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## Disordered to ordered phase transformation: correlation between microstructure and magnetic properties in Fe-Pd thin films

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### Abstract

The kinetics of the transformation from disordered to ordered phase has been studied in Fe<sub>56</sub>Pd<sub>44</sub> thin films deposited on silicon substrates by Joule evaporation. Subsequent thermal treatments at 550 °C in vacuum were performed at different annealing times, to promote the formation and the optimization of the ordered *L1*<sub>0</sub> FePd phase. X-ray diffraction allowed to investigate the microstructure of the samples (grain size, volume fraction of the transformed phase, order parameter of the ordered phase, and average order of the alloy), whereas magnetic measurements allowed us to evaluate coercivity and to independently estimate the volume fraction of the ordered *L1*<sub>0</sub> phase. The kinetics of the transformation turned out to reach completion after 20 minutes of annealing time, with grain size growing with a power law with exponent equal to 8.24. The resulting monophasic film has a high density of defects, which contribute to the magnetic coercivity. A linear relationship between the coercive field and the average order parameter of the alloy has been established. Finally, the evolution of transformed phase obeys Avrami's law with the exponent  $\sim 0.4$ , indicating that the transformation is controlled by the nucleation mechanism without grain growth.

**Keywords:** Thin films, Fe-Pd, Order–disorder phenomena, Annealing, Kinetics, Microstructure,  $A1$ - $L1_0$  phase transformation, Coercivity.

## I. Introduction

$L1_0$  ordered ferromagnetic intermetallics, such as FePt, FePd and CoPt, are of technological interest due to their large uniaxial magnetocrystalline anisotropy ( $K_u$ ,  $6.6 \times 10^7$  erg/cm<sup>3</sup> for FePt,  $1.8 \times 10^7$  erg/cm<sup>3</sup> for FePd and  $9.4 \times 10^7$  erg/cm<sup>3</sup> for CoPt) [1], which confers them a high coercive field. However, the details of the microstructure of these materials turn out to affect coercivity in a complex way, involving the presence of the equiatomic disordered phase and how it transforms into the ordered one, including the development of defects such as c-axes variants, grain boundaries, antiphase boundaries (APBs), twins and polytwins. For example, large values of coercivity have been attributed to domain wall pinning at the interface between the ordered and the disordered phases and to the c-axes variant at the boundaries of the ordered phase [2]. The FePd system is extremely apt at studying domain-wall pinning by planar crystal defects and microstructural heterogeneities, as well grain size effects, which contribute to the increase of the coercive field [3-6], through the presence of twins, polytwins or stacking faults.

These defects develop during the disordered  $A1 \rightarrow$  ordered  $L1_0$  phase transformation, through processes that can be controlled by suitable thermal treatments. These can be grouped into direct heating of the substrate during deposition, and post-deposition annealing of as-prepared samples. For both groups several annealing protocols have been attempted, with different heating and cooling rates, annealing temperatures, and times, all of which affect the grain size and the formation of the structural defects during the disorder  $\rightarrow$  order phase transformation. As a consequence, also the magnetic properties are affected, allowing their optimization for applications in high-density recording, permanent magnets, spintronic devices, etc.

In Fe-Pd thin films, in situ annealing in electron microscopy has been exploited to study the role of the annealing temperature [7] and of the heating rates [8] for samples grown on single-crystal substrates such as MgO (001), inferring a slow rate of crystal growth. The magnetically hard  $L1_0$  phase can develop with variants in three different space orientations [5], which can induce twin structures. These, as well as stacking faults [6] and antiphase boundaries and twin boundaries [9] have been shown to improve the alloy coercivity. Once induced by the thermal treatments, the disorder  $\rightarrow$  order transition can be followed through physical properties such as magneto-crystalline anisotropy (which depends on the magnetic phases developing in the sample) and coercivity (which depends on magneto-crystalline anisotropy as well as microstructural features of the sample), and through the order parameter of the ordered phase [10,11]. The kinetics of such a transformation has been modelled by the Johnson-Mehl-Avrami-Kolmogorov (JMAK) approach [12], not only for FePd but also for FePt and CoPt systems

[2,13,14], that allows to obtain the volume fraction of the transformed (ordered) phase as a function of annealing time. The transformation can take place through nucleation of new grains, or growth (by diffusion or by accretion) of existing grains, along either 1, 2 or 3 dimensions [15, 16]. The grain growth follows a power law whose exponent  $n$  provides an indication of the mechanisms involved.

In turn, the grains size affects the magnetic coercivity following a complex pattern [17-20], where grain boundaries, density of grains with respect to the magnetic exchange length, crystalline anisotropy at the scale of the individual grain, and the single-domain / multi-domain boundary all play a role.

In this complex scenario, our paper aims at clearly identifying the links among microstructure, grain growth kinetics and magnetic properties in  $\text{Fe}_{56}\text{Pd}_{44}$  thin films. By exploiting suitable theoretical models on both microstructure and magnetic properties data, we have been able to investigate in details the mechanism of grain growth and the transformed phase (from disordered to ordered) fraction as a function of the annealing time. Laws correlating the coercivity of the ordered and disordered phases, the transformed phase volume fraction, and the order parameters of the  $L1_0$  phase and of the alloy as a whole have been found and experimentally validated.

## II. Experimental

$\text{Fe}_{56}\text{Pd}_{44}$  thin films, with a thickness of 80 nm, were grown on polycrystalline silicon substrates by thermal evaporation. The alloy was prepared mixing elemental Fe and Pd powders of 99.99% purity in a tungsten crucible placed in high vacuum chamber (base pressure  $10^{-5}$  Pa). The operation pressure and applied average current were fixed to  $7 \cdot 10^{-4}$  Pa and 124 A respectively. The chemical composition of the thin films was determined through energy dispersive X-ray analysis (EDX), with a relative error of 2% Pd, whereas their thickness was determined through scanning electron microscopy on samples mounted in cross section, with a relative error of 4%.

The alloy phase composition was studied using X-Ray diffraction (XRD) in a pseudo-parallel beam configuration with a glancing angle of  $1^\circ$  of the X-ray beam (CoK $\alpha$  radiation,  $\lambda = 0.1790307$  nm) on the sample surface.

Annealing has been performed in vacuum (base pressure  $2 \times 10^{-6}$  mbar) at a temperature of 550 °C for selected times. After the annealing, the samples have been cooled to room temperature by free cooling, still in vacuum.

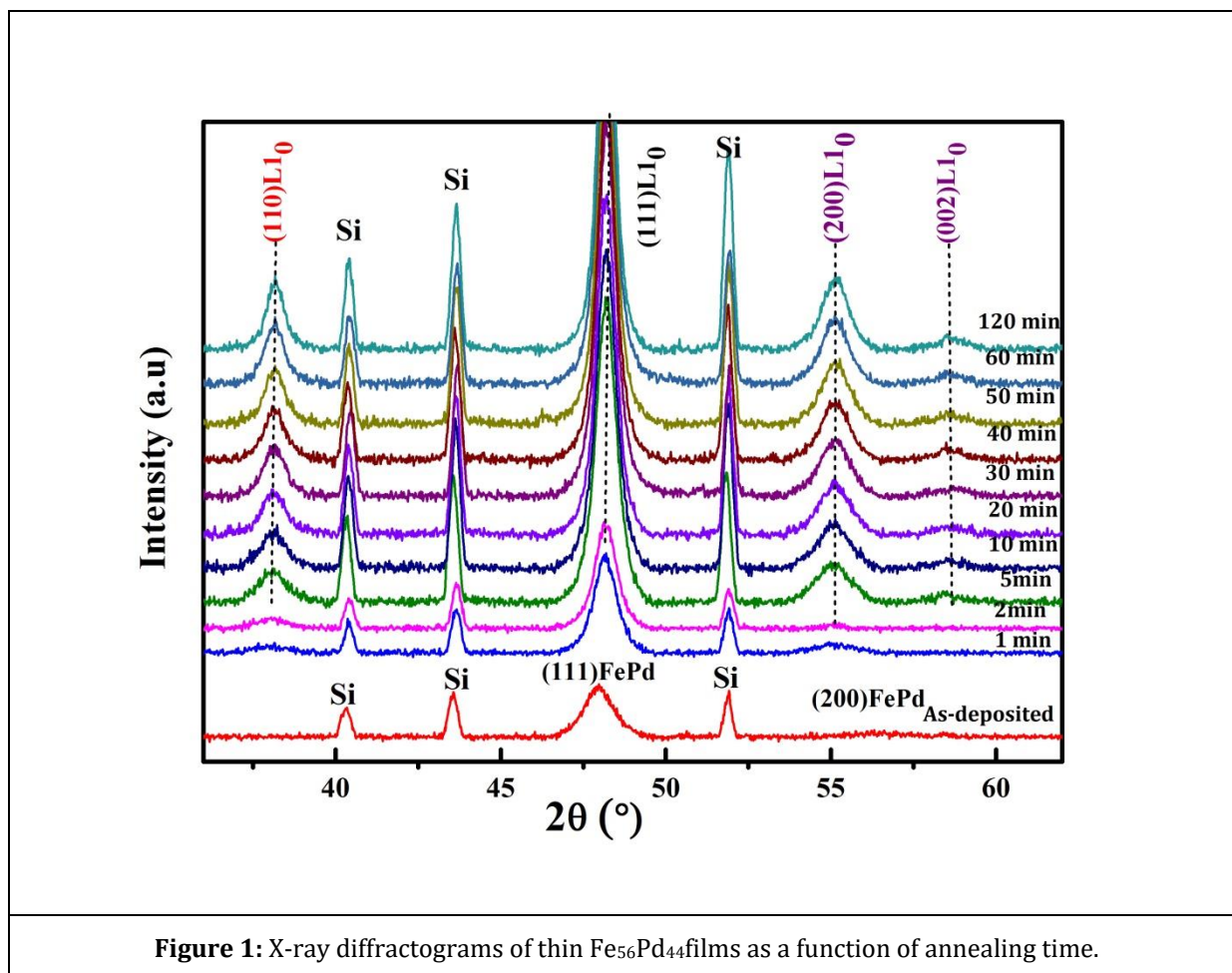
The magnetic properties of the as-prepared and annealed thin films have been measured by Alternating Gradient Field Magnetometry (AGFM), at room temperature with the magnetic field

applied in the sample plane (maximum value 1.5 kOe and 10 kOe for the as-deposited and annealed samples respectively).

