Chapter 7

The magnetoelastic coupling in $Gd_5(Si_xGe_{1-x})_4$

7.1 Introduction

In this chapter, we study the effect of the magnetic field on the magnetostructural transition in $Gd_5(Si_xGe_{1-x})_4$ alloys with $x \leq 0.5$. In particular, the variation of the transition field, H_t, with the transition temperature, T_t, is discussed as a function of x. This parameter, dH_t/dT_t, plays a key role in the scaling of ΔS, showing a different behaviour between the two compositional ranges ($x \leq 0.2$ and $0.24 \leq x \leq 0.5$) where the magnetostructural transition occurs. Moreover, dH_t/dT_t is related to the strenght of the magnetoelastic coupling: in these compounds, the value of ΔS measured when the transition is field-induced coincides with the value measured when it is induced by the application of pressure [1]. Therefore, through the Clausius-Clapeyron equation (Eq. 1.17), it is shown that (see section 6.3)

$$\frac{\Delta M}{\Delta V} = \frac{dT_t}{dH_t}\frac{dP_t}{dT_t} \quad . \tag{7.1}$$

Accordingly, a strong magnetoelastic coupling yields a small value of dH_t/dT_t.

7.2 $H - T$ diagram from magnetisation and DSC measurements

The systematic measurements of $Gd_5(Si_xGe_{1-x})_4$ samples (x=0, 0.05, 0.1, 0.18, 0.2, 0.25, 0.3, 0.365 and 0.45) are detailed in section 5.2 (magnetisation) and 5.3 (DSC under field).

From both sets of measurements -DSC and $M(H)$- the dependence of the transition temperature, T_t, on the transition field, H_t, can be evaluated independently.

