Low-temperature magnetic states in copper-manganese alloys

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We consider a broad spectrum of low-temperature states in copper-manganese alloys, from the kondo phase to the mixed "cluster glass + antiferromagnetism" state. A magnetic phase diagram is proposed for copper-manganese alloys in the impurity density range from 0.2 to 80 at.%.

INTRODUCTION

The surprising physical phenomena accompanying the localization of the magnetic moment of a paramagnetic impurity in a nonmagnetic matrix, such as the Kondo effect, the spin-glass state, or the local spin fluctuations, continue to attract great interest. The overlap of the magnetic-phase boundaries, the character of the phase transition, the nonergodicity, and the applicability of traditional molecular-field methods are but a few of the questions arising in the analysis of the magnetism of a nonmagnetic matrix containing magnetic impurities.[1–4]

Addition of manganese to copper, by virtue of the fact that manganese has two types of magnetic moment (ferro- and antiferromagnetic), and also by virtue of low Kondo temperature of the alloys ($T_k \sim 0.012$ K), leads to a broad spectrum of low-temperature magnetic states in copper-manganese alloys.[5,6]

Our task was to investigate the onset and coexistence of low-temperature magnetic states in copper alloyed with manganese at concentrations ranging from the one at which the Kondo regime is realized ($c_{\text{Mn}} \sim 0.05$ at.%) to the one corresponding to ordered antiferromagnetism ($c_{\text{Mn}} \gtrsim 80$ at.%).

EXPERIMENTAL TECHNIQUE

We investigated copper containing from 0.05 to 80 at.% manganese. Metals of purity not lower than 0.997 were melted in an induction furnace. The samples were wires 0.07–0.2 mm in diameters, foils 20–50 μm thick, and cylinders 2.5 mm in diameter and 1.5–10 mm high. The alloying and sample production technology is described in detail in Refs. 7–9. The magnetic susceptibility was determined by the compensated-transformer method in alternating fields of frequency from 10 to 1200 Hz and amplitude 0.1–5 G. The relative error in the measurement of χ did not exceed 3%. Measurements of the Hall emf and of the magnetoresistance in strong magnetic fields (see Refs. 7 and 9) up to $B = 15$ T at $T = 4.2$ K were carried out with the "Solenoid" setup of the General-Physics Institute of the USSR Academy of Sciences. The relative error in the measurement of the Hall emf and of the magnetoresistance did not exceed 3%. The electric resistivity and the thermoelectric power were investigated by the standard procedures described in Ref. 10. The temperature was determined with a Cu–CuFe thermocouple with absolute error up to 0.1°C. The ESR procedure developed by us for the investigation of spin glasses is described in Ref. 10.

RESULTS AND DISCUSSION

Figure 1 shows the temperature dependences of the magnetic susceptibility of a Cu + 8.1 at.% Mn alloy in an external magnetic field. A strong shift of T_f is evident in magnetic fields $H \sim 8 \cdot 10^4$ A/m — $\Delta T_f \approx 2.6$ K. Yet when the frequency of the alternating magnetic field used in the measurements is changed from 38 to 600 Hz the freezing-temperature shift is 0.15 K, i.e., $|\Delta T_f(\omega)| \ll |\Delta T_f(H)|$. Our results for $\Delta T_f(\omega)$ agree with the data of Ref. 11. The inset of Fig. 1 shows the suppression of the maximum of $\chi_{\text{a.c.}}$ by an external magnetic field. The deviation of $\Delta\chi/\chi_{\max} \propto H$, is proportional to H, i.e., the Edward-Anderson parameter α is equal to 1 (Ref. 12). Similar relations are observed in the alloys Cu + 1at.% Mn, Cu + 9at.% Mn, Cu + 10at.% Mn. Copper-manganese alloys with $c_{\text{Mn}} < 14$ at.% are of greatest interest for the comparison of the theory with experiment, since they are typical spin glasses with RKKY interaction.

