function of temperature. A decrease in the order degree leads to an increase in the strength of the amplitude dependent damping, due to the dislocation mobility increases. [doi:10.2320/matertrans.M2014397]

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1. Introduction

The degree of order and the defect configuration both influence the mobility of dislocations and grain boundaries in Fe-25 at%(Al + Si), Fe-15 at%(Al + Si),¹⁾ Fe-25 at%Al-8 at%Cr, Fe-25 at%Al-25 at%Cr²⁾ and in Fe-10 at%Si.³⁾ In Fe-Al-Si and Fe-Al-Cr alloys a decrease in the order degree after heating up to 1200 K and 1050 K, respectively leads to the appearance or enhancement of a damping peak at around 1000 K during the subsequent cooling measurement. For Fe-Al-Si alloys this peak is absent during the heating and it is related to the aluminium and silicon solute grain boundary relaxations.¹⁾ The temperature of this peak depends on the order degree of the alloy. The lower is the D0₃ order-disorder transition temperature, the lower is the peak temperature.¹⁾ On the other side, for Fe-Al-Cr alloys the intensity of the peak recorded during the cooling is around three times larger than the value measured during heating, this peak being related to the solute grain boundary relaxation of the aluminium and chromium.²⁾

In binary Fe-Si alloys a higher degree of order (B2) accompanied by a grain boundary blocking led to a smaller peak height of the grain boundary solvent peak which appears at around 800 K.³⁾ The decrease of the order degree by heating the sample above 1223 K enhances the dislocations and grain boundaries mobility making easier the recovery of the as-quenched dislocations. This leads to a rearrangement of grain boundary dislocations revealed through the increase in the grain boundary peak intensity.³⁾

In fact, in ordered alloys independently of the type of order, D0₃ or B2, the mobility of dislocations and grain boundaries is markedly reduced, which is shown through small values of damping. In contrast, when the order decreases or disappears after annealing, the dislocation and grain boundary mobility increases which results in an increase in damping as compared with the ordered state.²⁾

In alloys and lightly coldworked metals, the damping measured at increasing amplitudes is at first a constant, then increases sharply above amplitudes of around 10⁻⁶.^{4,5)} This behaviour has been explained by Granato and Lücke as due to the breaking away of dislocations loops from pinning impurities.^{5,6)} Under increasing applied stress, the lengths of dislocations between pinning points bow out until the dislocation escapes from pinning points. This mechanism of break-away of dislocation from weak pinning points give rise to the appearance of amplitude dependent damping (ADD) being ε_c the transition strain value between the linear and non-linear damping behaviour.⁴⁻⁸⁾ The critical strain, ε_c can be related to the microyield stress.⁸⁻¹⁰⁾ Contrary to macroscopic flow stress measurements, in ADD measurements the dislocations are only forced to overcome the weak obstacles, solutes, and not the strong ones.⁴⁻⁹⁾ A relation between the macroscopic and microscopic yield stress can be made under determined conditions,^{9,11,12)} but it will be not discussed in the current work. Nevertheless, ADD can be also used for studying heavy plastically worked materials by considering the average free length of dislocations.^{9,13)} Therefore, the order changes occurring in alloys as a function of temperature should be described by means of ADD since the decrease in the dislocation mobility in the ordered lattice. It leads to smaller bow outs and then smaller amount of breaking away processes due to the smaller swept areas by the dislocation movement. Indeed, the aim of this work is to show that ADD is a powerful tool for viewing the order behaviour as a function of temperature. For this purpose, ADD and neutron thermodiffraction studies have been coupled.

2. Experimental

2.1 Samples

Samples of Fe-Al-Cr, Fe-Al-Si and Fe-Si were used in this work. The following sample composition were used for ternary alloys (at%), Fe-25Al-8Cr, Fe-25Al-25Cr, Fe-12Al-12Si and Fe-6Al-9Si.^{1,2)} Binary Fe-Si samples were taken

*Corresponding author, E-mail: olambri@fceia.unr.edu.ar

Table 1 Description of studied Ternary (T) and binary (B) samples.

