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## Magnetization and magnetostriction of $(\text{Co}_{1-x}\text{Ni}_x)_{75}\text{Si}_{15}\text{B}_{10}$ amorphous alloys ( $x = 0, 0.4$ )

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**Résumé.** — La variation thermique de l'aimantation  $M$  et de la magnétostriction  $\lambda_s$  a été étudiée dans deux alliages amorphes de composition  $(\text{Co}_{1-x}\text{Ni}_x)_{75}\text{Si}_{15}\text{B}_{10}$ , afin d'analyser  $\lambda_s(T)$  à l'aide des fonctions de corrélation de spin à « un-ion » et à « deux-ions ». L'alliage de cobalt ( $x = 0$ ) présente un vrai comportement à un-ion de 0 K jusqu'à  $T_c$ , tandis que l'alliage Co-Ni ( $x = 0,4$ ) s'écarte notablement de ce comportement au-dessus de 220 K : cette dépendance en température surprenante confirme des données expérimentales antérieures obtenues dans cette même série.

**Abstract.** — The temperature dependence of the magnetization  $M$  and of the magnetostriction  $\lambda_s$  has been investigated in two amorphous  $(\text{Co}_{1-x}\text{Ni}_x)_{75}\text{Si}_{15}\text{B}_{10}$  alloys, in order to analyse  $\lambda_s(T)$  in terms of one-ion and two-ion spin correlation functions. The Co alloy ( $x = 0$ ) exhibits a pure single-ion behaviour from 0 K up to  $T_c$ , while the Co-Ni alloy ( $x = 0.4$ ) deviates markedly from this behaviour above 220 K : this surprising temperature dependence is in agreement with previous high temperature data in this series, but cannot be ascribed to a 2-ion contribution.

### 1. Introduction.

The temperature dependence of the magnetostriction constant  $\lambda_s$  in most amorphous alloys has been found to be correctly described by the one-ion spin correlation function,  $\hat{I}_{5/2}[\mathcal{L}^{-1}(m)]$  where  $m = M(T)/M(0)$  is the reduced magnetization,  $\mathcal{L}$  is the Langevin function, and  $\hat{I}_{5/2}$  is a modified hyperbolic Bessel function [1]. The agreement was perfect for pure iron-metalloid amorphous alloys  $\text{Fe}_{100-y}\text{M}_y$ . In cobalt-rich alloys,  $(\text{Co}_{1-x}\text{Fe}_x)_{80}\text{M}_{20}$ , O'Handley pointed out that the single-ion contribution  $\lambda_1$  to  $\lambda_s$  is just the linear combination of a positive term arising from Fe and a negative one from Co atoms, but he suggested that a two-ion contribution,  $\lambda_2$ , does exist also and becomes relatively very important when  $\lambda_1$  cancels (for  $x$  values near 0.06). Following Callen [2], he wrote

$$\lambda_s(T) = \lambda_1 \times \hat{I}_{5/2}[\mathcal{L}^{-1}(m)] + \lambda_2 m^2. \quad (1)$$

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Up to now, all is correct but unfortunately equation (1) has never been checked correctly : first, the temperature dependence of the magnetostriction constant of the amorphous alloy  $\text{Co}_{80}\text{B}_{20}$  was only discussed qualitatively as a competition between a negative single-ion contribution and a positive two-ion one [1]. In fact, the data taken from reference [1] are consistent with a negative single-ion term, and (if it exists) a much smaller negative two-ion term up to 550 K, while strong anomalies appear above as will be shown in figure 5. Later on, a quantitative analysis of equation (1) was attempted by a number of researchers : assuming  $\hat{I}_{5/2}[\text{L}^{-1}(m)] \approx m^3$ , they have plotted  $\lambda_s(T)/m^2$  against  $m$  :

$$\lambda_s(T)/m^2 = \lambda_1 m + \lambda_2 . \quad (2)$$

Such an analysis was used e.g. for discussing the temperature dependence of  $\lambda_s$  in  $(\text{Co}_{1-x}\text{Ni}_x)_{75}\text{Si}_{15}\text{B}_{10}$  alloys above room temperature [3]. Surprisingly, these data were more or less in agreement with the predictions of equation (2), although this equation was certainly irrelevant to this temperature range since near  $T_c$ ,  $\hat{I}_{5/2}[\text{L}^{-1}(m)] \approx 3/5 m^2$ . Hence,  $\lambda_s(T)/m^2$  should be equal to  $\lambda_1 m + \lambda_2$  at very low temperature and equal to  $3/5 \lambda_1 + \lambda_2$  (it should no longer vary) near  $T_c$ .