Substantially different dependences of the freezing temperature T_f on the external magnetic field have by now been observed. The premises of the theories using a molecular field with infinite interaction radius (in particular, the theories of Edwards and Anderson (EA) and of Sherrington and Kirkpatrick for the Ising and Heisenberg models) lead to a quadratic dependence of the freezing temperature on H (Refs. 13–16):

$$\Delta T_f = T_f^{(0)} - T_f^{(H)} \propto g(S, H) \propto H^2, \tag{1}$$

where S is the impurity spin, $g(S, H)$ is the order parameter in the *EA* theory, and $T_f^{(H)}$ is the spin-glass freezing temperature in an external magnetic field. According to Néel's classical theory of superparamagnetism,[17] the superparamagnetic-cluster blocking temperature $T_B^{(H)}$ is given by

$$[T_B^{(H)}/T_B^{(0)}]^{1/2} = 1 - H/H_A, \tag{2}$$

where H_A is the magnetic anisotropy field. In the modified superparamagnetism theory $T_B^{(H)}$ is identified with T_f of spin glass.[18] In later studies, e.g., in Parisi's theory,[19] it follows from the equation for the stationary point

$$2g_S(\tau - g_S) + {}^2/_3 g_S^3 = H^2, \tag{3}$$

$$g_S = g_S(S, H), \tag{4}$$

where τ is the coefficient of $\text{Sp}Q_{\alpha\beta}$ and $Q_{\alpha\beta}$ is the order parameter, that

$$\Delta T_f \sim g_S \sim H^{2/3}. \tag{5}$$

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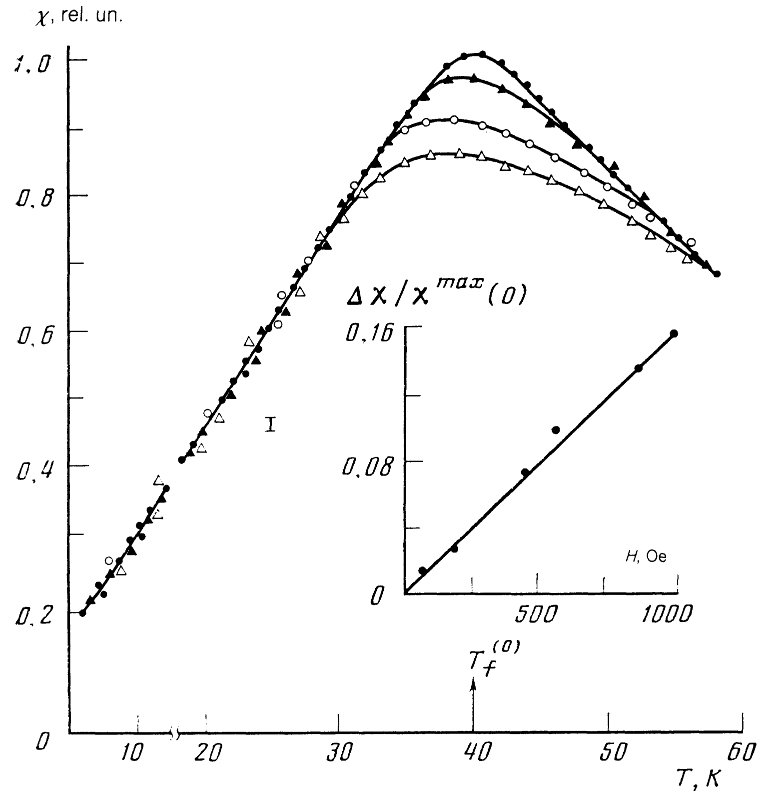

FIG. 1. Effect of external magnetic field on the magnetic suceptibility of copper-manganese alloys: ● – $H = 0$, ▲ – 400 A/m, ○ – 4800 A/m, △ – 8.10^4 A/m.

Obviously, an empirical dependence of the temperature of the maximum of the magnetic susceptibility of spin glasses on the external magnetic field could decide whether they can be adequately described by the existing theories.

Cogent conclusions can be deduced from the agreement between the Néel superparamagnetism theory and the experimental $\chi^H(T)$ observed for a large class of substances, ranging from typical copper-manganese spin glasses to percolation systems such as the alloy Cu + 51.2 at.% Ni (Ref. 20). For most spin glasses, the shift of the freezing temperature in an external magnetic field is negative. In spin glasses with mixed exchange interaction, however, for example in palladium-nickel-manganese alloys, and also in the amorphous gadolinium-aluminum magnetic, the shift of T_f as a function of H can reverse sign in a certain critical magnetic field peculiar to the particular substance.[21,22]

