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PRESSURE EFFECT ON THE CURIE TEMPERATURE OF  $Dy_2Fe_{17-y}Al_y$  COMPOUNDS †

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ABSTRACT

The pressure variation of the Curie temperature  $T_c$  has been measured up to 15 kbar by means of the ac susceptibility. The pressure changes of  $T_c$  are negative, large and not linear, the  $T_c(p)$  curves being concave. The initial values of  $dT_c/dp$  are remarkably large being equal to  $-6.0 \text{ Kkbar}^{-1}$  for  $Dy_2Fe_{17}$ .

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**I n t r o d u c t i o n.** Large values for the spontaneous magnetostriction ( about  $1.6 \cdot 10^{-2}$  ) were observed by Givord *et al* [ 1 ] for  $Y_2Fe_{17}$  and  $Gd_2Fe_{17}$  and by Radwański for  $Dy_2Fe_{17}$  [ 2 ] . The magneto-volume effect ( including the pressure dependence of the magnetic parameters ) is intensively investigated on both crystalline and amorphous materials because it is believed to be a key for understanding the origin of magnetism in 3d metals ( Heine *et al* [ 3 ] ).

**E x p e r i m e n t.** Samples of  $Dy_2Fe_{17-y}Al_y$  series were prepared in an arc-melting furnace under argon atmosphere with Dy of 3N purity and Fe and Al of spectral purity. X-ray diffraction measurements confirmed that the material consisted of one single phase. The pressure dependence of the Curie temperature was determined from the temperature dependence of the ac susceptibility. The measurements were carried out at various pressures up to 15 kbar ( 1.5 GPa ) in the Politechnical University, Cracow. The temperature of the samples was measured with a copper-constantan thermocouple. The susceptibility was measured by an inductance magnetometer placed inside the Be-Cu pressure cell. The pressure vessel was connected by the capillary tube with the gas compressor IF-012A made by UNIPRESS Warsaw, Poland. Gaseous helium was used as the pressure transmitting medium. The pressure was measured by means of a manganine pick-up coil. Powdered samples ( about  $10^{-4} \text{ kg}$  ) were placed inside a teflon holder which was mounted in a secondary coil. This coil was placed in a weak variable magnetic field of about  $10^{-3} \text{ T}$  and frequency 273 Hz.

**R e s u l t s.** The pressure dependence of the ac susceptibility was measured for five samples with  $y = 0, 1.5, 3, 5$  and  $6$  of the  $Dy_2Fe_{17-y}Al_y$  series. The ac susceptibility starts to increase with increasing temperature and afterwards drops through a maximum just before the Curie point. The maxima are very broad for  $Dy_2Fe_{17}$  but become more pronounced with increasing Al concentration. The sharp drop confirms the single phase of the measured samples. The Curie temperature,  $T_c$ , is defined by the inflection point of the susceptibility curve. The value for  $T_c$  of  $Dy_2Fe_{17}$  obtained in this way is in satisfactory agreement with the value of the Curie temperature given by Buschow [4] but is 20 K lower compared to the value reported by Piusa *et al* [5] from the temperature dependence of the magnetization. The external pressure does not change the shape of the susceptibility curve but causes a significant reduction of  $T_c$ . The shift of  $T_c$  versus pressure is shown in Fig. 1. The reduction of  $T_c$  over 15 kbar is remarkably large and amounts to 73 K for  $Dy_2Fe_{17}$ . The pressure dependence is nonlinear; with the increase of pressure the reduction decreases. The same behaviour was observed for  $Er_2Fe_{17}$  by Brouha *et al* [6]. Moreover, with the increase of the Al concentration the Curie temperature becomes less sensitive to pressure. The initial slope of the  $T_c(p)$  curves and the average values,  $(T_c(15 \text{ kbar}) - T_c(0))/15 \text{ kbar}$ , are presented in Fig. 2 together with  $dT_c/dp$  and  $d \ln T_c / d \ln V$  as a function of  $y$ .

The Stoner-Wohlfarth theory for itinerant-electron ferromagnetism [7] supplies a relation for the pressure dependence of  $T_c$ :

$$dT_c/dp = 5/3 \kappa T_c - \alpha / T_c \quad (1)$$

where  $\kappa$  is the compressibility and  $\alpha$  a coefficient being dependent on microscopic parameters of the electronic band structure. This relation indicates that the pressure dependence of  $T_c$  decreases with increa-