So, the aim of the present work was

- i) to study the temperature dependence of  $\lambda_s$  in a pure Co-metalloid amorphous alloy ( $x = 0$ ), in order to complete down to 4.2 K the high temperature data taken from reference [3] and to compare with previous experiments ;
- ii) to repeat the same study from 4.2 K up to  $T_c$  in the alloy with a nickel content  $x = 0.4$ , in order to check the surprising behaviour observed in reference [3] with the similar alloys ( $x = 0.34, 0.37, 0.39, 0.41$ ).

## 2. Experimental.

The melt alloys of the expected compositions have been rapidly quenched into ribbons  $30 \mu\text{m} \times 10.2 \text{ mm}$  in cross section. Both surfaces have been checked to be amorphous. The chemical analysis gave a composition slightly different from the nominal one for  $x = 0$ , namely  $\text{Co}_{74.7}\text{Si}_{14.6}\text{B}_{10.7}$ .

For analysing correctly equation (1), we have measured in the same samples the thermal variations of the magnetic moment  $\sigma$  by the extraction method, of the linear thermal expansion coefficient  $\alpha_T$  and of the magnetostriction constant  $\lambda_s$  using a tube-type capacitance dilatometer. Two methods were used for measuring  $\lambda_s$  : first, the cylinder method [4] gave us accurately the forced magnetostriction  $d\lambda_{\parallel}/dH$  and the longitudinal magnetostriction  $\lambda_{\parallel}$  ( $\lambda_{\perp}$  could not be measured due to the large demagnetizing field) ; then TSUYA's method [5] using a thin parallelepiped gave us  $\lambda_{\parallel}$  and  $\lambda_{\perp}$ , hence  $\lambda_s = 2/3(\lambda_{\parallel} - \lambda_{\perp})$ .

The magnetic moment has been measured from 4.2 K up to 380 K in a magnetic field up to 4 MA/m. The plot  $\text{Ln}(\sigma)$  vs.  $\text{Ln}(H_i)$  gives a straight line for  $T = T_c = 320 \text{ K}$  and a critical exponent  $\delta = 4.6$ . The plot from  $T_c$  up to 380 K of  $-\partial \text{Ln}(T)/\partial \text{Ln}(\chi T)$  vs.  $T$  where  $\chi$  is the magnetic susceptibility gives a straight line as predicted in reference [6], defining  $T_c = 319 \text{ K}$  and a critical exponent  $\gamma = 1.54$ . Hence, we deduce  $\beta = \gamma/(\delta - 1) = 0.43$ . These values are consistent with  $\beta = 0.42 \pm 0.04$  and  $\delta = 5 \pm 0.4$ , the critical exponents given in the literature for  $(\text{Co}_{0.6}\text{Ni}_{0.4})_{75}\text{P}_{16}\text{B}_6\text{Al}_3$  [7]. So, the spontaneous magnetic moment was derived near  $T_c$  by plotting  $\sigma^{3.6}$  vs.  $H/\sigma$ . The temperature dependence of the magnetic moment is given in figure 1 for the two samples, together with data taken from reference [3] assuming the density,  $d$ , to be 8 460 and 8 340  $\text{kg m}^{-3}$  respectively for  $x = 0$  and 0.39. The consistency of the two sets is satisfactory except at room temperature where our values are slightly larger.

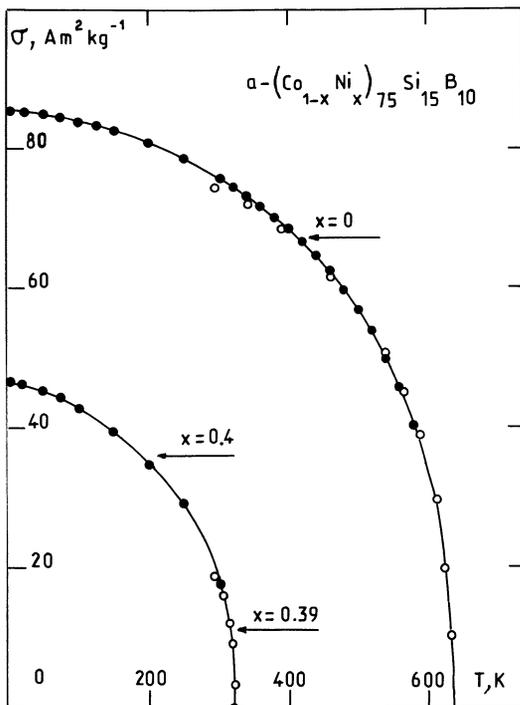


Fig. 1. — Temperature dependence of the magnetic moment in the amorphous  $(Co_{1-x}Ni_x)_{75}Si_{15}B_{10}$  alloys. Open circles : data taken from reference [3], for  $x = 0$  and  $0.39$ . Full circles : our data for  $x = 0$  and  $0.4$ .