We call attention to the disparity between the experimental data and theoretical estimates of the suppression $\Delta\chi/\chi_{\max}$ of the magnetic susceptiblity by an external magnetic field in the Néel and EA theories.[16,20,23] A theoretical estimate of $\Delta\chi/\chi_{\max}$ for the alloy Cu + 1 at.% Mn in a magnetic field $H = 2.4 \cdot 10^5$ A/m yields 1% as against the experimental 12% (Ref. 24). For Cu + 8.1 at.% Mn in a field $H = 8 \cdot 10^4$ A/m the ratio $\Delta\chi/\chi_{\max}$ is likewise 1% and the experimental estimate ~18%. It appears that the molecular-field theories with infinite interaction radius fail to describe spin glasses because they use a nonrealistic description of the spin-glass state in real alloys.

The analysis of various types of magnetic ordering in solids, undertaken with an aim at describing the "spin-glass" state, either reduces to an approach based on the concentration of the magnetic impurity, or is based on assuming specific types of interaction.[25-28] With increasing concentration of the magnetic impurity, a transition takes place from the interaction between isolated spins (the Kondo regime) to direct exchange interaction for a disordered magnet. Neglecting the presence of an annealed Kondo effect at $T > T_k^0$, where T_k^0 is the Kondo temperature of the interacting impurities, the maximum iron concentration for the Kondo regime in gold-iron alloys is ~0.032 at.%.[29] The equivalent critical iron concentration for copper-iron alloys is 0.0034 at.%.[30,31] In copper-manganese alloys the characteristic maximum on the temperature dependence of the electric resistivity occurs at a manganese content $c_{\mathrm{Mn}} < 0.05$ at.%.[32] The maximum of the magnetic susceptibility at the spin-glass "freezing" temperature is realized starting with $c_{\mathrm{Mn}} \sim 0.2$ at.%.[33] It can thus be arbitrarily assumed that at $c_{\mathrm{Mn}} < 0.05$ at.% the Kondo regime is realized in copper-manganese alloys in the case of weak interaction of the localized magnetized moments. In the interval 0.05 at.% $\leqslant c_{\mathrm{Mn}} \leqslant 0.2$ at.%, there exists apparently a Kondo regime if the interaction between clusters is strong. When the manganese content exceeds 0.2 at.% a spin-glass phase exists in the Cu-Mn alloys. It is intermixed with a Kondo phase up to $c_{\mathrm{Mn}} \sim 8$ at.% since, for example for Cu + 8 at.% Mn, a maximum of magnetic susceptibility is observed[23,25] at $T_f \sim 40$ K on top of a gigantic thermoelectric power $\sim 10^{-8}$ V/deg. It is curious that spin-glass and annealed-Kondo-effect phases coexist in copper-manganese alloys, since $T_K^{(0)} \sim 0.012$ K.[6]

The first to consider the overlap of the Kondo and spin-glass states was Larsen in noise theory.[34] The particular phase realized depended on the ratio of the parameters $\Delta_c/T_K^{\rm eff}$ and $T_M/T_K^{\rm eff}$ (Ref. 35), where Δ_c is the energy of the interaction between the impurities, $K_K^{\rm eff}$ is the effective temperature of the Kondo alloy, and T_M is the temperature of the electric resistivity maximum. The procedure developed by Fischer to calculate $\rho\ (T)$ in the two-spin-correlator approximation for the Kondo effect permits an estimate of the temperature T_M in the approximations of single relaxation time and for a quadratic density of two relaxation modes. It is of interest that noise theory[34] is a limiting case of the Suhl-Nagaoka approximation at $\Delta_c \gg T_K^0$. According to Ref. 34, the maximum-resistivity temperature T_M is

$$\frac{T_M}{T_K^{(0)}} = \exp\left[\frac{2S^2_{\rm eff}(T)}{T/(\partial S^2_{\rm eff}/\partial T)}\right]_{T=T_M}, \tag{6}$$

where $S_{\rm eff}$ is the effective spin of the impurity. Expression (6) is valid only for $T \gg T_K^{(0)}$ or $T_j \gg T_K^{(0)}$, where T_f is the spin-glass freezing temperature. To estimate the maximum-resistivity temperatures T_M we use Fischer's results[35]:

a) in noise theory

$$T_M^{(0)}/T_f = {}^1/_2 \ln(T_M^{(0)}/T_K), \tag{7}$$

b) in the approximation of a single relaxation time (8)

