Z. Physik B 23, 23 – 26 (1976)

Electrical Resistivity of Metastable Copper-Iron Solid Solutions

D. Korn and H. Pfeifle
Fachbereich Physik der Universität Konstanz

J. Niebuhr
Fachhochschule Ravensburg

Received October 7, accepted October 15, 1975

Abstract. Metallic solid solutions of CuFe are produced by simultaneous condensation of both components onto a sapphire substrate held at 80 K. The electrical resistivity of the CuFe films is measured as a function of temperature throughout the whole concentration range. Maxima in the residual resistivity appear at 20 at-% and 80 at-% Fe. A low temperature minimum in the electrical resistivity is observed on the Fe rich side of the system.

I. Introduction

Binary alloys, consisting of a magnetic and a nonmagnetic component have been extensively studied. The properties of localized magnetic moments in various matrices and the related low temperature anomaly in the electrical resistivity have been discussed in several review articles (see for example [1] and [2]). The investigations however are restricted by the limited solubility of the component with the localized magnetic moment in the host metal. No example exists, that such an alloy can be produced as a stable solution throughout the whole concentration range. At room temperature, for example the solubility of Fe in Cu lies far below 1 at-%. AuFe and PdFe are alloys with a wider range of solubility. Therefore the electrical resistivity of AuFe [3] and PdFe [4] has been measured in an extended concentration range but not throughout the whole system.

By the method of quench-condensation insoluble alloys can be forced into a solid solution over a limited temperature range. This method had been developed by Buckel and Hilsch [5]. Felsch [6] produced AuFe films between 0 and 100 at-% Fe in that way and investigated the electrical, magnetic and structural behaviour. At high Fe concentration the films grow amorphous at a condensation temperature of 20 K. Reviewing the results of analogous investigations [7] we obtain as an empirical rule that the amorphous phase will only occur at those compositions where the two components are insoluble under normal conditions. AgMn films, investigated with respect to their magnetic susceptibility [8] are polycrystalline and not amorphous as often assumed [9–11]. Feldtkeller [12] demonstrated that Cu films with an addition of 16 at-% Fe are polycrystalline, even when the temperature of the substrate is held at 4 K during evaporation. Kneller [13] produced CuFe films at room temperature throughout the whole concentration range and determined their structural and magnetic properties. These films are all crystalline and exist in the fcc phase up to 25 at-% Fe and in the bcc phase from 50 to 100 at-% Fe. According to Kneller the fcc solid solutions are paramagnetic whereas the bcc configuration is ferromagnetic. The saturation magnetization does not change according to the rigid band model, but rather each Fe atom carries a localized magnetic moment. This is in contrast to CuNi where single Ni atoms in Cu have no permanent magnetic moment.

The electrical resistivity of dilute CuFe has found an extended theoretical treatment [14, 15]. Experimentally, dilute bulk CuFe has been the subject of research since years [2]. More recently the electrical resistivity of dilute CuFe films has been investigated [16–19]. At low Fe concentration there exists the well-known minimum in the temperature dependence of the electrical resistivity. Towards increasing Fe concentration an additional maximum appears at a temperature below the minimum temperature [16]. This maximum is well-known e.g. in the systems AuFe [20] and AgMn [21] and is explained as a consequence of a decreasing amplitude of spin-flip scattering caused by ordering of the impurity spins [22]. The maximum cannot be found in bulk samples of CuFe because of the limited solubility. With increasing Fe concentration this maximum moves towards higher temperatures and ends up as a hump in the phonon contribution of the resistivity as Kaufmann showed [23]. This konvex bending of the $\rho(T)$-graph is well established in other transition alloy systems and is often used as criterion for the onset of the spinordering [3, 24].

In this paper we shall describe the extension of the measurements on CuFe films up to 100 at-% Fe. Together with the data of Kneller [13] about structure and magnetic behaviour they give us more insight in the behaviour of a binary alloy system consisting of a nonmagnetic component and a component with localized magnetic moment.

