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The Pole Effect of Ferromagnetic Thin Sheets and Stiffening of Fe-B Base Amorphous Alloys*

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Synopsis

Several kinds of ferromagnetic crystalline thin sheets have been made by cold rolling, and some ferromagnetic amorphous ribbons have been prepared from the melts by rapidly quenching method. At the fundamental mode, the so-called "pole effect" of thin sheets has become more pronounced with increasing magnetic field, but this effect has almost vanished below about 100 of the length to thickness ratio of the specimens even at the fundamental mode. The "pole effect" of thin sheets and amorphous ribbons has decreased with increasing the frequency of mode, becoming nearly zero at the third tone mode or more. Magnetic field dependence of Young's modulus for amorphous alloys has been almost saturated above 100 Oe at the third tone mode, and the temperature dependence of Young's modulus for $Fe_{80}B_{20}$ amorphous alloy has showed a stiffening below the Curie temperature. This behavior is very similar to the "pole effect", but the former is not related to the latter because the "pole effect" has been almost reduced by measuring at the third tone mode. Such a peculiar phenomenon has not been observed in Co-B amorphous alloys.

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I. Introduction

Magnetic properties of thin amorphous ribbons have been investigated by many researchers(1,2), and it has been shown that Fe-base amorphous alloys exhibit a large ΔE effect(3,4). Further, Berry and Pritchet(5) have for the first time demonstrated that a peculiar phenomenon, which they termed "the pole effect", is superimposed on the intrinsic magnetoelastic effect. Because of this effect, the elastic constants of specimens remarkably change the slope on passing through the Curie temperature at a low frequency mode(6). Török and Hausch(7) have also pointed out that the pole effect could introduce serious errors in the result of the elastic constant. We have investigated the elastic properties of many amorphous alloys, and elucidated that Fe-base, especially Fe-B, amorphous alloys exhibit a remarkable stiffening below the Curie temperature(8∿12). However, this behavior has been often confused with the pole effect. The so-called "pole effect" is generally affected by the shape of samples, especially its length and thickness.

In order to understand clearly the difference between the pole effect and the stiffening, we have studied the influence of magnetic field and temperature on Young's modulus for several crystalline and amorphous alloys and the results are reported in this paper.

II. Experimental

Both amorphous and crystalline alloy samples are used. Amorphous ribbons of $Fe_{80}B_{20}$*, $Fe_{75}Cr_5B_{20}$ and $Co_{70}B_{30}$ were prepared from the melt by a rapid quenching method. The crystalline samples used were $Fe_{53.4}Ni_{33.8}Cr_{12.8}$ elinvar alloy and $Ni_{96}Co_4$ alloy. The starting materials used were all high purity elements. The molten alloys were cast into an iron mold with 30 × 30 mm^2, and reduced to sheets of 5 mm thickness by hot forging. After annealing, the forged alloys were cold-rolled to different thicknesses ranging from 3.5 to 0.04 mm. Prior to measurements the samples were annealed in vacuum for 2 hrs. The annealing temperatures used were 200°C and 50° below the crystallization temperature for amorphous samples, and 1000°C for crystalline samples. Considering the strong stress-sensitivity to Young's modulus of the Ni-based alloy(13), the assembly consisting of the sample and

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* All the compositions are expressed in atomic per cent shown as subscripts.

the platinum wire were annealed in the spot-welded condition prior to measurements. Young's modulus was calculated using the measured resonant frequencies f_n ($400 \sim 2500$ Hz) of the specimens by using the modulated vibrator-controlled oscillator(14) with the flexural free-free modes of the samples.

