Vol. A52 (1977) *ACTA PHYSICA POLONICA* No 3

MAGNETIC PROPERTIES OF SOME NICKEL-COPPER-MANGANESE SOLID SOLUTIONS

By I. Nicoară

Department of Physics, University of Timişoara*

I. Pop and Gh. Ilonca

Department of Physics, Babeş-Bolyai University, Cluj**

(Received July 6, 1976)

Some magnetic and galvanomagnetic measurements were performed on $\mathrm{Ni}_x\,(\mathrm{Cu\text{-}Mn})_{1-x}$ solid solutions. These alloys in the concentration range $0.65 \leqslant x \leqslant 0.997$ are ferromagnetic under normal conditions and some of them perhaps ferrimagnetic, as it follows from the temperature dependence of the reciprocal magnetic susceptibility. Galvanomagnetic measurements, i. e., magnetoresistance anisotropy of these alloys indicated that Fermi surface topology presents open electronic orbits.

1. Introduction

The magnetic properties of nickel solid solutions are very interesting from the theoretical point of view, because for their explanation a combined localized and band model is required. In binary Ni–Cu alloys, as is well known, the conduction electrons of copper are gradually filling the $3d$ band of nickel, and the magnetic moment of the nickel atom decreases to zero with increasing copper concentration. If the manganese atoms get in the lattice of nickel-copper alloys, then the magnetic moments of the localized pair of manganese and nickel atoms could be antiferromagnetically coupled, resulting in a ferrimagnetic system. For this purpose we have prepared some nickel-copper-manganese single crystals and we have investigated their magnetic and galvanomagnetic properties (Table I).

---

* Address: Timişoara, B-dul V. Pîrvan Nr. 4. Universitatea din Timişoara, Roumania.
** Address: Cluj, Str. Kogălniceanu Nr. 1. Universitatea Babes-Bolyasi, Roumania.

TABLE I

Quality of the single crystals grown under different conditions and the characteristics of the studied samples

Number of samples	Concentration			Temperature gradient °C/cm	Growth velocity cm/h	Magnetic measurement*				Transverse magnetoresistance $\Delta\varrho_{\perp}/\varrho_0$	Quality of the crystals
						($T > \theta_c$)		($T < T_c$)			
	Ni at %	Cu at %	Mn at %			n(μB)	θ_c(K)	n(μB)	T_c(K)		
4	65	20	15	20	0.5			0.43	230		bad
5		15	20	20	0.5	1.91	320	0.57	240	positive	good with dendrites
6		10	25	50	2.4						good
				20	1.5	1.90	390	0.46	260		good with dendrites
				70	0.8						bad
7	70	20	10	60	0.5	1.61	380	0.49	300	negative	good with dendrites
				50	0.6						good with dendrites
8	70	10	20	60	2.4	1.46	415	0.45	280	negative	good with dendrites
9		5	25	60	2.4	1.55	590				bad
11	80	5	15	60	2.0	1.83	510	0.66	540		—
13		15	5	30	2.4	1.49	460				good
14	90	7	3	28	1.6	1.56	580				—
16	99	0.5	0.5	28	1.6	1.51	650			negative	very good
17	98	1	1	20	1.6						bad
				25	1.0			0.62	640	negative	good
				20	1.5						very good
23	99.7	0.15	0.15	14	1.0	1.49	645			negative	good
24	99.5	0.25	0.25	14	1.6	1.49	650			negative	good

* The Curie temperatures T_c and θ_c were determined by an extrapolation procedure of the magnetization curve, respectively, reciprocal magnetic susceptibility (the dotted curve) with a precision up to 5 K.

2. *Experimental procedure and sample preparation*

The phase diagram of the Ni–Cu–Mn system shows that the solid solution of (Cu+Mn) in Ni extends to 40 at%. The solid solutions Ni–Cu–Mn were prepared by melting the constituent materials using a sectioned graphite resistor [1] in vacuum (10^{-5}–10^{-6} torr) from 99.99% pure metals. As single crystals of these alloys were grown by a variant of the Bridgman method by controlled solidification in the crucible, Ni–Cu–Mn solid solutions were prepared by melting the proper stoichiometric proportions of these single crystals and remelting them repeatedly. This procedure improved the stoichiometry and homogenity of the single crystals.

The magnetic susceptibility and magnetization of saturation temperature dependence were determined using an induction-ballistic method [2] in the temperature range 77–1100 K and magnetic field strengts up to 16k Oe. The magnetic field dependence and anisotropies of the magnetoresistance were determined by using the four-probe method [3] at liquid nitrogen temperature. For the transversal magnetoresistance measurements the obtained single crystals were made in the shape of disks 6–8 mm in diameter and of thickness between 0.9–1.2 mm.

