

## Temperature Dependence of Magnetization of Amorphous TM<sub>70</sub>Cr<sub>5</sub>Si<sub>10</sub>B<sub>15</sub> (TM = Fe, Co, Ni) Alloys

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We report the salient features of the magnetic properties of amorphous TM<sub>70</sub>Cr<sub>5</sub>Si<sub>10</sub>B<sub>15</sub> (TM = Fe, Co, Ni) alloys. The temperature dependence of magnetization for amorphous ribbons were measured by a SQUID and a VSM from 5 K to 700 K under an external field of 10 kOe. Except Ni<sub>70</sub>Cr<sub>5</sub>Si<sub>10</sub>B<sub>15</sub> that shows a paramagnetic behaviour, both Fe and Co based amorphous alloys show a typical ferromagnetic thermo-magnetization curves. For these two ferromagnetic alloys, the saturation magnetization in the temperature range from 5 K to about 0.4  $T_c$  can be described by the Bloch relation,  $M_s(T) = M_s(0) [1 - BT^{3/2} - CT^{5/2}]$ . The spin wave stiffness constants and the range of exchange interaction were analyzed from the magnetization behaviour. The variation of the magnetic properties are discussed and compared with the composition of the alloys.

### 1. Introduction

A number of studies have been reported on the low temperature magnetic properties and the magnetic excitations of locally isotropic amorphous metallic ferromagnets over the last 10 years[1-4]. In amorphous structures the wave vector is not well defined and no easily definable Brillouin zone exists. However, much experimental evidence has shown that the long wavelength spin waves can be clearly and stably defined in amorphous alloys that have a topologically disordered type, which is manifested by the random close packing of the atomic spheres. In general, it has been found that such glasses exhibit well-defined longwavelength spin wave excitations with conventional ferromagnetic dispersion relation,

$$E_q = D(T)q^2 + \Delta \quad (1)$$

where  $q$  is the wave vector,  $D(T)$  is the spin wave (or exchange) stiffness constant and  $\Delta$  is an effective energy gap due to the dipole-dipole interactions[5]. There are many experimental evidences in the literature[1-4] showing that the decrease in magnetization with increasing temperature at low temperature in both crystalline and non-crystalline ferromagnets can be adequately described by Bloch's relation:

$$M_s(T) = M_s(0) [1 - BT^{3/2} - CT^{5/2}]$$

$$\text{or } \Delta M_s(T)/M_s(0) = BT^{3/2} + CT^{5/2} \\ = B_{3/2}(T/T_c)^{3/2} + C_{5/2}(T/T_c)^{5/2}. \quad (2)$$

Although both crystalline and non-crystalline ferromagnets follow the general predictions of Heisenberg model, details of the magnetization results for glassy alloys differ from those of crystalline ferromagnets. In metal-metalloid alloys the magnetic and electron transport properties of the amorphous materials are strongly affected by the type of element and its concentration because of the superposition of structural and compositional disorder. It is from the Bethe-Slater curve that the sign of the exchange integral between transition metals depends sensitively on their interatomic distance. Especially, Cr atoms indicates a negative exchange integral so it is interesting to study the TM(transition metal)-Cr alloy system. Recently, it was shown by the present authors[6], there exists antiferromagnetic (AFM) coupling in transition metal based amorphous alloy with Cr element. In order to get the better understanding of the magnetic properties, it is required to investigate the spin wave excitations and the temperature dependence of magnetization. In this paper, we report the effects of a transition metals such as Fe, Co and Ni in TM-Cr-Si-B alloy on their magnetic properties.

## 2. Experimental

Amorphous  $\text{TM}_{70}\text{Cr}_5\text{Si}_{10}\text{B}_{15}$  (TM = Fe, Co, Ni) alloys were prepared by the single roller melt-spinning method. The compositions of these amorphous alloys were confirmed by inductive coupled plasma spectroscopy analysis. The ribbons were 2 mm wide and 20–30  $\mu\text{m}$  thick. The amorphous state of the samples was verified by X-ray diffraction using Cu K $\alpha$  radiation. The temperature dependence of the saturation magnetization was measured from 5 K to 800 K in the external magnetic field of 10 kOe using a SQUID and a VSM. To minimize the effects of the demagnetizing fields, the magnetic field was applied in the plane of the ribbon pieces parallel to their length. The temperature was increased at a rate of 60 K/h to achieve sufficient accuracy in the measurement. In order to estimate the values of spectroscopic splitting  $g$  factor, FMR experiments were carried out at room temperature on a rounded sample of 1 mm in diameter using a Varian X-band electron resonance spectrometer. Specifically, the  $\text{TE}_{102}$  resonance mode in the rectangular cavity was employed.

