Indian J. Phys. **49**, 143-154 (1975)

Anisotropy in low-field transverse magnetoresistivity of nickel-copper alloys at room temperature

S. N. KAUL*

Department of Physics, Indian Institute of Technology, Kharagpur, West Bengal

(*Received* 25 *May* 1974, *revised* 12 *November* 1974)

Transverse magnetoresistivity measurements have been performed at room temperature on recrystallized plate samples of nickel and nickel-copper alloys with 9.38, 18.77 and 28.32 atomic percent copper in fields upto 10 KOe. Polar plots of 90.62 and 81.23 at.% nickel alloys at $H = 1.55$ KOe exhibit anisotropies in transverse magnetoresistivity characteristic of pure nickel samples while 71.68 at.% nickel alloy shows an isotropic behaviour. The general trend of these anisotropies is retained for all the samples even at $H = 7.75$ KOe although a negative minimum occurs instead of a positive maximum. At $H = 7.75$ KOe only the purest nickel sample shows marked anisotropy which is in close agreement with Marsocci's observation on single crystal nickel thin films. This close agreement suggests that spin orbit interaction can, in part, account for the observed anisotropy. Such an interaction fails to provide a satisfactory explanation for the anisotropies observed at $H = 1.55$ KOe. These anisotropies, however, lend themselves to a straight forward explanation if transport theory is modified slightly to include shape anisotropy. The experimental data when analysed in the light of such a theory yields the saturation value of transverse magnetoresistivity and a component of longitudinal magnetoresistivity.

1. INTRODUCTION

The magnetoresistivity of bulk single crystals has aroused considerable interest in a number of workers starting with Kaya (1928) and Döring (1938) to the investigations made by Fawcett & Reed (1962) and Marcus & Langenberg (1963). Similar investigations on wiskers, polycrystalline thin plates and single crystal thin films of nickel and nickel-based alloys include contributions due to a large number of investigators (Isin & Coleman 1965, Ehrlich *et al* 1967, Marsocci 1964, Walden & Cotellessa 1967, Chen & Marsocci 1972). Most of the above mentioned measurements have been performed on very pure samples, at very low temperatures and in very high magnetic fields, satisfying thereby the high-field condition namely, $\omega_c \tau > 1$ where ω_c is the cyclotron frequency and τ is the average relaxation time of an electron in a cyclotron orbit. In the high-field

*Present Address : Defence Metallurgical Research Laboratory, P.O. : DMRL, Hyderabad 500 258.

region, τ is very long compared with the characteristic time necessary to complete a cyclotron orbit and the effect of collisions upon the electronic motion is then a negligible perturbation compared with the effect of the topological features of the Fermi surface. Therefore, the main interest underlying all such investigations has been to map the Fermi surface and to study the size effects which come into play when the sample thickness becomes comparable with the electronic mean free path. Relatively less emphasis has been laid on the measurements carried out either in low magnetic fields on samples containing appreciable amounts of impurity or lattice imperfections or at high temperatrures, where phonon scattering gives a significant contribution. Alternatively, scanty experimental data on magnetoresistivity are presently available in the low-field region ($\omega_c\tau < 1$) where τ remains short enough and the topological features of the Fermi surface are not manifested in the electronic motion. The low-field magnetoresistivity should, therefore, provide a powerful tool to reveal dominant scattering processes in such materials.

Although the properties of the Fermi surface have become relatively well understood, considerable speculation still exists regarding the possible resistivity mechanism in the ferromagnetic metals. Several qualitative features of low-field magneto resistivity have been attributed mainly to main wall scattering—a possibility which has already been ruled out by the investigations made by Schwerer & Silcox (1971) over a large number of nickel specimens. The possible scattering mechanism, to which the low-field magnetoresistivity can be attributed, remains yet to be explored, especially when limited experimental data in this field region are presently available.

In the present communication, we report the detailed measurements of low-field transverse magnetoriesistivity performed on textured plate samples of nickel and nickel-copper alloys at room temperature. The misorientation effects in the low-field region have been fully taken care of and the results discussed in the light of existing theories modified slightly to include shape anisotropy.