Sample denomination	Sample composition	Ordering transition	
		Temperature, T/K	
TCr8	Fe-25 at% Al-8 at% Cr	$D0_3 \leftrightarrow B2$	$B2 \leftrightarrow bcc$
		833	973
TCr25	Fe-25 at% Al-25 at% Cr	$B2 \leftrightarrow bcc$	
		973	
TSi12	Fe-12 at% Al-12 at% Si	$D0_3$	
		Always ordered. Order decreases as temperature increases.	
TSi9	Fe-6 at% Al-9 at% Si	$D0_3 \leftrightarrow bcc$	
		983	
B10	Fe-10 at% Si	$B2 \leftrightarrow bcc$	
		973	

from commercial Fe-10 at% Si alloys supplied by NKK Corp.³⁾ A summary of used samples is shown in Table 1. For more details on used samples see Refs. 1–3). Samples were homogenized at 1323 K during 1 hour under high vacuum (10^{-5} Pa), followed by quenching into room temperature water. Metallographical studies show that the grain size of samples does not change with annealing up to 1273 K, under high vacuum. Mean grain size for ternary and binary alloys was around 90–100 μm and 2–3 μm , respectively.

2.2 Measurements

Neutron diffraction (ND) studies were performed both at D20 and D1B powder diffractometers in the Institute Laue Langevin (ILL), Grenoble, France. Spectra were obtained under vacuum (10^{-2} Pa) *in situ* during heating from room temperature up to around 1200 K. The heating rate was 3 K/min. The used samples were parallelepiped of about 2 mm \times 4 mm \times 20 mm. For Fe-Al-Cr and Fe-Al-Si alloys, measurements were performed at D1B using neutron wavelengths of $\lambda = 0.252$ nm and 0.128 nm, respectively. For Fe-Si alloys ND studies were performed at D20 with a neutron wavelength of $\lambda = 0.13$ nm.

Mechanical spectroscopy measurements (damping and natural frequency as a function of temperature or strain) were performed in an inverted torsion pendulum at frequencies close to 1 Hz under vacuum (about 10^{-5} Pa). The employed samples for ternary and binary alloys were bars of rectangular section of around 1 mm \times 2.2 mm \times 20 mm and 0.3 mm \times 3 mm \times 30 mm, respectively. The maximum strain on the surface of the samples was 5×10^{-5} .^{7,10)} Damping (Q^{-1}) and square frequency were measured with an error less than 2%.

Amplitude dependent damping, ADD, i.e. damping as a function of the maximum strain on the sample, ε_0 , was calculated from eq. (1),^{7,14,15)}

$$Q^{-1}(\varepsilon_0) = -\frac{1}{\pi} \frac{d(\ln(A_n))}{dn} \quad (1)$$

where A_n is the area of the n^{th} decaying oscillation and n is the period number. The decaying of the oscillations were