The frequencies f_n are given by

$$f_n = C_n(d/\ell^2)(E/\rho)^{1/2} \tag{1}$$

The mode constant C_n has the value $C_1 = 4.7296$ for the fundamental mode and $C_2 = 7.8529$, $C_3 = 10.9904$ for the second and third tones, respectively(15). The mode can be changed by selecting the nodal points of samples. The nodal points are 0.224ℓ and 0.7758ℓ for the fundamental vibration, 0.1321ℓ, 0.5ℓ and 0.869ℓ for the second tone and 0.0944ℓ, 0.3558ℓ, 0.6442ℓ, 0.9056ℓ for the third tone. Demagnetization of the sample was done at 50 Hz with a diminishing A.C. field of 200 Oe. The experiment of ΔE effect was carried out in a pair of Helmholtz coils designed for measuring at low and high temperatures. The fields were changed from 0 to 1000 Oe.

III. Results and discussion

Magnetic field dependence at room temperature of the fundamental mode f_1 and the second tone mode f_2 for various kinds of the length to thickness (ℓ/d) ratios of $Fe_{53.4}Ni_{33.8}Cr_{12.8}$ crystalline alloys are shown in Fig. 1. In the case of thin sheets, the frequency f_1 is not easily saturated due to the so-called "pole effect". The pole effect is associated with the following equation(6);

$$\frac{f_n^2}{f_{ns}^2} = 1 + (6\gamma_n M_s \ell^2/E_s d^2)H \tag{2}$$

where f_n is the mode frequency, f_{ns} the frequency attained at saturation in the imagined absence of the pole effect. The value of γ_n is equal to $\tanh\alpha_n$ (for n=1, 3, 5, ...) or $\coth\alpha_n$ (for n=2, 4, 6, ...) and α_n is the successive solution of $\tanh\alpha_n=\tan\alpha_n$ (for n=1, 3, 5, ...) or $\coth\alpha_n=\cot\alpha_n$ (for n=2, 4, 6, ...), M_s is the saturation magnetization, E_s Young's modulus at saturation field and H the strength of magnetic field. From eq. (2), it is clear that the pole effect becomes smaller the shorter and thicker the samples are. Actually, as seen in the figure, the increase in f_n is less marked for the thicker samples and samples with a ℓ/d ratio less than 100 scarecely show the pole

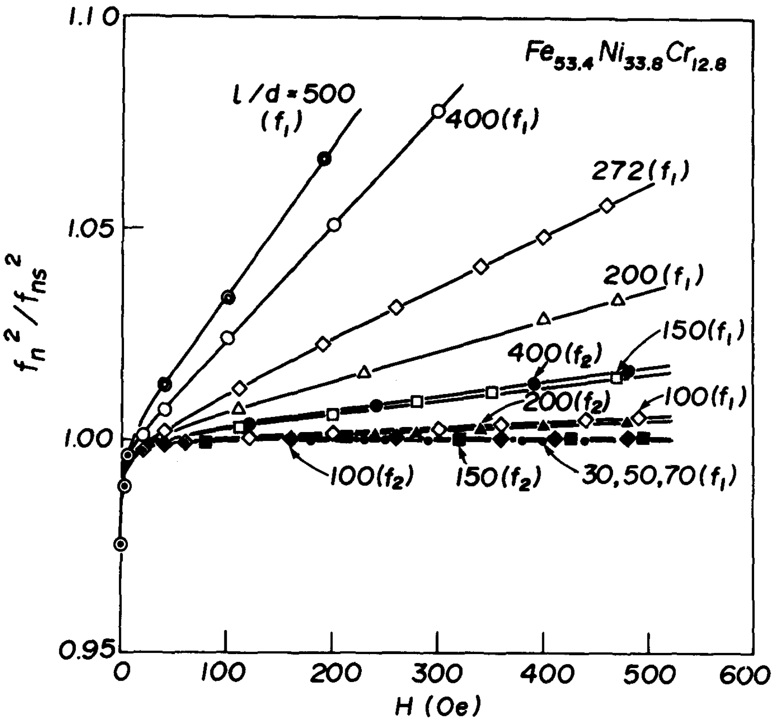

Fig. 1. The f_n^2/f_{ns}^2 for the annealed crystalline $Fe_{53.4}Ni_{33.8}Cr_{12.8}$ alloy, measured as a function of field strength for the fundamental and second modes.

