Ferromagnetic Domain Structure as Affected by the Uniaxial Anisotropy Induced in a 40 Percent Co-Ni Single Crystal

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Synopsis

We have found that the domain structure in the annealed state of a 40 percent cobalt-nickel single crystal is very fine and complicated as compared with those of ordinary ferromagnetic crystals but it becomes simpler and larger after quenching from above the Curie temperature. This can be explained as follows: Since the domain structure may in general be fine and complicated at temperatures just below the Curie temperature, domain walls must displace to establish a more stable domain configuration as the temperature lowers. But, this process is suppressed appreciably at relatively low temperatures, because, in solid solution, the ferromagnetic uniaxial anisotropy is induced, in compliance with the domain distribution, by the anisotropic distribution of atoms at high temperatures and the domain wall displacement can take place only by being accompanied by the redistribution of atoms which can not occur at low temperatures. While, in quenching, the specimen crystal is cooled down so rapid that the uniaxial anisotropy and hence the anisotropic distribution of atoms can not be induced, and thus the quenched specimen crystal behaves just as ordinary ferromagnetic crystals.

It is shown that these findings together with the results of considerations reported previously lead us to the conclusion that the perminvar-type magnetic properties are due to the stabilization of domain walls by the induced uniaxial anisotropy in f. c. c. solid solutions with cubic anisotropy constants of any sign and b. c. c. solid solutions with negative cubic anisotropy constants.

I. Introduction

Recently, we[1] have interpreted the uniaxial ferromagnetic anisotropy induced by magnetic annealing in cubic solid solution alloys of the substitutional type as a magnetocrystalline anisotropy energy resulting from the anisotropic distribution of atoms occuring during magnetic annealing. We[2] have shown, further, that, according to the above-mentioned idea, when a ferromagnetic solid solution is cooled slowly from above its Curie temperature in the absence of an externally applied magnetic field, the uniaxial anisotropy may be induced along the directions of magnetization vectors distributed in compliance with the domain structure, and then, since the rotation of the magneization vector from its original stabilized direction accompanies an increase in induced uniaxial anisotropy, moving domain walls may be affected by a restoring force, namely, domain walls may be stabilized.

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(1) S. Taniguchi and M. Yamamoto, Sci. Rep. RITU, **A6** (1954), 330. S. Taniguchi, Sci. Rep. RITU, **A7** (1955), 269.

(2) M. Yamamoto and S. Taniguchi, Nippon Kinzoku Gakkai-shi, **19** (1955), 127 (in Japanese). S. Taniguchi, Sci. Rep. RITU, **A8** (1956), 173.

The effect of this domain-wall stabilization upon the wall displacement is remarkable in face-centred cubic (f. c. c.) solid solution with the cubic anisotropy constant, K, of any sign and in body-centred cubic (b. c. c.) solid solution with a negative K, and these solid solutions show, in the annealed state, the following magnetic behaviors :-

(1) Wall displacements are very difficult to be done (the initial susceptibility is small),

(2) Wall displacements are nearly proportional to an increase in applied magnetic field as long as they are small (the susceptibility is nearly constant at low fields), but further increase of magnetic field is accompanied with rapid displacements of domain walls (the susceptibility increases rapidly),

(3) For the displacements of 180° walls, there is a critical field above which they displace irreversibly, and

(4) When an externally applied magnetic field is reduced to zero, non-180° walls eventually return back to their original positions, even if they once displace far from their original positions.

Since the induced uniaxial ferromagnetic anisotropy energy is usually of the order of 10^3 erg/cc,[3] the wall displacements may be controlled mainly by the restoring force resulted from the stabilization of domain walls due to the induced uniaxial anisotropy. Thus, if the above-mentioned solid solutions had, in their annealed state, fine and complicated domain structures as compared with "ordinary" ferromagnetic substances, they should have the following magnetic characteristics :- (a) The susceptibility is nearly constant until applied magnetic field reaches to the order of 1 Oe, above which the susceptibility rises rapidly, and (b) the coercive force is pretty large (of the order of 1 Oe), while the remanence is very small, the hysteresis curve being of a constricted form. Moreover, if it were so, the anomalous perminvar-type magnetic behavior[5] such as the increase of the initial susceptibility as well as the remarkable decrease of the range of constant permeability by alternating current demagnetization, the significance of heat treatment enhancing such magnetic characteristics, etc. can be explained completely[2,4]. It is hardly needless to say that, when the ferromagnetic solid solutions concerned are cooled rapidly from above their Curie temperatures, domain walls may not be stabilized, and so the initial susceptibility rises, the coercive force decreases, and the above-mentioned anomalous magnetic characteristics disappear. It is to be noted, further, that the solid solutions concerned respond remarkably to magnetic annealing.