3. *Experimental results and discussion*

The temperature dependence of reciprocal magnetic susceptibility is generally non-linear, as one can see from Fig. 1, where the results are presented for part of the investigated samples, i. e. for the alloys with greater concentrations of copper and manganese.

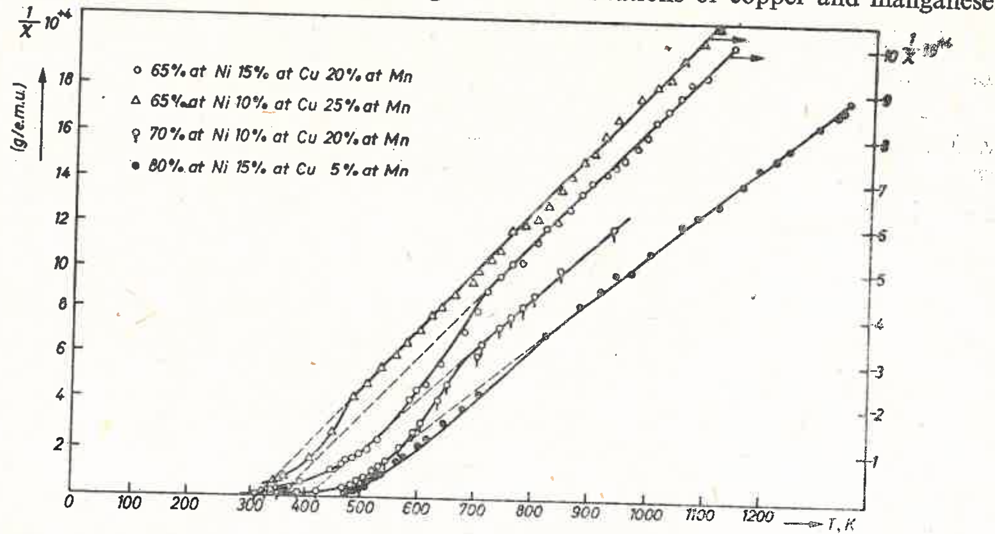

Fig. 1. Reciprocal of the magnetic susceptibility of Ni–Cu–Mn alloys vs temperature

The anomaly of thermal variation of reciprocal magnetic susceptibility have been corelated with the ferrimagnetic ordering of nickel and manganese magnetic moments below the Curie temperature.

The samples with low copper and manganese concentration are ferromagnetic, as one can see from Fig. 2, where the temperature dependence of specific magnetization is presented for the sample 98 at% Ni — 1 at% Mn — 1 at% Cu at different magnetic field intensities.

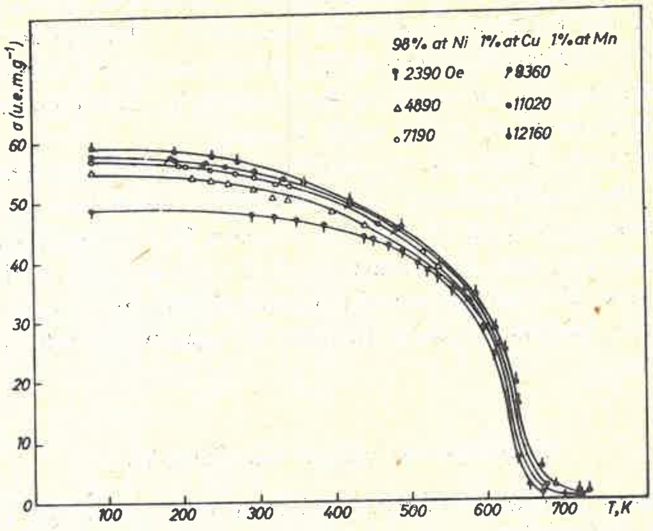

Fig. 2. Magnetization curves vs temperature

In the paramagnetic region, the reciprocal magnetic susceptibility has a linear temperature dependence.

In the magnetic ordering temperature region for all investigated samples the thermal variation of spontaneous magnetization has been determined. This is presentated in Fig. 3, for the samples 98 at% Ni – 1 at% Cu — 1 at% Mn and 80 at% Ni — 5 at% Cu — 15 at% Mn.

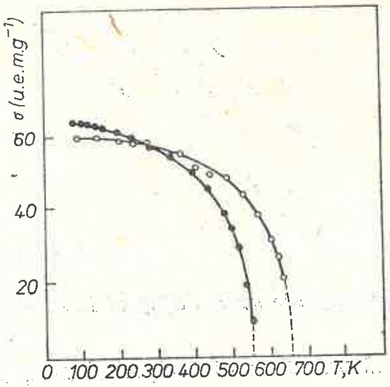

Fig. 3. Spontaneous magnetization as a function of temperature for various samples: ● 80 at % Ni — 5 at % Cu — 15 at % Mn; ○ 98 at % Ni — 1 at % Cu — 1 at % Mn

Similar results were obtained also for other samples. The spontaneous magnetization versus $T^{3/2}$ obeys a linear dependence in the temperature range $0 \leqslant T \leqslant 0.5\,T_c$ as can see from Fig. 4, for samples 65 at% Ni — 15 at% Cu — 20 at% Mn; 70 at% Ni — 20 at% Cu — 10 at% Mn and 70 at% Ni — 10 at% Cu — 20 at% Mn.