effect, and the f_1 is easily saturated above 100 Oe. Therefore, it is confirmed that the pole effect vanishes by using a thick sample with a ℓ/d ratio of 100 even at the fundamental mode f_1. Figure 2 shows the temperature dependence of Young's modulus for $Fe_{53.4}Ni_{33.8}Cr_{12.8}$ crystalline elinvar alloy, and it can be seen from the figure that there is no temperature dependence around room temperature at zero field (curve d). However, the extraordinary increase in the Young's modulus of the sample with the ratio 500 is observed below the Curie temperature at the fundamental mode f_1 under magnetic field of 500 Oe (curve a). This behavior can be attributed to the pole effect. This

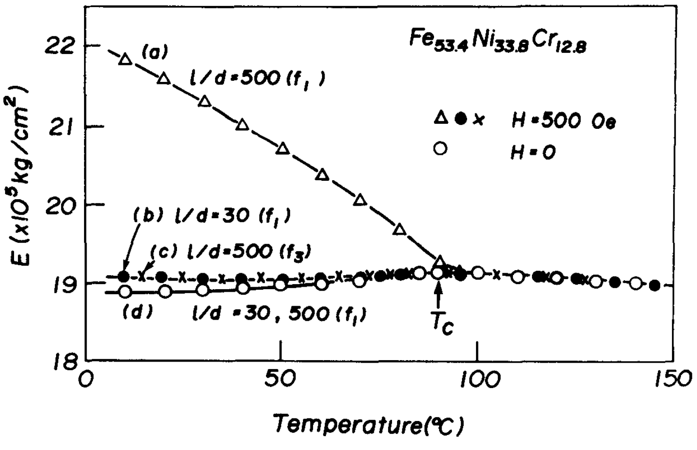

Fig. 2. Temperature dependence of the Young's modulus for two different length-thickness ratios and overtones for the same sample shown in Fig. 1. The magnetic fields are zero (o) and 500 Oe (△, ● , x).

effect is almost reduced by measuring at the third tone mode f_3 (curve c, cross sign). In Fig. 1, it was pointed out that the pole effect is absent when samples with the ratio 100 or less are used even at the fundamental mode f_1. As shown in the figure, the curve of the sample with the ratio 30 measured at f_1 in the magnetic field of 500 Oe (curve b, closed circle) is completely superposed on the curve c measured at f_3. It should be noted that the value at saturated magnetic field is much smaller than the value extrapolated linearly above the paramagnetic range, showing the softening phenomenon. Figure 3 shows the change in the fundamental mode f_1 of $Fe_{53.4}Ni_{33.8}Cr_{12.8}$ crystalline alloy as a function of the ℓ/d ratio. Measurements were carried out at various strengths of the magnetic field at the fundamental mode f_1. The value of f_1 increases above the ratio 100 with increasing magnetic field strength. It is clear from the figure that there is hardly any dependence on the field strength below the ratio 100, indicating the absence of the pole effect. From the results of Figs. 1, 2 and 3, it is concluded that the pole effect is almost reduced by using the sample with the ℓ/d ratio less than 100, or measuring at the third tone or more. Berry and Pritchet(6) have pointed out that the theoretical slope ratio γ_n/γ_1 becomes 0.0185 at n=3. This means that the frequency change scarcely occurs at the third tone mode. Hence, our experimental results are consistent with the theoretical calculation.