(3) S. Chikazumi and T. Oomura, J. Phys. Soc. Japan, **10** (1955), 842 (Fe-Ni). M. Yamamoto, S. Taniguchi, and K. Aoyagi, Read at the magnetism-section meeting of Phys. Soc. Japan, Nov. (1956) (Ni-Co). T. Nagashima, T. Yamamoto, and Y. Nakamura, Read at the magnetism-section meeting, Oct. (1955) (Ni-Co) and July (1956) (Fe-Co). L. Marechal, J. de phys., **16** (1955), 122 (Fe-Co). J. L. Snoek and J. Smith, Cited by G. W. Rathenau, Rev. Mod. Phys., **25** (1953), 55 (Fe-Ni-Co).

(4) M. Yamamoto, S. Taniguchi, and K. Aoyagi, Phys. Rev., **102** (1956), 1295 ; Oyo-Butsuri, **26** (1957), 639.

(5) Cf. R. M. Bozorth, *Ferromagnetism*, D. van Nostland, (1951), p. 160.

Now, the above-mentioned supposition that the domain structure in the annealed state of a solid solution showing the perminvar-type magnetic properties may be fine and complicated might appear, for the first sight, to be inconsistent with available theory and experiments concerning the ferromagnetic domain structure. But, since almost all of investigations of magnetic domain structures were so far made on pure metals,(6) we can not decide, from the results of these investigations, whether the above-mentioned supposition is correct or not. Previously, Bozorth and Walker(7) observed that the domain structure of a 60 percent Co-Ni alloy single crystal which shows well-defined perminvar characteristics in the annealed state(8) was fine and complicated as compared with that of silicon iron crystals, and they ascribed the cause of this phenomenon to the fact that the magneto-crystalline anisotropy constant of the alloy is negative. However, theory of the magnetic domain structure indicates that, if the shape of the specimen crystal is appropriate, the domain size is nearly independent of the number of directions of easy magnetization. It is to be noticed, further, that the very complicated domain structure observed by them is not stable energetically, since such a domain structure containing many non-180° walls accompanies an increase in magnetoelastic energy.

In order to prove the above-mentioned supposition, we have made the observation of domain structures, in the annealed state and in the state quenched from above the Curie temperature, of a single crystal of 40 percent cobalt-nickel alloy which is face-centred cubic solid solution showing well-defined perminvar characteristics.(8) We have also observed the domain structure in the state annealed in the presence of externally applied magnetic field to ascertain the perfectness of the specimen crystal used and also to know the domain structure of pure metal crystal having the same material constants and external shape as the specimen crystal.(9) The reasons why we chose 40 percent Co-Ni alloy as the specimen are:- (1) It shows well-defined perminvar characteristics in the annealed state, (2) it is easier to observe the domain structure with this alloy crystal since it has a fairly large magnetocrystalline anisotropy (about -6×10^4 erg/cm^3)(10), and (3) quenching from above the Curie temperature is conveniently done since it has a not so high Curie temperature (about 800°C)(11).

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(6) Silicon iron containing about 4 percent Si employed frequently for the study of the ferromagnetic domain structures may be regarded as equivalent to pure iron, since it is a body-centred cubic solid solution with a positive magnetocrystalline anisotropy constant, containing a small quantity of silicon.

(7) R. M. Bozorth and J. G. Walker, Phys. Rev., **79** (1950), 888.

(8) M. Yamamoto, S. Taniguchi, and K. Hoshi, Nippon Kinzoku Gakkai-shi, **17** (1953), 615; Sci. Rep. RITU, **A6** (1954), 39. M. Yamamoto and S. Taniguchi, Nippon Kinzoku Gakkai-shi, **19** (1955), 645 and 648; Sci. Rep. RITU, **A8** (1956), 280.

(9) Cf. III (4).

(10) M. Yamamoto, Nippon Kinzoku Gakkai-shi, **11** (1947), No. 11-12; **13** (1949), No. 6; Sci. Rep. RITU, **A4** (1952), 14.

(11) R. M. Bozorth, *Ferromagnetism*, D. van Nostland, (1951), p. 276.

II. Specimen Crystal and Experimental Procedure

(1) *Specimen crystal*

The specimen crystal used is a single crystal of 40 percent Co-Ni alloy, shaped into a rectangular-rod form of about $1.5 \times 0.5 \times 0.3$ cm^3, of which the rod axis is along the [111] direction, namely, one of the directions of easy magnetization and of which wide side surfaces, narrow side surfaces, and end surfaces are, $(\bar{1}10)$, $(\bar{1}\bar{1}2)$, and (111) planes, respectively (Fig. 1). It was prepared by the following procedure:- The mixture of electrolytic nickel and electrolytic cobalt in an alumina crucible was melted by a vacuum Tammann furnace and then the melt was solidified from its lower end by lowering the crucible through the furnace at the rate of 1~2 cm/hr. The single crystal thus produced was etched with conc. HNO_3 and its crystal orientation was determined by the light figure method.[12] Then, it was cut parallel to the desired crystal planes using an etching cutter[13] and the cut surfaces were polished by emery papers. Finally, after cold-worked surface layer was removed by electrolytic polishing described later, the specimen crystal was annealed at 1200°C for an hour in a hydrogen stream.