### III. Results

#### a) Microstructure

X-ray diffraction patterns of the studied  $\text{Fe}_{56}\text{Pd}_{44}$  thin films are shown in **Figure 1**, for the as-deposited sample and for those annealed at 550 °C for different annealing times. The diffractogram of the as-deposited sample shows the presence of the fundamental peaks (111) and (200) of the face-centered cubic (fcc) disordered FePd phase. In addition to the fundamental peaks, (110) super-lattice line and the splitting of the (200) peak into the (200) and (002) peaks have been observed for all the annealed films. The presence of the superstructure peak is a signature of the transformation of the disordered fcc FePd phase into the ordered  $L1_0$  FePd phase being underway.



**Figure 1:** X-ray diffractograms of thin  $\text{Fe}_{56}\text{Pd}_{44}$  films as a function of annealing time.

**Table 1** summarizes the lattice constants of the fcc (disordered) and  $L1_0$  (ordered) phases deduced from the X-ray diffractograms using the Bragg equation. The lattice parameter of the disordered FePd phase has been calculated from the (111) peak, whereas for the ordered  $L1_0$

FePd both the (111) and (200) peaks have been taken into account. The obtained lattice constants are consistent with those reported in literature for  $L1_0$  FePd films [21-23], being  $3.82 \text{ \AA} < a < 3.86 \text{ \AA}$  and  $3.71 \text{ \AA} < c < 3.75 \text{ \AA}$ . Moreover, the tetragonal ratio  $c/a$  is equal to 0.945, as expected [24,25].

**Table 1**

Structural parameters as a function of the annealing time (at 550 °C): lattice parameters of the fcc and  $L1_0$  phases, average crystallite size  $\langle D \rangle$ , tetragonal ratio  $c/a$  (for the  $L1_0$  phase) and degree of chemical ordering  $S_{tetr}$ .

Lattice constants (Å)						
Annealing time (min)	Fcc FePd	$L1_0$ FePd		Tetragonal ratio	Degree of chemical ordering	Grain size (nm)
	$a=b=c$ $\pm 0.005 \text{ \AA}$	$a=b$ $\pm 0.005 \text{ \AA}$	$c$ $\pm 0.005 \text{ \AA}$	$c/a$ $\pm 0.002$	$S_{tetr}$ $\pm 0.003$	
0	3.814	-----	-----	-----	0	-----
1	-----	3.865	3.680	0.952	0.937	10.1
2	-----	3.868	3.672	0.949	0.964	11.4
5	-----	3.867	3.666	0.948	0.976	12.0
10	-----	3.871	3.658	0.945	1.004	12.9
20	-----	3.871	3.660	0.946	0.999	14.4
30	-----	3.871	3.658	0.945	1.004	14.8
40	-----	3.867	3.660	0.946	0.991	14.9
50	-----	3.870	3.659	0.945	1.000	15.9
60	-----	3.873	3.655	0.944	1.000	16.0
120	-----	3.870	3.659	0.945	1.000	17.7

The order parameter  $S_{tetr}$  is calculated by means of the model given by Roberts [26], which has shown a good correlation with data of different  $L1_0$  phases (AuCu, CoPt) [27]:

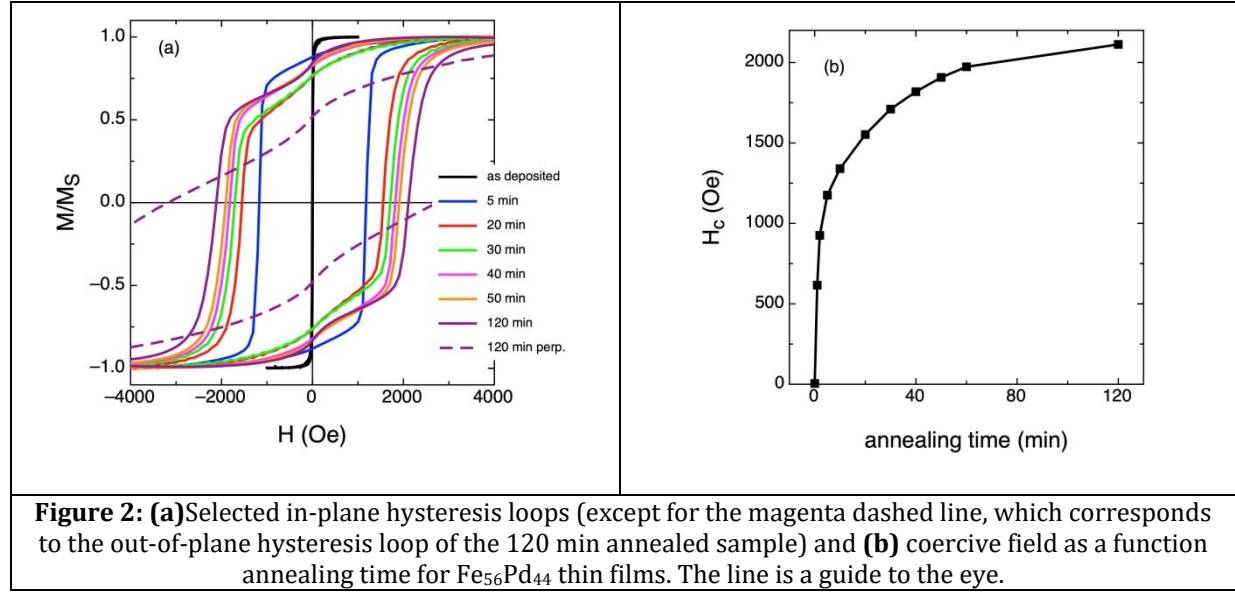
$$S_{tetr}^2 = \frac{(1-\frac{c}{a})}{(1-\frac{c}{a})_{S=1}} \quad (1)$$

**Table 1** shows the evolution of the degree of chemical ordering as a function of the annealing time. The average crystallite size  $\langle D \rangle$  was calculated from the measured width of the (111) peak profile using the Scherrer equation, after subtraction of the instrumental contribution to peak broadening.

### b) Magnetic properties

**Figure 2** shows the hysteresis loops at room temperature of the as-prepared and annealed samples, and the corresponding coercive fields as a function annealing time. All the hysteresis loops are measured in the in-plane configuration, except for the one shown with the magenta dashed line in Figure 2a, that corresponds to the sample annealed for 120 min measured with the field in the out-of-plane direction. The hysteresis loop shape of the as-deposited sample is characteristic a soft ferromagnet, with a coercive field of 5 Oe, in agreement with XRD results that revealed the presence of only the fcc phase. Concerning the annealed samples, as the annealing time increases the loops become wider, with typical characteristics of hard ferromagnets. The comparison of the in-plane and out-of-plane loops of the sample annealed for 120 min reveals that the transformed  $L1_0$  phase grows without any preferential direction: the magnetic remanence in the in-plane configuration is still rather high, and higher than that in the out-of-plane configuration. This is compatible with a disordered distribution in space of the easy

axes of the  $L1_0$  crystals, and is also confirmed by the Mössbauer data reported in [9]. It is worth remarking that the choice of a different substrate (e.g.  $\text{SiO}_2$  or  $\text{MgO}$ ) would have probably led to a different orientation of the  $L1_0$  grains, and therefore resulted in different values of the coercivity [28] and different loop shapes, even though the kinetics of transformation discussed in the following, that should mostly depend on temperature and time, would probably have remained the same. Finally, in the case of the annealed samples, especially for the longer annealing times, a small kink in the in-plane hysteresis loops is observed close to the remanence; this kink is often attributed to the presence of a residual soft fcc phase even in the annealed samples, or to defects in the hard  $L1_0$  phase that determine a local magnetic softening [29].



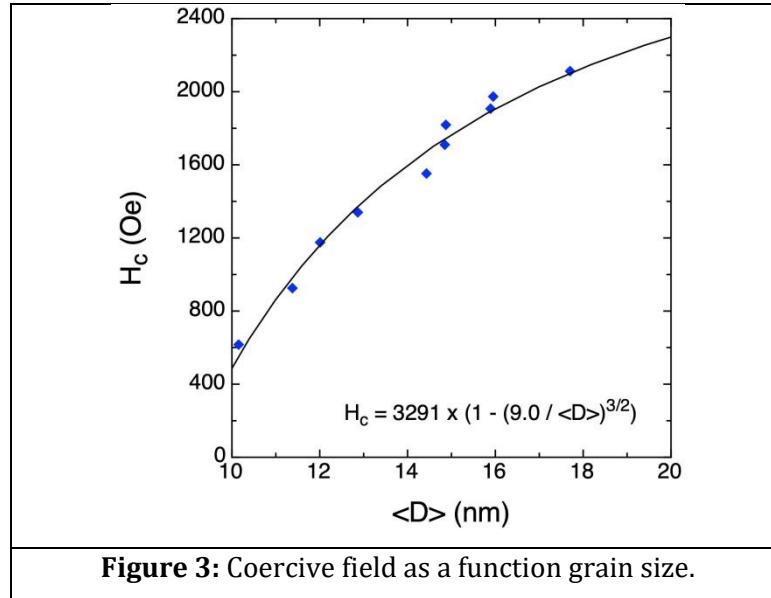
The coercive field monotonically increases with annealing time (**Figure 2b**), with an abrupt development of coercivity in the first few minutes of the thermal treatments. Initially, this is due to the transformation of disordered fcc phase into the  $L1_0$  one, followed by the appearance of structural defects (antiphase boundaries, twin boundaries, order/disorder phase boundaries). **Table 1** clearly shows that the disordered fcc phase was completely transformed into the ordered  $L1_0$  after only 10 minutes of heat treatment, when  $S_{tet}$  is already equal to 1. Beyond 10 minutes, the coercivity continues to increase by the effect of structural defects [9].