2. Experimental Details

The present work employed moderately pure nickel samples in the form of sheets, procured from E. Merch (nominal purity, 99.9%) and International Nickel Ltd. (99.99%), in addition to Johnson Matthey spectrographically pure specimens of nickel and nickel-copper alloys used in earlier work (Dutta Roy & Subrahmanyam 1969). All the samples were cut to rectangular shape ($25\times5\times0.3$ mm^3) with their length along the rolling direction and were annealed for 24 hours at 850°C in a vacuum of 10^{-5} torr. The design of the sample holder and current measuring circuit was essentially the same as used by Dutta Roy & Subrahmanyam (1969) but the present voltage measuring circuit employed a galvanometer amplifier which combined the advantages of the design details recommended

by various workers (Preston 1946, MacDonald 1947, Muir 1964), and provided a better merit figure ($\sim 10^{-9}$ volts) and a greater simplicity in measurement than achieved earlier. Extra provision for rotating the sample in the external magnetic field was made by fixing a vernier on the top of the sample holder. Great care was exercised while aligning the sample in the external field since the low-field magnetoresistivity behaviour is very much sensitive to the orientation of the sample current with respect to the magnetic field as pointed out by Berger & DeVroomen (1965) and Schwerer & Silcox (1971). The correct alignment of the sample in the external field was ensured by aligning the sample till the values of magnetoresistivity at $\theta = 0°$ and $\theta = 90°$ were reproduced at $\theta = 180°$ and $\theta = 270°$ respectively, where θ is the angle between the plane of the sample plate and the direction of magnetic field, which lies in a plane perpendicular to the sample plane. In order to ensure the reproducibility of the experimental data, the rotation of the sample in the external field was well guided throughout. The standardization of the apparatus was done by measuring the field variation of transverse magnetoresistivity of a nickel wire of diameter 0.061 cm and length 2.5 cm : the transverse magnetoresistivity at 3 KOe was always found to be 1.26×10^{-2}, which is within two per cent of the value deduced from Englert's data quoted by Chikazumi (1964).

3. Experimental Results

3.1. *Pure Nickel Samples*

The polar plots of transverse magnetoresistivity have been recorded at room temperature for three pure nickel samples labelled as sample No. 1 (Johnson Matthey), sample No. 2 (E' Merch) and sample No. 3 (International Nickel Ltd.) in figures 1*a* and 1*b* at $H = 1.55$ KOe and 7.75 KOe respectively. The curves for all the three samples in figure 1*a* indicate a positive and a negative maximum at $\theta = 90°$ and 60° respectively. Figure 1*b* reveals that a negative minimum occurs instead of a positive maximum and the anisotropy is marked only in the purest nickel sample (sample No. 1). Figure 2 depicts the field variation of transverse magnetoresistivity along (a) $\theta = 90°$ and (b) $\theta = 60°$ directions. The field dependence of transverse magnetoresistivity along $\theta = 60°$ bears a marked similarity with the field variation along $\theta = 0°$ except for the fact that magnetoresistivity along $\theta = 0°$ direction saturates at relatively lower magnetic fields. The field variation along $\theta = 0°$ and 60° directions resembles the usual behaviour of a typical ferromagnet whereas along $\theta = 90°$ magnetoresistivity passes through a positive maximum at low fields and almost saturates at high fields.