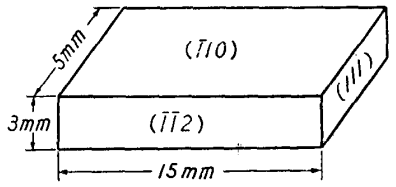


Fig. 1. Dimensions and shape of the specimen crystal used.

(2) *Experimental procedure*

The method of electropolishing employed to obtain flat crystal surfaces suitable for the domain-pattern observation is exactly the same as that which was used previously by one of the authour (Yamamoto) and Iwata[14] when they studied the domain patterns on nickel single crystals. Electropolished crystal surfaces were washed with hot water and then dried quickly. Domain patterns revealed by magnetic colloid[15] dropped onto the crystal surfaces were observed under a metallographic microscope using a bright-field illumination, and photomicrographs of domain patterns were taken on 35 mm films. In order to accentuate the patterns as well as to observe domain wall displacements, magnetic field was applied at right angles to the crystal surface by the use of a small permanent magnet. The demagnetization of the specimen crystal was done between the poles of a small electromagnet of which the alternating magnetic field was gradually diminished.

(3) *Heat treatments*

The specimen crystal was subjected to the following three kinds of heat-treatments in a vacuum. The first is slow cooling after one hour of annealing at

(12) M. Yamamoto, Nippon Kinzoku Gakkai-shi, **5** (1941), 214; Sci. Rep. Tôhoku Univ., **31** (1943), 121. Light figures of Ni-Co alloy crystals will be reported shortly by M. Yamamoto and J. Watanabé.

(13) M. Yamamoto and J. Watanabé, Nippon Kinzoku Gakkai-shi **19** (1955), 38; Sci. Rep. RITU, **A8** (1956), 230.

(14) M. Yamamoto and T. Iwata, Sci. Rep. RITU, **A5** (1953), 433.

(15) W.C. Elmore, Phys. Rev., **54** (1938), 1092; **62** (1942), 486.

1,000°C, the second is rapid cooling from an appropriate temperature above the Curie point (about 800°C), and the last is slow cooling in an externally applied magnetic field or magnetic annealing.

In order to cool the specimen crystal as rapid as possible in a vacuum, a special apparatus as shown in Fig. 2 was deviced. In an evacuated heating tube (quartz)

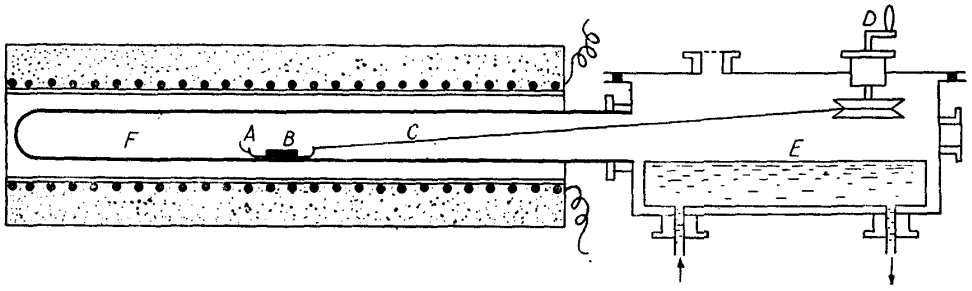


Fig. 2. Quenching apparatus employed.

A :	Nickel boat	D :	Handle
B :	The specimen	E :	Water-cooled copper plate
C :	Thin nichrom wire	F :	Quartz tube

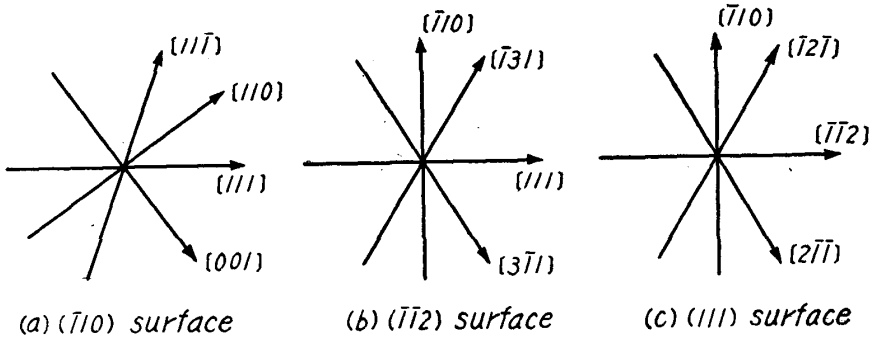
of a nichrome furnace, a small nickel boat on which the specimen crystal lies, is brought, as fast as possible, onto a water-cooled copper plate by pulling a nichrome wire attached to it. Thus, after rapid cooling, the domain pattern observations can be made without re-electropolishing the specimen crystal which may affect the domain structure through magnetic field due to the current passed.

Magnetic annealing was done by placing the specimen crystal in a solenoid set in a nichrome furnace and by applying magnetic field of 560 Oe from 800°C in the way of cooling at the rate of 200°C/hr after one hour's heating at 1,000°C.