The coercivity is also linked to the grains size, which increases as a function of the annealing time, as illustrated in **Figure 3**. Different models link the coercive field  $H_c$  to the size of the grains  $D$ , such as the one by Kneller and Luborsky [20] that accounts for an increase of  $H_c$  with  $D$ :

$$H_c = H_A \left[ 1 - \left( \frac{D_p}{D} \right)^{3/2} \right] \quad (2)$$

where  $H_A = 2K_1/M_s$  is the anisotropy field and  $D_p$  is the critical grain size for the transition from super-paramagnetic to single-domain (SD) regime. Expression (2) is valid for an assembly of randomly oriented non-interacting SD particles with  $D_p < \langle D \rangle < D_{MD}$  where  $D_{MD}$  is the critical size for a transition from single-domain to multiple-domain regime.  $D_p$  and  $D_{MD}$  of FePd have been theoretically calculated, and are in the range 5 nm to 40 nm [1]. The experimental data in **Figure 3** were fitted with equation (2), giving  $H_A^{fit} = 3291$  Oe and  $D_p^{fit} = 9.0$  nm. For our samples, the grains are therefore all the time above the superparamagnetic limit, as expected. Thus, the

magnetic structure present in Fe<sub>56</sub>Pd<sub>44</sub> thin film is characterized by weakly exchange-coupled SD grains. This has been deduced for the case of the FePt thin films [14]. On the other hand, the obtained value of the anisotropy field  $H_A^{fit}$  is reduced by a factor of 10 compared to the theoretical value  $H_A^{th} = 33000$  Oe [1], obtained assuming  $H_A^{th} = 2K_1 / M_S$ ,  $K_1 = 1.8 \times 10^7$  erg/m<sup>3</sup> and  $M_S = 1100$  emu/cm<sup>3</sup> [1]. This reduction is similar to that obtained in FePt systems [14], where it was attributed to local compositional fluctuations, structural defects and the misorientation of the anisotropy axis from grain to grain. In our present study, we also attribute this reduction to the presence of structural defects [9].



#### IV. Determination of the transformed phase fraction

##### a) From microstructure data

In order to calculate the transformed phase volume fraction from XRD data, a relationship between the order parameter and the volume fraction of the  $L1_0$  phase has been devised. It is useful to remind that the departure from perfect order in a binary superlattice can be described by the long-range order parameter  $S$ , which is atomistically defined as a function of the fraction of atoms occupying the correct lattice sites (e.g. the face-centre positions), and the fraction of such sites in the lattice. However,  $S$  of a real sample cannot be experimentally determined directly from this definition since the number of atoms occupying a particular lattice site is not accessible. There are two options for the experimental determination of  $S$  [27]: i) comparing the integrated intensity of a superlattice and a fundamental line in the experimental powder diffraction pattern of the sample (let us call this  $S_{int}$ ); and ii) evaluating the extent of tetragonality of the  $L1_0$  crystals by assuming a model that relates the  $c/a$  ratio to the degree of order (let us call this  $S_{tetr}$ ). The two approaches have been directly compared in [30] or can be compared from the data reported in studies about other alloys (FePt and CoPt) with  $L1_0$



ordering [31]. The comparison shows that: i) for the same sample, the two approaches give different values of  $S$ , with  $S_{tetr}$  being always larger than  $S_{int}$ , and ii) the relative difference decreases as  $S$  increases. One reason of this can be found in the fact that, while the  $c/a$  ratio is deduced from the peak positions, and hence does not depend on the relative proportion of the ordered and disordered phases, the intensity ratio between superlattice and fundamental lines depends both on the ordering degree of the crystals and on the amount of ordered phase formed. In the choice of the intensity ratio as an indicator of the degree of long-range order of an alloy, one has to assume that the order-disorder transformation occurs continuously and homogeneously throughout the alloy, meaning that it starts with a small order parameter in all the grains composing the sample and the order gradually builds up with time (or temperature) of annealing. By contrast, other mechanisms of ordering can be observed, especially in films grown close to room temperature, namely the discontinuous (or inhomogeneous) transformation, occurring by nucleation and growth of ordered domains along grain boundaries [32-34]. In this case, especially in the early stages of the transformation, the sample is composed of a mixture of ordered and disordered grains, and the degree of long-range order of the chemically ordered regions (i.e. the  $L1_0$  phase) can be better described by  $S_{tetr}$ . In the latter case, the volume average of the order parameter has to be taken into account, which is  $S_{alloy} = f_v S_{tetr}$ . The volume fraction of the ordered phase formed (transformed fraction):

$$f_v = \frac{V_{L1_0}}{V_{sample}} \quad (3)$$

can be calculated from XRD data as a product of  $S_{tetr}$  and the intensity ratio between superlattice and fundamental lines [35]:

$$f_v = \frac{1}{S_{tetr}^2} \frac{\left(\frac{I_s}{I_f}\right)_{meas}}{\left(\frac{I_s}{I_f}\right)_{calc}} \quad (4)$$

being  $I_s$  the integrated intensity of a superlattice peak and  $I_f$  the integrated intensity of a fundamental peak, and  $S_{tetr}$  is the previously defined order parameter. Subscript *meas* indicates experimentally measured intensities, while *calc* denotes the intensities calculated from the intensity equation for a diffraction peak [36]:

$$I = |F^2| m L_p(\theta) T(\theta) \quad (5)$$

where  $F$  is the structure factor,  $m$  is the multiplicity factor,  $L_p(\theta)$  and  $T(\theta)$  are the Lorenz-polarisation factor and the temperature factor respectively, dependent on the Bragg angle  $\theta$ . This factorisation allows to take into account both the incomplete ordering of the  $L1_0$  phase and the increase in amount of this phase occurring during a discontinuous transformation.

From the X-ray diffractograms of the annealed samples, we have therefore chosen the fundamental (111) and the super-lattice (110) peaks in order to determine the volume fraction: the (111) peak was fitted with a Pseudo-Voigt type function, i.e. a linear combination of a

Lorentzian  $L(2\theta, H)$  and a Gaussian  $G(2\theta, H)$ , whereas the super-lattice (110) peak was fitted with a Gaussian function. These peaks have good statistics compared to the peak (200) and give a better fit. The fittings were performed with the Fityk software (version 0.9.8) [37].

Since it is assumed that after 120 minutes of annealing the phase transformation is complete ( $f_v = 1, S_{tetr} = 1$ ), from equation (4) it follows that, at this annealing time,

$$\left(\frac{I_s}{I_f}\right)_{meas,120\ min} = \left(\frac{I_s}{I_f}\right)_{calc,120\ min} \quad (6)$$

thus the transformed fraction of the  $L1_0$  phase at each annealing time  $t$ , denoted by  $f_v(t)$ , can be written as:

$$f_v(t) = \frac{1}{S_{tetr,t}^2} \frac{\left(\frac{I_s}{I_f}\right)_{meas,t}}{\left(\frac{I_s}{I_f}\right)_{meas,120\ min}} \quad (7)$$

The values of the transformed fraction are given in **Table 2**.

### b) From magnetic properties

The transformed phase fraction can also be calculated from the hysteresis loop data, in an independent way, using the following expression [38]:

$$H_c = f_v^{soft} H_c^{soft} + f_v^{hard} H_c^{hard} \quad (8)$$

where  $H_c^{soft}$  ( $H_c^{hard}$ ) and  $f_v^{soft}$  ( $f_v^{hard}$ ) are the coercive field and the volume fraction of the soft (hard) phase, respectively. It is important to remark that equation (8) considers the two soft and hard phases magnetically decoupled. In the case of the studied disorder  $\rightarrow$  order phase transformation, equation (8) can be written as:

$$H_c(t) = H_c^{fcc} + (H_c^{L1_0} - H_c^{fcc}) f_v^{L1_0}(t) \quad (9)$$

with  $f_v^{fcc}(t) + f_v^{L1_0}(t) = 1$ .

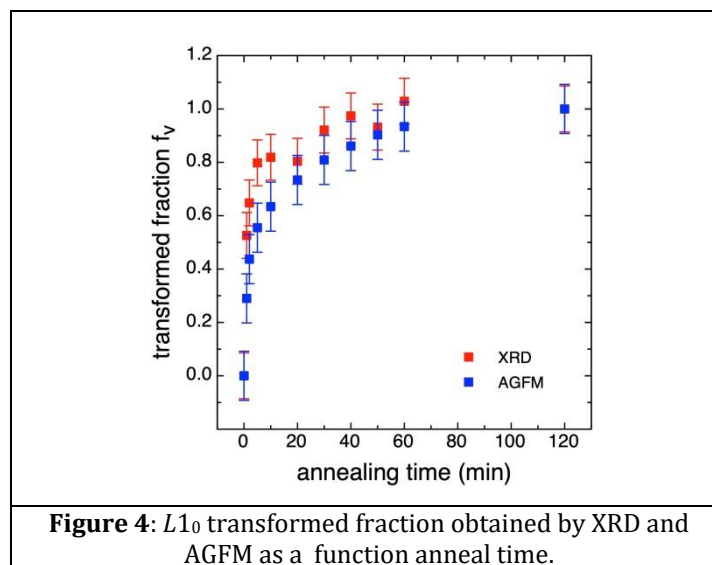
From equation (9) we finally deduce the transformed fraction of the  $L1_0$  phase:

$$f_v^{AGFM}(t) = f_v^{L1_0}(t) = \frac{H_c(t) - H_c^{fcc}}{H_c^{L1_0} - H_c^{fcc}} \quad (10)$$