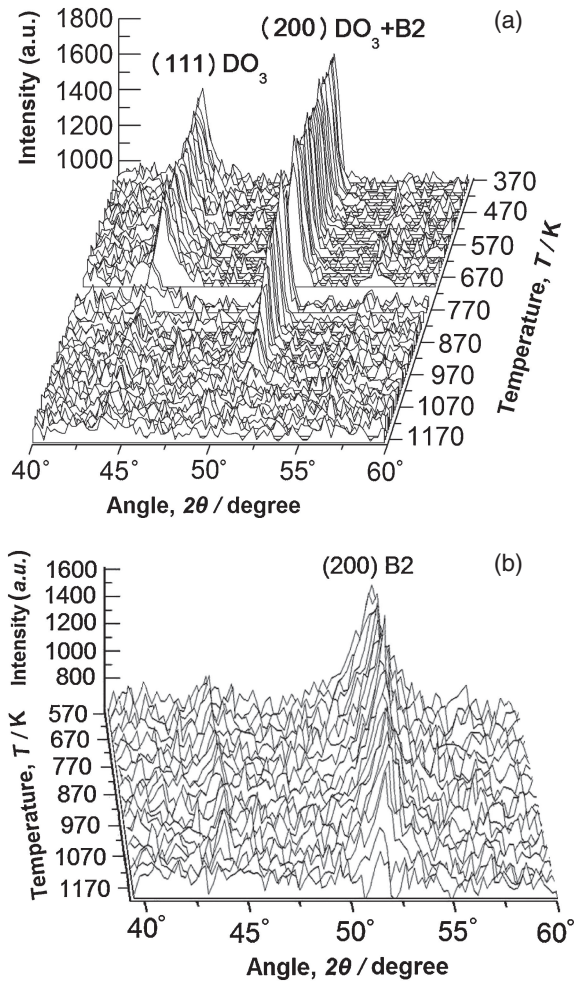


Fig. 1 “*in situ*” neutron thermo-diffraction patterns measured during heating for as-quenched samples, (a) Fe-25 at% Al-8 at% Cr (TCr8 sample), (b) Fe-25 at% Al-25 at% Cr (TCr25 sample).

performed at constant temperature ($T \pm 0.5$ K) from room temperature up to different maximum temperatures above 1000 K, depending of the alloy type, with a heating rate of 1 K/min. Polynomials were fitted to the curve of the logarithm of the decaying areas of the torsional vibrations as a function of the period number by means of Chi-square fitting. Subsequently the eq. (1) was applied. Polynomials of degree higher than 1 indicate that Q^{-1} is a function of ε_0 , leading to the appearance of ADD effects, as it can be inferred easily. This procedure allows obtaining the damping as a function of the maximum strain (ε_0) from free decaying oscillations.^{7,14,15)} The degree of fitted polynomials was smaller than 3.

The strength of ADD effects is measured through S parameter, such that^{7,14,15)}

$$S = \frac{\Delta Q^{-1}}{\Delta \varepsilon_0} \quad (2)$$

ΔQ^{-1} being the damping change measured when the amplitude changes $\Delta \varepsilon_0$. In fact, S is an average slope value of Q^{-1} against ε_0 curves, within whole the strain interval.

3. Results and Discussion

Neutron diffraction studies reveal that all as-quenched samples have different order state at room temperature and

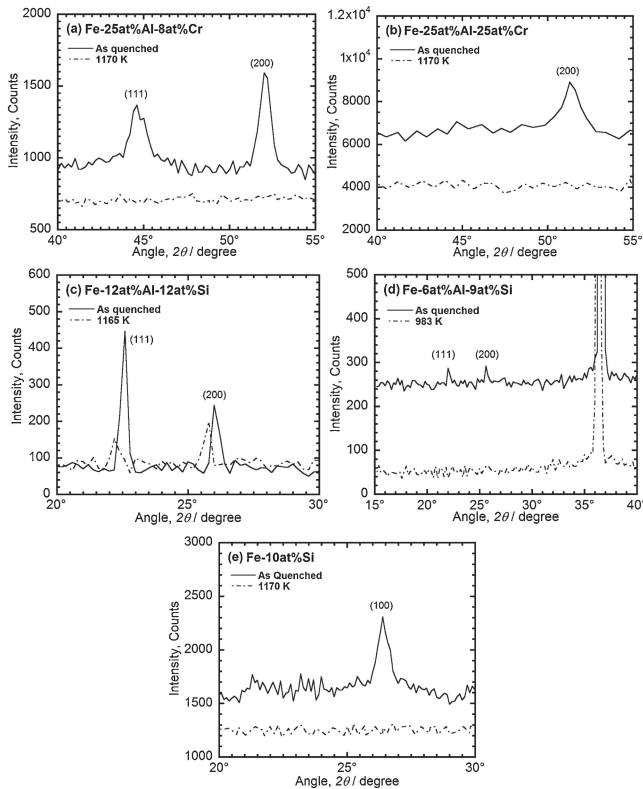


Fig. 2 Temperature dependence of the neutron diffraction patterns for samples exhibiting order: TCr8, TCr25, TSi12, TSi9 and B10 samples.

different order evolution as a function of temperature. Figures 1(a) and 1(b) show a three-dimensional picture (intensity, 2θ and temperature) of the phase evolution observed “*in situ*” at the diffractometer for samples TCr8 and TCr25, respectively. No changes in the full width at half maximum of the diffraction peaks were found for all the group of studied samples, which indicate that changes in the grain size do not occur during heating; in agreement with the metallurgical observations.