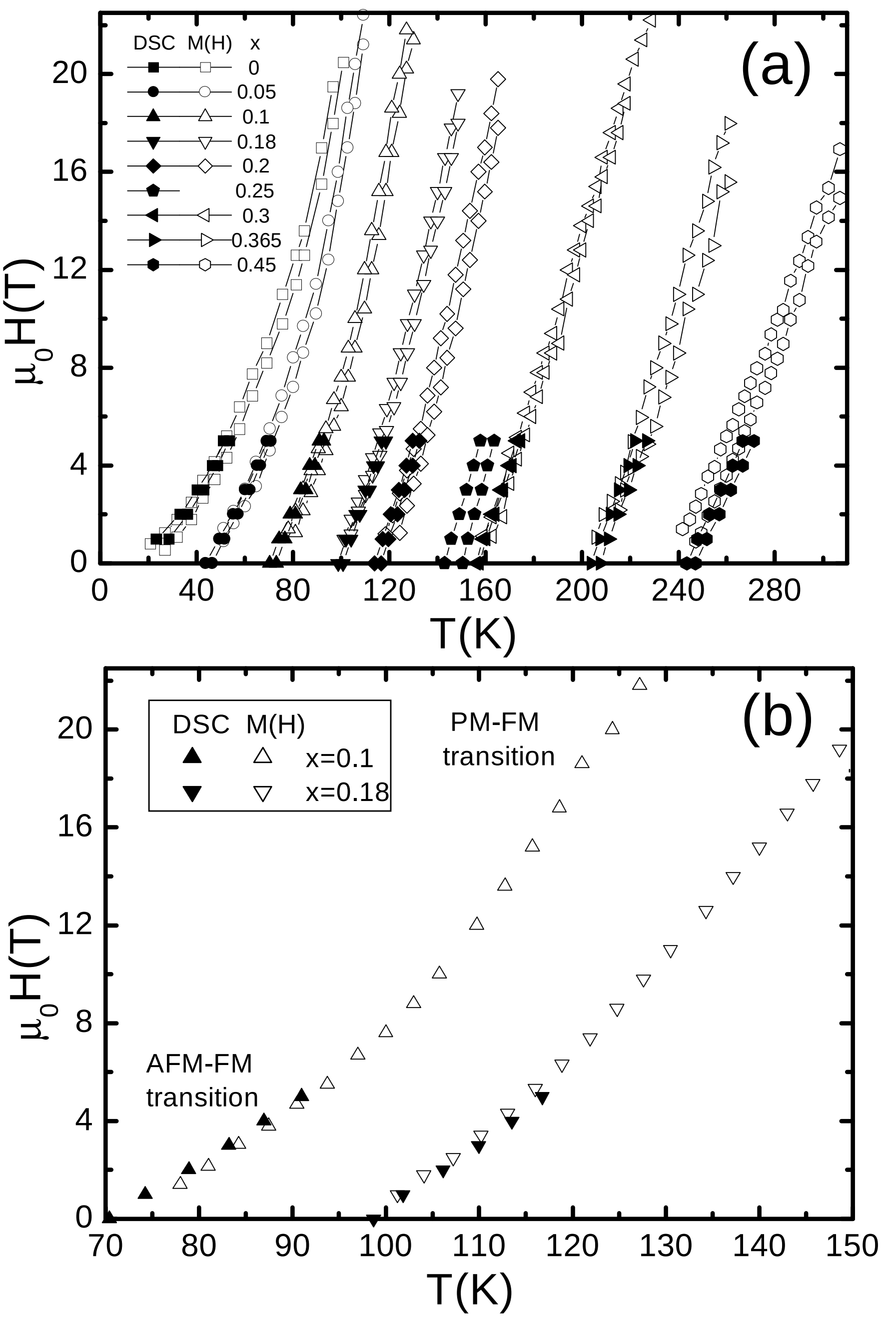

Figure 7.1: (a) Transition field, H_t, as a function of the transition temperature, T_t, for $Gd_5(Si_xGe_{1-x})_4$ (from x=0 to x=0.45) obtained from magnetisation isotherms (increasing and decreasing H) and DSC isofield data (cooling and heating). (b) Detail of panel (a) showing $H_t(T_t)$ for x=0.1 and x=0.18, on increasing H and cooling.

From magnetisation isotherms, $H_t(T)$ is defined at each temperature as the field corresponding to the inflection point within the transition region. Due to the hysteresis between increasing and decreasing field, two different values of H_t are obtained. From DSC, $T_t(H)$ is estimated at each applied field as the peak position in the dQ/dT curves. Due to the thermal hysteresis, two different values of T_t are obtained (see Table 6.1). Figure 7.1 (a) displays the transition field as a function of the transition temperature obtained from both DSC and $M(H)$ curves. Notice the good agreement between isofield and isothermal data. T_t values at zero field obtained by $M(H)$ (extrapolated) and DSC are displayed in Table 7.1 for all compositions, being in agreement with the phase diagram (Fig. 2.2). Interestingly, for $0.24 \leq x \leq 0.5$, where only the PM-to-FM transition occurs, $H_t(T_t)$ shows a linear behaviour over the whole field range, while for $x \leq 0.2$, the slope of $H_t(T_t)$ varies progressively from a low-field value (AFM-FM transition) to a high-field value (PM-FM transition). This effect is illustrated in Fig. 7.1 (b), which shows a detail of Fig. 7.1 (a) for x=0.1 and x=0.18 curves. Such a progressive change in the slope is due to the fact that, at high fields, the magnetostructural transition overlaps the second order PM-AFM transition (Fig. 5.5 (c)), giving rise to a unique PM-FM transition.

7.3 dH_t/dT_t and magnetoelastic coupling

Figure 7.2 compiles, for all compositions, the values of the slope, dH_t/dT_t, as a function of x, determined from the data in Fig. 7.1. For $x \leq 0.2$, two limiting values of dH_t/dT_t corresponding to the low and high field regimes are displayed, while a single value of dH_t/dT_t is found for $0.24 \leq x \leq 0.5$. Datum for x=0.5 is taken from Ref. [2]. We note the linear dependence of dH_t/dT_t on x, which is decreasing for the PM-FM transition (solid line in Fig. 7.2), while it is increasing for the AFM-FM transition (dashed line in Fig. 7.2). Both lines meet at the composition range where the second-order transition disappears ($0.2 < x < 0.24$), in agreement with the phase diagram (Fig. 2.2 and Ref. [3]). The value of dH_t/dT_t for x=0 at high fields is lower than expected because a field higher than 23 T (the maximum available in the present work) must be applied to fully induce the PM-FM transition. Values of dH_t/dT_t obtained from DSC and $M(H)$ measurements for all compositions are displayed in Table 7.2 and compared with values given in literature.

The strength of the magnetoelastic coupling is associated with the field dependence of T_t (*i.e.*, a strong magnetoelastic coupling yields a small value of dH_t/dT_t) as demonstrated in the introduction of this chapter. Consequently, the decrease in dH_t/dT_t with increasing x for the PM-FM transition indicates a strengthening of the magnetoelastic coupling. This may be explained by considering that FM

x	ID	Heat T.	$T_t(H=0)$ (K)			
			DSC		$M(H)$	
			cool.	heat.	incr. H	decr. H
0	#1	NO	~13*	~20*	~15*	~22*
0.05	#1	T4+Q	43.8	46.5	44.5	45.9
0.1	#1	NO	70.4	73.1	73.2	76.5
0.18	#1	NO	-	-	103.7	106.1
		T4	98.7	100.9	97.8	99.5
0.2	#1	NO	113.9	116.6	114.6	120.5
0.25	#2	NO	143.0	150.5	-	-
0.3	#2	NO	169.7	177.5	-	-
		T4+Q	156.1	157.3	156.6	160.4
0.365	#3	NO	200.7	204.5	204.3	209.3
0.45	#7	NO	247.2	252.3	245.1	252.2
		T4	243.5	247.1	238.0	244.9

Table 7.1: Transition temperature at zero field, $T_t(H=0)$, at the first-order transition obtained by extrapolating $T_t(H)$ obtained from $M(H)$, and also by DSC at H=0 for all measured samples. *These values are valid after the low-temperature FM phase has been induced irreversibly in the x=0 compound by the application of a high enough magnetic field (see section 2.4.1).