II. Experimental Details

The alloy is prepared by melting the two pure components together in an Al_2O_3 crucible under a protective argon atmosphere. The ingot is then filed into powder and stepwise evaporated from a tungsten filament held at about 1,700 °C. The evaporation rate is about 10 Å per second. The evaporation takes place in an UHV cryostat. The residual gas measured at some distance of the tungsten filament has a pressure of 10^{-8} Torr and consists mainly of H_2O (checked with mass analyzer). The thickness of the films, determined by means of an interference microscope, lies around 500 Å for all films. The temperature is controlled both by an Allen-Bradley 100 Ω resistor and a Pt-resistor and can be varied between 2 K and 350 K with an accuracy of 1%. The electrical resistivity is measured with a conventional four electrode dc method and has a relative error of 10^{-5}. All measurements have been done with a current of 10^{-4} A.

The absolute value of the specific resistivity was calculated out of the optical film thickness, which leads to an error of about 5% if one compares the films among themselves. To compare the resistivities obtained in this manner to any bulk samples, another systematic error must be taken into account. The densities of the film material and the bulk material can differ by a certain amount. This fact together with other reasons causes a bigger value of the optical thickness in comparison to the thickness calculated from the thermal part of the resistivity. That has been pointed out by Bassewitz and Minnigerode [25]. Because of the high concentrations we had no means to determine the film thickness out of the thermal part of the resistivity. To be consistent throughout the whole concentration range, we used the optical thickness even for the pure Fe and Cu films which leads to different temperature coefficients of our Cu and Fe films with respect to tabulated bulk values. It should be mentioned that the pure Cu and Fe films are prepared in the same manner as the alloys and not evaporated from the bulk.

III. Experimental Results

Fig. 1 shows the typical behaviour and the heat treatment of CuFe film after evaporation. After deposition of the film at 80 K the resistivity is measured as a function of temperature below 80 K. Then the film is annealed up to 160 K and again the resistivity is determined below 160 K before annealing up to 350 K. Below the annealing temperature the measurements are reversible. The irreversible part of the resistivity as a function of temperature gives no indication of a phase change and annealing has to a good approximation no influence onto the temperature coefficient of the resistivity. To discuss the electrical resistivity of the CuFe films we choose the annealing stage of 160 K. At this annealing temperature the lattice defects are reduced in comparison to the annealing temperature of 80 K. On the other hand the atoms of both components are still randomly distributed over the lattice sites. In Fig. 2 we see the electrical resistivity of these polycrystalline CuFe films in the concentration range from 0 at-% to 100 at-% Fe. At 6 at-% Fe a hump in the resistivity function can be seen at about 70 K. With increasing Fe concentration this hump moves towards higher temperatures and the positive temperature coefficient of the resistivity rises simultaneously until it reaches its maximum at about 30 at-% Fe. Beyond 30 at-% Fe the temperature coefficient decreases and ends up in a nearly constant value between 60 at-% and 100 at-% Fe. In the region

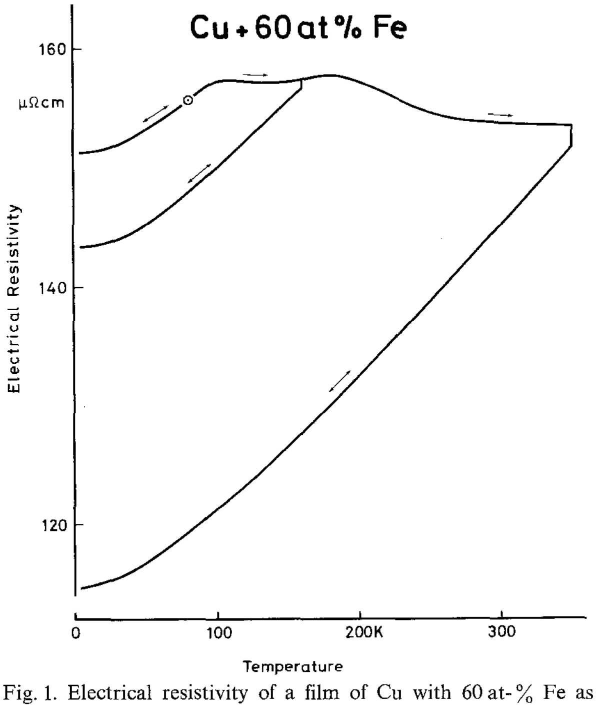

Fig. 1. Electrical resistivity of a film of Cu with 60 at-% Fe as a function of temperature between 2 K and 350K. ⊙ denotes the point of condensation