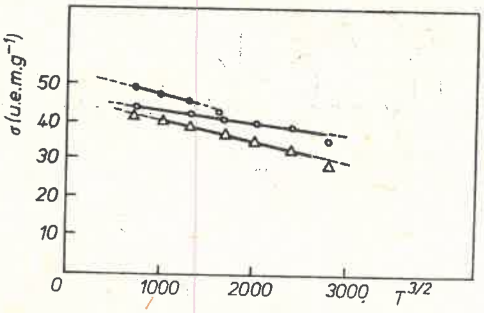

Fig. 4. Spontaneous magnetization as a function of $T^{3/2}$; ● 65 at % Ni — 15 at % Cu — 20 at % Mn; ○ 70 at % Ni — 20 at % Cu — 10 at % Mn; △ 70 at % Ni — 10 at % Cu — 20 at % Mn

The linear dependence σ vs $T^{3/2}$ means that the spin wave model at low temperature is well verified for all investigated samples ferro- and ferrimagnetically ordered, i. e. the spontaneous magnetization follows the relationship

$$\sigma_s = \sigma_0[1-\alpha T^{3/2}].$$

4. *Magnetoresistance measurements*

Magnetoresistance is a criterion [4, 5] well-suited for assessing the part played by s–d exchange interaction in the electrical resistivity; its temperature and field dependence is controlled by the response of localized magnetic moments to an external magnetic field. It is therefore of interest to determine in what way the effect of a magnetic field influences the resistivity. Magnetoresistance measurements have been previously used to study the anomalous s–d scattering [6–9].

The anisotropy and field dependence of the transverse magnetoresistance in applied field up to 18 kOe and 77 K have been measured. The anisotropy of the transverse magnetoresistance $\left[\frac{\Delta\varrho_\perp}{\varrho_0} = \frac{\varrho_H-\varrho_0}{\varrho_0}\right]$ for the field in the cut plane is illustrated in Fig. 5 and 6, for two typical samples: 70 at% Ni — 20 at% Cu — 10 at% Mn and 99 at% Ni — 0.5 at% Cu — 0.5 at% Mn.

This is characterized by the minimum centered on the major symmetry axes. From the anisotropy and magnetic field dependence of the magnetoresistance of the samples we have obtained clear evidence that their Fermi surfaces have open orbits.

The field dependence of the $\frac{\Delta\varrho_\perp}{\varrho_0}$ is presented in Fig. 7 and 8.

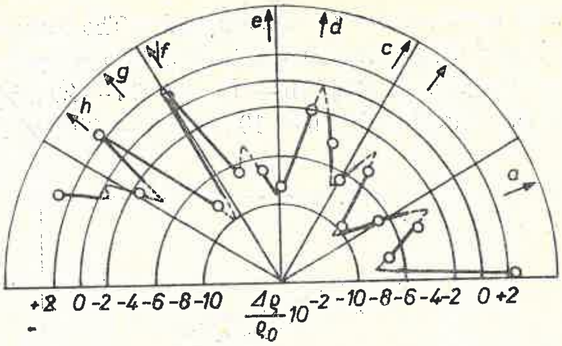

Fig. 5

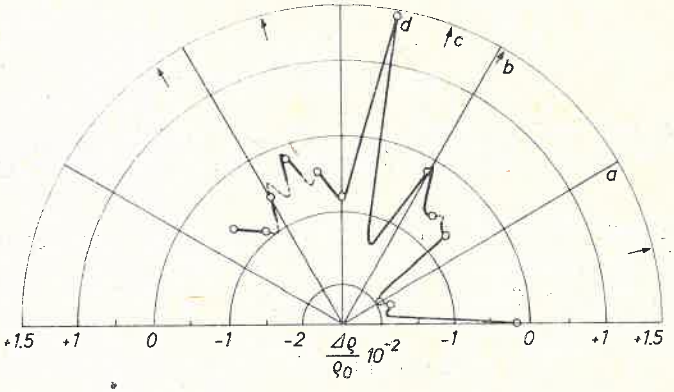

Fig 6

Fig. 5 and 6. Anisotropy of the magnetoresistance $\left(\frac{\Delta\varrho}{\varrho}\right)$ of 70 at % Ni — 20 at % Cu — 10 at % Mn and 99 at % Ni — 0.5 at % Cu — 0.5 at % Mn, respectively, at a temperature 77 K in magnetic field 18k Oe. The arrows indicate the field directions for which the field dependence of the $\frac{\Delta\varrho}{\varrho_0}$ is shown in Fig. 7 and 8