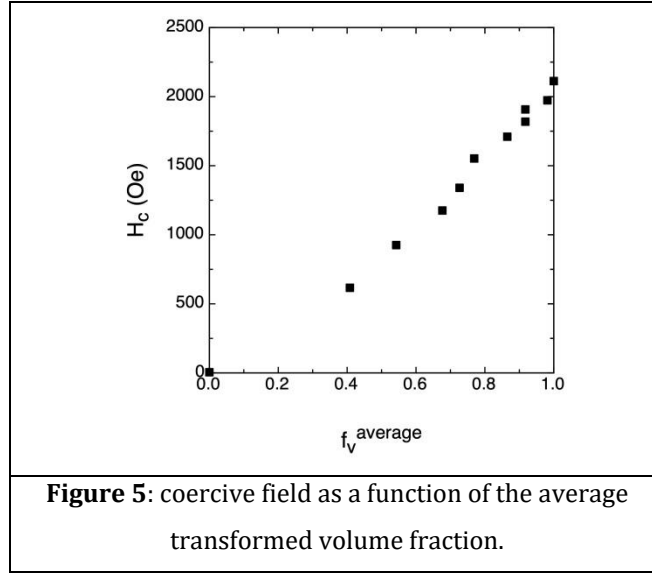
$H_c^{fcc}$  is the coercive field of the disordered FePd phase, obtained from the hysteresis loop of the as-prepared sample (5 Oe, as discussed previously), and  $H_c^{L1_0}$  is the coercive field of the ordered  $L1_0$  FePd phase, taken from the sample where the transformation is complete after annealing at 550 °C for 120 minutes (and therefore 2113 Oe). The values of the transformed fraction of the  $L1_0$  ordered phase as a function of the annealing time are given in **Table 2**.

**Figure 4** shows the transformed fraction of the  $L1_0$  phase as a function of annealing time obtained with the two independent methods, using the values reported in **Table 2**. As it can be clearly seen, the results are in very good agreement. It is worth remarking that  $f_v$  has been determined exploiting only rather common experimental techniques, instead of the much more complex and expensive TEM and Mössbauer spectrometry investigations usually required for such purposes [2,13].

This result allows us to correlate the coercive field of the samples annealed for different times to their respective transformed volume fractions, as shown in **Figure 5**, that shows a remarkable linear dependence of the two quantities, as also already observed in [2,13]. In Figure 5, an average value for the transformed fraction, obtained from the two techniques, is used.



It is worth underlining that transformed fraction values obtained with the two techniques, although remarkably in agreement, are not coincident, as X-ray diffraction and magnetometry are two extremely different techniques, investigating very different aspects of the samples. From their similar behaviour one can infer that the processes of phase transformation can be studied, with comparable results, both from structural and magnetic measurements. If other techniques were available for studying the same phenomena, it is expected that they would provide similar results, the discrepancies being due to the details of the experimental techniques that way differently the numerous samples details.



**Figure 5:** coercive field as a function of the average transformed volume fraction.

**Table 2**

Coercive field  $H_c$ , and transformed fraction obtained by XRD data  $f_v^{XRD}$  and by AGFM data  $f_v^{AGFM}$ , with their average value  $f_v^{average}$ .

Time(min)	0	1	2	5	10	20	30	40	50	60	120
$H_c$ (Oe)	5	616	925	1176	1340	1552	1710	1819	1907	1973	2113
$f_v^{XRD}$	0	0.526	0.648	0.798	0.819	0.804	0.921	0.974	0.932	1.029	1.000
$f_v^{AGFM}$	0	0.290	0.437	0.555	0.634	0.734	0.809	0.861	0.903	0.934	1.000
$f_v^{average}$	0	0.408	0.542	0.676	0.726	0.769	0.865	0.917	0.917	0.981	1.000

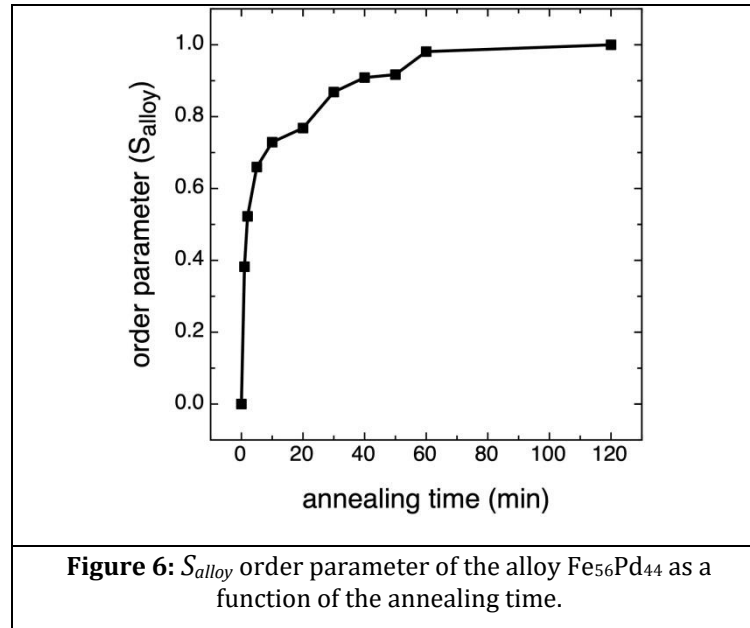
From these results, we can therefore assume that equation (10) is valid in the generalized case of disordered  $\rightarrow$  ordered phase transformations, with uncoupled phases, allowing the determination of the transformed fraction of the ordered phase.

### c) Order parameter of the alloy

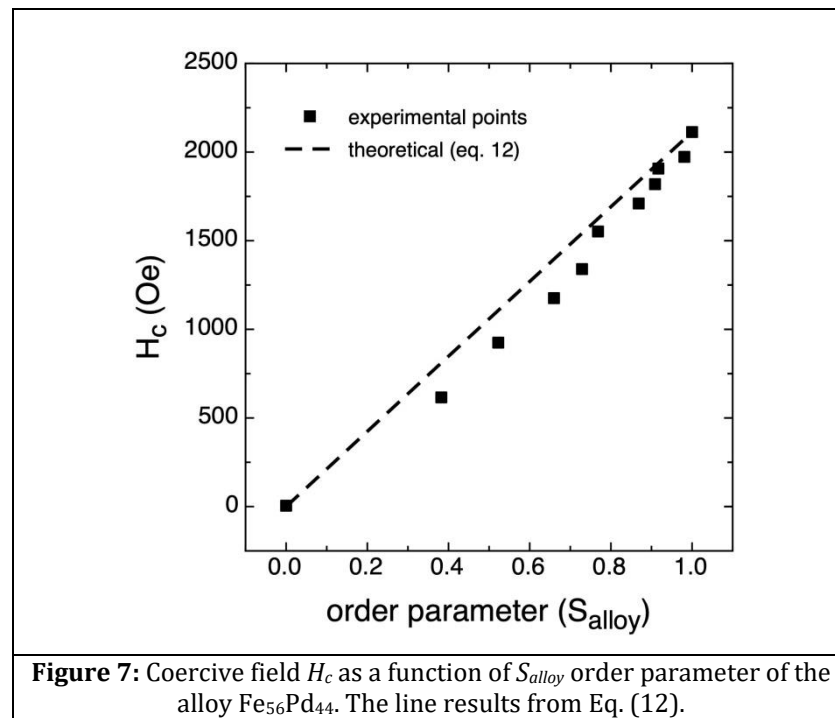
The order parameter of the alloy as a whole,  $S_{alloy}$ , can now be calculated with the following expression [34, 39]:

$$S_{alloy} = S_{tetr} f_v^{average} \quad (11)$$

where  $S_{tetr}$  and  $f_v^{average}$  values can be found in **Tables 1** and **2**. Its evolution as a function of the annealing time is reported in **Figure 6**. After approximately 10 minutes of annealing,  $S_{tetr}$  becomes equal to 1, whereas the ordering parameter of the alloy is still lower than unity. This is because of the progressive growth of the average volume fraction  $f_v$  of the ordered  $L1_0$  phase in the alloy, indicating that for certain annealing times the disordered fcc and ordered  $L1_0$  phases coexist. The transformation of the former into the latter is a first order transition. It is worth remarking that, as already discussed for Figure 2a, an order parameter close to 1 at long annealing times ensures that a well-developed  $L1_0$  phase is present, but the anisotropy axes of the  $L1_0$  crystals are oriented in space in a disordered way, that does not give rise to a dominant perpendicular anisotropy.



It is now possible to correlate the evolution with annealing time of the coercive field and of the average order parameter of the  $\text{Fe}_{56}\text{Pd}_{44}$  alloy, as shown in **Figure 7**.



The use of  $S_{\text{alloy}}$  as the order parameter against which the coercivity values should be analysed is in agreement with [2], that suggests that a microstructural factor relating to the ordered volume fraction and that scales similarly to it should be taken into account. The linear relationship between  $H_c$  and  $S_{\text{alloy}}$  shown in Figure 7 supports this choice.

The linear relationship shown in Figure 7 can be written as follows:

$$H_c(t) = \frac{H_c^{L1_0} - H_c^{fcc}}{S_{L1_0}} S_{alloy}(t) + H_c^{fcc} \quad (12)$$

It is worth remarking that the theoretical line calculated with equation (12) is completely determined by available experimental data, and is not the result of a fitting procedure. It is also in very good agreement with the experimental values. An almost linear relationship between the coercive field and the mean order parameter of the alloy is therefore clearly highlighted.