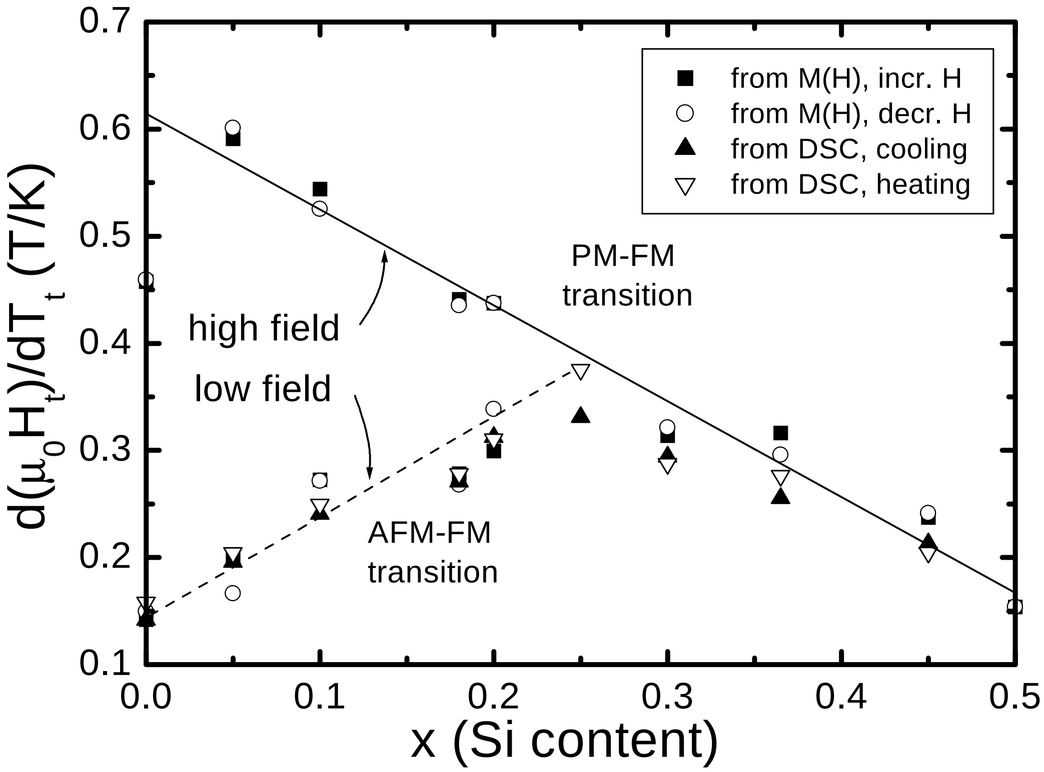

Figure 7.2: Slope of $H_t(T_t)$ calculated from data in Fig. 7.1. For x=0.25, 0.3, 0.365, 0.45 and 0.5 (the latter from Ref. [2]) a single slope is obtained, which corresponds to the PM-FM transition. For x=0, 0.05, 0.1, 0.18 and 0.2 two limiting slopes are obtained: a low-field value (associated with the AFM-FM transition) and a high-field value (associated with the PM-FM transition). Solid and dotted lines are a guide to the eye.

x	$d(\mu_0 H_t)/dT_t$ (T/K)						
	DSC		M(low H)		M(high H)		
	cool.	heat.	inc. H	dec. H	inc. H	dec. H	Literature
0	0.142	0.158	0.142	0.149	0.458	0.460	0.125 [4]
0.05	0.196	0.204	0.202	0.166	0.591	0.601	
0.08							0.294 [5]
0.1	0.241	0.250	0.273	0.271	0.544	0.525	0.27 [3], 0.26 [3]
0.18	0.271	0.278	0.278	0.268	0.441	0.435	
0.2	0.312	0.311	0.299	0.338	0.437	0.437	
			$M(H)$				
			inc. H		dec. H		
0.25	0.331	0.376	-		-		
0.3	0.294	0.287	0.314		0.321		
0.365	0.255	0.277	0.316		0.296		
0.375							0.28 [6, 7], 0.25 [8],
0.43							0.23 [9]
0.45	0.213	0.205	0.237		0.241		0.21 [1], 0.22 [10]
0.5							0.154 [2], 0.18 [11], 0.14 [12]