## 3. Results and Discussion

Figure 1 shows a typical temperature-dependence of saturation magnetization in amorphous  $\text{Fe}_{70}\text{Cr}_5\text{Si}_{10}\text{B}_{15}$  and  $\text{Co}_{70}\text{Cr}_5\text{Si}_{10}\text{B}_{15}$  alloys with  $H = 10$  kOe. We obtained values of the saturation magnetizations at 0 K,  $M_s(0) = 152.8$  emu/g ( $\text{Fe}_{70}\text{Cr}_5\text{Si}_{10}\text{B}_{15}$ ) and 79.5 emu/g ( $\text{Co}_{70}\text{Cr}_5\text{Si}_{10}\text{B}_{15}$ ), respectively, by extrapolating  $M_s$  to 0 K via a simple spin wave approximation from the temperature dependence of the saturation magnetization,  $M_s(T)$ . The saturation magnetization of Fe-based amorphous alloy was much higher than that of Co-based alloys as was already observed in the other similar systems[7]. The Curie temperatures were determined using the Arrot plots from the isofield magnetization curve. Curie temperatures of Fe and Co based amorphous alloy samples are about 600 K and 730 K. The saturation magnetization behaviour of  $\text{Ni}_{70}\text{Cr}_5\text{Si}_{10}\text{B}_{15}$  alloy looks like paramagnetic as we can see in fig. 2. This result indicates that Ni acts as non-magnetic atom in an amorphous state. Figure 3 shows the  $T^{3/2}$  dependence of the magnetization. The reduced magnetization is plotted versus  $T^{3/2}$  below about  $0.4 T_c$ . There is a good relationship between the reduced magnetization and  $T^{3/2}$  at lower temperature, as expected from eq. (2). The Bloch coefficients B and C were obtained from fig. 3 by least square fitting. The values of B and C are  $24.2 \times 10^{-6} \text{K}^{-3/2}$  and  $1.2 \times 10^{-8} \text{K}^{-5/2}$  for  $\text{Fe}_{70}\text{Cr}_5\text{Si}_{10}\text{B}_{15}$  alloy and  $17.9 \times 10^{-6} \text{K}^{-3/2}$  and  $0.95 \times 10^{-8} \text{K}^{-5/2}$  for  $\text{Co}_{70}\text{Cr}_5\text{Si}_{10}\text{B}_{15}$  alloy, respectively. By conventional linear spin wave theory, the Bloch coefficient B is related to the spin wave stiffness constant  $D$  through

$$D = (2.612)^{2/3} [g \mu_B / B M_s(0)]^{2/3} [\kappa_{eff} / 4\pi]. \quad (3)$$

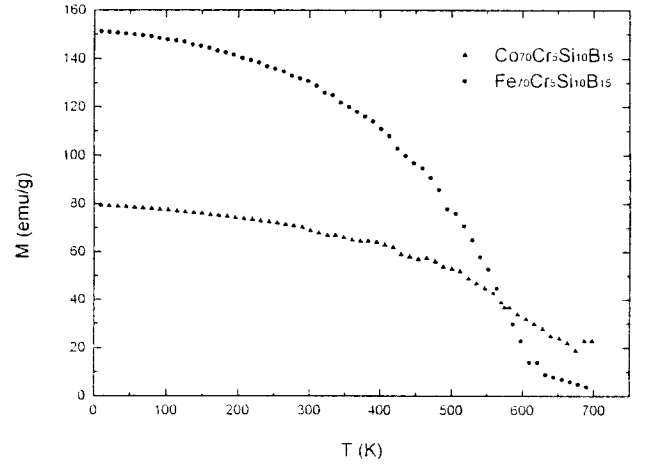


Fig. 1. The temperature dependence of magnetization for amorphous  $\text{Fe}_{70}\text{Cr}_5\text{Si}_{10}\text{B}_{15}$  and  $\text{Co}_{70}\text{Cr}_5\text{Si}_{10}\text{B}_{15}$  alloys.