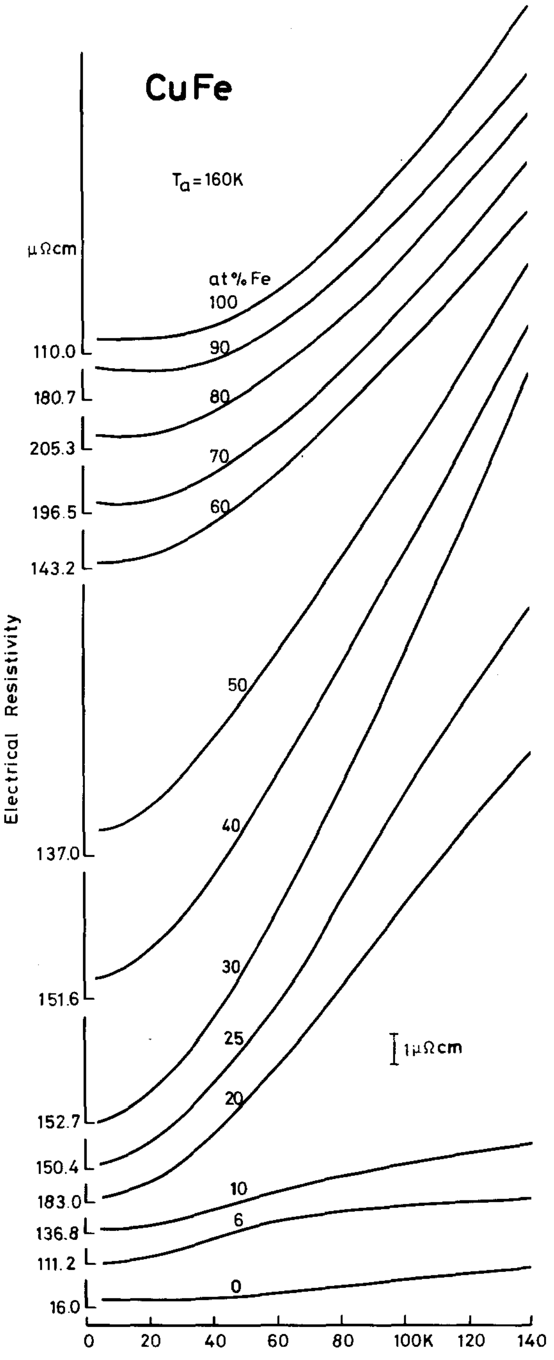

Fig. 2. Electrical resistivity of CuFe films as a function of temperature for Fe concentrations between 0 and 100 at-%. Annealing temperature $T_a = 160$ K

between 10 at-% and 50 at-% Fe the resistivity between 2 K and 40 K can be described by a T^2-law. At high Fe concentrations (70 at-%–90 at-% Fe) in Fig. 2 we observe a minimum at about 13 K. Starting at pure Fe, where the minimum is still weakly apparent, the depth of the minimum $\Delta\rho = \rho(2K) - \rho(T_{\min})$ rises with increasing Cu concentration and reaches a