$$T_M^{(1)}/T_f = {}^1/_2 \ln(T_M^{(1)}/T_K) - 1,$$

c) for two relaxation modes

$$S^2_{\rm eff} \approx \frac{T\chi_0(T)}{\alpha T_f} \ln \frac{\alpha T_f + T}{T}, \tag{9}$$

where α is a parameter that depends on the character of the magnetic impurity and χ_0 is the static magnetic susceptibility. For copper-manganese alloys we have $\alpha = 2.2$ (Ref. 35). Substituting (9) in (6) and leaving out the intermediate calculations, we obtain

$$T_M^{(2)}/T_K = \exp 2/[1 - z/(1+z)\ln(1+z) + T_M^{(2)}\chi_0'/\chi_0], \tag{10}$$
$$z = \alpha T_f/T.$$

Since $T_M\chi_0'/\chi_0 \ll 1$, we have ultimately

$$T_M^{(2)}/T_K = \exp 2/[1 - z/(1+z)\ln(1+z)]. \tag{11}$$

The experimental and calculated values of the temperatures $T_{(i)}$ of the resistivity maximum and of the temperature of the maximum of the magnetic susceptibility $\chi_{\rm a.c.}$ are listed in Table I. It follows from the results that noise theory does not describe satisfactorily the concentration dependence of the temperature of the resistivity maximum, since it overestimates the values of $T_M^{(0)}$ compared with experiment. This is

Table I. Temperatures of maximum resistivity and the magnetic suceptibility ($\chi_{\rm a.c.}$) of copper-manganese alloys[8,25,32,33,35–43]

c, at.%	$T_M^{\rm exp}$, K	$T_f^{\rm exp}$, K	$T_M^{(0)}$, K	$T_M^{(1)}$, K	$T_M^{(2)}$, K	T_0, K
0,05	8–10	–	–	–	–	–
0,10	5–6	–	–	–	–	–
0,15	2.4	–	–	–	–	–
0,20	–	3.0	11.0	7.0	8.0	–
0,24	10	2.8	10.6	6.8	7.8	18
0,48	11	4.8	17.6	12.4	9.5	20
0,50	13	5.0	18.0	13.0	10.0	–
0,51	13	5.1	18.2	13.2	10.3	22
0,60	–	6.0	–	–	–	–
0,70	17	8	28	24	21	–
1,0	27	10.0	42	30	28	–
1,15	–	11.5	44	32	29	–
1,2	–	11.0	46	36	30	–
1,4	–	13	52	40	34	27
1,5	–	12	50	38	39	–
1,6	45	13	55	46	42	–
2,0	53	15	69	49	42	–
2,4	–	19	88	64	75	–
2,7	70	18	80	68	63	–
3,0	–	19	88	64	75	–
3,2	75	21	108	73	84	–
4,5	100	27	124	106	100	–
5,0	–	28	129	112	102	–
5,4	–	27	124	106	100	40
6,3	144	33	156	136	128	–
8,0	159	39	192	149	135	–
9,7	190	44	221	188	177	–
10,0	198	50	242	196	181	–
14,0	232	60	310	235	213	–
18,0	298	75	375	280	175	–
19,6	300	–	–	–	–	–
22,0	321	92	470	355	282	–
25,0	343	108	510	384	302	–
28,4	364	126	592	444	360	–
36,0	368	–	–	–	–	–
45,0	396	184	870	650	525	–
60,0	400	164	665	586	486	–
68,0	388	133	520	390	315	–
78,2	340	82	344	258	205	–
92,0	–	18	67	49	42	–

apparently due to the different treatments of the Kondo effect in the Larsen and Fischer theories.[34,35,44]

In noise theory we have $T_M \to T_K^{(0)}$ as $c^{\mathrm{imp}} \to 0$, while in the Suhl-Nagaoka approximation[44] $T_M \to 0$ at a vanishingly small magnetic-impurity concentration.

The experimental $T_M(c_{\mathrm{Mn}})$ dependence is best described for $c_{\mathrm{Mn}} < 14$ at.%, thereby permitting a prediction of the experimental temperatures of the maximum resistivity of copper-manganese alloys. It appears that a manganese concentration $c_{\mathrm{Mn}} \sim 14$ at.% is the limit for the spin-glass state in copper-manganese alloys.