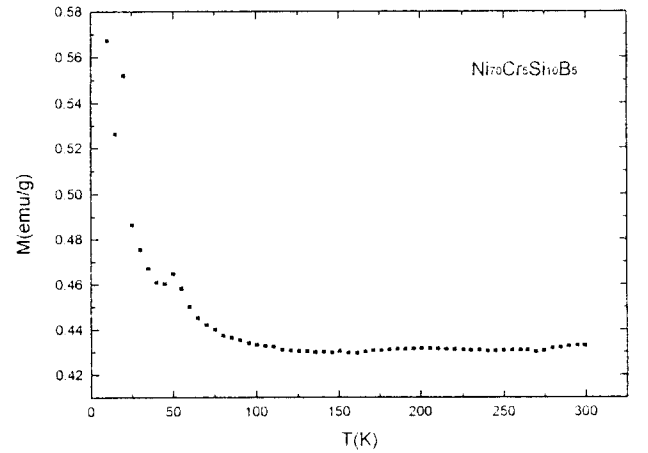


Fig. 2. The temperature dependence of magnetization for  $\text{Ni}_{70}\text{Cr}_5\text{Si}_{10}\text{B}_{15}$  alloy.

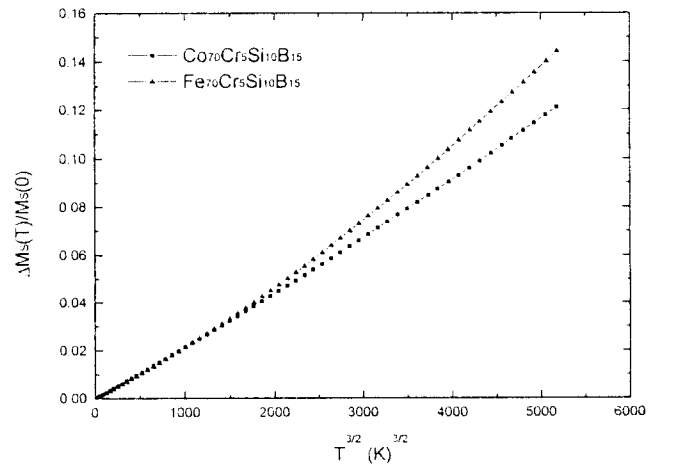


Fig. 3.  $T^{3/2}$  temperature dependence of magnetization for  $\text{Fe}_{70}\text{Cr}_5\text{Si}_{10}\text{B}_{15}$  and  $\text{Co}_{70}\text{Cr}_5\text{Si}_{10}\text{B}_{15}$  alloys.

Here  $g$  is spectroscopic splitting factor,  $\mu_B$  is Bohr magneton and  $\kappa_B$  is the Boltzmann constant. The spectroscopic splitting factor  $g$  represents the relative contribution of orbital magnetic moment to total magnetic moment. The theoretical description of the FMR signal is based on the analysis of the free energy density. From our spectroscopic measurement we estimate the values of  $g$  to be 2.20 and 2.11 for  $\text{Fe}_{70}\text{Cr}_5\text{Si}_{10}\text{B}_{15}$  and  $\text{Co}_{70}\text{Cr}_5\text{Si}_{10}\text{B}_{15}$  alloys, respectively. This result indicates that the contribution of orbital magnetic moments in  $\text{Fe}_{70}\text{Cr}_5\text{Si}_{10}\text{B}_{15}$  alloy is larger than for the  $\text{Co}_{70}\text{Cr}_5\text{Si}_{10}\text{B}_{15}$  alloy. The values of spin wave stiffness constant obtained from eq. (3) are 117.2 and 198.8 meV  $\text{\AA}^2$  for  $\text{Fe}_{70}\text{Cr}_5\text{Si}_{10}\text{B}_{15}$  and  $\text{Co}_{70}\text{Cr}_5\text{Si}_{10}\text{B}_{15}$  alloys, respectively. The  $D$  value of  $\text{Co}_{70}\text{Cr}_5\text{Si}_{10}\text{B}_{15}$  alloy is much higher than  $\text{Fe}_{70}\text{Cr}_5\text{Si}_{10}\text{B}_{15}$  alloy. This result indicates a "magnetic hardening" of the exchange interaction. In order to get the mean square range of exchange interaction, we used following relation[5],

$$\langle r^2 \rangle = (16/3 \kappa_B) [\xi(3/2)/\xi(5/2)] [CD/B] \quad (4)$$

where  $\xi(3/2)$  and  $\xi(5/2)$  are the Riemann zeta functions. The values of  $\langle r^2 \rangle$  are 7.01 and 12.8  $\text{\AA}^2$  for Fe and Co-based alloys, respectively. This is consistent with the spin wave constant, which show the value of the Co-based alloy is larger than for Fe-based alloy. Therefore, our results show that "magnetic hardening" in  $\text{Co}_{70}\text{Cr}_5\text{Si}_{10}\text{B}_{15}$  alloy is due to the larger exchange energy between Co atoms. In order to clarify the exchange energy for our samples we estimated the exchange integral. Exchange integral  $J$  is described in terms of the Curie temperature[8]

