Susceptibility of copper-nickel alloys

V. E. Rode, A. Bluschke, B. N. Kraĭnov, and A. I. Leĭchenko
Moscow State University
(Submitted March 27, 1972)
Zh. Eksp. Teor. Fiz. **64**, 548-552 (February 1973)

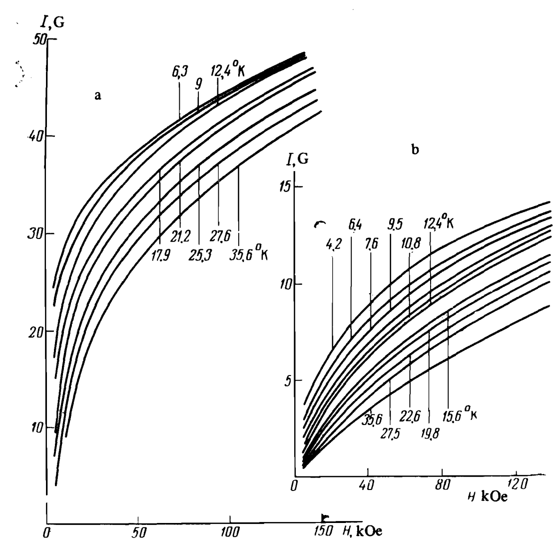
The field dependence of the magnetic moment of copper-nickel alloys in pulsed magnetic fields up to 150 kOe is measured at various temperatures above the Curie point. Six alloys from the critical concentration region are investigated. The contribution from magnetic clusters is taken into account in the treatment of the magnetic-moment curves for the alloys investigated. The cluster behavior in a magnetic field is described by a Langevin-gas model. The experimental curves are quite consistent with the theoretical curves and the term due to the clusters is important. The mean magnetic moment per cluster is $7\mu_B$ and the effective number of clusters per cm^3 is of the order of 10^{20}. In order of magnitude, the value of $\chi_d(0)$ is 10^{-4} emu/cm^3. The large values of the alloy susceptibilities in the region studied can be attributed to a transition of the system through the critical state when the condition for existence of ferromagnetism is not satisfied.

We have measured the field dependence of the magnetic moment of copper-nickel alloys in pulsed magnetic fields of intensity up to 150 kOe at different temperatures above the Curie point. The Curie temperatures and the measurement temperature intervals for each alloy are listed in the table. We have investigated six alloys with compositions close to the critical concentration corresponding to the transition of the system from the ferromagnetic into the paramagnetic state. The method for obtaining the magnetic field and for the measurements were described earlier[1]. The accuracy with which the magnetic moment was measured in a strong field was 2%. All the alloys were subjected to homogenizing annealing for 96 hours at 1100°C. The homogeneity of the composition of the alloys was verified with the aid of an x-ray microanalyzer. The impurity content in the samples was less than 0.01%.

Measured values of the Curie temperature T_c of alloys of the Cu-Ni system are given in a number of articles[2-4]. However, in view of discrepancies between data pertaining to alloys having Curie temperatures ranging from 2 to 40°K, it was necessary to perform independent measurements of T_c of the investigated samples. The temperature dependence of the initial susceptibility in a weak alternating magnetic field was plotted with an x-y recorder. The temperature at which the initial susceptibility reached the maximum value was identified with the Curie temperature of the alloy. The obtained values of T_c agree well with the data of Robbins, Claus, and Beck[2].

The figure shows the dependence of the magnetic moments of two samples on the magnetic field at different temperatures. Experimental curves for other alloys are similar in form. The tendency of the obtained curves to saturate in strong fields, and also data by others[5] confirming the presence of a inhomogeneous distribution of the magnetization in the Cu-Ni alloys, lead to the need for introducing the susceptibility χ_{cl} connected with the magnetic clusters.

The susceptibility of the investigated paramagnetic alloys in a strong field can be represented, in analogy with the work of Foner et al.[6], in the form of the following sums:



Field dependence of the magnetic moment of the C-Ni alloy at different temperatures: a—sample with 43.8 at. % Ni + 56.2 at. % Cu, b—sample with 39.2 at. % Ni + 60.8 at. % Cu.