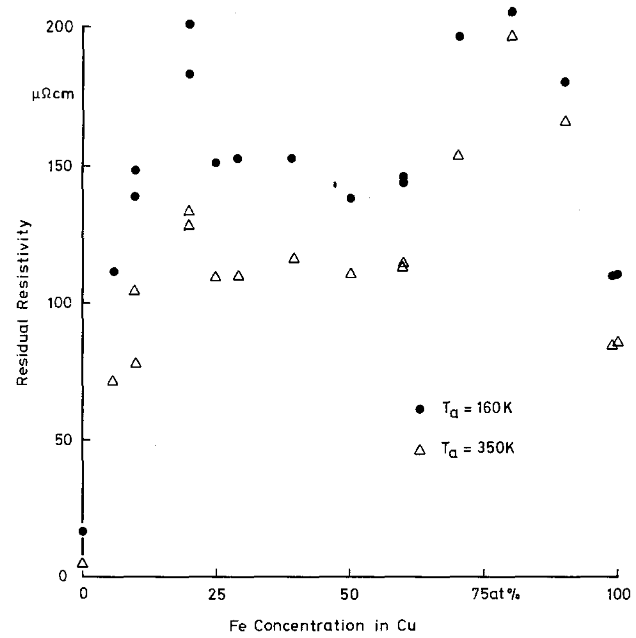

Fig. 3. Residual electrical resistivity of CuFe films as a function of Fe concentration for two different annealing temperatures

maximum at that concentration where the residual resistivity has a broad peak.

Those minima in a magnetic matrix are known since long time. Meissner and Vogt [26] report about a resistivity minimum in polycrystalline Co. Single crystalline whiskers of Cr [27] have also resistivity minima. Kuzmenko *et al.* [28] observed resistivity minima in pure Fe films. As Yoshii *et al.* [29] demonstrated, those minima depend on the vacuum conditions during evaporation. The additional minima between 5.5 at-% and 8.5 at-% Fe published by Kaufmann [23], could not be verified with our CuFe films.

In Fig. 3 is shown the residual resistivity as a function of Fe concentration. At $T_a = 160$ K the residual resistivity rises very fast with the Fe concentration, reaches a maximum at about 20 at-% Fe and drops abruptly to a nearly constant value till a broad second peak occurs around 80 at-% Fe. On annealing the residual resistivity does not change considerably when going from 80 to 160 K, but annealing up to 350 K (Fig. 3) lowers the first sharp maximum and scarcely influences the second peak.

IV. Discussion

The residual resistivity of binary alloys without miscibility gap is normally explained by Nordheim's rule, which however does not hold in the case of *s*- and *d*-

conduction electrons [30] nor in the case of two different scattering mechanisms. The existence of a localized magnetic moment and a doublepeaked virtual bound state of the Fe in very dilute CuFe alloys is well established (see for a comprehensive listing [1, 31]). We, however, see no possibility to discuss the resistivity maxima in CuFe in terms of the virtual bound state model and we shall try another explanation.

The residual resistivity of the fcc Cu films increases with the number of Fe atoms because of their potential and spin scattering. Due to the indirect interaction and the low temperature the directions of the Fe spins are fixed but nevertheless randomly disordered with respect to each other. At about 20–25 at-% Fe the limit is reached where because of statistical reasons the Fe atoms form cluster and their spins interact directly. At that concentration the alloy film becomes ferromagnetic and changes its structure from fcc to bcc. The parallel alignment of the Fe spins causes the resistivity to drop. An increase in temperature will shift the resistivity to a higher value due to the spin-disordered state at high temperature. This explains why the temperature coefficient of the resistivity reaches its maximum at higher Fe concentrations than the residual resistivity. Adding more Fe to Cu the ordering temperature moves far beyond the temperature range set by this experiments and the temperature coefficient keeps its nearly constant value up to 100 at-% Fe.

This maximum in the residual resistivity and the shifted maximum in the temperature coefficient have been measured in bulk AuFe by Sundahl *et al.* [32]. For these alloys the same explanation holds. PdFe shows a maximum in the residual resistivity as a function of concentration as well [4].

The second broad peak at about 80 at-% Fe has been unknown for this type of binary alloys till now. One possible explanation may be that at the percolation limit for Cu in bcc Fe, which is at about 25 at-% Cu in Fe [33], the copper atoms do not act exclusively as centers of scattering, but constitute a current path through the film. This would correspond to an additional conduction band in the picture of the band model. As the whole film does not show a deviation from the bcc phase [13] the statistical Cu clusters beyond the percolation limit should have a bcc structure too. However, there is a remarkable anomaly in the lattice constant and the coercive force at about 12 at-% Cu in Fe. This would mean, that the structure at this concentration reaches a high degree of disorder. Disorder does not change the temperature coefficient appreciably but increases the residual resistivity, a behaviour which is consistent with our results.