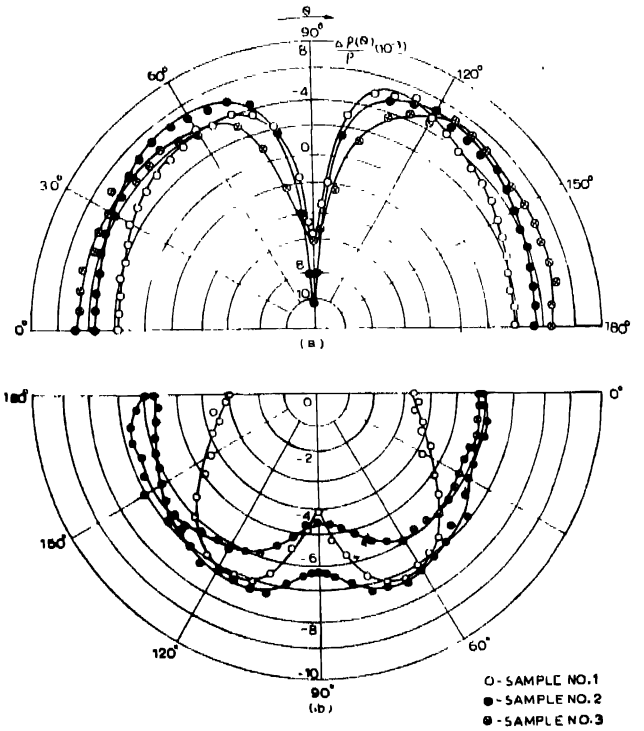

Fig. 1. The magnetoresistivity as a function of the angle θ between the magnetic field direction and the plane of nickel strips at room temperature (300°K) and at (a) 1.55 KOe, (b) 7.75 KOe.

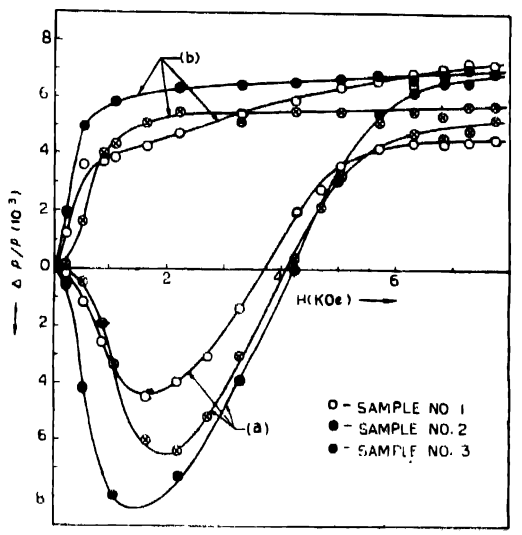

Fig. 2. The field dependence of magnetoresistivity of pure nickel samples for the directions of (a) $\theta = 90°$ and (b) $\theta = 60°$ magnetoresistivity variation in Fig. 1 (a).

3.2. *Nickel-Copper Alloys*

Figures 3*a* and 3*b* are the graphical manifestation of the angular dependence of magnetoresistivity $(\Delta\rho/\rho)$ for nickel copper alloys at $H = 1.55$ KOe and 7.75 KOe respectively; nickel data has been included for comparison. The salient features of these polar diagrams can be summarized as follows.

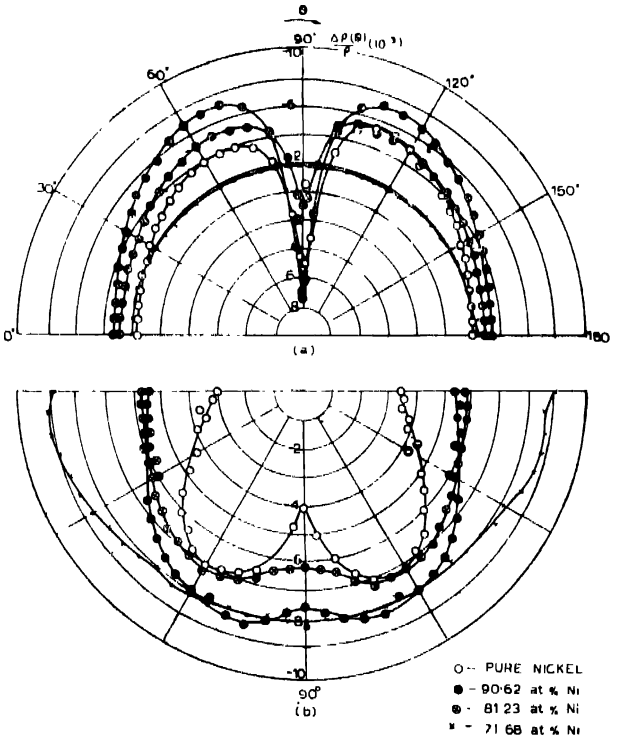

Fig. 3. Polar plot of magnetoresistivity of nickel-copper alloys at room temperature (300°K) and at magnetic fields (a) 1.55 KOe, (b) 7.75 KOe.

