The Origin of High Magnetic Permeability in "Hardperm" of the Ni-Fe-Nb System*

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Electrical resistivity ρ , crystal magnetic anisotropy constant K_1 , saturation magnetostriction constant λ_s , and the ΔE effect, $\Delta E/E_0$ have been measured to investigate the origin of high permeability in the Ni-Fe-Nb system. The investigation shows that the value of ρ for Ni-Fe-Nb alloys increases with decrease in cooling rate and decreases with increase in cold reduction, indicating the formation of the K state in these alloys.

With Nb addition, the absolute values of K_1 and λ_s and the value of $\Delta E/E_0$ decrease rapidly and become very small near the composition which shows the highest initial permeability μ_0 . These results indicate that the high permeability of Ni-Fe-Nb alloys is probably caused by the very small values of K_1 , λ_s and $\Delta E/E_0$, that is, by the appearance of magnetoelastically isotropic state.

(Received December 27, 1975)

I. Introduction

The alloys which have widely been used as high permeability alloys are Permalloy-type alloys, Sendust, Hardperm, etc. The high permeability of alloys in the former two system has hitherto been ascribed mainly to the extraordinarily small values of both saturation magnetostriction constant λ_s and crystal magnetic anisotropy constant K_1 ^{(1)~(3)}.

Recently the present author discovered, with Masumoto and Murakami, that the addition of Nb to Ni-Fe alloys raised not only the permeability but also the hardness, and a generic name "Hardperm" is given to these alloys⁽⁴⁾⁽⁵⁾. This paper describes an investigation of the origin of high permeability in

Hardperm by measurements of electrical resistivity, saturation magnetostriction, crystal magnetic anisotropy and the ΔE effect.

II. Experimental

Alloying materials Ni, Fe and Nb containing the impurities (800 or 3200 g in total weight) listed in Table 1 were vacuum-melted in a high frequency induction furnace using about 0.2% Mn as deoxidizer. The resulting ingot was reduced to rod specimens of 2 mm diameter and 100 mm length for measurements of electrical resistivity ρ , saturation magnetization I_s , λ_s and Young's modulus E . For K_1 measurements, single crystals were prepared under argon atmosphere by the Tamman-Bridgmann method. The optimum descending rates for growth of the single crystals ranged from 10 to 2 mm/hr, which were made smaller the larger the Nb content. The single crystals were etched in a boiling saturated ferric chrolide solution and their planes was determined by the light figure method. The crystals were then cut to plates 1.5~2.5 mm in thickness parallel to the

* This paper presented at the 1975 Spring Meeting of the Japan Institute of Metals. Reported originally in Japanese in J. Japan Inst. Metals, 40 (1976), 682; The 92nd report from the Research Institute of Electric and Magnetic Alloys.

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Table 1 Analytical results of metals used.

Metal	Co	Fe	Mn	Si (wt %)	Cu	Ta	C	S
Electrolytic Ni	0.180	0.003	0.000 ₁	0.000 ₄	0.001 ₈	—	0.002	0.000 ₃
Electrolytic Fe	—	—	0.006	0.005	0.004	—	0.005	0.005
Metalic Nb	—	0.002	0.001	0.002	—	0.015	0.023	—

(100) plane by use of the crystal goniocutter. While correcting the deviations from the (100) plane with emery abrasive paper, the plates were disk shaped to have a size of 1 mm thickness and 8 mm diameter and then polished in chromium oxide to remove the surface layer strained. They were electrolytically polished to a thickness of about 0.8 mm using a 9:1 solution of phosphoric acid and chromium trioxide. The disk-shaped specimens so prepared showed the deviations from the (100) plane to be within 1° . The measurement of torque curves was started from the [001] direction which was confirmed by the light figure method and the X-ray back Laue method.