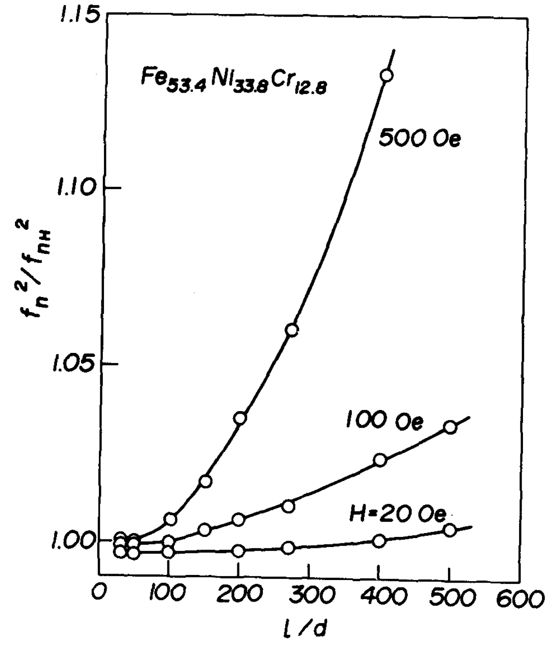

Fig. 3. Dependence of the f_n^2/f_{nH}^2 on ℓ/d ratio at various magnetic fields for the crystalline $Fe_{53.4}Ni_{33.8}Cr_{12.8}$ alloy with the fundamental mode f_1.

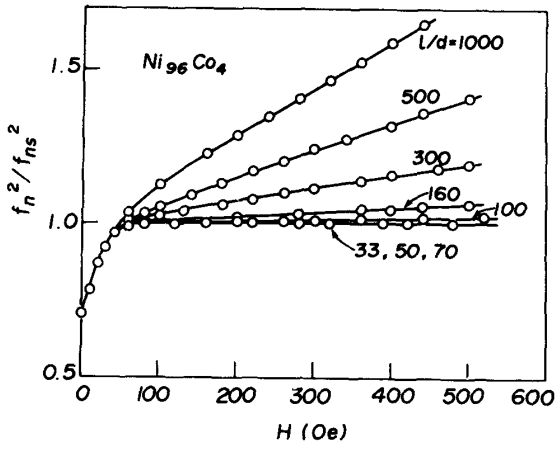

Fig. 4. Dependence of the f_n^2/f_{ns}^2 on the magnetic field for annealed crystalline $Ni_{96}Co_4$ alloy at the fundamental mode f_1. The length-thickness ratio was changed from 1000 to 33.

Figure 4 shows the change in frequency for the $Ni_{96}Co_4$ alloy as a function of magnetic field at the fundamental mode f_1. This alloy also exhibits a large pole effect when the sample is very thin and long, and its effect is almost absent below the ratio of 100.

Field dependence of the relative change of square of frequencies for amorphous $Fe_{80}B_{20}$ alloy is shown in Fig. 5. The samples were

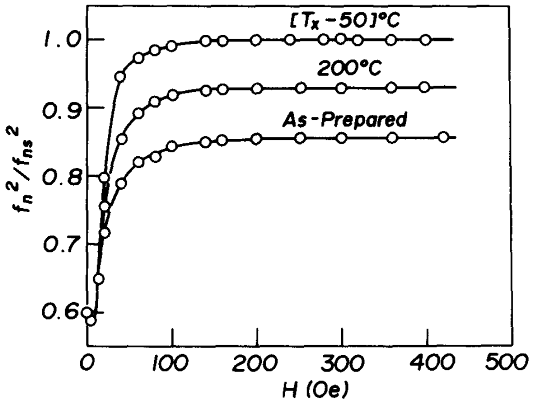

Fig. 5. Dependence of the f_n^2/f_{ns}^2 on magnetic field for amorphous $Fe_{80}B_{20}$ alloy in the (a) as-prepared, (b) annealed at 200°C and (c) annealed at a temperature 50° lower than the crystallization temperature T_x.

annealed for 2 hrs at 200°C or at a temperature 50° lower than the crystallization temperature, T_x. Measurements were made at room temperature by using the third tone mode f_3 in order to reduce the pole effect. These curves are almost saturated above 100 Oe. The relative change in square of frequencies increases with increasing annealing temperature and the value of the sample annealed at (T_x-50)°C is larger than that of as-prepared sample by a factor of 1.5. This behavior has already been observed in $Fe_{82}B_{18}$ amorphous alloy by us(9). Next, we will show in more detail the effect of annealing on the ΔE effect for $Fe_{80}B_{20}$ amorphous alloy. Figure 6 shows the variation of the ΔE effect with annealing temperature. Annealing was carried out for 2 hrs and measurements were made at room temperature. The ΔE effect is generally defined by $\Delta E/E_D = (E_S-E_D)/E_D$, where E_D and E_S are Young's modulus at zero and saturated magnetic fields, respectively. The value of $\Delta E/E_D$ increases with increasing annealing temperature and decreases drastically just at the crystallization temperature, T_x. Similar behavior for $Fe_{82}B_{18}$ and $Fe_{86}B_{14}$ amorphous alloys was reported by us(9). Such an increment by annealing may be due to the relaxation of the internal stresses induced in the samples during rapid quenching from the melt.