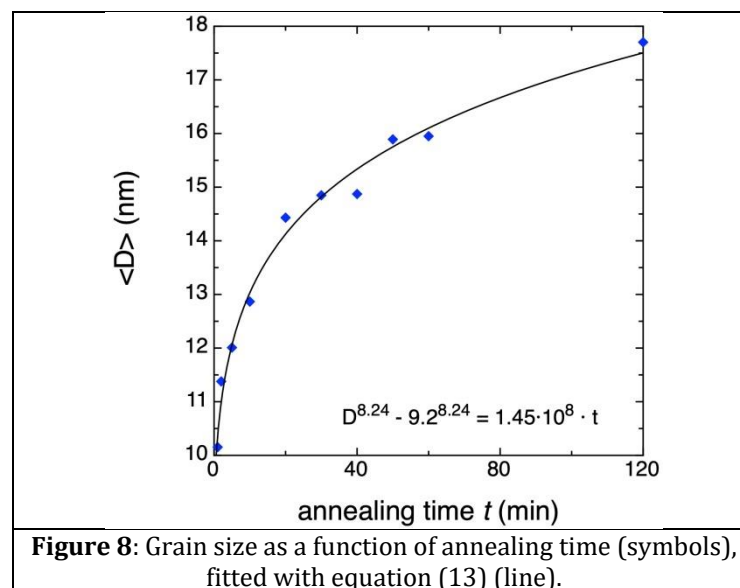
## V. Kinetics of grain growth and phase transformation

### a) Kinetics of grain growth

The kinetics of the transformation of the disordered fcc phase into the ordered  $L1_0$  one, in the studied polycrystalline  $\text{Fe}_{56}\text{Pd}_{44}$  thin films, can be followed by examining the evolution of the grain size as a function of annealing time. The crystallite size  $\langle D \rangle$  calculated from XRD data and reported in **Table 1** is plotted in **Figure 8** as a function of annealing time. It can be clearly seen that the size of the grains varies slowly with the annealing time. Indeed, the evolution of grain growth is described by the normal grain growth equation which is expressed as [14,40,41]:

$$D^n - D_0^n = ct \quad (13)$$

where  $n$  is the grain-growth exponent,  $D_0$  is the initial grain size, and  $c$  is the growth rate, that is given by the following expression:  $c = c_0 \exp(-E_a/k_B T)$  where  $E_a$  is the activation energy for grain growth,  $T$  is the temperature and  $k_B$  is the Boltzmann constant. To determine the parameters  $c$  and  $n$  for our thin film sample, the experimental data of **Figure 8** have been fitted with equation (13), obtaining:  $c = 1.45 \times 10^8 \text{ nm}^{8.24}/\text{min}$  and  $n = 8.24$ .



**Figure 8:** Grain size as a function of annealing time (symbols), fitted with equation (13) (line).

Such a value of the grain growth exponent is compatible with similar findings in literature either for FePd or FePt systems: values in the 8-11 range are specific for slow grain growth rates [14,42], whereas smaller values, in the 3-4 range, are associated to much faster grow rates [31,42]. During the grain growth process, a disordered alloy undergoes microstructure changes through a combination of two processes, i.e. the ordering of the atoms in the alloy, and the recrystallization of the grains. The latter develops through nucleation of new grains, and through their growth. In fact, in our case the evolution of grain size as a function of annealing time (**Figure 8**) can be divided into two main parts corresponding to different microstructures. In the first part, approximately up to 20 minutes of annealing time, the grains size increases from 10 to 14 nm. Beyond 20 minutes, in the second part, the variation of the grains size is approximately equivalent (up to  $\sim 4$  nm), but it takes place over a much longer annealing time interval. This difference is due to the high density of defects, particularly, the grain boundaries, in the as-deposited film, which create a significant driving force contributing to nucleation as well as ordering. This phenomenon leads to the phase transformation of fcc structure phase towards the tetragonal structure  $L1_0$  in the  $Fe_{56}Pd_{44}$  alloy. Indeed, during this interval of time, the density of the grain boundaries decreases in the  $Fe_{56}Pd_{44}$  alloy. At  $t = 20$  min, the alloy reaches a transformed fraction of 76.9 % (see **Table 2**) and an average order parameter  $S_{alloy}$  of 0.769. From this instant of time, new kinds of defects will be created in the ordered alloy, such as antiphase walls, twins and polytwins, leading to a different kinetics [9].

Furthermore, as depicted in **Figure 8**, the shape of the curve indicates that the growth of grain as a function of time is a continuous process. The same shape of the curve was obtained for FePt thin films [14,43]. However, this continuous process takes place according to different mechanisms, where the reduction of grain boundaries (associated to the initial grain growth) is at a certain instant of time suppressed, and followed by the development of new microstructure defects. Several mechanisms responsible for the suppression of grain growth have been suggested in literature, such as the presence of a second phase which induces the trapping of the movement of the grain boundaries [44], the appearance of grain boundaries etching on the surface of the thin film [45], or the correlation between adjacent grains which accelerates the stagnation of grain growth [46]. All these mechanisms can in principle induce the slowdown of the grain growth process in our samples.

#### b) Kinetics of $A1 \rightarrow L1_0$ phase transformation

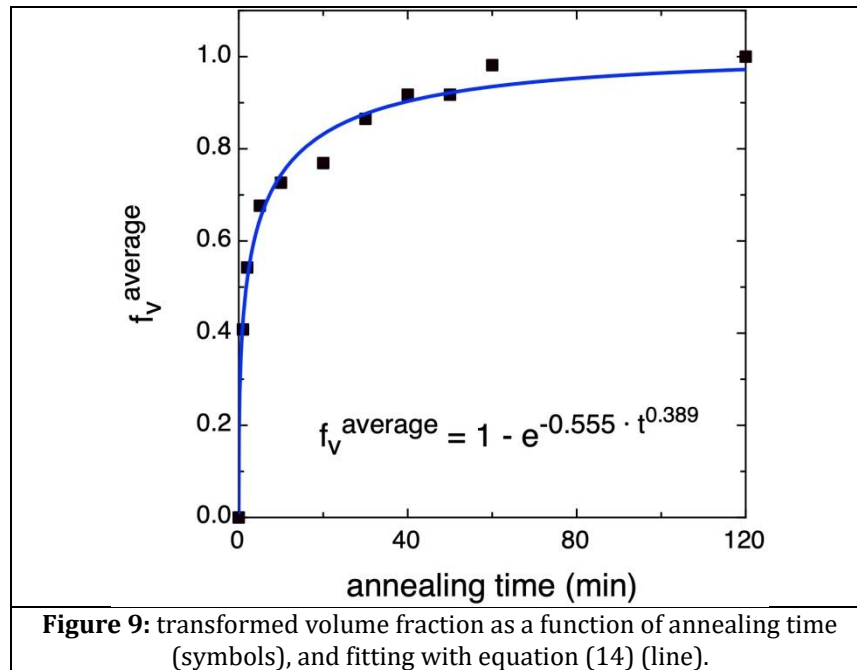
The Johnson-Mehl-Avrami-Kolmogorov (JMAK) theory has been used to study the kinetics of the phase transformation from the disordered  $A1$  phase to ordered  $L1_0$  phase. This theory is based on the nucleation and growth mechanisms, where the nucleation sites are randomly distributed to form grains. Thus, the concept of nucleation-growth transformation can be

expressed using the evolution of the transformed fraction  $f_v(t)$  of the ordered phase as a function of the annealing time [12]:

$$f_v^{average}(t) = 1 - e^{-kt^{n_A}} \quad (14)$$

where  $k$  is the overall rate constant that depends on temperature, and the index  $n_A$  is the exponent of Avrami that includes contributions from growth and nucleation as well as the dimensionality of growth (i.e. 1D, 2D or 3D).

**Figure 9** reports the average values of the transformed fraction  $f_v^{average}$  as a function of annealing time, together with the fit exploiting equation (14) using  $k$  and  $n_A$  as free parameters.



**Figure 9:** transformed volume fraction as a function of annealing time (symbols), and fitting with equation (14) (line).

The fit gave  $n_A = 0.39 \pm 0.03$  and  $k = 0.555 \text{ min}^{-n_A}$ . The quality of the fitting shows that Avrami law is quite well followed by the experimental data.

The Avrami exponent  $n_A$  of equation (14) describes the kinetics of transformation of the A1 phase into the  $L1_0$ , and can be expressed as the sum of two terms: a nucleation condition term ( $a$ ), and one related to the growth process, combining the growth dimensionality ( $b$ ) and the growth mechanism ( $c$ ):  $n_A = a + b * c$  [47]. The nucleation process has three different modes: **1**) continuous, where the nucleation speed does not vary with time (i.e. the nucleation rate is constant) and  $a$  takes the value of 1; **2**) instantaneous, where site saturation occurs and  $a$  takes the value of 0; and **3**) with decreasing speed over time. The effect of the growth process on the Avrami exponent is factorised in order to take into account the possible combinations of the growth mechanism ( $c = 0.5$  for diffusion-controlled growth and  $c = 1$  for interface-controlled



growth), and the dimensionality of the growth in space ( $b = 1D, 2D$  or  $3D$ ). The different possible combinations of the Avrami exponent are summarised in **Table 3** [48].

**Table 3**  
*Theoretical values of Avrami exponent  $n_A$  [48]*

Growth mechanism	Nucleation mechanism	Dimensionality growth		
		1D	2D	3D
Interface controlled growth	Constant nucleation rate	$n_A=2$	$n_A=3$	$n_A=4$
	Instantaneous nucleation	$n_A=1$	$n_A=2$	$n_A=3$
	Decreasing nucleation rate	$n_A=1-2$	$n_A=2-3$	$n_A=3-4$
Diffusion controlled growth	Constant nucleation rate	$n_A=1.5$	$n_A=2$	$n_A=2.5$
	Instantaneous nucleation	$n_A=0.5$	$n_A=1$	$n_A=1.5$
	Decreasing nucleation rate	$n_A=0.5-1.5$	$n_A=1-2$	$n_A=1.5-2.5$

In our case, the Avrami exponent  $n_A \sim 0.4$  that we obtained for our FePd system indicates that the  $A1 \rightarrow L1_0$  phase transformation is diffusion controlled, is one-dimensional (1D) and proceeds with nucleation rate decreasing with time. Values of the same order of magnitude (0.3) have been obtained for FePt systems [13,34], where heterogeneous nucleation mechanisms have been invoked. Such values smaller than 1 are typical of thin film systems, whereas bulk alloys are characterized by Avrami exponent values between 2 and 4 [49].