Values for ρ , I_s , λ_s and E were measured by the potentiometric method, the ballistic galvanometer method, the optical roller method and the vibrator controlled oscillator system, respectively. The K_1 value was derived from the torque L determined by the recording torque magnetometer. The L is expressed in the form of

$$L = -\frac{\partial E_a}{\partial \theta} = -\frac{K_1}{2} \sin 4\theta, \quad (1)$$

where E_a is crystal magnetic anisotropy energy and θ is an angle between I_s and the [001] direction. Since the measured torque curve has the tendency to deviate from eq. (1) with decrease in K_1 , the coefficient of each term separated by Fourier analysis was calculated to obtain the value of K_1 .

III. Results

The electrical resistivity ρ of the specimens was measured after heating at 1150°C for 3 hr in pure hydrogen atmosphere, followed by cooling at various rates from 600°C to room temperature. Figure 1 shows the relations between variation of ρ , $\Delta\rho/\rho$ and cooling rate for the alloy specimens C-1~C-6 with nearly constant Ni content. The value of $\Delta\rho/\rho$ for specimen C-1 without Nb addition decreases with decrease in cooling rate, but the specimens with Nb addition show an increase in $\Delta\rho/\rho$ with decrease in cooling rate. The variations of ρ with cold reduction in area at room temperature are shown in Figs. 2 and 3 for the same speci-

Fig. 1 Variation of electrical resistivity ρ with cooling rate from 600°C after heating at 1150°C for 3 hr for Ni-Fe-Nb alloys: C-1 (79.65% Ni, 20.35% Fe), C-2 (79.43% Ni, 17.60% Fe, 2.97% Nb), C-3 (79.57% Ni, 15.41% Fe, 5.02% Nb), C-4 (79.58% Ni, 13.36% Fe, 7.0% Nb), C-5 (79.65% Ni, 11.39% Fe, 8.96% Nb) and C-6 (79.43% Ni, 10.74% Fe, 9.92% Nb).

Fig. 2 Variation of electrical resistivity ρ with cold reduction in area for the alloys shown in Fig. 1: Rapidly cooled ($9^\circ\text{C}/\text{sec}$) after heating at 1000°C for 1 hr.

mens which were subjected to rapid cooling ($9^\circ\text{C}/\text{sec}$) and slow cooling ($240^\circ\text{C}/\text{hr}$), respectively, after heating at 1000°C for 1 hr in pure hydrogen atmosphere. In all the specimens alloyed with Nb, the values of $\Delta\rho/\rho$ decrease with reduction in area. The slow cooling prior to cold working results in a marked increase in

Fig. 3 Variation of electrical resistivity ρ with cold reduction in area for the alloys shown in Fig. 1: Slowly cooled (240°C/hr) after heating at 1000°C for 1 hr.

$\Delta\rho/\rho$ as compared with the rapid cooling, indicating that the degree of formation of the K state is greater in the case of slow cooling.

Figure 4 shows the equi-value curves of the highest values of initial permeability μ_0 obtained previously with Ni-Fe-Nb alloys by the present

Fig. 4 Highest initial permeability μ_0 , crystal magnetic anisotropy constant K_1 and saturation magnetostriction constant λ_s for Ni-Fe-Nb alloys cooled from 600°C at optimum rates for μ_0 after heating at 1150°C for 3 hr.

author and collaborators⁽⁴⁾ and of crystal magnetic anisotropy constant K_1 and saturation magnetostriction constant λ_s obtained with the compositions designated by closed circles when cooled at an optimum rate for μ_0 from 600°C after heating at 1150°C for 3 hr. It is evident from the figure that the absolute values of K_1 and λ_s are very small near the composition for which μ_0 attains the highest value of 100000.

Figure 5 shows the relations between K_1 , cooling rate and Nb content for single crystal specimens, with nearly constant Ni contents of $79.21 \sim 79.85\%$, which were heated at 1150°C for 3 hr under pure hydrogen atmosphere and then cooled at various rates. The values for K_1 greatly vary depending on cooling rate, but its absolute value decreases with increase in Nb content. In Fig. 6 are shown the relations between K_1 , cooling rate and Fe content for single crystal specimens with nearly constant Nb content ($8.81 \sim 9.02\%$) which were cooled at various rates after heating at 1150°C for 3 hr in pure hydrogen atmosphere. An inspection of this figure indicates that the specimens exhibit negative maximum values of K_1 near the composition $\text{Ni}_3(\text{Fe}, \text{Nb})$ for all the cooling rates.