Figure 2 shows the neutron diffraction patterns for ternary TCr8, TCr25, TSi12 and TSi9 and binary B10 samples both for the as-quenched state and after an annealing *in situ* at the diffractometer, where the order disappears or decreases; depending of the sample type. The evolution of the order degree as function of temperature for all the used samples in this work is in agreement with previous works and phase diagrams.^{1,2,16} Table 1 summarizes for each alloy type the order behaviour as a function of temperature, determined from ND studies.

As stated previously, ADD studies are very sensitive for revealing information about the interaction processes of dislocation with pinning points. Consequently, it will be shown in the present work, that the effects of the change in the mobility of dislocations due to the modification on the order degree or order type can be finely determined from ADD tests. Figure 3 shows some curves of the whole group of the damping against maximum strain amplitude measured at different (constant) temperatures, T , during heating up to 1050 K for a TCr8 sample. The strength of amplitude dependent damping curves, S , increases as temperature increases, due to the increase in the dislocation mobility

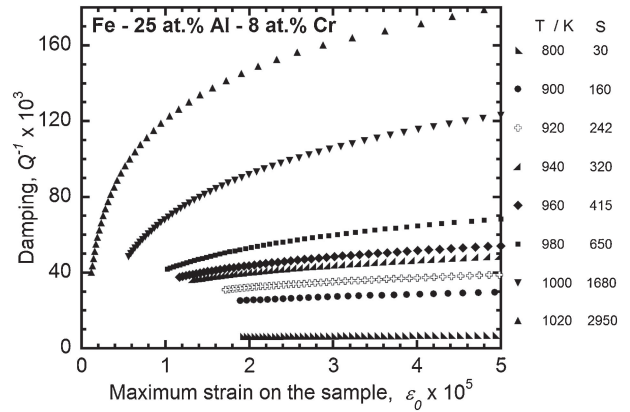


Fig. 3 Damping as a function of the maximum amplitude strain, ϵ_0 , on a type TCr8 sample measured at several constant temperatures during the heating. The temperatures and S are shown at the side.

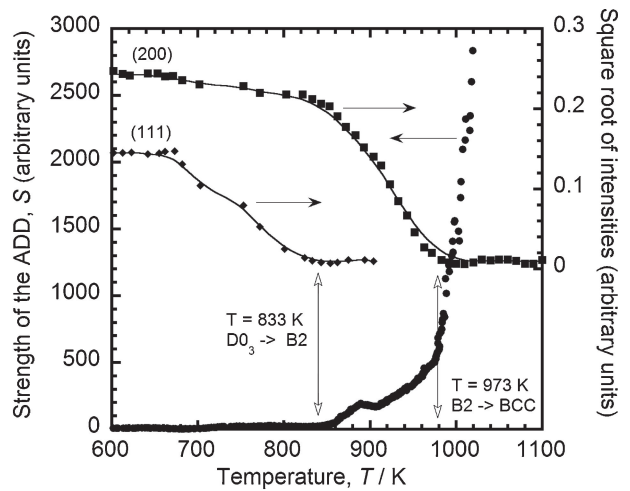


Fig. 4 Strength of the amplitude dependent damping, S , as a function of temperature (left axis) and square roots of intensities of the diffraction peaks (right axis) for a type TCr8 sample. Vertical arrows indicate the transitions temperatures.

with temperature. In fact, the increase in S clearly reveals that the dislocation line sweeps larger areas during its bowing giving rise to a larger amount of thermally assisted break away processes.