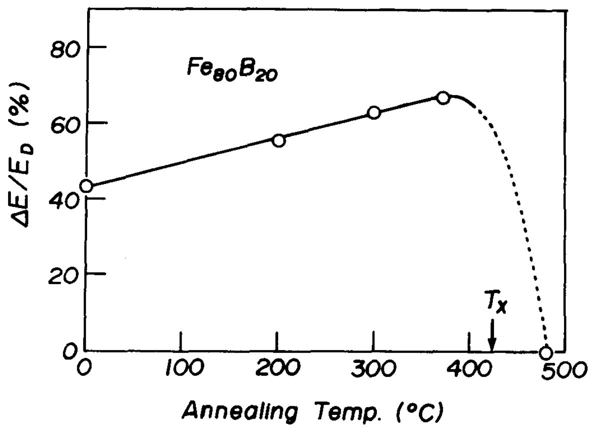

Fig. 6. Variation of the ΔE effect with annealing temperature for the amorphous $Fe_{80}B_{20}$ alloy. T_x is the crystallization temperature of the alloy.

Figure 7 shows the temperature dependence of Young's modulus at zero and 500 Oe for as-prepared $Fe_{80}B_{20}$ and $Fe_{75}Cr_5B_{20}$ amorphous alloys. Since the Curie temperature T_c is very close to the crystallization

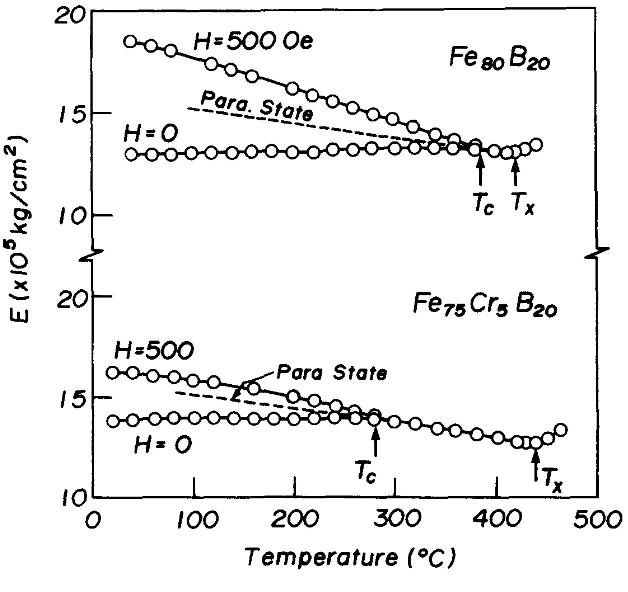

Fig. 7. Temperature dependence of the Young's modulus at the third mode under the field of zero and 500 Oe for as-prepared $Fe_{80}B_{20}$ and $Fe_{75}Cr_5B_{20}$ alloys.