Figure 7 shows the effect of Nb content on

Fig. 5 Effect of cooling rate after heating at 1150°C for 3 hr and Nb content on crystal magnetic anisotropy constant K_1 for $79.21 \sim 79.85\%$ Ni-Fe-Nb alloys.

Fig. 6 Effect of cooling rate after heating at 1150°C for 3 hr and Fe content on crystal magnetic anisotropy constant K_1 for Ni-Fe-8.81~9.02% Nb alloys.

Fig. 7 Effect of Nb content on saturation magnetostriction constant λ_s and ΔE effect, $\Delta E/E_0$ for 79.21~79.85% Ni-Fe-Nb alloys cooled at optimum rates for μ_0 after heating at 1150°C for 3 hr.

λ_s for the rod specimens with 79.21~79.85% Ni which were cooled at an optimum rate for μ_0 after heating at 1150°C for 3 hr in pure hydrogen atmosphere and also on the ΔE effect, $\Delta E/E_0$, where E_0 is Young's modulus in the demagnetized state. It is shown that these values become smaller rapidly with Nb content. In Fig. 8 is shown the effect of Fe content on λ_s and $\Delta E/E_0$ for the rod specimens with 8.81~9.02% Nb which were cooled at an optimum

Fig. 8 Effect of Fe content on saturation magnetostriction constant λ_s and ΔE effect, $\Delta E/E_0$ for Ni-Fe-8.81~9.02% Nb alloys cooled at optimum rates for μ_0 after heating at 1150°C for 3 hr.

rate for μ_0 after heating at 1150°C for 3 hr in pure hydrogen atmosphere. These two figures indicate that the values λ_s and $\Delta E/E_0$ are very small near the composition for which μ_0 attains the highest value.

IV. Discussion

Now, let us examine the origin of high permeability revealed by Ni-Fe-Nb alloys in terms of the K state and magnetoelastic conditions of the alloys.

1. K state

Ni-Fe binary alloys always show a decrease in ρ depending on cooling rate, but the addition of Nb to the alloys increases the ρ value as shown in Fig. 1. This proves that although the K state is not present in Ni-Fe alloys, there is the initial stage of ordering with a very small anti-phase domain in Ni-Fe-Nb alloys as pointed out by Davies and others for Fe-Al alloys^{(6)~(9)}. As shown in Figs. 2 and 3, cold working always increases the value of ρ in

Ni-Fe alloys irrespective of its degree and exhibits its strong effect. In the case of Ni-Fe-Nb alloys, however, cold working decreases the value of ρ , indicating that the decrease in ρ due to destruction of the K state is greater than its increment by cold working. The slowly cooled Ni-Fe-Nb alloys are well developed in the K state compared with the rapidly cooled ones and the magnetoelastically isotropic arrangement of their atoms is considered to cause high permeability. Although the relation between the K state and high permeability can be further clarified by microstructural investigations, this approach has been proved extremely difficult as revealed by many X-ray and neutron diffraction studies on the addition of Cr, Mo, V and W to Ni-Fe alloys^{(10)~(15)}.

2. Magnetoelastic condition

Let us first consider the 180° domain wall displacements, since in general the initial permeability μ_0 is more closely related to reversible displacement of domain wall than domain rotation.

Let us now define μ_0 when the internal stress σ_i takes the form of $\sigma_0 \cos 2\pi x/l$ and consequently the internal energy varies from place to place. We obtain

$$\mu_0 = \frac{16I_s^2 l}{3\lambda_s \sigma_0 \sqrt{kT_c}} \sqrt{\frac{aK_1}{b}}, \quad (2)$$

where l is the distribution wavelength of σ_i , i.e. the domain size, k is Boltzmann's constant, a is a lattice constant, and T_c is the magnetic transformation temperature.