The polar diagrams at $H = 1.55$ KOe of 90.62 and 81.23 at.% of nickel indicate pronounced positive and negative maxima characteristic of pure nickel while 71.68 at.% nickel alloy shows nearly an isotropic behaviour. The polar plots at $H = 7.75$ KOe show that all these alloy samples retain their low-field behaviour except for the occurence of a negative minimum instead of a positive maximum.

Figure 4 shows the field variation of magnetoresistivity $(\Delta\rho/\rho)$ along (a) $\theta = 90°$ and (b) $\theta = 60°$ directions for the purest nickel sample and its alloys. Like pure nickel, the field variation of magnetoresistivity for the alloy samples along $\theta = 0°$ resembles closely the variation along $\theta = 60°$ direction. For

90.62 and 81.23 atomic per cent nickel alloys magnetoresistivity along $\theta = 90°$ exhibits a positive maximum characteristic of pure nickel whereas for 71.68 atomic per cent nickel alloy instead of this positive maximum a small negative minimum occurs and thereafter transverse magnetoresistivity increases almost linearly with the external magnetic field upto the highest field available.

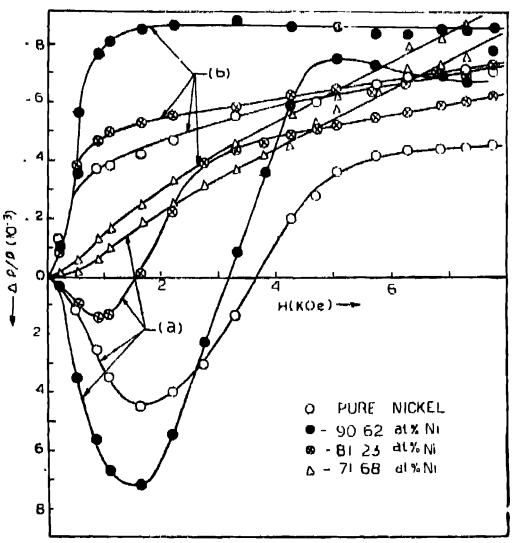

Fig. 4. The field dependence of magnetoresistivity of nickel-copper alloys for (a) $\theta = 90°$ and $\theta = 60°$ directions of the polar plot Fig. 3(a).

3.3. *Experimental Accuracy*

The measuring system imposed a limit on the resolution of $(\Delta\rho/\rho)$ of order 10^{-4}, however, slight misalignment while rotation of the sample caused errors which amounted to about 2% in the total value of $(\Delta\rho/\rho)$ in the polar plots. The experimental accuracy for the magnetoresistivity field dependence data was limited solely by the measuring system since the samples in those two directions were almost perfectly aligned by the procedure described in section 2.

4. Theoretical Background and Discussion

Similar anisotropies in magnetoresistivity have been observed, in the past, by Kaya (1928) and Doring (1938) in bulk nickel single crystals and more recently by Marsocci (1964), Walden *et al* (1967) and Chen *et al* (1972) in single crystal and polycrystalline thin films of nickel and nickel-based alloys. It is customary to explain these anisotropies on the basis of a domain theory within the frame work of which magnetoresistivity in any crystal direction is written in the form of a power series in the direction cosines of current and magnetization with respect to the crystal axes. Marsocci (1965), extending Smit's (1951) calculations,

has explained the observed anisotropy in transverse magnetoresistivity partly in terms of spin-orbit interaction. Both spin-orbit and domain theories are, however, inadequate to account for the observed anisotropies at $H = 1.55$ KOe where magnetoresistivity passes through both positive and negative values.