temperature T_x for the $Fe_{80}B_{20}$ amorphous alloy, the slope of the paramagnetic Young's modulus cannot be obtained accurately. However, by the addition of a small amount of Cr, the difference between T_c and T_x increases, and so the slope of the paramagnetic Young's modulus for the $Fe_{75}Cr_5B_{20}$ amorphous alloy can be determined accurately. The same slope was used for the $Fe_{80}B_{20}$ amorphous alloy. It is well known that at zero magnetic field, the $Fe_{80}B_{20}$ amorphous alloy exhibits elinvar characteristics in a wide temperature range. While, at the applied magnetic field, the pole effect is also observed at the fundamental mode f_1, but this effect is not present at the third tone mode f_3, and Young's modulus at room temperature is slightly larger than the paramagnetic Young's modulus extrapolated above the Curie temperature. Then, the stiffening at saturated magnetic field is not so clear from Fig. 7. Next we examined the stiffening of Young's modulus for the $Fe_{80}B_{20}$ amorphous sample, because Young's modulus at saturated magnetic field increases with increasing annealing temperature as shown in Fig. 6. Figure 8 shows the temperature dependence of Young's modulus for $Fe_{80}B_{20}$ amorphous alloy preannealed at 200°C for 2 hrs. In the zero magnetic field, elinvar characteristics are also observed. However, a large pole effect is observed at the fundamental mode f_1 in the field of 500 Oe. It should be noted that Young's modulus at the

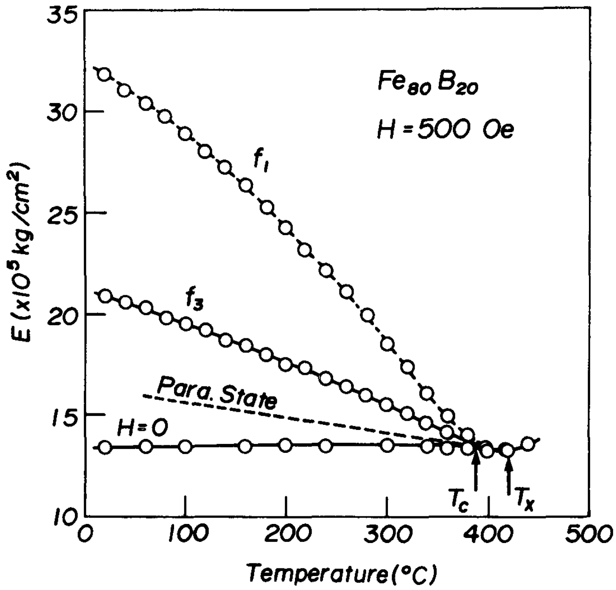

Fig. 8. Temperature dependence of the Young's modulus at the fundamental and third mode under the field of zero and 500 Oe for the annealed $Fe_{80}B_{20}$ alloy.

third tone mode f_3 is remarkably larger than that of the value of Young's modulus extrapolated above the Curie temperature. This slope indicated by the broken line in the figure was obtained by the same procedure as for Fig. 7 by using $Fe_{75}Cr_5B_{20}$ amorphous alloy annealed at 200°C for 2 hrs. Since the pole effect is almost absent at the third tone mode, this large stiffening seems to be the intrinsic behavior of $Fe_{80}B_{20}$ amorphous alloy. Such a peculiar phenomenon was earlier observed in many Fe-base amorphous alloys.

Figure 9 shows the temperature dependence of Young's modulus for $Co_{70}B_{30}$ amorphous alloy preannealed at 200°C for 2 hrs. These alloys take a large negative temperature coefficient even at zero magnetic field and show no elinvar characteristics. The pole effect is also observed at the fundamental mode f_1. However, the stiffening is not observed at the third tone mode f_3 in saturated magnetic field, and the curve almost overlaps with the curve measured at zero magnetic field.

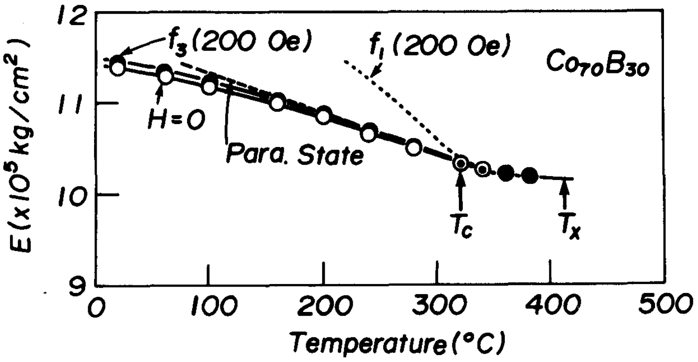

Fig. 9. Temperature dependence of the Young's modulus at the fundamental and third mode under the field of zero and 500 Oe for the annealed amorphous $Co_{70}B_{30}$ alloy.