Calculation of the scattering $\hat{T}$ matrix in nonstationary perturbation theory for the *s-d* model up to terms of order JV and J^3V lead to two contributions to the thermoelectric power—a Kondo contribution and a resonant contribution due to the spin-glass state.[38] Since the contributions to $S(T)$ are of opposite sign, $S(T_0) \equiv 0$ at the point T_0. It can be seen from the table that the temperature at which the thermoelectric power of Cu–Mn alloys reverses sign depends linearly on the manganese concentration. We have previously shown[23] that approximation of the excitation spectrum by either oscillatory or relaxing modes is unsatisfactory. The relaxing-mode approximation[23] is applicable only in a very narrow manganese-concentration interval $(0.25\ \text{at.\%} \leqslant c_{\mathrm{Mn}} \leqslant 0.5\ \text{at.\%})$.

Returning to the table, note that for all manganese concentrations the experimental temperatures of the resistivity maximum exceed the temperatures of the maximum of $\chi_{a.c}$: $T_M^{\exp} > T_f$. This arrangement of the characteristic points is typical of classical spin glasses and agrees with that predicted in Fischer's theory.[45] The experimental dependences of the characteristic temperatures on the impurity concentration are described by the following expressions:

$$T_M \sim \begin{cases} c, & c_{\mathrm{Mn}} < 6\ \text{at.\%} \\ c^{\alpha}, & 6\ \text{at.\%} \leqslant c_{\mathrm{Mn}} \leqslant 28\ \text{at.\%}\ , \end{cases} \tag{12}$$

$$0.5 \leqslant \alpha \leqslant 0.7$$

$$T_i \sim \begin{cases} c, & c_{\mathrm{Mn}} < 3\ \text{at.\%} \\ c^{1/2}, & 3\ \text{at.\%} \leqslant c_{\mathrm{Mn}} \leqslant 16\ \text{at.\%}\ . \end{cases} \tag{13}$$

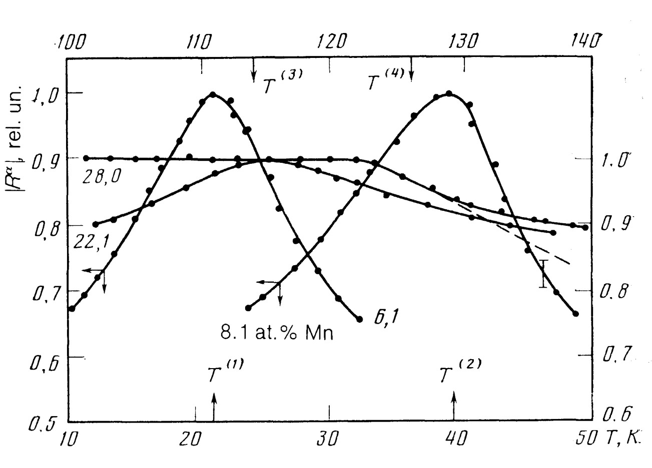

FIG. 2. Temperature dependence of the anomalous-Hall-effect "constant" of copper-manganese alloys in a field $H = 0.04$ T.

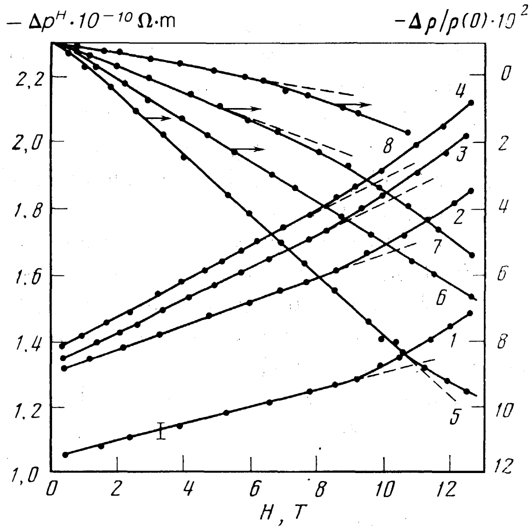

FIG. 3. Isothermal field dependences of impurity Hall resistance and of the transverse magnetoresistance of copper-manganese alloys at $T = 4.2$ K. Curves 1.5 – 6.1; 2.6 – 8.1; 3.7 – 22.1; 4.8 – 28.0 at.% Mn.