Next, when part of the 90° domain walls are most strongly restricted by non-ferromagnetic impurities, the domain walls are curved parabolically with increase in magnetic field strength⁽¹⁶⁾. Then, μ_0 is given by

$$\mu_0 = \frac{4\pi I_s^2}{3} \sqrt{\frac{a}{K_1 k T_c}} \frac{s^2}{b}. \quad (3)$$

Here, s^2/b is the quantity which is related to the domain dimension and its relation with the ΔE effect is given by⁽¹⁷⁾

$$\frac{\Delta E}{E_0} = \frac{3\lambda_s^2 E_0}{8} \sqrt{\frac{a}{K_1 k T_c}} \frac{s^2}{b}. \quad (4)$$

Hence, eq. (3) becomes

$$\mu_0 = \frac{32\pi I_s^2}{9\lambda_s E_0} \left(\frac{\Delta E}{E_0} \right). \quad (5)$$

As can explicitly be seen from eqs. (2) and (5), it is evident that μ_0 is closely related to K_1 , λ_s and $\Delta E/E_0$.

The values of μ_0 calculated from eqs. (2) and (5) for 79.21~79.85% Ni-Fe-Nb alloys and Ni-Fe-8.81~9.02% Nb alloys are shown in Figs. 9 and 10 respectively, with the observed values for comparison. For the calculations,

Fig. 9 Initial permeability μ_0 for 79.21~79.85% Ni-Fe-Nb alloys.

Fig. 10 Initial permeability μ_0 for Ni-Fe-8.81~9.02% Nb alloys.

our observed values of K_1 , I_s , λ_s and $\Delta E/E_0$ were used while estimating σ_0 at 3×10^8 dyn/cm² from the measured data for the ΔE effect⁽¹⁸⁾ and l at 10^{-3} cm from a domain powder pattern for Ni-Fe alloys⁽¹⁹⁾. As can be seen from the figure, the variations the calculated and observed values of μ_0 with alloy composition are quite similar to each other.

As discussed above, the addition of Nb to Ni-Fe alloys leads to the formation of the K state and the lowering of absolute of K_1 and λ_s and the ΔE effect. As a consequence, the alloys become magnetoelastically isotropic. The remarkably high permeability of these alloys may be attributed to increased domain wall mobility⁽²⁰⁾⁽²¹⁾.

V. Conclusions

Measurements of electrical resistivity, saturation magnetization, saturation magnetostriction, crystal magnetic anisotropy and Young's modulus were performed on rod specimens and (100) plane single crystal specimens of Hard-perm in the Ni-Fe-Nb system, in order to investigate the origin of high permeability of the alloys. Prior to measurements, all the specimens were heated at 1150°C for 3 hr in pure hydrogen atmosphere and then cooled from 600°C at various rates.

(1) The electrical resistivity of Ni-Fe-Nb alloys increases with decreasing cooling rate and decreases with increasing cold reduction in area, thus indicating the formation of the K state.

(2) The addition of Nb to Ni-Fe alloys results in a marked decrease of absolute values of crystal magnetic anisotropy constant K_1 and saturation magnetostriction constant λ_s and of the ΔE effect, $\Delta E/E_0$. These values become extremely small near the composition for which the initial permeability attains the highest value. The variation in $\Delta E/E_0$ with composition is quite analogous to that of λ_s .

(3) Ni-Fe-Nb alloys become magnetoelastically isotropic as the formation of the K state greatly reduces the absolute values of K_1 and λ_s and the value of $\Delta E/E_0$. The resulting ease of domain wall displacement is considered as the origin of the high permeability of the

alloys.

Acknowledgments

The author is grateful to Dr. Hakaru Masumoto, Director of the Research Institute of Electric and Magnetic Alloys, for his advice and encouragement and to Messrs. Yuetsu Murakami and Shizuo Kadowaki of the same Research Institute for their cooperation. Thanks are also due to Prof. Hideo Saito and Dr. Hiroaki Morita of Tohoku University for their assistance in measurements of crystal magnetic anisotropy.

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