Table 7.2: dH_t/dT_t obtained from $M(H)$ and DSC under magnetic field for all measured samples. For $x \leq 0.2$, two limiting values are obtained from magnetisation data. Values from different references are also compiled for comparison.

exchange interactions are stronger for increasing x, as suggested by the magnetic phase diagram, where T_t increases linearly with x (Fig. 2.2). The fact that dH_t/dT_t for the PM-FM transition has continuous behaviour, although the PM phase is monoclinic for $0.24 \leq x \leq 0.5$ and orthorhombic-II for $x \leq 0.2$, suggests that the magnetoelastic coupling is weakly dependent on the actual crystallographic structure. Concerning the AFM-FM transition, and taking into account that the structural transition is the same (for $x \leq 0.2$) or very similar (for $0.24 \leq x \leq 0.5$) to that occurring in the PM-FM case, the increase in dH_t/dT_t with x may be related to the fact that the transition involves two ordered magnetic phases (FM and AFM). Fig. 7.2 thus summarizes the behavior of the first-order transition in $Gd_5(Si_xGe_{1-x})_4$ as a function of x, T and H.

The behaviour of dH_t/dT_t with x is relevant in the scaling of $|\Delta S|$ which appears in $Gd_5(Si_xGe_{1-x})_4$ alloys (Chapter 6): taking into account the Clausius-Clapeyron equation and as ΔM always decreases with T, $|\Delta S|$ thus increases with T_t for $x \leq 0.2$ when the AFM-FM transition takes place, due to the larger increase of dH_t/dT_t with T_t as compared to the decrease in ΔM with T_t. In contrast, $|\Delta S|$ decreases with T_t for the PM-FM transition, since the increase in dH_t/dT_t with T_t is not large enough as to overcome the decrease in ΔM. Therefore, although the main feature of the scaling of $|\Delta S|$ with T_t is not only determined by dH_t/dT_t vs T_t, the particular dependence of dH_t/dT_t on x and H enables the scaling.

7.4 Conclusions

The variation of the transition field with the transition temperature, dH_t/dT_t, has been studied in $Gd_5(Si_xGe_{1-x})_4$ for all the range of compositions where the first-order transition occurs, $0 \leq x \leq 0.5$. Taking into account the behaviour of dH_t/dT_t as a function of x and that ΔM decreases monotonously with T_t, it is shown that dH_t/dT_t governs the scaling of ΔS with T_t reported in Chapter 6, giving further evidence that the origin of this scaling is the magnetoelastic nature of the transition. Moreover, two distinct behaviors for dH_t/dT_t have been found on the two compositional ranges where the magnetostructural transition occurs, thus showing the difference in the strength of the magnetoelastic coupling of this system.

Bibliography

[1] L. Morellon, P. A. Algarabel, M. R. Ibarra, J. Blasco, B. García-Landa, Z. Arnold, and F. Albertini, Phys. Rev. B **58**, R14721 (1998).

[2] A. Giguère, M. Földeàki, B. Ravi Gopal, R. Chahine, T. K. Bose, A. Frydman, and J. A. Barclay, Phys. Rev. Lett. **83**, 2262 (1999).

[3] L. Morellon, J. Blasco, P. A. Algarabel, and M. R. Ibarra, Phys. Rev. B **62**, 1022 (2000).

[4] E. M. Levin, K. A. Gschneidner, Jr., and V. K. Pecharsky, Phys. Rev. B **65**, 214427 (2002).

[5] V. K. Pecharsky and K. A. Gschneidner, Jr., Adv. Cryog. Eng. **43**, 1729 (1998).

[6] E. M. Levin, V. K. Pecharsky, and K. A. Gschneidner, Jr., Phys. Rev. B **62**, R14625 (2000).

[7] E. M. Levin, V. K. Pecharsky, and K. A. Gschneidner, Jr., J. Magn. Magn. Mater. **231**, 135 (2001).

[8] E. M. Levin, V. K. Pecharsky, K. A. Gschneidner, Jr., and P. Tomlinson, J. Magn. Magn. Mater. **210**, 181 (2000).

[9] V. K. Pecharsky and K. A. Gschneidner, Jr., Appl. Phys. Lett. **70**, 3299 (1997).

[10] L. Morellon, J. Stankiewicz, B. García-Landa, P. A. Algarabel, and M. R. Ibarra, Appl. Phys. Lett. **73**, 3462 (1998).

[11] V. K. Pecharsky and K. A. Gschneidner, Jr., Phys. Rev. Lett. **78**, 4494 (1997).

[12] E. M. Levin, V. K. Pecharsky, and K. A. Gschneidner, Jr., Phys. Rev. B **60**, 7993 (1999).