sing  $T_c$ -values since the second term usually dominates for Fe alloys. Eq. (1) holds in the  $Dy_2Fe_{17-y}Al_y$  system up to  $y = 3$ . A value of  $2400 \text{ K}^2 \text{ kbar}^{-1}$  for the parameter  $\alpha$  is deduced from the experimental values of  $dT_c/dp$  in this region. This value is much larger than the value of  $1700 \text{ K}^2 \text{ kbar}^{-1}$  for a typical Invar system as  $Fe_{65}Ni_{35}$ , for instance [8]. The relation (1) is not applicable for higher Al concentrations. There, both the Curie temperature and the value of  $dT_c/dp$  significantly decrease. A large value for  $dT_c/dp$  of  $-3 \text{ K kbar}^{-1}$  is still present for  $y = 6$  although a magnetic contribution to the thermal expansion is hardly detectable [9]. In Table 1, some characteristic parameters are collected: the spontaneous magnetostriction, the Curie temperature and its pressure coefficient, the high-field susceptibility and the coefficient  $\alpha$ . It is seen that the compound  $Dy_2Fe_{17}$  is characterized by a large value of  $dT_c/dp$ . The large values for the high-field susceptibilities, evaluated from magnetization measurements up to 7 T on polycrystalline samples [5] are mainly due to effects arising from the magnetocrystalline anisotropy [14] and/or the bending of the antiparallel moment configuration [15].

We note that eq. (1) is not able to describe correctly the pressure dependence of  $T_c$ . By integrating this equation, neglecting the first, usually small, term one gets  $T_c(p)/T_c(0) = (1 - p/p_{\text{crit}})^{0.5}$ . However, the experimental data do not follow this expression for all investigated compounds of the  $Dy_2Fe_{17-y}Al_y$  system.

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Table 1. Some magnetovolume effect parameters of  $Dy_2Fe_{17-y}Al_y$  system together with those of other amorphous and crystalline Invar alloys. The values marked by  $^+$  were accounted with  $K = 0.8 \text{ Mbar}^{-1}$  [ 6 ] and by  $^{++}$  with  $K = 0.6 \text{ Mbar}^{-1}$  like for Fe. The parameter  $\alpha$  was calculated from eq. ( 1 ).

Compound	$T_c$ (K)	$dT_c/dp$ (Kkbar $^{-1}$ )	$d \ln T_c / d \ln V$	$\omega_m$ (0)	$\chi_{hf}$ (Am $^2$ /Tkg)	$\alpha$ (K $^2$ kbar $^{-1}$ )	References
$Dy_2Fe_{17}$	371	- 6.0	20.2 $^+$	16.2 $10^{-3}$	0.90	2400 $^+$	this work, 9,5
$Dy_2Fe_{15.5}Al_{1.5}$	428	- 5.0	14.6 $^+$	10.9	0.90	2400 $^+$	this work, 9,5
$Dy_2Fe_{14}Al_3$	465	- 5.0	13.4 $^+$	7.7	0.90	2560 $^+$	this work, 9,5
$Dy_2Fe_{12}Al_5$	391	- 4.0	12.8 $^+$	2.5	1.50	1760 $^+$	this work, 9,5
$Dy_2Fe_{11}Al_6$	273	- 3.0	13.7 $^+$	0.9	2.20	910 $^+$	this work, 9,5
$Er_2Fe_{17}$	310	- 4.1	16.5	-	-	1400 $^+$	6
$Fe_{74}Ni_{26}$	491	- 3.5	19.6 $^{++}$	18.0	0.18	1900 $^{++}$	8, 10, 11
a- $Fe_{75}Ti_{25}$	238	- 6.1	32.0 $^{++}$	4.5	0.42	1500 $^{++}$	11
a- $Fe_{83}B_{17}$	600	- 2.6	5.4 $^{++}$	20.0	0.11	1920 $^{++}$	11
$Fe_{72}Pt_{28}$	505	- 3.3	8.2 $^{++}$	14.0	0.11	1900 $^{++}$	12
$ZrFe_2$	630	- 2.0	4.0 $^{++}$	10.0	0.03	1660 $^{++}$	6, 13

FIGURES CAPTIONS

Fig.1. Pressure shift of the Curie temperature of  $\text{Dy}_2\text{Fe}_{17-y}\text{Al}_y$  system:  $\bullet$ ,  $y=0$ ;  $\circ$ ,  $y=1.5$ ;  $+$ ,  $y=3$ ;  $\ominus$ ,  $y=5$  and  $\circ$ ,  $y=6$ .

Fig.2. The pressure parameters of the  $\text{Dy}_2\text{Fe}_{17-y}\text{Al}_y$  system versus the Al concentration,  $y$ : a) the pressure coefficient of  $T_c$ , initial ( $\bullet$ ) and the average value for the pressure between 0 and 15 kbar ( $\circ$ ); b) the pressure and logarithmic volume dependence of  $T_c$  calculated from the initial ( $\ominus$  and  $\bullet$ ) and the average (0-15 kbar) relative change of  $T_c$  ( $\circ$ ).