We can therefore summarise the evolution of the microstructure of our  $Fe_{56}Pd_{44}$  thin films upon heating: the fcc disordered phase initially present gradually transforms into the ordered  $L1_0$  one by means of an inhomogeneous process involving both nucleation and growth of new grains of the ordered phase in the fcc matrix and its progressive ordering. After 10 minutes of annealing, the ordering transformation is complete, as demonstrated by the ordering parameter  $S_{tet}$  of the  $L1_0$  phase (see **Table 1**), but the volume fraction of the transformed phase is still sensibly lower than unity, reaching a value close to 1 only after 60 minutes of annealing (see **Figure 9**). Mössbauer investigations [9] revealed that in these films antiphase boundaries and twin boundaries develop. As the activation energy of the lattice diffusion and of the disorder-order transformation for alloys with  $L1_0$  atomic ordering are typically of the same order of magnitude (see e.g. [42,43] for a comparison) and since the antiphase domains growth is due to the diffusion of vacancies at the antiphase boundaries [50], we can infer that the order-disorder transformation in the  $Fe_{56}Pd_{44}$  alloy is mainly due to the role of vacancies. Their diffusion at grain boundaries and at antiphase boundaries induces grain size growth, in agreement with **Figure 8**.

## Conclusions

1. The evolution of the grain size as a function of the annealing time obeys the law of normal grain growth  $D^n - D_0^n = ct$ , where  $n$  is the growth exponent equal to 8.24. Correspondingly, the average grain sizes  $\langle D \rangle$  are smaller than the critical grain size  $D_{MD}$  for multi-domain FePd particles ( $D_{MD} = 40$  nm) even after prolonged annealing (120 min). This result indicates that although the Fe<sub>56</sub>Pd<sub>44</sub> thin films alloy is characterized by a high density of defects, the growth of the grains is slow. Some defects, such as grain boundaries, were developed during processing, while others, such as twinned domains, twin boundaries, and antiphase boundaries, were developed during heat treatment.
2. The volume fraction of the transformed  $L1_0$  phase has been calculated using the following expression:

$$f_v^{L1_0}(t) = \frac{H_c(t) - H_c^{fcc}}{H_c^{L1_0} - H_c^{fcc}}$$

which we propose as valid in the generalized case of disordered  $\rightarrow$  ordered phase transformations, allowing the determination of the transformed fraction of the ordered phase from purely magnetic measurements.

3. We examined the kinetics of the  $A1 \rightarrow L1_0$  transformation in the Fe<sub>56</sub>Pd<sub>44</sub> alloy annealed at 550 °C for up to 120 min. We found that the transformation obeys to Avrami's law  $f_v(t) = 1 - \exp(-kt^{n_A})$ , where  $f_v$  is the transformed volume fraction. The obtained Avrami exponent is equal to  $n_A \sim 0.4$ . This value indicates that the phase transformation is controlled by the nucleation mechanism, with a decreasing nucleation rate, which is a consequence of rapid transformation. The low value of the Avrami exponent is due to the presence of structural defects.
4. An almost linear relationship between the coercive field and the average order parameter of the alloy has been established:  $H_c(t) = \frac{H_c^{L1_0} - H_c^{fcc}}{S_{L1_0}} S_{alloy}(t) + H_c^{fcc}$

The evolution of the coercivity with annealing time obtained through this formula is determined exclusively from available experimental data, and is in rather good agreement with the measured coercivity values. An almost linear relationship between the coercive field and the average order parameter of the alloy is therefore clearly highlighted.

## Acknowledgments

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### Data availability

All the data are available from the corresponding author upon reasonable request.

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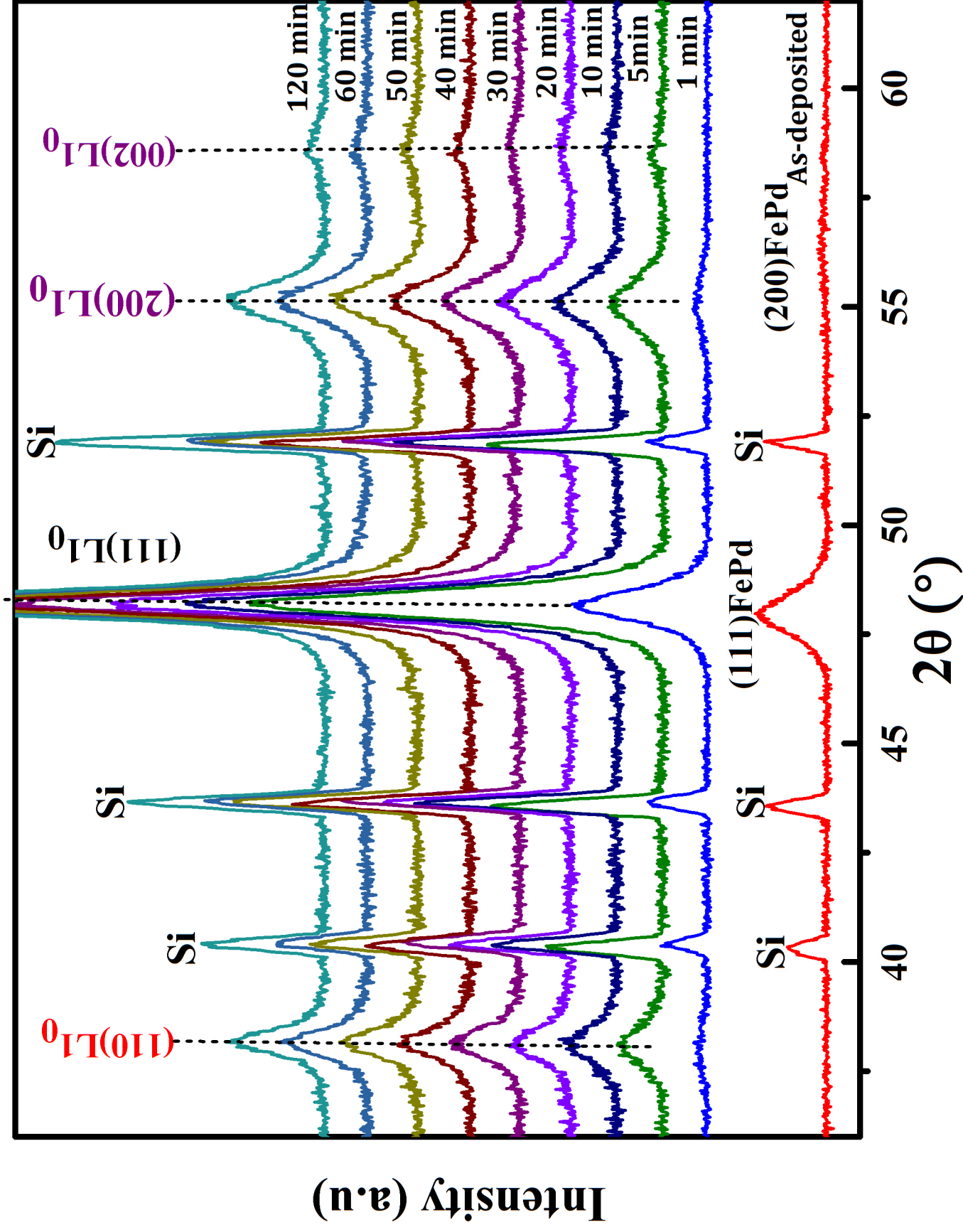
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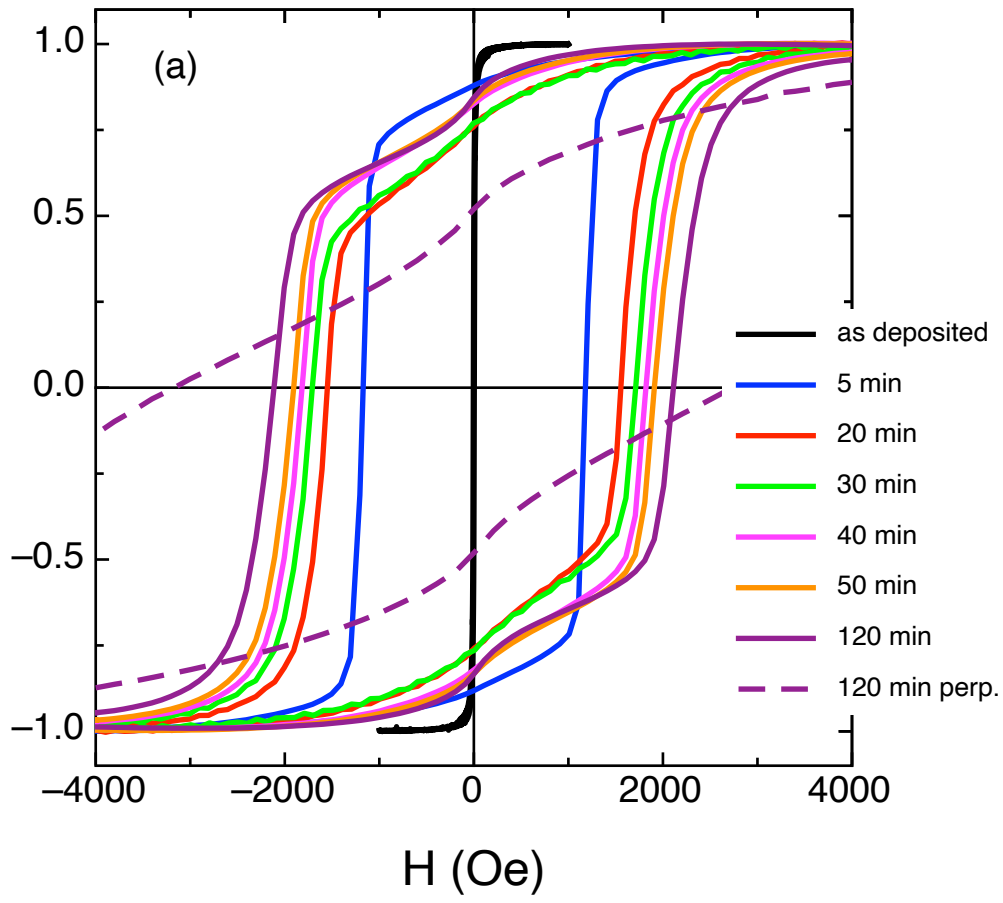
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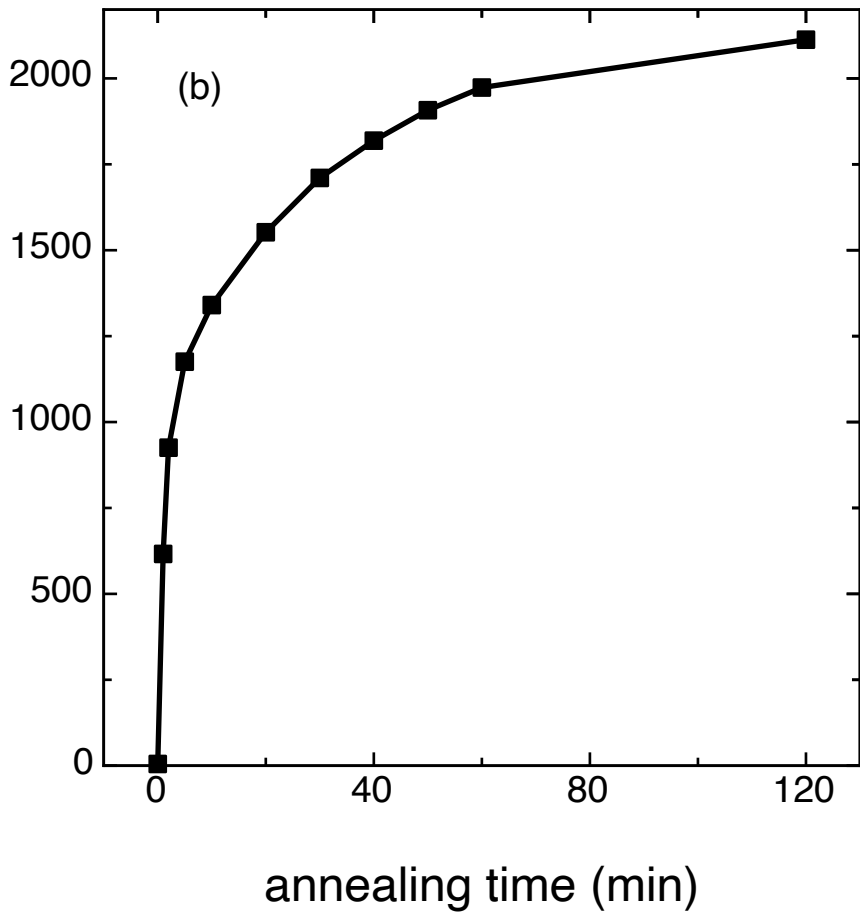