In the text that follows, intention is to show that the observed anisotropies and the field dependence data for all the samples can be easily explained on the basis of a theory incorporating shape anisotropy.

The anisotropy observed in transverse magnetoresistivity of the purest nickel sample at $H = 7.75$ KOe is in close agreement with the observations of Marsocci (1964) in single crystal nickel thin film at 12 KOe lying in a plane perpendicular to the direction of current which was parallel to [100] direction. The polar diagram of Marsocci (1964) when the current was parallel to [110] direction shows entirely different characteristics. This comparison evidently suggests that in the textured nickel plates employed for the present investigation [100] direction lies along the length of the plate and the current is parallel to this direction. The same argument holds for 90.62 and 81.23 atomic per cent nickel alloys whose back reflexion photographs reveal considerable texture reminiscent of pure nickel samples. This point of view is further supported by the detailed X-ray analysis, quoted, by Bozorth (1951) and Barrett & Massalski (1966), which shows that in nickel and nickel-based alloys (001) [100] texture develops after recrystallization.

The shape of the present samples and their considerable texture suggest that both shape as well as magnetocrystalline anisotropy play a dominant role in aligning the spontaneous magnetization along one of the easy directions of magnetization. Ample evidence exists in literature (Pippard 1963, Kittel 1963, Anderson & Gold 1963, Smit 1951) supporting the thesis that the appropriate field acting on a conduction electron in a ferromagnet is the magnetic induction $B(= H+4\pi M_s)$ and not the applied field H. Thus, all the subsequent arguments are based on the magnetic induction B instead of the magnetic field H. In the case of cubic metals, the solution of the Boltzmann transport equation in relaxation time approximation employing the procedure of Jones & Zener (1934*a*) in the low-field limit leads to the following expression for magnetoresistivity (Ziman 1960).

$$\Delta\rho/\rho = B^2/\rho\{\rho_1^{(2)}+\rho_2^{(2)}\Sigma\,\alpha_i^2\beta_i^2+\rho_3^{(2)}(\Sigma\,\alpha_i\beta_i)^2\}, \qquad (1$$

where α_i and β_i are the direction cosines of the current and magnetic induction vectors, referred to the crystal axes; ρ is the resistivity in zero magnetic field and $\rho_1^{(2)}$, $\rho_2^{(2)}$, $\rho_3^{(2)}$ are integrals (over K-space) which involve various second-order transport coefficients. For the direction of current along [100] direction, this expression reduces to

$$\Delta\rho/\rho = B^2/\rho\{\rho_1^{(2)}+(\rho_2^{(2)}+\rho_3^{(2)})\beta_1^2\}. \qquad \ldots\ (2)$$

The saturation values of magnetoresistivity for longitudinal, $(\Delta\rho_{\parallel s}/\rho)$ and transverse, $(\Delta\rho_{\perp s}/\rho)$ are obtained when the vector B aligns itself parallel or perpendicular to the direction of current. The coefficients appearing in eq. (2) when expressed in terms of the saturation values of longitudinal and transverse magnetoresistivity yield the following expression for the magnetoresistivity (Kaul & Dutta Roy 1972).

$$\Delta\rho/\rho = (\Delta\rho_{\perp s}/\rho)(B/B_s^g)^2 + [(\Delta\rho_{\parallel s}/\rho) - (\Delta\rho_{\perp s}/\rho)(B_s^a/B_s^g)^2](B/B_s^a)^2\beta_1^2, \quad \ldots \quad (3)$$

where B_s^a and B_s^g are the saturation values of B along [100] and any general direction in (100) plane respectively and include demagnetization factors relevant to these directions. Eq. (3) readily reduces to the well known expression for the total magnetoresistivity change if we do not distinguish between the values of B in different directions.