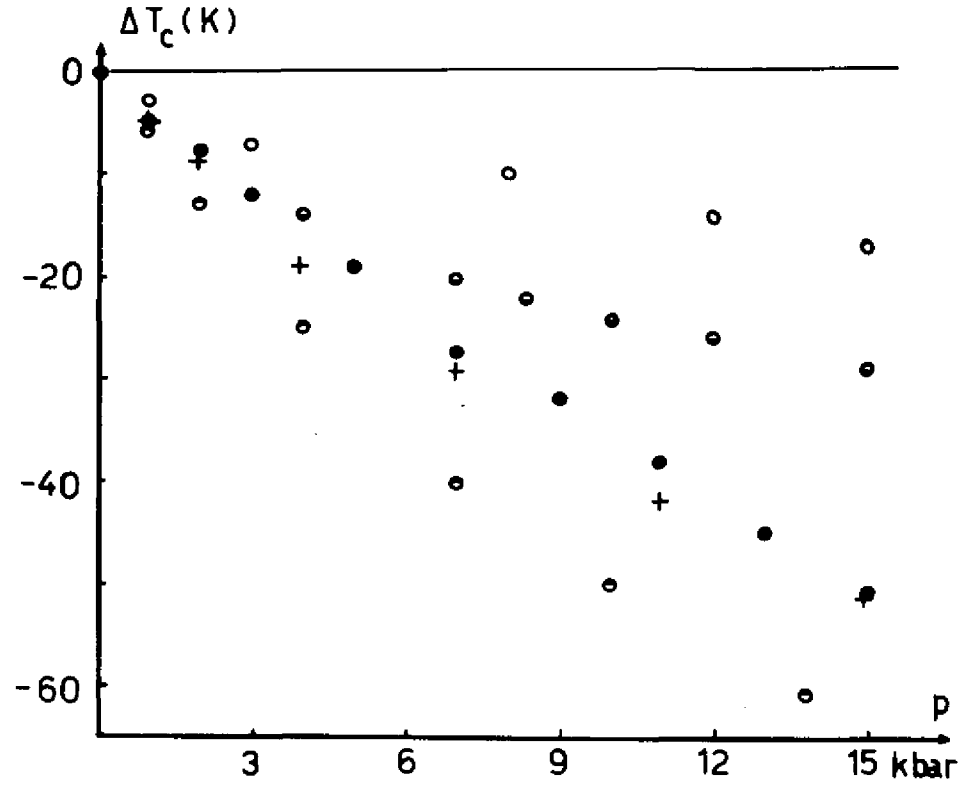


Fig.1

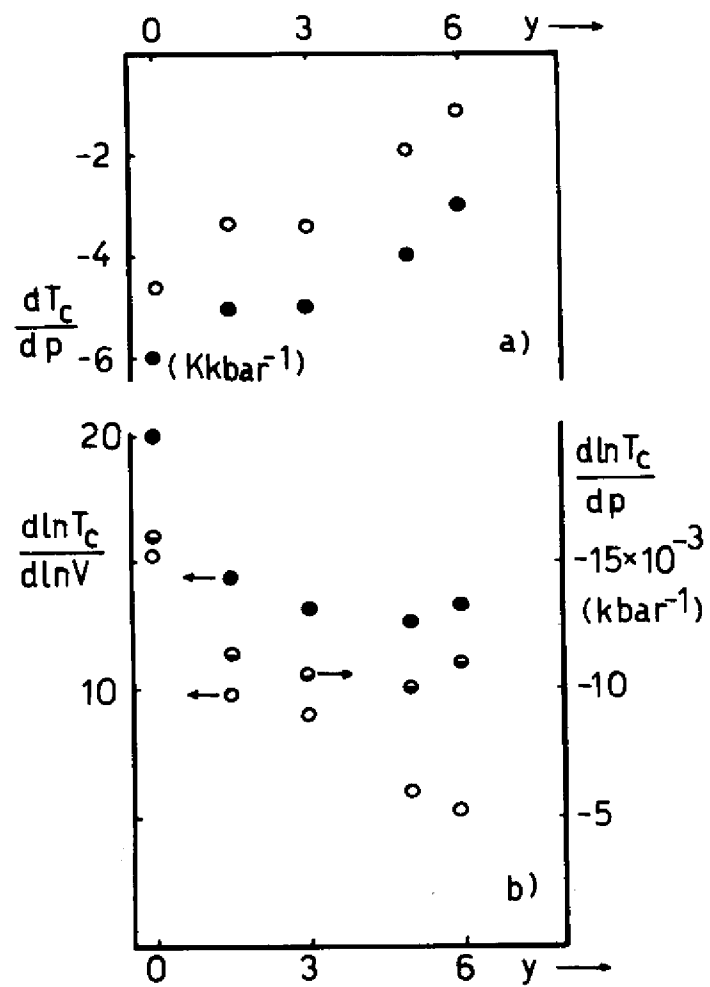


FIG. 2