The linear thermal expansion coefficient  $\alpha_T$  has been measured from 20 K to 300 K and is plotted vs. the temperature in figure 2a for the sample with  $x = 0$  ; the expansivity is given in figure 2b. Above 300 K,  $(\delta\ell/\ell)_T$  is obtained by extrapolating the low temperature curves. Using these data together with those taken from figure 1, it is possible to derive accurately the temperature dependence of the magnetization :

$$M(T) = \sigma(T) \times d_0 \times 1/[1 + 3 \times (\delta\ell/\ell)_T] \tag{3}$$

where  $d_0$  is the density at 0 K and  $(\delta\ell/\ell)_T$  the expansivity at the temperature  $T$  taken from figure 2.

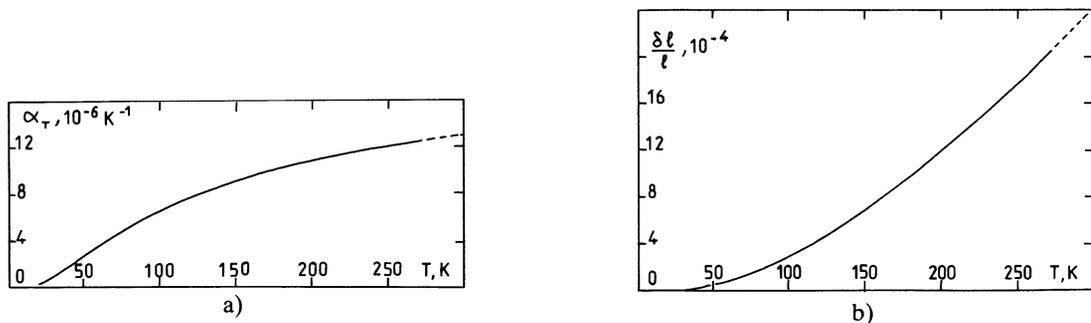


Fig. 2. — Temperature dependence of the thermal expansion for  $Co_{75}Si_{15}B_{10}$  ; a : the linear thermal expansion coefficient ; b : the expansivity measured from 20 K up to room temperature.

The thermal variation of the magnetostriction coefficient  $\lambda_s$  of both alloys is given in figure 3. For comparison, we have given also the temperature dependence of  $\lambda_{||}$  as measured with the cylinder method : due to the anisotropy of the demagnetized state,  $|\lambda_{||}|$  appears to be about 20 % larger than  $|\lambda_s|$  and its temperature dependence is slightly different also. The one-ion model (Eq. (1) with  $\lambda_2 = 0$ ) predicts the temperature dependence given by the full line in figure 3.

Last, the forced magnetostriction  $d\lambda/dH$  is given in figure 4 as a function of the temperature. Data for  $x = 0.4$  indicate a  $\Lambda$ -like peak near  $T_c = 320$  K, while for  $x = 0$  data do not vary significantly up to 300 K, due to the higher Curie Temperature,  $T_C \approx 640$  K.