From the present results, it is concluded that the pole effect is observed in all crystalline and amorphous thin specimens at low frequency modes, and the stiffening is observed in only elinvar-type amorphous alloys. Török and Hausch have pointed out that the pole effect could introduce serious errors in the results of the elastic constant and also commented that the giant ΔE effect obtained by us is caused by the pole effect(7). However, the field dependence of the relative change in Young's modulus is easily saturated above 100 Oe as shown in Fig. 5 of the present paper and in Fig. 2 of our previous paper(9). This indicates that our values of the ΔE effect do not contain the pole effect. Therefore, it is concluded that stiffening is an intrinsic characteristic of the Fe-base amorphous alloys, though its origin is still not clear. In the case of crystalline pure Ni, a little stiffening is observed at saturated magnetic field(16). This behavior has been connected with the spontaneous volume magnetostriction by Hausch and Warlimont(16). However, Fe-B amorphous alloys show invar characteristics and their spontaneous volume magnetostriction is opposite in sign to that of pure Ni(17,18). As such, the stiffening of $Fe_{80}B_{20}$ amorphous alloy could not be explained by their expression. Crystalline pure Gd shows a similar large spontaneous volume magnetostriction(19) compared with that of Fe-B amorphous alloys and its Young's modulus exhibits a remarkable stiffening below the Curie temperature and this behavior was discussed in terms of RKKY interaction. However, the temperature dependence of Young's modulus shows the stiffening even at zero magnetic field, although the curve for the $Fe_{80}B_{20}$ amorphous alloy shows no stiffening at zero magnetic field, indicating elinvar characteristics as shown in Fig. 7. Therefore, the origin of stiffening for $Fe_{80}B_{20}$ amorphous alloy may be different from that of Ni or Gd crystalline metals.

Finally we would like to comment on the anomalous temperature dependence of Young's modulus for $Fe_{53.4}Ni_{33.8}Cr_{12.8}$ crystalline alloy shown in Fig. 2. Generally, the temperature dependence of Young's modulus for Fe-base alloys with the composition limit for the martensitic transformation exhibits a similar behavior as that of $Fe_{53.4}Ni_{33.8}Cr_{12.8}$ alloy as shown in Fig. 2 at saturated magnetic field. In other words, the value of Young's modulus is lower than that of the paramagnetic value extrapolated above the Curie temperature. This phenomenon was explained by Hausch and Warlimont(20) as due to the contributions of linear magnetostriction, forced volume magnetostriction and exchange interaction. On the other hand, recently, the possible contribution of softening to the anomalous elastic properties has been discussed. Shibata and Fujita(21) have reported that Fe-Ni-C alloys having an M_s temperature above the Curie temperature indicate a remarkable softening even in a paramagnetic temperature range. In contrast to such a behavior, it should be noted that the $Fe_{80}B_{20}$ amorphous alloy, which has no relationship with the martensitic transformation, shows no such softening at saturated magnetic field. On the other hand it indicates stiffening as shown in Fig. 7.

Summary

Magnetic field and temperature dependence of Young's modulus for several kinds of crystalline and amorphous alloys with different length to thickness ratios have been investigated, and the pole effect and stiffening have been discussed. The results obtained are summarized as follows:

(1) Thin sheets show a remarkably large pole effect at the fundamental mode f_1, but specimens with a ratio less than 100 do not exhibit the pole effect even at f_1.

(2) The pole effect is almost absent when measurements were done at the third tone mode f_3 or more.

(3) Magnetic field dependence of Young's modulus for the amorphous alloys measured at the third tone mode f_3 is almost saturated above 100 Oe at room temperature.

(4) In the case of $Fe_{80}B_{20}$ amorphous alloy annealed at 200°C for 2 hrs, remarkable stiffening is observed below the Curie temperature. This behavior is distinguished from the pole effect by measuring at the third tone mode or more.

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