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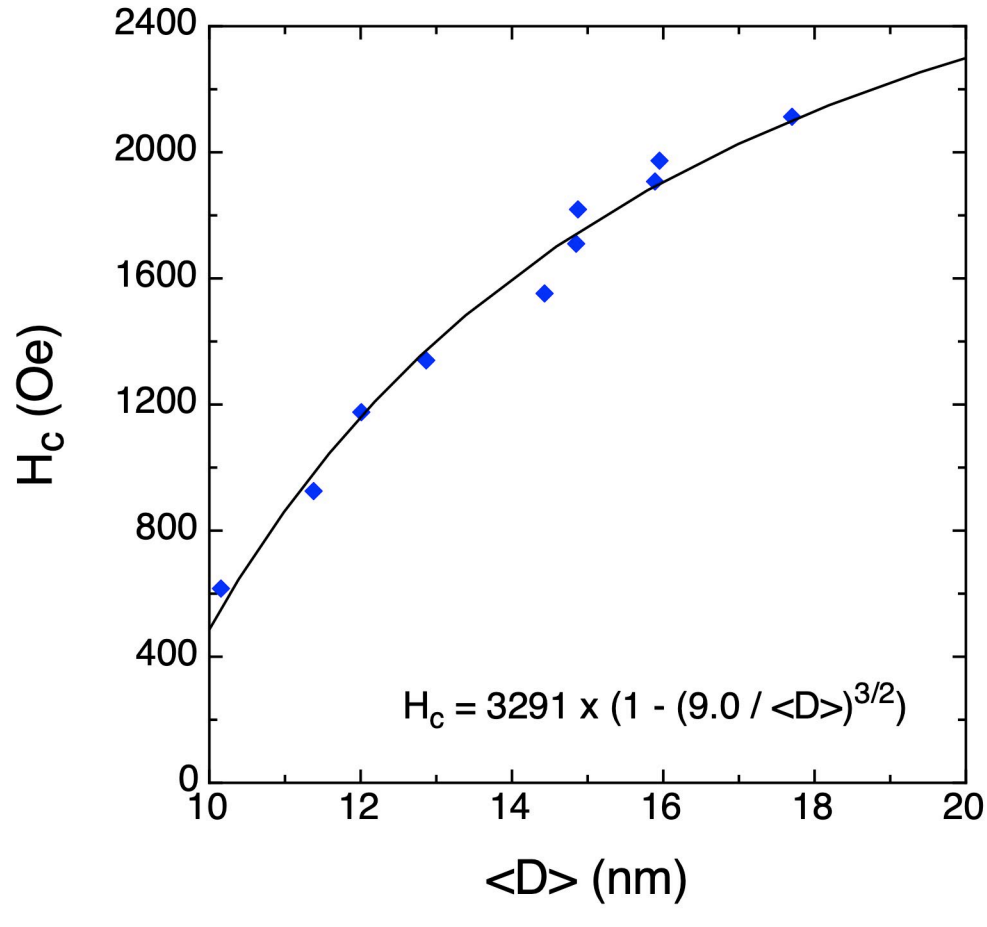