If the magnetic field is applied along any general direction in (100) plane, both $\Delta\rho_{\parallel s}/\rho$ and $\Delta\rho_{\perp s}/\rho$ contribute to the total magnetoresistivity; also the demagnetization factor varies from unity along [001] to zero along [010] giving rise to the difference in the values of B for different directions of the external magnetic field. Therefore, one expects an angular variation of magnetoresistivity when the magnetic field is rotated in (100) plane. When the field is applied along [001] direction, i.e., $\theta = 90°$ on the polar plot, due to large shape anisotropy, the magnetic induction vector B prefers to orient itself initially along a-axis, where according to eq. (3) only longitudinal magnetoresistivity contributes and then finally along the external field direction—a situation where transverse magnetoresistivity is obtained. Thus, the positive maximum in the field dependence curve corresponds to a fraction of the saturation value of the longitudinal magnetoresistivity $(\Delta\rho_{\parallel}/\rho)$. Such a behaviour is confined to an angular width of about 10° from [001] direction for all the samples in each quadrant. Outside this region, the demagnetization factors along the external field direction start becoming comparable to that along [100] direction and consequently the vector B directly orients itself along the external field direction instead of choosing the above mentioned path. The saturation value of transverse magnetoresistivity is achieved at very low-fields, if the external field is applied along a direction d (figure 5) i.e., along the projection of the vector B on the (100) plane, in which case B and current vector I are coplanar with the external magnetic field H and the demagnetization factors being comparable, the magnetic induction vector B directly orients along H. This direction of the external magnetic field could be distinguished as $\theta = 60°$ direction in the polar plots. The transverse magnetoresistivity saturates at relatively lower value of the magnetic field in case magnetic field is applied along the breadth of the sample i.e., [010] direction or $\theta = 0°$ in the polar plot, since the demagnetization factor along this direction is almost zero and is nearly the same as that for [100] direction, therefore, whole

of the external field is effective in rotating B along its own direction. The value of magnetoresistivity obtained by extrapolating the saturated portion of the magnetoresistivity field dependence curve along $\theta = 60°$ has been taken to represent the saturation value of transverse magnetoresistivity ($\Delta\rho_{\perp T}/\rho$) along that direction.

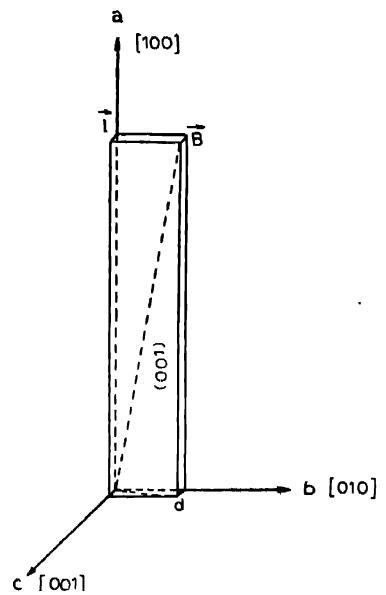

Fig. 5. Orientation of the magnetic induction vector with respect to three crystal axes, a (length), b (breadth) and c (thickness).

On the basis of above analysis, the values of $\Delta\rho_{\parallel}/\rho$ and $\Delta\rho_{\perp s}/\rho$ for nickel and its alloy samples have been computed from experimental data and tabulated in table 1.

Table 1. Electrical resistivity, longitudinal and transverse magnetoresistivity of nickel and nickel-copper alloys at room temperature (300°K).

Samples	ρ ($\mu\Omega$ cm)	$\Delta\rho_{\parallel}/\rho$ (10^{-3})	$\Delta\rho_{\perp s}/\rho$ (10^{-3})
Sample No. 2	9.94	8.5	−6.1
Sample No. 3	9.50	6.6	−5.2
Sample No. 1	8.46	4.5	−3.6
90.62 at. % Ni	17.66	7.3	−8.6
81.23 at.% Ni	25.20	1.4	−5.1
71.68 at. % Ni	41.60		−1.4

A glance at this table reveals the following interesting features :

(i) Both the longitudinal and transverse magnetoresistivity pass through a maximum at 90.62 atomic percent of nickel, although longitudinal magnetoresistivity falls more rapidly with copper concentration than its transverse counterpart. Figure 6 is the graphical manifestation of the above observation. Such an observation closely resembles similar investigations on the longitudinal magnetoresistivity of polycrystalline nickel-copper alloy samples by Masumoto & Shirakawa quoted by Bozorth (1951) and by Walden & Cotellessa (1967) on the transverse magnetoresistivity of single crystal thin films of the same alloy system.