The Hall effect is known to be one of the most sensitive methods for the investigation of systems with anomalous scattering of conduction electrons. Nonetheless, the number of investigations in which so sensitive a procedure was used is small.[37,46] In the alloy Cu + 4.6 at.% Mn (Ref. 46) at $B = 0.3$ T the effect was practically unobservable, and was apparently totally suppressed by the strong magnetic field. Köster[37] observed anomalies of R^H at manganese concentrations higher than 10 at.%. The temperature dependences of the anomalous-Hall-effect constant of copper-manganese alloys are shown in Fig. 2. The anomalies of $R\alpha$ are observed at temperatures close to the spin-glass freezing temperatures: $T(i) \approx T_f$. With increasing manganese content in the copper-manganese alloys, the maxima of $R\alpha$ degenerate into inflection points, apparently as a result of the increased role of the antiferromagnetic interaction.

The Béal-Monod-Weiner spin component is revealed

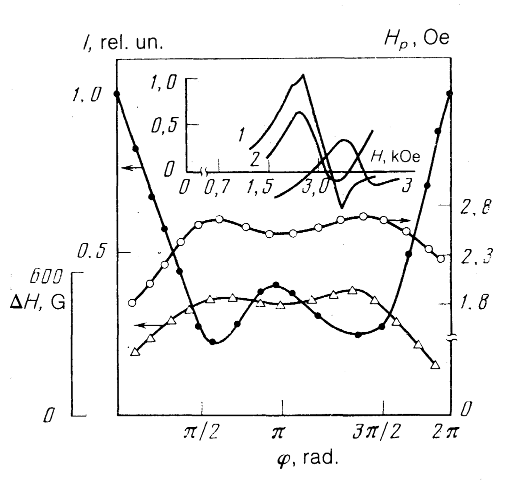

FIG. 4. Dependences of the signal intensity (●), linewidth (△), resonant field H_r (○), and ESR spectrum of Cu + 6.1 at.% Mn alloy on the rotation angle in a constant magnetic field $H = 0.7$ T. Inset: ESR spectra of Cu + 6.1 at.% Mn alloy $1 - \varphi = 0$, $2 - \varphi = \pi/2$, $3 - \varphi = \pi$.

on the temperature dependences of the impurity Hall resistance (Fig. 3) by the deviation of $\rho^H(B)$ from linearity in magnetic fields $B \approx (8\text{–}10)$ T and by the fact that the transverse magnetoresistance is negative.[7,10]

We consider now the ESR method. Typical ESR spectra and plots of the signal intensity, of the resonant field, and of the ESR linewidth for the alloy Cu + 6.1 at.% are shown in Fig. 4. The anisotropy of the ESR signals can apparently be attributed to the large value of the exchange anisotropy in copper-manganese alloys.[47] The freezing temperatures obtained from the $I(T)$ curves are close to the T_f values determined from the maxima of the magnetic susceptibility.[23]

Quenched Cu-Mn alloys containing less than 14 at.% impurity exhibit a behavior typical of spin glasses. If the freezing is in an external magnetic field, the ESR signal intensity increases with increasing H, while the amplitude of the resonant field and the ESR line width decrease simultaneously. The existence of a spin-glass state in this impurity concentration range is convincingly conformed by neutron-scattering and spin-echo experiments.[48,49] All the investigated copper-manganese alloys with 4 at.% $\leqslant c_{\rm Mn} \leqslant$ 14 at.% are characterized by a g-factor shift proportional to the impurity density $\Delta H(g) = KC$, where $K = d[\Delta H(g)]/dc = 360\ G/$at.% Mn. A similar shift was observed[50] for copper-manganese alloys containing 0.2 to 4 at.% manganese. The g-factor shift in Cu–Mn alloys has a hyperbolic temperature dependence and vanishes at a temperature $T = T_0$ that rises with increasing manganese concentration, a fact attributed to the larger role of the antiferromagnetic interaction.[50] In fact, at a manganese content $\gtrsim$ 14 at.% the ESR spectra of copper-manganese alloys reveal, beside the spin-glass phase signal, also a signal due to the "short-range-antiferromagnetism" phase, see Fig. 5. The amplitude of this signal is practically independent of the freezing magnetic field. A neutron-diffraction phase of the short-range antiferromagnetism in coppermanganese alloys is revealed by the presence, on top of the usual FCC-lattice reflection lines, of additional reflections from the (110) and (201) planes.[51]