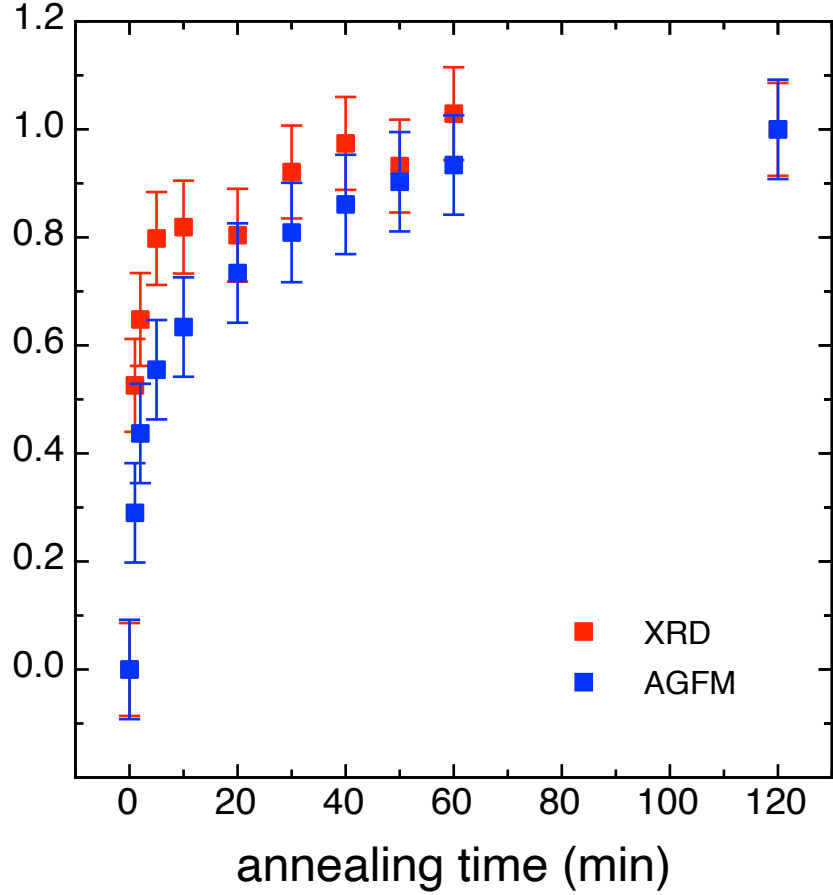
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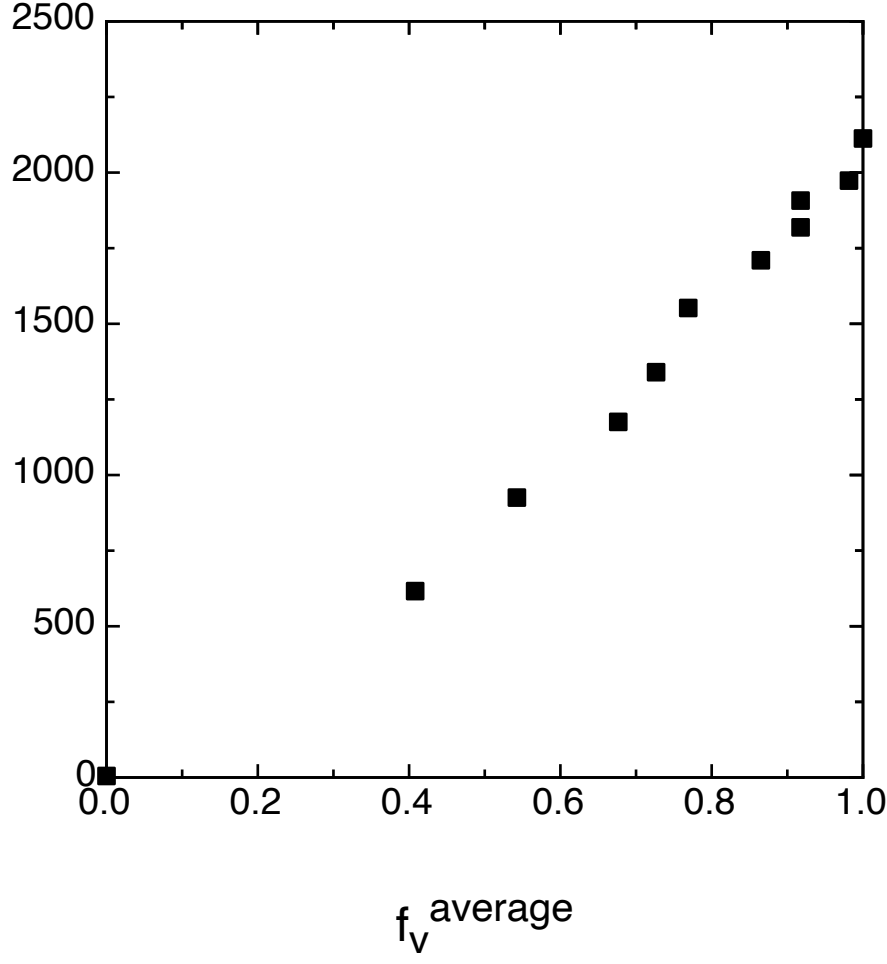
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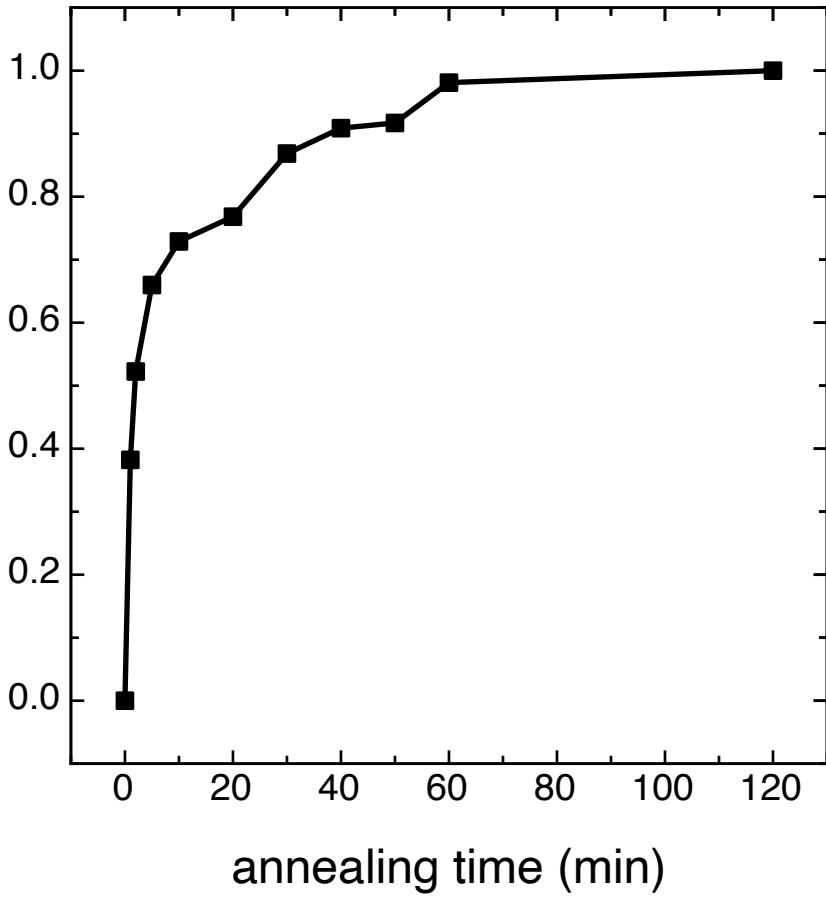
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$H_c$  (Oe)

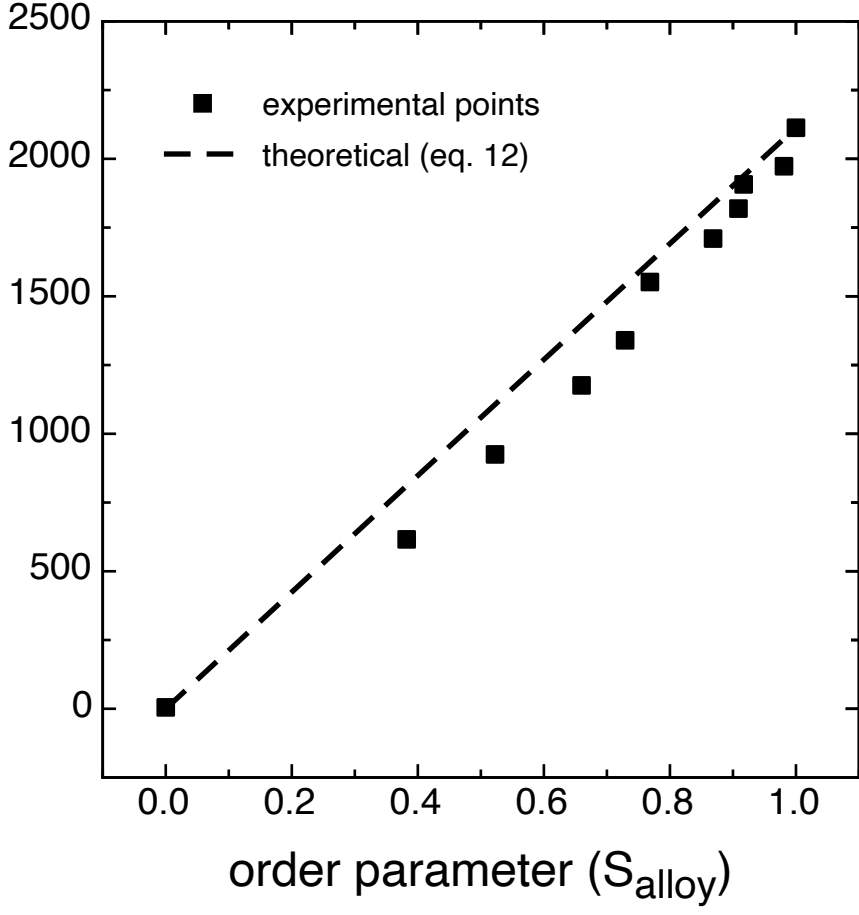


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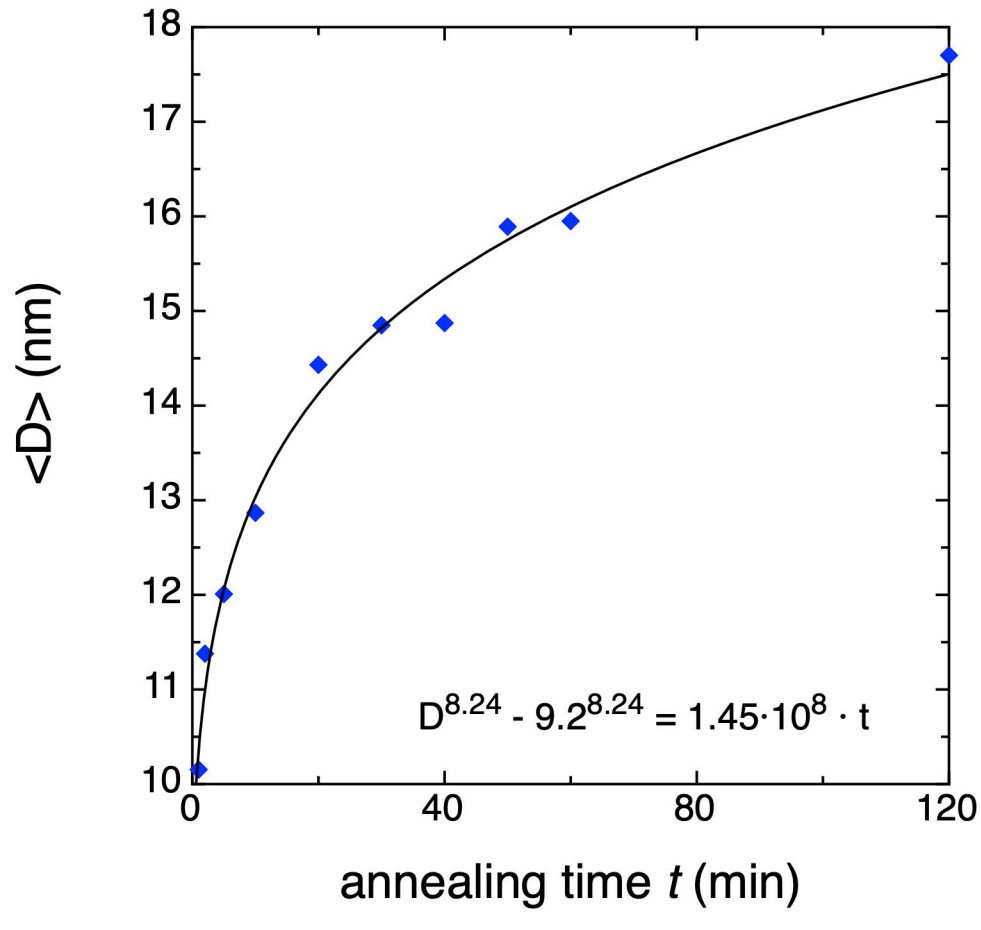
order parameter ( $S_{\text{alloy}}$ )



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