Numerical values of the parameters, determined by least squares*

Alloy composition, at,%		T_c, °K	ΔT, °K	C, 10^5	$-B\cdot10^{16}$	$\chi_d(0)\cdot10^5$	$-\beta\cdot10^{12}$	$\Delta\chi_d(0)\cdot10^5$	$\Delta\beta\cdot10^{12}$	μ, μ_B	$N\cdot10^{-19}$, cm^{-3}	$\chi_d^f(0)\cdot10^5$
Ni	Cu											
39,2	60,8	—	4.2—35.6	13.5	14,4	13.1	11.0	0,4	0,6	7	7,1	—
43,8	56,2	3,4	6,3—35,6	14,7	15,3	14,2	10,8	0,4	0,4	7	7,1	—
44,0	56,0	3.8	9,2—30,4	14,1	19,5	13,6	14,3	1.1	0,8	7	7,1	13
45,0	55,0	6.6	10,8—25,4	14,3	21,4	13,8	15,5	1,4	1,4	7	7,1	11
45.7	54,3	15	27,5—38,7	18,6	28,5	18,1	15,7	1,7	0,7	7	7.2	12

*Here ΔT is the temperature interval of the measurement; $\Delta\chi_d(0)$ and $\Delta\beta$ are the variances of the corresponding parameters. The quantities for which no dimensionality is given are in electromagnetic units.

$$\chi=\chi_s+\chi_d+\chi_{VV}+\chi_{dia}+\chi_{cl}, \quad (1)$$

where χ_S is the Pauli spin susceptibility of the s-electrons, χ_d is the Pauli spin susceptibility of the d-electrons, χ_{dia} is the diamagnetic susceptibility of the electrons of the filled shells and of the conduction electrons, and χ_{VV} is the van-Vleck paramagnetic susceptibility. To obtain information on the band structure of the investigated alloy it is necessary to separate from the sum (1) the Pauli susceptibility χ_d of the d-band electrons. We can estimate the contributions of χ_S, χ_{VV}, and χ_{dia} in (1).

The paramagnetic susceptibility of the s-band electrons χ_S is 0.9×10^{-6} emu/cm^3 for Ni[6]. A theoretical calculation of the van-Vleck susceptibility χ_{VV} for Ni, performed by Shimizu[7], gives a value 1.1×10^{-5} emu/cm^3, which also agrees with estimates made in[6]. We assume that the orbital susceptibility of the investigated alloys is equal to the van-Vleck susceptibility of Ni multiplied by the Ni concentration in the alloy, since the d-band of Cu is completely filled and should make no contribution to χ_{VV}.

The diamagnetic susceptibility of the investigated alloys can be estimated on the basis of measurements of the magnetic susceptibility of the Cu^+ ion, whose d-band is filled. The measured ionic susceptibility of Cu is $\chi_{dia}=-2.5\times10^{-6}$ emu/cm^3[8]. A calculation of the diamagnetic susceptibility of Ni[9], in which the entire electron shell of the atom is taken into account with the exception of the 4s electron, yields a value -3.3×10^{-6} emu/cm^3. Analogous calculations for Cu[9] give a value -3.4×10^{-6} emu/cm^3. We assume that the experimentally determined diamagnetic susceptibility of the Cu^+, which equals -2.5×10^{-6} emu/cm^3, can be regarded as the upper limit of χ_{dia} of the investigated alloys, since in Ni the d-band is not completely filled and it is known that it is precisely the d-electrons that make the principal contribution to the diamagnetic susceptibility.

Thus,

$$\chi_s+\chi_{VV}+\chi_{dia}\leqslant0.9\cdot10^{-6}+1.1\cdot10^{-5}\cdot0.5+2.5\cdot10^{-6}\approx1\cdot10^{-5},$$

and the maximum contribution from these susceptibilities is less than 10% of the experimental susceptibilities.

The expression for χ_d is[10]:

$$\chi_d=\chi_d(0)\ (1+\beta H^2), \quad (2)$$

where

$$\chi_d(0)=2\mu_B^2N(\varepsilon_F)\left[1-J_{eff}N(\varepsilon_F)\right]^{-1}, \quad (2a)$$

$$\beta={}^1/_2\nu\mu_B^2\left[1-J_{eff}N(\varepsilon_F)\right]^{-3},\quad \nu=\left[\frac{N''}{N}-3\left(\frac{N'}{N}\right)^2\right]_{\varepsilon=\varepsilon_F}. \quad (2b)$$

Here $N(\epsilon)$ is the density of states, J_{eff} is the energy of the exchange interaction, and ϵ_F is the Fermi energy.

The average magnetic moment per cluster is 8–12 μ_B[5], so that the behavior of the clusters in the magnetic field can be described with the aid of the simple model of a Langevin gas. The effective field of the interaction between the clusters was not taken into account, since the fields in which the investigations were performed were strong. Thus, the magnetic moment connected with the presence of clusters can be represented in the form

$$M_{cl}=N\mu L\left(\frac{\mu H}{kT}\right)=N\mu\,\mathrm{cth}\frac{\mu H}{kT}-\frac{NkT}{H} \quad (3)$$

where μ is the average magnetic moment per cluster, N is the effective number of clusters per cm^3, $L(x)=\coth x-1/x$ is the Langevin function, and K is Boltzmann's constant.