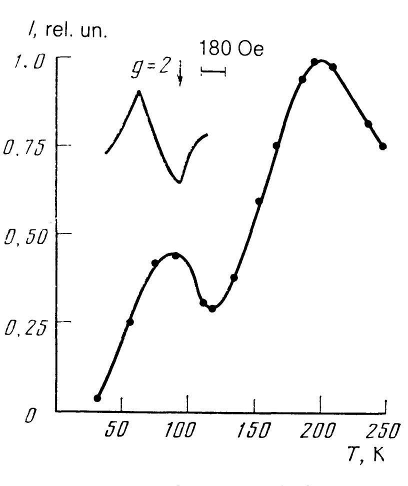

FIG. 5. Intensity of ESR signal of Cu + 22 at.% Mn alloy, annealed at 800 °C for 1 h and quenched in water, and again annealed at 450 °C for 5 h.

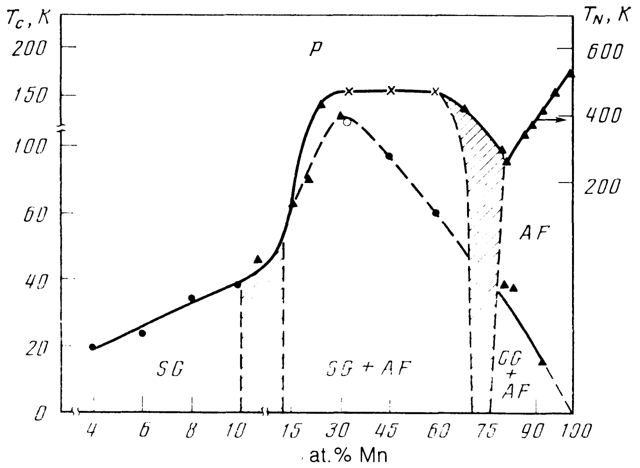

FIG. 6. Magnetic phase diagram of copper-manganese alloys. Phases: *P*—paramagnetic, *SG*—spin glass, *AF*—antiferromagnetic, *CG*—cluster glass, T_c—phase-transition point, T_N—Neel temperature, ●—ground state, annealed at 800 °C for 1 h and quenched in water, ×—antiferromagnetic-ordering state, ○—from measurements of the magnetic suceptibility, ▲—data of Refs. 52 and 53.

Antiferromagnetic-ordering annealing (5 h at 450 °C) raises the temperature of the transition into the short-range antiferromagnetism phase (see Fig. 6), suppressing partially or fully the spin-glass phase with increase of manganese concentration in copper-manganese alloys. Preliminary magnetic annealing (MA) in magnetic fields from 3 to 100 kOe suppresses the short-range antiferromagnetism phase either partially (in weak magnetic fields $B \lesssim 0.3$ T) or fully (at $B > 3$T). The spin-glass state signal is not suppressed even in magnetic fields $B \approx 10$ T (MA). From our standpoint, the maximum concentration at which short-range antiferromagnetism appears is $c_{\rm Mn}$ = 14 at.%, and not 25 at.% as proposed by Beck. The short-range antiferromagnetism realized in copper-manganese alloys is apparently a mixture of spin glass with antiferromagnetism (mictomagnetism) in the manganese concentration interval from 14 to 60 at.%.

The possibility of existence of micotomagnetism in copper-manganese alloys at intermediate impurity concentration is determined from measurements of the temperature dependences of the magnetic susceptibility, and also from results of model-based calculations and neutron-diffraction investigations.[54,55] The mictomagnetic state includes in this case a spin-glass phase formed of ferromagnetic clusters that interact with one another via the RKKY mechanism, and an incipient antiferromagnetic order due to the short-range antiferromagnetic exchange interaction.

Evidence favoring the existence of mictomagnetism is provided by the simultaneous influence of strong magnetic field up to 10 T on the anomalies of the ESR spectrum at the freezing and Neel points. The shaded region in Fig. 6 corresponds to the absence of a clearly pronounced phase transition—from 70 to 80 at.% Mn. At a manganese concentration higher than at.% a mixture of cluster-glass and antiferromagnetism phases (CG + AF) is produced in the Cu–Mn alloys. This is an inhomogeneous anti-asperomagnetic state, as attested by the characteristic shapes of the plots of the weak-field magnetization vs temperature.[52,53]

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