This work has been sponsored by the "Deutsche Forschungsgemeinschaft".

References

1. Rizzuto, C.: Rep. Progr. Phys. **37**, 147 (1974)
2. van Dam, J.E., van den Berg, G.J.: phys. stat. sol. (a) **3**, 11 (1970)
3. Mydosh, J.A., Ford, P.J., Kawatra, M.P., Whall, T.E.: Phys. Rev. B **10**, 2845 (1974)
4. Abramova, L.T., Federov, G.V., Volkenshagen, N.V.: Phys. Met. Metallogr. **33**, Nr. 3, 70 (1972)
5. Buckel, W., Hilsch, R.: Z. Physik **146**, 27 (1956)
6. Felsch, W.: Z. angew. Physik **29**, 217 (1970)
7. Korn, D., Pfeifle, H., Zibold, G.: Z. Physik **270**, 197 (1974)
8. Korn, D.: Z. Physik **214**, 136 (1968)
9. Kok, W.C., Anderson, P.W.: Phil. Mag. **24**, 1141 (1971)
10. Kok, W.C.: Phil. Mag. **30**, 351 (1974)
11. Vogt, E.: phys. stat. sol. (a) **28**, 11 (1975)
12. Feldtkeller, E.: Z. Physik **157**, 65 (1959)
13. Kneller, E.F.: J. appl. Phys. **35**, 2210 (1964)
14. Kondo, J.: Solid state Physics Vol. **23**, Eds. F. Seitz, D. Turnbull. New York: Academic Press 1969
15. Fischer, K.: phys. stat. sol. (6) **46**, 11 (1971)
16. Korn, D.: Z. Physik **238**, 275 (1970)
17. Hauser, J.J., Hamann, D.R., Kammlott, G.W.: Phys. Rev. B **3**, 2211 (1971)
18. Knorr, K., Lin, C.S., Leslie, J.D.: Solid State Comm. **10**, 637 (1972)
19. Silverman, P.J., Briscoe, C.V.: Solid State Comm. **16**, 367 (1975)
20. Gerritsen, A.N.: Physica **23**, 1087 (1957)
21. Gerritsen, A.N., Linde, J.O.: Physica **17**, 573 (1951)
22. Silverstein, S.D.: Phys. Rev. Lett. **16**, 466 (1966)
23. Kaufmann, K.: Z. Physik B **22**, 117 (1975)
24. Mydosh, J.A., Budnick, J.I., Kawatra, M.P., Skalski, S.: Phys. Rev. Lett. **21**, 1346 (1968)
25. v. Bassewitz, A., v. Minnigerode, G.: Z. Physik **181**, 368 (1964)
26. Meissner, W., Vogt, G.: Ann. Phys. **7**, 892 (1930)
27. Semenenkov, Y.Y., Tutrov, V.I.: Phys. Lett. Metallogr. **27**, 2, 148 (1969)
28. Kuzmenko, V.M., Lazarev, B.G., Sudovtsov, A.I., Melnikoff, V.I.: JETP **31**, 230 (1970)
29. Yoshii, T., Yamakawa, K., Fujita, E.: J. Phys. Soc. Jap. **37**, 572 (1974)
30. Bambakidis, G., Smith, R.J., Otterson, S.A.: phys. stat. sol. (a) **26**, 53 (1974)
31. Grüner, G.: Adv. Phys. **26**, 6, 941 (1974)
32. Sundahl, R.C., Chen, T., Sivertsen, J.M., Sato, T.: J. appl. Phys. **37**, 1024 (1966)
33. Sato, H., Arrott, A., Kikucki, R.: J. Phys. Chem. Solids **10**, 19 (1959)

Prof. Dr. D. Korn
Dipl.-Phys. H. Pfeifle
Fachbereich Physik der Universität Konstanz
D-7750 Konstanz
Postfach 7733
Federal Republic of Germany

Prof. Dr. J. Niebuhr
Fachhochschule Ravensburg
D-7980 Ravensburg
Deisenfangstraße 31
Federal Republic of Germany