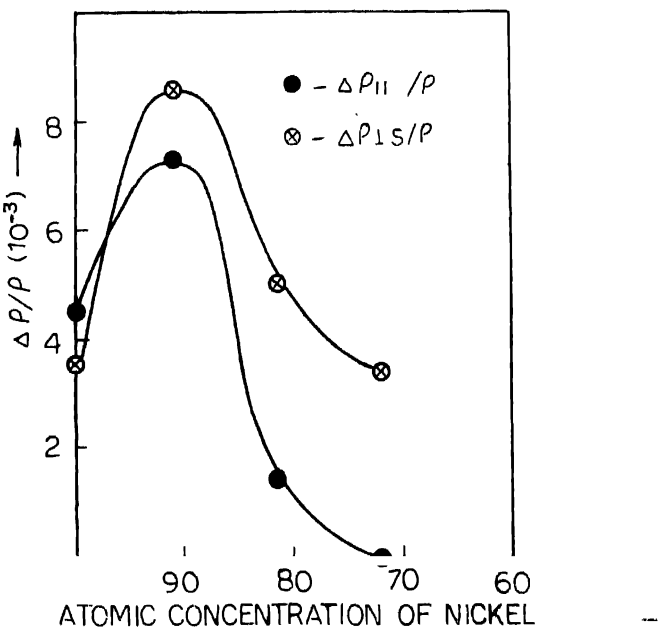

Fig. 6. Variation of $\Delta\rho_{\parallel}/\rho$ and $\Delta\rho_{\perp s}/\rho$ with nickel concentration.

(ii) For 71.68 at. % nickel alloy the longitudinal magnetoresistivity drops to a very small value suggesting thereby that the vector B lies very close to the current direction so that the application of the field in $\theta = 90°$ direction results in the orientation of B along the field direction as the external field strength is progressively increased. This argument also provides a simple explanation for the observed isotropic polar diagram for this alloy, since all the directions of the magnetic field in the (100) plane are equivalent once B is along the current direction. Such a situation can arise only when the sample is nearly polycrystalline in which case the shape anisotropy is alone effective

in orienting the vector B along [100] direction. This conclusion is well supported by the back-reflection photograph which indicates pronounced pollycrystalline rings with very faint Laue spots.

(iii) Both the longitudinal and transverse magnetoresistivity show a higher value for moderately pure nickel samples than the purest nickel sample, an observation in agreement with similar observation made by Smit (1951).

In the derivation of eq. (3) we have assumed that the sample behaves as a single magnetic domain and the magnetoresistivity behaviour is totally due to the rotation of the domain magnetization. We now show that the generality of above results is not lost even though the sample is a multi-domain one.

When the field H is applied in a direction normal to the plane of the sample and it is not yet strong enough to saturate the sample so as to form a single magnetic domain, the domains which build up tend to have their directions of magnetization in the plane of the sample due to the demagnetization field. When H is increased the domains with direction of magnetization along a-axis i.e., [100] direction grow at the expense of the unfavourably oriented domains till the sample becomes a single domain with direction of magnetization along [100] direction and thereafter the magnetization is pulled toward the direction of H. In such a case also a positive maximum corresponding to $\Delta\rho_{\|}/\rho$ is expected. The demagnetization factors along 'd' and [100] directions being comparable, application of H along d direction directly yields the transverse magnetoresistivity. The above mentioned point of view is shared with Taylor *et al* (1968) who attributed the lwo-field magnetoresistivity behaviour of iron to the domain wall motion and subsequent magnetization rotation.