III. Experimental Results

(1) *($\bar{1}10$) domain patterns in the annealed state and in the quenched state.*

The arrangement of principal crystallographic directions including the directions of easy magnetization <111> in the ($\bar{1}10$) surface is shown in Fig. 3(a). In the ($\bar{1}10$) surface, there are two directions of easy magnetization, one of which is the [111] direction parallel to the rod axis and the other is the [11$\bar{1}$] direction making



(a) ($\bar{1}$10) surface (b) ($\bar{1}\bar{1}$2) surface (c) (111) surface

Fig. 3. Arrangement of important crystallographic directions including the directions of easy magnetizations <111> in the ($\bar{1}$10), ($\bar{1}\bar{1}$2), and (111) surfaces of the specimen crystal.

an angle of 71° to the former. Photo. 1[16] shows a domain pattern on the $(\bar{1}10)$ surface observed in the annealed state, which is devided in a complicated way by 180° walls parallel to the [111] and $[11\bar{1}]$ directions, 109° walls parallel to the [110] direction, and 71° walls parallel to the [001] direction (cf. Fig. 4).

On the contrary, domain patterns observed in the state quenched from above the Curie temperature show a simple configuration which is mainly composed of 180° domains magnetized parallel to the [111] direction, of which the size is very

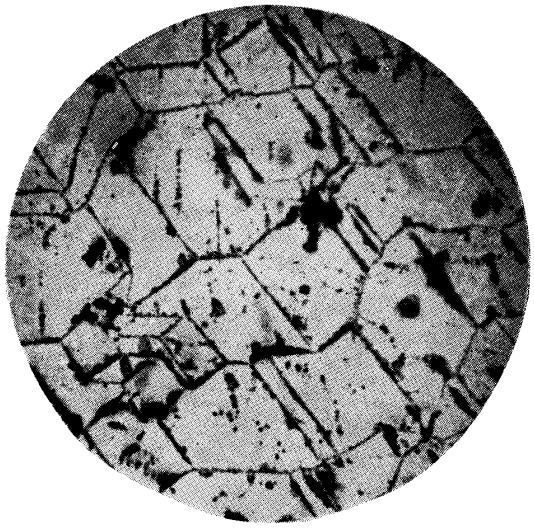


Photo. 1. $(\bar{1}10)$ domain pattern in the annealed state. Weak normal field is absent ($H_n=0$). ×280

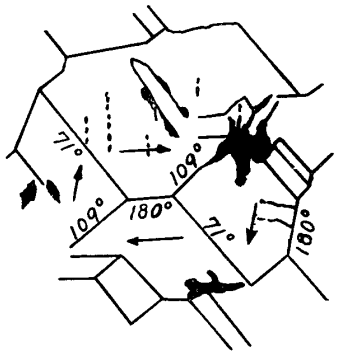


Fig. 4. Interpretation of the $(\bar{1}10)$ domain pattern of Photo. 1.

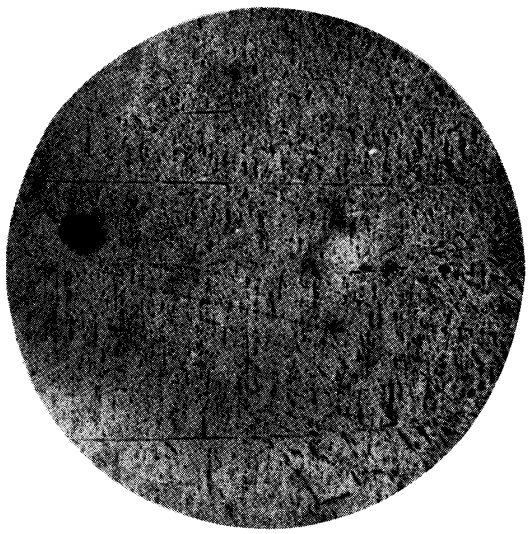


Photo. 2. $(\bar{1}10)$ domain pattern in the state quenched from above the Curie temperature. Weak normal field is applied upwards to the crystal surface ($H_n=+$). ×80

Photo. 3. $(\bar{1}10)$ domain pattern in the state quenched from 850°C. $H_n=+$. ×80

(16) The photomicrographs and figures of domain patterns on the $(\bar{1}10)$, $(\bar{1}\bar{1}2)$, and (111) surfaces are placed in the positions corresponding to Figs. 3 (a), (b), and (c), respectively.