Thus, in the reduction of the experimental data the magnetic moment of the Cu-Ni alloys takes the form

$$M(H,T)=CH+BH^3+N\mu\,\mathrm{cth}\frac{\mu H}{kT}-\frac{NkT}{H}, \quad (4)$$

where $c=\chi_S+\chi_{VV}+\chi_{dia}+\chi_d(0)$ is the part of the susceptibility independent of the field and of the temperature, and $B=\chi_d(0)\beta$.

The reduction of the plots of the field dependence of the magnetic moment of the alloys at each value of the temperature was carried out by least squares with the aid of computer. We separated the parameters C, B, and N, and μ, and consequently also $\chi_d(0)$ and β. The experimental curves are described sufficiently well by expression (4), and the term due to the clusters is significant. The average magnetic moment per cluster was found to be 7 μ_B, and the effective number of clusters per cm^3 was of the order of 10^{20}. None of the parameters varied very strongly with the temperature. The average values of the parameters for different temperatures and their rms deviations from the mean value are listed in the table. For comparison, the table gives the values of $\chi_d^f(0)$ obtained by us for the same alloys at temperatures below the Curie points[1]. In order of magnitude, $\chi_d(0)$ is equal to 10^{-4} emu/cm^3, i.e., it is larger by one order of magnitude than $\chi_d(0)=0.6\times10^{-5}$ emu/cm^3 for pure nickel, and larger than $\chi_d(0)=3\times10^{-5}$ emu/cm^{-5} of the alloy containing 66 at.% Cu[1].

The large susceptibility of the alloys in the investigated concentration region can be attributed to the transition of the system through a critical state, at which the condition for the existence of ferromagnetism ceases to be satisfied. According to a theoretical prediction[11,12], when the concentration c approaches the critical concentration c_0 the susceptibility of alloys should increase like $(c-c_0)^{-1}$. An exact verification of this assumption is not possible in the present study, since we do not know the exact value of the critical concentration c_0. It is of interest to compare (2a) with Stoner's criterion

$$1-NJ_{eff}<0$$

for the onset of ferromagnetism[13]. Since the denominator in (2a) is small at concentrations close to c_0, one should expect large values of the susceptibility, as was indeed obtained in the experiment.

The values of the coefficient β agree in order of magnitude with the estimates obtained from band theory for Pd: $\beta\leq0.99\times10^{-11}$ Oe^{-2}[10], but is negative in sign. Apparently the Fermi energy of Cu-Ni alloys lies in the region where the curvature $N(\epsilon)$ is small or negative. The experiment value of β for the alloy $Pd_{0.95}Rd_{0.05}$[14] is very close to the value of β obtained for the Cu-Ni alloys.

[1] A. Blüschke, B. N. Krainov, W. E. Rode, and S. A. Finkelberg, Phys. Stat. Sol. (a), 10, 223 (1972).

[2] C. G. Robbins, H. Claus, and P. A. Beck, J. Appl. Phys., 40, 2269 (1969).

[3] A. J. P. Meyer and C. Wolff, Acad. Sci., Paris, **246**, 576 (1958).

[4] A. Arrott, Phys. Rev., 108, 1394 (1957).
[5] T. J. Hicks, B. Rainford, J. S. Kouvel, G. G. Low, and J. B. Comly, Phys. Rev. Lett., **22**, 531 (1969). J. S. Kouvel and J. B. Comly, Phys. Rev. Lett., **24**, 598 (1970).
[6] S. Foner, A. J. Freeman, N. A. Blum, R. B. Frankel, E. J. McNiff, Jr., and H. C. Praddaude, Phys. Rev., 181, 863 (1969).
[7] M. Shimizu, T. Takahashi, and A. Katsuki, J. Phys. Soc. (Japan), 18, 801 (1963).
[8] S. V. Vonsovskiĭ, Magnetizm. Magnitnye svoistva dia-, para-, ferro-, antiferro- and ferrimagnetikov (Magnetism. Magnetic Properties of Para-, Ferro-, Antiferro-, and Ferrimagnets), Nauka, 1971, p. 168.
[9] J. H. M. Stoelinga, R. Gersdorf, G. de Vries, Physica, **41**, 457 (1969).
[10] E. P. Wohlfarth, Phys. Lett., **22**, 280 (1966).
[11] J. Mathon, Proc. Roy. Soc., **306**, 355 (1968). J. Mathon and E. P. Wohlfarth, Phys. Stat. Sol., 30, K131, (1968).
[12] C. Hargitai, S. Shtrikman, and E. P. Wohlfarth, Phys. Lett., **39A**, 87 (1972).
[13] E. C. Stoner, Proc. Roy. Soc., **A165**, 372 (1938); **A169**, 339 (1939).
[14] S. Foner and E. J. McNiff, Phys. Lett., **29A**, 28 (1969).

Translated by J. G. Adashko
59