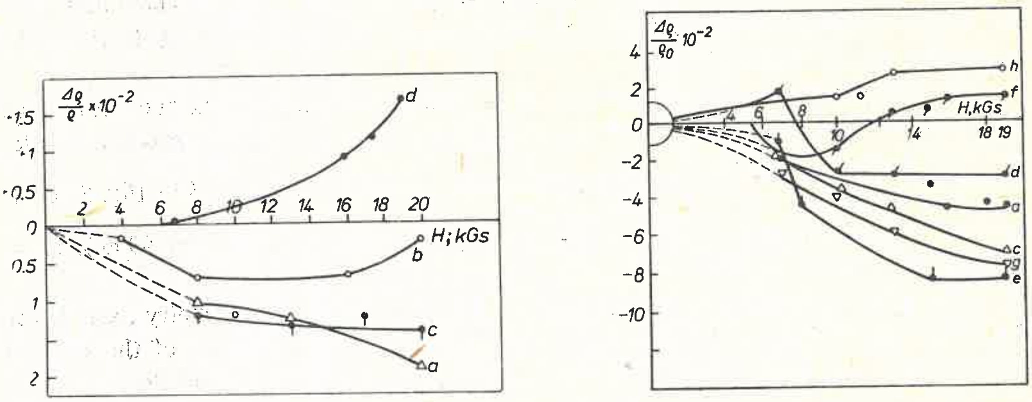

Fig. 7, 8. Magnetoresistance $\frac{\Delta\varrho}{\varrho_0}$ as a function of magnetic field

The magnetoresistance is saturated with an increasing field for a general field direction. But then more rapid linear increase is due to the effect on the conductivity tensor of open cyclotron orbits. The complicated behaviour of the magnetoresistivity below 8–10 kOe is probably a consequence of the magnetocrystalline anisotropy and is not relevant to the present paper.

The saturation of $\frac{\Delta\varrho_\perp}{\varrho_0}$ for an arbitrary field direction shows that the Ni–Cu–Mn alloy behaves like an uncompensated metal for which $\omega_c\tau > 1$ at the highest fields H, ω_c being the cyclotron frequency $\left(\frac{eH}{m^*c}\right)$ of carriers of effective mass m^* and τ the carrier relaxation time.

The smallness of the magnetoresistance and the fact that in does not active the ideal quadratic field dependence, associated with open orbits [10], indicate that $\omega_c\tau$ is still only of the order of unity for a field of 18 kOe even in these samples. The field dependence approaches the expected saturation at a minimum (Fig. 7, 8) while for neighboring field directions the open orbits produce a field dependence which is more rapid than linear.

From these measurements, an anomalous magnetoresistance results, i. e. a negative magnetoresistance (Fig. 7, 8). These results suggest that the ferromagnetic behaviour seems not to be interpretable only in terms of a collective electron model. The presence of anomaly in magnetoresistance of the ferromagnetic Ni–Cu–Mn solid solutions also suggest that localized moments exist in these compounds.

We assume that the most important contribution to the negative magnetoresistance for the experimental temperature and field is due to anomalous scattering of the conduction electrons by Mn impurities with the localized magnetic moment.

The magnetoresistance of Ni–Cu–Mn alloys is well described by the *s–d* exchange model between the conduction electrons and the localized moments [11]. The magnetic field dependent part of the anomalous *s–d* scattering gives a negative contribution to the magnetoresistance.

REFERENCES

[1] M. Zăgănescu, I. Farcas, D. Nicoară, *Analele Univ. Timişoara. Seria şt. Fiz.-Chim.* **VII. p2,** 245 (1969).

[2] I. Pop, V. I. Chechernikov, *Pribory Tekh. Eksper.* **5**, 180 (1964).

[3] I. Nicoară, Thesis, Babeş-Bolyai University, Cluj 1973.

[4] G. J. Van den Berg, Proc. 9-th. International Conf. Low Temp. Phys., Part B, Plenum Press, New-York 1965, p. 933.

[5] M. T. Béal-Monod, R. A. Weiner, *Phys. Rev.* **170**, 552 (1968).

[6] A. N. Gerritsen, *Physica* **19**, 61 (1953).

[7] G. J. Los, A. N. Gerritsen, *Physica* **23**, 633 (1957).

[8] P. Monod, *Phys. Rev. Lett*, **19**, 1113 (1967).

[9] H. Rohrer, *Phys. Rev.* **174**, 583 (1968).

[10] L. M. Lifschitz, M. Ya. Azbel, M. I. Kaganov, *Zh. Eksp. Teor. Fiz.* **31**, 63 (1956).

[11] I. Nicoară, *Phys. Status Solidi* (b) **77**, *K*17 (1976).