(a) $H_n=0$

(b) $H_n=+$

(c) $H_n=-$

Photo. 4. ($\bar{1}10$) domain pattern in the state quenched from 900°C. ×80

Photo. 5. ($\bar{1}\bar{1}2$) domain pattern in the annealed state. $H_n=0$. ×280

Photo. 6. ($\bar{1}\bar{1}2$) domain pattern in the state quenched from 850°C. $H_n=0$. ×80

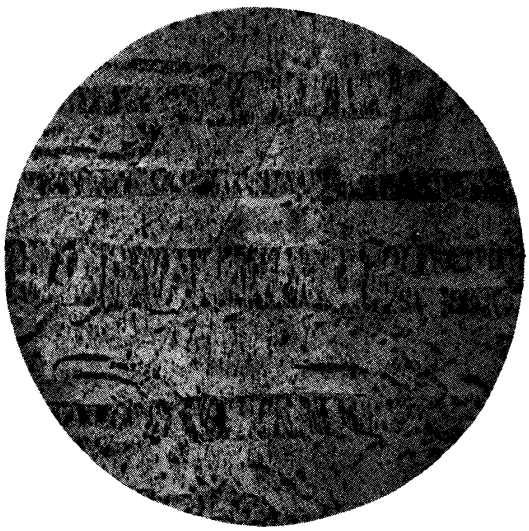


Photo. 7. ($\bar{1}\bar{1}2$) domain pattern in the state quenched from 950°C. $H_n=+$. ×80

much larger than that of 180° domains in the annealed state, as seen, for example, from Photo. 2. It is to be noted, however, that, in the state quenched from 850°C, long and slender domains bounded by 71° walls — they are called, for simplicity, 71° domains in the following — predminate in the major part of the crystal surface, as shown in Photo. 3, and on raising the quenching temperature the area occupied by 71° domains diminishes rapidly, as seen from Photo. 4, there being almost no difference between quenching from 900°C and that from 950°C. This may possibly be due to the fact that, with our quenching apparatus, during the specimen crystal is brought from the furnace onto the water-cooled copper plate, its temperature inevitably lowers to such an extent that rapid cooling from 900°C may acutually be that from just above the Curie temperature (about 800°C). Furthermore, it is to be noted that, in Photos. 3 and 4, 71° walls predominate as compared with 109° walls, which may be attributed to the fact that the energy of 71° walls is less than that of 109° walls, as the caluculation of wall energies by Lilley[17] shows.

The domain pattern in the annealed state (Photo. 1) is so stable that it is not altered by alternating demagnetization and domain walls do not nearly at all displace from their original positions by a weak magnetic field applied. On the contrary, in the quenched state domain walls are very easy to displace. For example, domain patterns in the same part on the $(\bar{1}10)$ surface in the quenched state observed when (a) no field was applied, (b) a weak normal field produced by a small permanent magnet was applied, and (c) the sense of the weak normal field was reversed, are shown, respectively, in Photo. 4(a), 4(b), and 4(c), from which it may be obvious that domain walls, in particular 180° walls, displace easily from their original positions. In the annealed state, however, domain walls did not displace by an applied field of the same order as that applied in the case of Photo. 4.

(2) *$(\bar{1}\bar{1}2)$ domain patterns in the annealed state and in the quenched state.*

The arrangement of principal crystallographic direction including the directions of easy magnetization $<111>$ in the $(\bar{1}\bar{1}2)$ surface is shown in Fig. 3(b). In the $(\bar{1}\bar{1}2)$ surface, there is only one direction of easy magnetization $[111]$ parallel to the rod axis. It may be expected, therefore, that only 180° domains magnetized parallel to the direction of easy magnetization lying in the surface are found on this surface, as in cases of the $(\bar{1}\bar{1}2)$ surface of nickel crystal and of the (110) surface of silicon-iron crystal. But, as seen from Photo. 5, the domain pattern observed acutually on the $(\bar{1}\bar{1}2)$ surface in the annealed state involves only a few 180° domains locally and the major part of the surface is composed of domains bounded by curved vein-like walls. The domains enclosed by curved walls are elongated along nearly $[\bar{1}31]$ and $[3\bar{1}1]$ directions (cf. Fig. 3(b)) and radiate small subsidiary domains, making a complicated structure. These domains may be regarded as closure domains composed of domains magnetized parallel to three directions of easy magnetization $[\bar{1}\bar{1}\bar{1}]$, $[\bar{1}\bar{1}1]$, and $[\bar{1}11]$ which are not contained in the surface.[18] Such a fact that the major part of flat $(\bar{1}\bar{1}2)$ surface is composed

(17) B. A. Lilley, Phil. Mag., **41** (1950), 792.

(18) T. Iwata and M. Yamamoto, Sci. Rep. RITU, **A6** (1956), 293, Section IV.

of domains magnetized parallel to the directions of easy magnetization which are not contained in the surface has never been found with pure nickel crystal[18] and gives a direct evidence for the fact that, as stated before in the introduction, in a solid solution alloy the uniaxial ferromagnetic anisotropy is induced in compliance with the distribution of magnetic domains at the time of the heat treatment.

In the state quenched from 850°C, the major part of the crystal surface are occupied by 180° domains magnetized parallel to the [111] direction of easy magnetization, but these 180° domains contain many fine island-like domains and, moreover, 180° walls are considerably wavy, as shown in Photo. 6. These island-like domains may be interpreted as closure domains produced by the intersection of long and slender 71° domains as seen on the $(\bar{1}10)$ surface (Photo. 3) with this crystal surface $(\bar{1}\bar{1}2)$ (cf. Fig. 5). On raising the quenching temperature above 900°C, the island-like domains disappear, corresponding to the above-mentioned rapid decrease of 71° domains on the $(\bar{1}10)$ surface. The width of 180° domains in the state quenched from 950°C (Photo. 7) are far larger than that in the annealed state (Photo. 5).