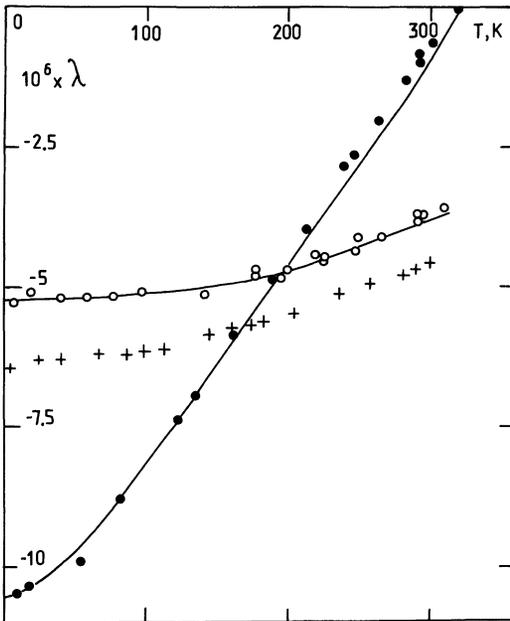


Fig. 3

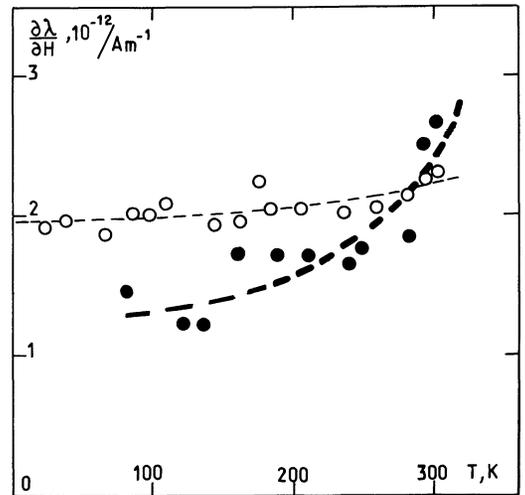


Fig. 4

Fig. 3. — Temperature dependence of the magnetostriction constant. Solid lines : prediction of the single-ion theory. Full circles :  $x = 0.4$ . Open circles :  $x = 0$  ( $\lambda_s$  measured with a parallelepiped). Crosses :  $x = 0$  ( $\lambda_{||}$  measured with a cylindrical sample, see text).

Fig. 4. — Temperature dependence of the forced magnetostriction in  $(Co_{1-x}Ni_x)_{75}Si_{15}B_{10}$  amorphous alloys. Open circles :  $x = 0$ . Full circles :  $x = 0.4$ .

### 3. Discussion.

The temperature dependence of  $\lambda_s$  as given in figure 3 seems to indicate a single-ion behaviour for the alloy  $x = 0$  and a slight deviation for  $x = 0.4$  above 200 K. But the  $\lambda_s(T)$  plot is not very convenient for testing accurately equation (1). We have recently proposed to plot  $y$  vs.  $t$  with :

$$t = m^2 / \hat{I}_{5/2} [\mathcal{L}^{-1}(m)] \quad y = \lambda_s(T) / \hat{I}_{5/2} [\mathcal{L}^{-1}(m)] \quad (4)$$

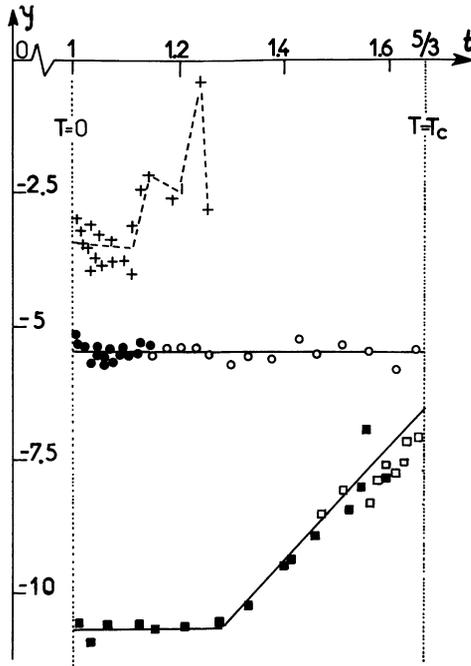


Fig. 5. — Analysis of the temperature dependence of the magnetostriction constant  $\lambda_s$  in various amorphous alloys : plot  $y(t)$  for  $\text{Co}_{80}\text{B}_{20}$  after [1] (crosses) and  $(\text{Co}_{1-x}\text{Ni}_x)_{75}\text{Si}_{15}\text{B}_{10}$  alloys :  $x = 0$ , after [3] (open circles) ;  $x = 0$ , this work (full circles) ;  $x = 0.34$ , after [3] (open squares) ;  $x = 0.4$ , this work (full squares).

where  $t$  will vary from unity at 0 K up to  $5/3$  at  $T_c$  [8]. Such a plot is a straight line :

$$y = \lambda_1 + \lambda_2 t \tag{5}$$

whenever the temperature dependence of  $\lambda_s$  can be described within Callen's model [2], i.e. as the sum of a single-ion and a two-ion contribution, where  $\lambda_1$  and  $\lambda_2$  are temperature independent.