$$T_c = 2 \langle Z \rangle J S (S + 1) / 3 \kappa_B \quad (5)$$

where  $Z$  is the average number of nearest neighbours of Fe or Co and  $S$  is the spin moment. The values of exchange integral  $J$  are 63 and 179 K for  $\text{Fe}_{70}\text{Cr}_5\text{Si}_{10}\text{B}_{15}$  and  $\text{Co}_{70}\text{Cr}_5\text{Si}_{10}\text{B}_{15}$  alloys, respectively. From the relation of  $M(\mu_B) = 2 x S \mu_B$  [9], we can calculate  $S$ , where  $M(\mu_B)$  and  $x$  are the magnetic moment of sample and concentration of transition metal, respectively. The magnetic moment of the alloy arises from the total contribution of the moment of Fe ( $\mu_{Fe}$ ) and Co ( $\mu_{Co}$ ), which is aligned ferromagnetically. The  $S_{Fe}$  and  $S_{Co}$  are found to be 0.90 and 0.49 for  $\text{Fe}_{70}\text{Cr}_5\text{Si}_{10}\text{B}_{15}$  and  $\text{Co}_{70}\text{Cr}_5\text{Si}_{10}\text{B}_{15}$  alloy, respectively. These values are smaller than the theoretical value of 1.11 and 0.86 in the pure Fe and Co, respectively. This result implies that amorphous  $\text{Fe}_{70}\text{Cr}_5\text{Si}_{10}\text{B}_{15}$  and  $\text{Co}_{70}\text{Cr}_5\text{Si}_{10}\text{B}_{15}$  alloys have a asperomagnetic spin structure due to random anisotropy. Especially, it indicates that the spread angle of conical spin structure for  $\text{Co}_{70}\text{Cr}_5\text{Si}_{10}\text{B}_{15}$  alloy is much larger than  $\text{Fe}_{70}\text{Cr}_5\text{Si}_{10}\text{B}_{15}$  alloy one. In the asperomagnetic structure, the moments are distributed at random but have some long-range preferred orientation in direction. In other words, an asperomagnet has a spontaneous magnetization. It can be

thought as a random ferromagnet. On the other hand,  $D/T_c$ , which measures the range of the exchange interaction, were observed to be 0.20 and 0.27 meV  $\text{\AA}^2/\text{K}$  for Fe and Co based alloys. However, these values are much smaller than 0.63 meV  $\text{\AA}^2/\text{K}$  in crystalline nickel. In eq. (2), the normalized coefficients  $B_{3/2}$  and  $C_{5/2}$  are defined as  $BT_c^{3/2}$  and  $CT_c^{5/2}$ , respectively. For the crystalline ferromagnet, Ni, the ratio  $C_{5/2}/B_{3/2}$  is of the order 1[10]. In noncrystalline ferromagnets, the ratio  $C_{5/2}/B_{3/2}$  provides a measure of the range of the exchange interaction[11]. The smaller values of  $C_{5/2}/B_{3/2}$  suggest that the mean exchange interaction has a shorter range in noncrystalline ferromagnets. The ratios of our sample are 0.32 and 0.36 for  $\text{Fe}_{70}\text{Cr}_5\text{Si}_{10}\text{B}_{15}$  and  $\text{Co}_{70}\text{Cr}_5\text{Si}_{10}\text{B}_{15}$  alloy, respectively. This implies that the  $T_c^{3/2}$  term dominates in amorphous solids for a much larger temperature range than that in crystalline solids.

#### 4. Conclusion

The temperature dependence of the magnetization of  $\text{Fe}_{70}\text{Cr}_5\text{Si}_{10}\text{B}_{15}$  and  $\text{Co}_{70}\text{Cr}_5\text{Si}_{10}\text{B}_{15}$  amorphous alloys apparently follows the prediction of spin wave theory but  $\text{Ni}_{70}\text{Cr}_5\text{Si}_{10}\text{B}_{15}$  alloy shows a paramagnetic behaviour. The spin wave stiffness constant and the range of the exchange interaction of Co based alloy is much higher than for the Fe based alloy. This result indicates a "magnetic hardening" of the exchange interaction by higher exchange energy.

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