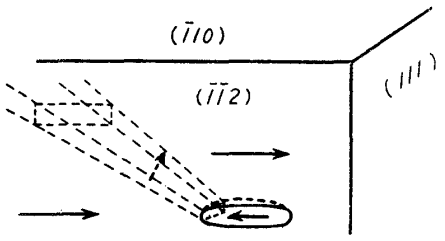


Fig. 5. Interpretation of island-like 180° domains in the $(\bar{1}\bar{1}2)$ domain pattern in the state quenched from 850°C.

It is to be noted, further, that the complicated domain pattern in the annealed state is extremely stable and is not altered by alternating current-demagnetization, just as in the case of the $(\bar{1}10)$ domain pattern mentioned above.

(3) *(111) domain patterns in the annealed state and in the quenched state.*

The arrangement of the principal crystallographic directions in the (111) surface are shown in Fig. 3(c), which indicates that any direction of easy magnetization does not lie in this crystal surface. It may be expected, therefore, that the domain pattern on the (111) surface is remarkably complicated as compared with those on the $(\bar{1}10)$ and $(\bar{1}\bar{1}2)$ surfaces, just as already observed with nickel crystals.[14,18] In fact, not only in the annealed state (Photo. 8) but also in the quenched state (Photo. 9), domain patterns on the (111) surface are equally composed of three sets of thick lines running along $<\bar{1}\bar{1}2>$ directions (cf. Fig. 3(c)) and fine structure between these thick lines.

(4) *$(\bar{1}10)$, $(\bar{1}\bar{1}2)$ and (111) domain patterns in the state annealed in an externally applied magnetic field.*

On annealing single crystal of ferromagnetic cubic solid solution in an externally applied magnetic field, the uniaxial ferromagnetic anisotropy having the direction of easy magnetization parallel to the direction of field applied during annealing or its nearby direction is induced uniformly over the whole of the specimen crystal, so that the domain structure composed mainly of 180° domains magnetized parallel to the direction of easy magnetization determined by both the induced uniaxial

Photo. 8. (111) domain pattern in the annealed state. $H_n = +$. ×280

Photo. 9. (111) domain pattern in the state quenched from 850°C. $H_n = +$. ×280

ferromagnetic anisotropy energy and the proper cubic magnetocrystalline anisotropy energy may be expected. Since, in the present experiment, magnetic field parallel to the rod axis of the specimen crystal, namly, one of the directions of easy magnetization determined by the cubic magnetocrystalline anisotropy alone was applied during annealing, 180° domains magnetized parallel to this direction ought to be formed. Also closure domains may naturally be formed in both ends of the specimen crystal.[19] But, although the closure domains involve the induced uniaxial ferromagnetic anisotropy energy in addition to the magnetoelastic energy, unlike the case of pure metal crystals, the width of main 180° domains may be roughly the same, though a little narrower, as that in pure metal crystals with the same shape, size and material constants as our alloy specimen crystal[20].

$(\bar{1}10)$, $(\bar{1}\bar{1}2)$, and (111) domain patterns in the state annealed in an externally applied magnetic field of 560 Oe are shown, respectivly, in Photos. 10~13, which show that the size of domains are of nearly the same order as that in the quenched state. It is to be noted that such a domain pattern as shown in Photo. 13 was observed near the end parts of the $(\bar{1}10)$ surface. This may not be the tree pattern caused by an slight inclination of the crystal surface from the $(\bar{1}10)$ plane but may be a domain pattern caused by the fact that magnetic field applied during annealing was not strong enough to saturate the end parts of the specimen crystal.

(19) The induced uniaxial ferromagnetic anisotropy is not so large that the domain structure may be similar to that of cubic crystals rather than that of cobalt crystal.

(20) The width of main 180° domains is $1/\sqrt{2}$-fold of that in the absence of the induced uniaxial anisotropy energy, when the magnetoelastic energy is equal to the uniaxial anisotropy energy. The influence of the induced uniaxial anisotropy on the width of the domains through a change in wall energy may be small, since the induced uniaxial anisotropy is far smaller than the cubic magnetocrystalline anisotropy in 40 percent Co-Ni alloy.