It should be stressed, that the behaviour of the strength of ADD as a function of temperature allows easily revealing the temperatures where the order state is modified. Indeed, Fig. 4 shows the behaviour of S as a function of temperature for a TCr8 sample, during the first heating from 600 K up to 1050 K. In addition, the evolution of the squared root of the intensities for the (200) and (111) reflections, which is proportional to the order parameter (η),¹⁷ is also shown in Fig. 4. S exhibits a slope change at the temperatures where the order state is modified. The first increase in S appears for the transformation $D0_3 \rightarrow B2$ at 833 K and the second one appears for the transition $B2 \rightarrow bcc$ at 973 K. In fact, dislocations should move in pairs in the ordered lattice, which leads to a decrease in their mobility. A decrease in the order degree enhances the mobility of the dislocation line, allowing sweep larger areas by bowing. It leads to a larger amount of weak pinning points inside the swept area, where

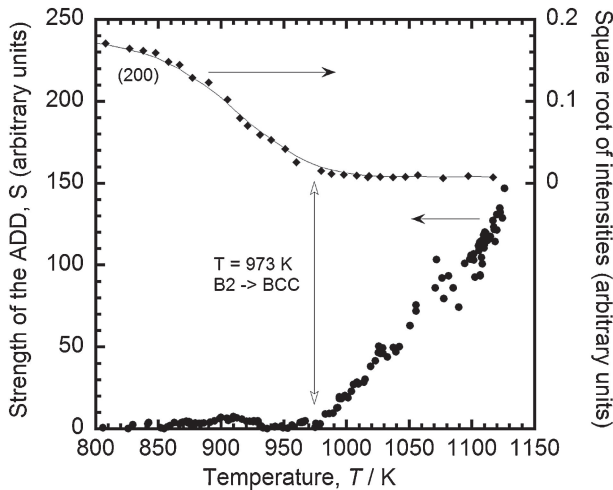


Fig. 5 Strength of the amplitude dependent damping, S , as a function of temperature (left axis) and square root of intensities of the diffraction peaks (right axis) for a type TCr25 sample. Vertical arrow indicates the transition temperature.

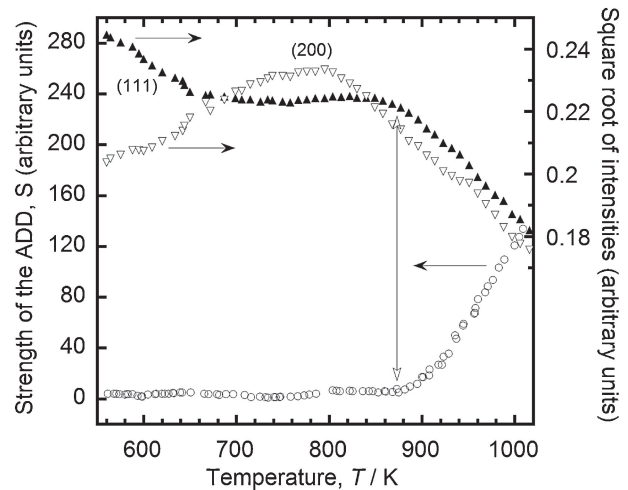


Fig. 6 Strength of the amplitude dependent damping, S , as a function of temperature (left axis, circles) and square roots of intensities of the diffraction peaks (right axis, triangles) for a type TSi12 sample. Vertical arrow indicates the transition temperature.

the dislocation could escape during its movement, giving rise to the increase in S values. The sharp S increase above 973 K is promoted by the increase in the dislocation mobility in the disordered phase, which is in agreement with the transition $B2 \rightarrow bcc$. Besides, the peak-shaped in S at around 890 K can be related to the change in mobility for dislocations between the transition $D03 \rightarrow B2$.¹⁸⁾

The behaviour of S for a TCr25 sample, which exhibits B2 order at room temperature, is shown in Fig. 5. S shows a slope change at 973 K which is well related to the transition $B2 \rightarrow bcc$. Indeed, the evolution of the square root of the intensities corresponding to the (200) reflection as a function of temperature exhibits the disappearance of the ordered state at around 973 K, (see diamonds in Fig. 5). Again, an excellent agreement between the change of slope in S and the falling of intensities can be observed.