5. Conclusion

The conclusions that are arrived at on the basis of the present experimental investigation on nickel and nickel-copper alloys are summarized as follows :

(i) Although the spin-orbit interaction explains, in part, the anisotropy observed at $H = 7.75$ KOe in the low-field transverse magnetoresistivity of samples in question, it utterly fails to account for the anisotropy observed at $H = 1.55$ KOe where transverse magnetoresistivity as a function of angle between the external magnetic field and the sample plane passes through both positive and negative values. These anisotropies, however, lend themselves to a straightforward explanation in the light of a transport theory modified slightly to include shape anisotropy.

(ii) The low-field magnetoresistivity behaviour (field dependence of transverse magnetoresistivity along different directions of the polar plot) is mainly due to domain wall motion at very low fields and subsequent magnetization rotation at higher fields.

Acknowledgment

The author wishes to thank Prof. S. K. Dutta Roy for his stimulating discussions which have gone a long way to improve the text of the present manuscript. Thanks are also due to Mr. B. K. Samantaray for taking the back reflection photographs and to Shri S. Majumdar for his technical assistance.

References

Anderson J. R. & Gold A. V. 1963 *Phys. Rev. Letters* **10**, 227.
Barrett C. S. & Massalski T. B. 1966 *Structure of Metals* (McGraw-Hill), p. 570.
Berger L. & de Vroomen A. R. 1965 *J. Appl. Phys.* **36**, 2777.
Bozorth R. M. 1951 *Ferromagnetism* (D. Van Nostrand Company, Inc., New York), p. **42**, 761.
Chen T. T. & Marsocci V. A. 1972 *J. Appl. Phys.* **43**, 1554.
Chen T. T. & Marsocci V. A. 1972 *Solid State Commun.* **10**, 783.
Chikazumi S. 1964 *Physics of Magnetism* (John Wiley and Sons, Inc., New York), p. 421.
Doring W. 1938 *Ann. Physik* **32**, 259.
Dutta Roy S. K. & Subrahmanyam A. V. 1969 *Phys. Rev.* **177**, 1133.
Ehrlich A. C., Huguenin R. & Rivier D. 1967 *J. Phys. Chem. Solids* **28**, 253.
Ehrlich A. C. & Rivier D. 1968 *J. Phys. Chem. Solids* **29**, 1293.
Fawcett E. & Reed W. A. 1962 *Phys. Rev. Letters* **9**, 336.
Fawcett E. & Reed W. A. 1963 *Phys. Rev.* **131**, 2463.
Fawcett E. & Reed W. A. 1964 *J. Appl. Phys.* **35**, 754.
Isin A. & Coleman R. V. 1965 *Phys. Rev.* **137A**, 1609.
Jones H. & Zener C. 1934*a* *Proc. Roy. Soc.* (London) **A145**, 268.
Kaul S. N. & Dutta Roy S. K. 1972 *Nuclear Physics & Solid State Symposium* (Chandigarh), **15C**, 31.
Kaya S 1928 *Sci. Repts. Tohoku. Imp. Univ. First Ser.* **17**, 1027.
Kittel C. 1963 *Phys. Rev. Letters* **10**, 331.
MacDonald D. K. C. 1947 *J. Sci. Instr.* **24**, 232.
Marcus S. M. & Langenberg D. N. 1963 *J. Appl. Phys.* **34**, 1367.
Marsocci V. A. 1964 *J. Appl. Phys.* **35**, 774.
Marsocci V. A. 1965 *Phys. Rev.* **137A**, 1842.
Muir W. B. 1964 *Rev. Sci. Instr.* **35**, 408.
Pippard A. B. 1963 *Proc. Roy. Soc.* (London), **A272**, 192.
Preston J. S. 1946 *J. Sci. Instr.* **23**, 173.
Schwerer F. C. & Silcox J. 1971 *J. Phys. Chem. Solids* **32**, 199.
Smit J. 1951 *Physica* **17**, 612.
Taylor G. R., Isin A. & Coleman R. V. 1968 *Phys. Rev.* **765**, 621.
Walden R. H. & Cotellessa R. F. 1967 *J. Appl. Phys.* **38**, 1335.
Ziman J. M. 1960 *Electrons and Phonons* (Clarendon Press, Oxford, England), p. 504.