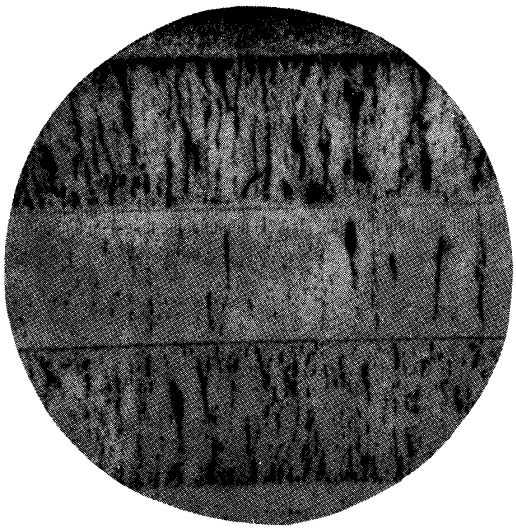


Photo. 10. $(\bar{1}10)$ domain pattern in the state annealed in magnetic field applied along the [111] direction of easy direction. $H_n = +$. ×100

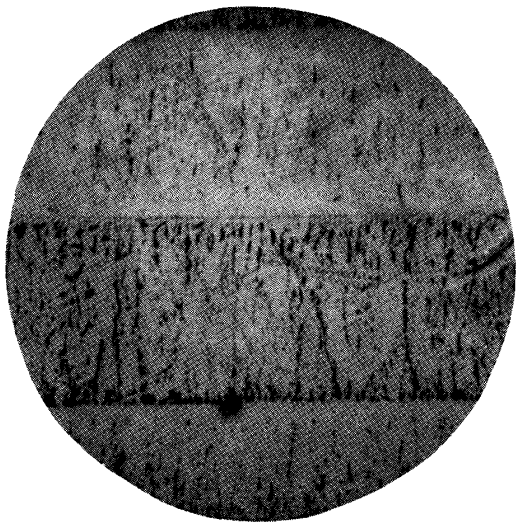


Photo. 11. $(\bar{1}\bar{1}2)$ domain pattern in the same state as Photo. 10. $H_n = +$. ×100

Photo. 12. (111) domain pattern in the same state as Photo. 10. $H_n = +$. ×100

Photo. 13. $(\bar{1}10)$ domain pattern observed near the ends of the surface in the same state as Photo. 10. $H_n = +$. ×100

IV. Considerations

In a long and slender single crystal of ferromagnetic cubic metal of which the rod axis is parallel to one of the directions of easy magnetization, the domain structure with 180° domains magnetized parallel to the rod axis is most favourable energetically. Non-180° domains, if they are, can exist only in the end parts of the specimen crystal, since they are accompanied with the magnetoelastic energy. Thus, with a rod single crystal of 40 percent Co–Ni alloy with a negative magnetocrystalline anisotropy constant, which is long along one of the <111> directions, it may be expected that 180° domains magnetized parallel to the rod axis are

predominant and hence such 180° domains are found on its $(\bar{1}10)$ and $(\bar{1}\bar{1}2)$ surfaces. Actually in the annealed state, however, domain patterns on these surfaces (Photos. 1 and 5) are very complicated and fine, and the domain distribution seems to be independent of the shape and size of the specimen crystal, as mentioned before. Also, in the $(\bar{1}10)$ surface, two kinds of domains magnetized parallel to two directions of easy magnetization lying in this surface exist together (Photo. 1), though, it may be supposed that, in the interior of the specimen crystal, the directions of magnetization vectors in domains are distributed nearly equally along four directions of easy magnetization. Moreover, as mentioned before, the domain structure in the annealed state is very stable and is affected little by alternating-current demagnetization. These facts indicate the stabilization of domain walls due to the induced uniaxial ferromagnetic anisotropy, which is supported by the fact that domain walls become easy to move after quenching the specimen crystal. Further, we have observed that, by quenching, domains become larger, in particular 180° domains magnetized parallel to the rod axis of the specimen crystal increase in volume, the domain structure becoming simpler (Photos. 2~4), indicating that both the wall and the magnetoelastic energies are reduced. This indicate that the domain structure in the annealed state are never an energetically stable one.

Then, why such an energetically unfavourable domain structure is realized in the annealed state? On cooling a ferromagnetic crystal of a finite size from above its Curie temperature, the spontaneous magnetization arises and the crystal is devided into domains when the temperature has attained the Curie temperature. Just below the Curie temperature where both the magnetoelastic and wall energies are very small, the domain structure may be very complicated and fine because of thermal effects or because of inevitable defects (internal stress, non-uniformity of the alloy composition, etc.). But, as the temperature further lowers, such a complicated and fine domain structure may become unstable since both the magnetoelastic and wall energies increase, so that an increase in volume of domains and their rearrangement may take place, resulting a domain structure such as observed usually at room temperature. Now, in a solid solution such as 40 percent Co–Ni alloy in which the uniaxial ferromagnetic anisotropy is induced, since wall displacements must always accompany the redistribution of atoms, wall displacements may be comparatively easy at high temperatures but they may be suppressed as the temperature lowers or as the rate of diffusion of atoms becomes small, and at room temperature domain walls cannot take the energetically lowest position. On the contrary, when such a solid solution is quenched from above its Curie temperature, it is cooled keeping the state in which atoms are distributed at random, so that the situation becomes exactly the same as in pure metals, namely, the domain structure in the quenched state may, ideally, become equivalent to that which would be realized in pure metals with the same material constants as the solid solution. In other words, on quenching the solid solution from above its Curie temperature, the size of domains, especially that of 180° domains, becomes large.