Figure 6 shows on the left axis the behaviour of S as a function of temperature for a TSi12 sample. The behaviour of the square root of intensities for (111) and (200) reflections corresponding to a TSi12 sample is also shown in the figure on the right axis. The (200) intensity shows an increase below around 800 K that can be attributed to an initial increase of the B2 nearest neighbours order (inverted empty triangles). The as-quenched sample recovers the B2 order degree since the $B2 \rightarrow A2$ transition is well above the measured temperature range. On the other side, the $D0_3$ order evolves mainly near the equilibrium value of the $D0_3$ order parameter since the $D0_3 \rightarrow B2$ transition is at a lower temperature.¹⁾

It is interesting to note that for the temperatures where the reflection (111) (full triangles) starts to decrease, over around 860 K, S starts to increase (empty circles). As said before, a decrease in the order degree leads to an increase in the dislocation mobility allowing larger bowings of the dislocation line which are able of interacting with a larger amount of weak pinning points, leading to an increase in S values. In a similar way, a TSi9 sample which is $D0_3$ ordered at room temperature changes to bcc at around 983 K and the S curve (not shown in Fig. 6) starts to increase markedly with

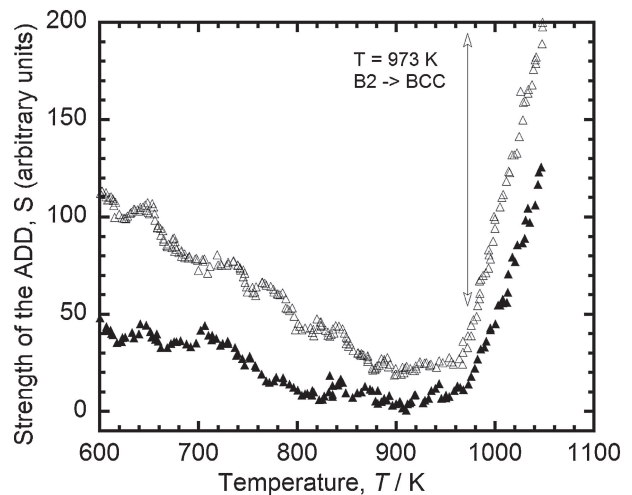


Fig. 7 Strength of the amplitude dependent damping, S , as a function of temperature for a type B10 sample. Full triangles: first heating. Empty triangles: second heating. Vertical arrow indicates the transition temperature.

temperature, in a comparable way to that found in previous Figures.

Figure 7 shows the behaviour of S for B10 sample, measured during two consecutive heating runs. The decrease in S values up to around 900 K during the first heating (full triangles) could be related to the recovery of order after the quenching process.^{3,19)} In contrast, from around 973 K onwards, which corresponds to the temperature where the sample transforms from $B2 \rightarrow bcc$, see Fig. 2 and Table 1, S starts to increase markedly. This kind of behaviour is similar to the previous shown for the ternary alloys.

For the second heating run (empty triangles), the S values become higher on the whole temperature range since recovery of as-quenched both dislocations and point defects took place during the first heating plus cooling run, enhancing the dislocation mobility.^{3,19)}

It should be pointed out that the behaviour of S as a function of temperature follows the changes in the order state

during the warming of the sample, i.e. the modification of the order state is viewed at the temperature where it occurs. In contrast, usual damping measurements as a function of temperature could reveal the appearance of order changes after successive heating and cooling runs up to temperatures over the order-disorder temperature transition. Consequently, through the study of the S behaviour as a function of temperature, the order changes and the dislocation arrangement for a given annealing treatment; can be determined simultaneously.

4. Conclusions

The usefulness of the strength of the amplitude dependent damping as a tool for viewing the order changes as a function of temperature was determined in several iron based alloys. An excellent correlation between the order behaviour, determined from neutron diffraction studies, and the strength of the amplitude dependent damping was found. A decrease in the order degree leads to an increase in the strength of the amplitude dependent damping, due to the dislocation mobility increases.

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