Our data taken from figure 3 are analysed in figure 5 in terms of  $y(t)$  : for  $x = 0$ , they are perfectly consistent with the high temperature data ( $T > 300$  K) taken from reference [3], and, since  $\lambda_2 = 0$ , they prove that the single-ion model is relevant for describing the magnetostriction of a pure Co-metalloid amorphous alloy, as was the case for Fe-metalloid alloys [1]. Here, we must mention that the idea of a noticeable two-ion contribution to  $\lambda_s$  in Co-metalloid alloys arises from two early papers by Jagielinski *et al.* [9] and by O'Handley [1] : these two works were published in 1977 and 1978 when only narrow ribbons were available, and the experiments were not as reliable as presently. In reference [9],  $\lambda_s$  vs.  $T$  for  $\text{Co}_{78}\text{Si}_8\text{B}_{14}$  was given to be temperature independent up to 200 K and to change rapidly from  $-5 \times 10^{-6}$  at 200 K to  $-2.5 \times 10^{-6}$  at 300 K, and for  $\text{Fe}_{78}\text{Si}_8\text{B}_{14}$  anomalies were also observed at about 150 K that have never been confirmed ; these anomalies in the thermal variations of  $\lambda_s$  are correlated with anomalies in the temperature dependence of the forced magnetostriction given by the same author later on in the series  $(\text{Co}_{1-x}\text{Ni}_x)_{78}\text{Si}_8\text{B}_{14}$  [10] : at 0 K, we can derive from his data  $d\lambda_{||}/dH = 2.1 \times 10^{-12}/\text{Am}^{-1}$  and  $1 \times 10^{-12}/\text{Am}^{-1}$  respectively for  $x = 0$  and 0.5, values in a good agreement with the ours, but the temperature

dependence of  $d\lambda/dH$  he found was a continuous decrease up to 300 K in contradiction with our figure 4.

On the other hand, the data for  $\text{Co}_{80}\text{B}_{20}$  taken from reference [1] are plotted in figure 5 : for  $t < 1.12$ ,  $y$  remains constant within experimental uncertainties (with a possible very small negative slope) while above  $t = 1.15$ , namely for  $T > 500$  K,  $y$  suddenly increases but no linear variation can reasonably be invoked. The rapid evolution of  $\lambda_s$  above 500 K could be explained either by problems in the strain gauges or by some structural changes due to heating the sample at temperatures approaching the crystallisation temperature which is much lower than  $T_c \approx 850$  K in this alloy.

Let us now discuss the case of the Co-Ni amorphous alloys : we give in figure 5  $y(t)$  for a sample  $x = 0.4$ . Due to the low  $T_c$  value, it is possible to draw the whole curve from 5 K up to  $T_c$  : the lower part is horizontal, thus suggesting again only a single-ion origin for  $\lambda_s$ . But for  $t > 1.3$ , i.e.  $T > 220$  K, we observe a strong deviation from the single-ion behaviour : experimental errors are not likewise since the magnetization was carefully determined (see Sect. 2), and previous data obtained by quite a different method in reference [3] for  $x = 0.34$  exhibit the same behaviour (open squares in Fig. 5).

We must emphasize that this linear variation of  $y$  vs.  $t$  above 220 K cannot be ascribed to a two-ion effect ; indeed, this one is not observed below this temperature, and moreover the critical exponent for the magnetostriction should be  $2\beta$  if the temperature dependence of the magnetostriction was defined only by the spin-correlation functions : in the present case, it appears to be larger than  $2\beta$ .

The magnetoelastic coupling per nickel atom appears to be the same sign but about 4 times larger than that of cobalt atoms : the anomalous behaviour of  $\lambda_s$  above 220 K could be explained within the framework of the ferrimagnetic single-ion model, where :

$$\lambda_s = (1-x) \lambda_{\text{Co}} \hat{I}_{5/2}[\mathcal{L}^{-1}(m_{\text{Co}})] + x \lambda_{\text{Ni}} \hat{I}_{5/2}[\mathcal{L}^{-1}(m_{\text{Ni}})] \quad (6)$$

for  $x = 0.4$ ,  $(1-x) \lambda_{\text{Co}} = -3.1 \times 10^{-6}$  and  $x \lambda_{\text{Ni}} = -7.4 \times 10^{-6}$ , hence the nickel contribution is dominant and a decrease of  $m_{\text{Ni}}$  with the temperature more rapid than  $m = x m_{\text{Ni}} + (1-x) m_{\text{Co}}$  could explain the deviation from the ferromagnetic single-ion model, namely an horizontal line in figure 5. This assumption is supported by the fact that the alloy with  $x = 1$  is non-magnetic, and for  $x = 0.4$ , the nickel is polarised by the magnetic cobalt so its magnetic moment decreases rapidly when the cobalt magnetization decreases near  $T_c$ .

As a conclusion, the temperature dependence of the magnetostriction constant  $\lambda_s$  of  $\text{Co}_{75}\text{Si}_{15}\text{B}_{10}$  is correctly described by the ferromagnetic one-ion model. This model is not relevant to the case of the nickel-substituted alloy for  $x = 0.4$  above 220 K : the ferrimagnetic one-ion model seems to be more plausible. Last, the magnetoelastic coupling per nickel atom appears to be the same sign but 4 times larger than that of cobalt atoms.

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