Indeed, the size of domains in our specimen crystal quenched from 900°C is

of nearly the same order as that which may be expected with pure metal crystal specimens having the size comparable to that of our crystal specimen. Actually, however, the domain structure in the state quenched from above the Curie temperature (Photos. 6 and 7) is not necessarily one of the energetically lowest states, namely, the state in which all of 180° domains are lined up along the rod axis as in the state annealed in an externally applied magnetic field (Photos. 10 and 11), but one which involves, besides 180° domains magnetized parallel to the rod axis, a pretty large number of 180° domains magnetized parallel to the other direction of easy magnetization, namely the $[11\bar{1}]$ direction. This is analogous to the domain structure in the thermally demagnetized state of an iron rod single crystal with the rod axis parallel to one of the <100> directions, which involves a pretty large number of domains with magnetization vectors parallel to the other <100> directions which are not parallel to the rod axis. In the case of iron single crystal, however, it can be seen from magnetostriction curves, that, if it is demagnetized by alternating field parallel to the rod axis, magnetization vectors of nearly all of domains are lined up along the rod axis. In the case of a 40 percent Co–Ni alloy single crystal quenched from above its Curie temperature, on the contrary, after alternating current demagnetization the group of 180°domains with magnetization vectors parallel to the $[11\bar{1}]$ direction appear again as a whole in nearly the same positions as before the demagnetization, though the positions of individual 180° walls deviates from their initial ones. This might be connected with the fact that the occurence of the uniaxial ferromagnetic anisotropy can not completely be suppressed by such rapid cooling as we have done or with the fact that this $[11\bar{1}]$ direction is by chance the direction along which our specimen crystal grew from the melt.

Now, in this way, the observation of the fact that the specimen crystal which may be regarded as magnetically perfect in the quenched state has an anomalously fine and complicated domain structure in the annealed state has verified our supposition mentioned in the introduction, that the domain structure in the annealed state of a solid solution showing the perminvar-type anomalous magnetic properties are fine and complicated. The perminvar-type magnetic properties can, therfore, be explained by the stabilization of domain walls as due to the uniaxial ferromagnetic anisotropy energy caused by the anisotropic distribution of atoms induced by annealing a cubic ferromagnetic solid solution in an externally applied magnetic field. And, it may be considered that such an anomalously fine and complicated domain structure is an "anomalous one" which can be observed only in the annealed state, the influence of the material constants and others for this domain structure being small. This is in harmony with the fact that the perminvar type anomalous magnetic properties have been found, to a more or less extent, in a wide range of composition in various ferromagnetic alloy systems.

V. Summary and Conclusion

We have studied the ferromagnetic domain structure and its change with heat-treatment in a 40 percent Co–Ni single crystal by observing, with the

powder-pattern technique, the domain patterns on the $(\bar{1}10)$, $(\bar{1}\bar{1}2)$, and (111) surfaces of the specimen crystal, which is long along the [111] direction. We have found that the domain structure is fine and complicated in the annealed state, but that, in the state quenched from above the Curie temperature, domains take the same size and structure as predicted from the magnetic domain theory and also as observed with pure metal crystals. In the state annealed in an externally applied magnetic field, we have also observed the same domain structure as predicted from the domain theory and also as observed in the state quenched from above its Curie temperature.

These experimental facts suggest the following conclusions. The fine and complicated domain structure in the annealed state is not proper to this alloy crystal or is not due to defects in the specimen crystal, but is characteraristic to the annealed state. At temperatures just below the Curie temperature, the domain structure may generally be very fine and complicated possibly due to the thermal agitation or a few defects which may be inevitable in our specimen crystal but may not affect the domain structure at room temperature, since both the wall and magnetostrictive energies are very small. And, in pure metal, as the temperature lowers, this domain structure may transform gradually to a simple and large-scaled structure such as observed at room temperature. But, in the case of an alloy such as 40 percent Co–Ni alloy, in which a uniaxial ferromagnetic anisotropy can be induced along the direction of easy magnetization determined by the magneto-crystalline anisotropy, if it is cooled slowly to a temperature at which the diffusion of atoms become impossible, the transformation of the domain structure may naturally be stopped before taking the one to be taken at room temperature, if it is quenched the domain structure becomes to be the same as that of pure metal since it may be brought to the room temperature before the uniaxial ferromagnetic anisotropy is induced, and if it is annealed in an externally applied magnetic field, the domain structure composed only of 180° domains magnetized parallel to the direction of the field applied during annealing is frozen.

Thus, the fine and complicated domain structure in the annealed state as the condition necessary to explain the perminvar-type anomalous magnetic properties as resulted from the stabilization of domain walls due to the induction of the uniaxial ferromagnetic anisotropy has been verified experimentally. Cousequently, it can also be concluded that the perminvar-type anomalous magnetic properties are in fact common to face-centred cubic solid solutions with the cubic anisotropy constant, K, of any sign and body-centred cubic solid solutions with a negative K, both of the substitutional type.

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